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ARTICLE

Micro paddy lysimeter for monitoring solute transport in paddy environment

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Abstract The micro paddy lysimeter (MPL) was developed and evaluated for its performance to simulate solute transport in paddy environment under laboratory conditions. MPLs were constructed using soil collected from Field Museum Honmachi of Tokyo University of Agriculture and Technology, Japan. For the physical characteristics of the hardpan layer, parameters such as thickness, and soil aggregate size, affecting the percolation rate were studied. For the plow layer, two types of plow soils, sieved and un-sieved soils were compared. The sieved soil plow layer was produced by mixing air-dried soils of different aggregate sizes of $D > 9.50$, $9.50 \geq D > 4.75$, $4.75 \geq D > 2.0$ mm and $D \leq 2.0$ mm at 47.1, 19.5, 20.6, and 12.8%, respectively. The un-sieved plow layer soil was directly used after collecting from the field. Inert tracer was applied to ponding water with controlled boundary conditions to evaluate the reproducibility of the soil hydraulic characteristics. HYDRUS-1D was used to evaluate the movement of bromide tracer in the MPL. The proposed conditions of the MPL were that the hardpan layer can be made from soil aggregates smaller than 0.425 mm with 2 cm thickness and that the plow layer can be prepared with sieved or un-sieved soils. With these conditions, the obtained results proved that MPLs can be a useful tool to simulate solute transport in paddy environment.

Keywords Micro paddy lysimeter · Hardpan · Plow layer · Tracer · Paddy field · Solute transport · HYDRUS-1D

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Introduction

In the world, more than 22% of the agricultural land with approximately 150 million hectares is paddy fields (Ghosh and Bhat 1998). The intensive use of agrochemicals for rice cultivation has been responsible for making paddy fields as a significant contributor of non-point source pollution. Many studies have been conducted to monitor the fate and transport of agrochemicals in paddy fields. Nevertheless, observations of the fate and transport of agrochemicals under field conditions are usually expensive, laborious and time consuming. In many studies, lysimeters have been proven to be effective tools in assessing, predicting and simulating flow and solute transport in the soil. However, most lysimeter studies have been carried out using upland soils; for example, in the laboratory, with disturbed or undisturbed soil columns (Mallawatantri et al. 1996), or in the field, with monolithic soil columns (Meissner et al. 1999; Malone et al. 2000; Owens et al. 2000). In Japan, a pioneering research using paddy lysimeters to study the runoff pattern of 21 herbicides in paddy water was reported by Maru (1990). Watanabe et al. (2008)

were also used outdoor lysimeters to develop mitigation techniques for controlling pesticide runoff from paddy fields. Micro paddy lysimeter (MPL) has been developed to simulate pesticide fate and transport in paddy environment (Nhung et al. 2009). The MPL can perform multiple experiments per year since it is independent from natural conditions such as weather and locations. In addition, the MPL preparation and experimental procedure is quick, inexpensive, and less laborious compared with that of field experiments. Although the MPL can simulate well the fate and transport of pesticide in the paddy water, pesticide leaching pattern in the MPL is still not well understood (Nhung et al. 2009). Detailed procedure for the preparation of paddy soil layers and the evaluation of solute transport phenomena for the MPL experiment ought to be documented.

Therefore, the objectives of this study were to construct a MPL having similar hydraulic properties as the actual paddy field by preparing appropriate plow and hardpan layers, and to evaluate performance of the MPL for simulating solute transport in paddy environment.

Materials and methods

Field monitoring

The field monitoring was performed in a paddy plot (10 × 30 m) at Field Museum (FM) Honmachi of Tokyo University of Agriculture and Technology (TUAT) in Tokyo, Japan, for two crops seasons from May 2005 to August 2005 and May 2006 to July 2006. The purpose of this field experiment was to investigate physical characteristics of paddy soils in the top 20-cm layer. The monitored parameters included soil bulk density, soil aggregate distribution, percolation rate, hydraulic head distribution, and hydraulic conductivity.

For analyzing soil physical properties, three soil samples of the top 15 cm paddy soil surface were taken by PVC rings with diameter of 16 cm from three different spots in the experimental paddy plot. The soil aggregate distribution was determined right after puddling by wet sieving method (West and Dumbleton 1975). Bulk density, saturate water content, hydraulic conductivity values were also determined right after puddling, 10 and 30 days after puddling.

An empty transparent plastic lysimeter (50 × 50 × 50 cm) was installed 20-cm depth from surface soil inside the paddy plot. On three sides of lysimeter, four mini piezometers were horizontally installed in the lysimeter to measure the hydraulic head distribution of surrounded plow soil in the monitored paddy plot (Fig. 1). Ten PVC rings (15-cm diameter × 20-cm high) were installed at 10

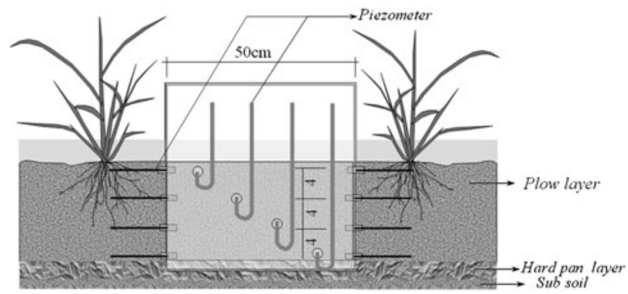

Fig. 1 Lysimeter design and installation in paddy field

different spots in the paddy plot for monitoring the percolation rate.

MPL experiments

MPL experiments were carried out to investigate the hydraulic properties and solute transport in upper paddy soil layers. The soil used in the experiment was collected from the same farm reported in the previous section.

For each experiment, three plastic MPLs of the size of 25 × 18 × 25 cm were used (Fig. 2). Each MPL consisted of four different layers. From bottom to top, they are glass bead layer of 1 cm (1 mm in diameter), hardpan layer of 1.5–3.5 cm (semi-permeable soil layer), plow layer of 14–15.5 cm (paddy field topsoil), and surface water layer of 3–4 cm, respectively. Total of five sets of experiment were conducted, namely, S1 to S5. S1 and S2 were used to examine the effect of hardpan layer on percolation rate of MPL by changing the soil aggregate size and thickness of hardpan layer. S3 was used to evaluate solute transport phenomena of the MPL condition optimized in S1 and S2. S1, S2, and S3 used sieved soil for the preparation of plow layer (Fig. 3). S4 was used to evaluate effect of the mixing time or puddling preparation on the percolation rate (Fig. 4) and S5 was used to evaluate solute transport phenomena upon optimized condition of hardpan layer and plow layer in S4. S4 and S5 used un-sieved soil for the

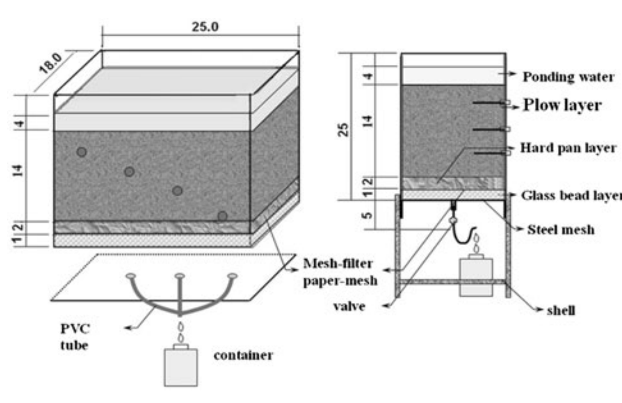

Fig. 2 MPL design and installation in laboratory (unit: cm)

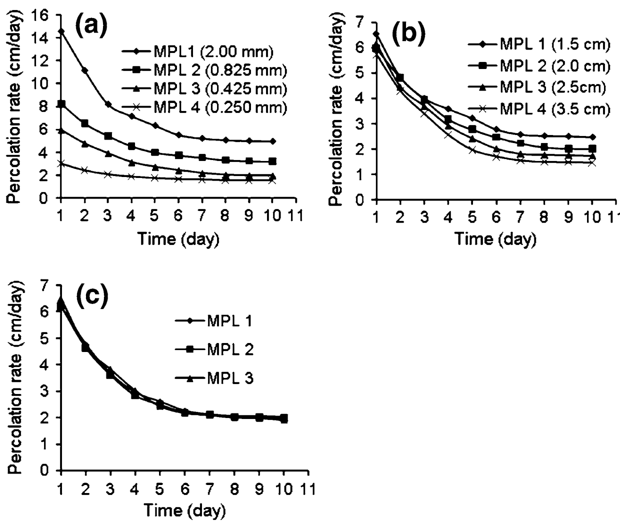

Fig. 3 Effect of soil aggregate size (**a**) (S1), thickness (**b**) (S2), of hardpan layer on percolation rate in sieved soil MPL and reproducibility of percolation rate in triplicate sieved soil MPLs (**c**) (S3)

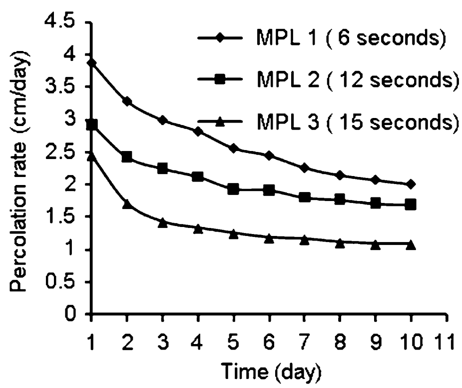

Fig. 4 Effect of mixing time of plow layer on percolation rate in unsieved MPL (S4)

plow layer. A peristaltic pump (Cole-Parmer 7553-80, BioSurplus, Inc., USA) was installed to control the percolation rate for S3 and S5. All experiments were conducted at room temperature. The soil aggregate distribution of plow layer in MPL was determined right after mixing by wet sieving method (West and Dumbleton 1975). Bromide tracer was applied in S3 and S5 by pouring 1-cm depth of bromide solution to three MPLs. After most of the 1-cm depth tracer solution infiltrated into the plow layer to produce a pulse application of the tracer, water was then added and maintained. The ponding water depth for S3 and S5 were kept constant at 3 and 4 cm hereafter, respectively.

S1 and S2 experiments

Two sets of MPL experiments were prepared to investigate the effect of hardpan layer properties. Paddy field soils were air-dried for 3 days and then were sieved into different aggregate sizes for later use. Rice straws and stones were manually removed during sieving. In S1, hardpan layers were prepared from air-dried soils passed though different sieve sizes $D \leq 2.0$, $D \leq 0.825$, $D \leq 0.425$, and $D \leq 0.25$ mm, respectively, where D (mm) is the effective diameter of the soil aggregate, in order to investigate the effect of the soil aggregate size of hardpan layer on the percolation rate in the MPL. In S2, hardpan layers were prepared with different thicknesses of 1.5, 2.0, 2.5, and 3.5 cm using the appropriate soil aggregate size (based on the results of S1) in order to evaluate the effect of hardpan thickness on the percolation rate.

As an initial condition, designed bulk density of 0.97 and corresponding porosity of 0.61 were first determined by pre-experiment. In the pre-experiment, the ratio of soil and water was optimized for the physical condition of plasticity to maintain the water saturation and workability for preparation of hardpan layer. Note that porosity and soil water content may be different depending on soil texture. Hardpan soil was saturated with appropriated amount of water and left for 24 h. The soil mixture was then mixed well right before MPL installation. Upon MPL installation, a glass bead layer (0.1-cm diameter and porosity of 0.41) was first placed on the bottom above stainless mesh (0.425 mm) then the hardpan layer soil was placed over the glass bead layer with a glass fiber filter in between to prevent the downward movement of hardpan soil. Care was taken to seal all the edges and corners to avoid preferential flow.

After creating the hardpan layer, the plow layer was installed in the MPL. The plow layer was prepared by mixing fixed proportions of different air-dried soil aggregates $D > 9.50$, $9.50 \geq D > 4.75$, $4.75 \geq D > 2.0$ and $D \leq 2.0$ mm at 47.1, 19.5, 20.6, and 12.8%, respectively, similar to the distribution of the actual field soil before puddling. The mixed soil was soaked in plenty of water for 24 h before being manually mixed and transferred to the MPL. The soil was filled up to 15.5 cm above the hardpan layer and water was added up to the prescribed level. The bulk density and the porosity of the plow soil layer were 0.92 g/cm^3 and 0.63, respectively. The MPL was left still for 24 h to achieve initial soil settlement and then the drainage valve was opened to drain with gravity about 10 days until the percolation rate was stabilized. The paddy water level was kept constant by using Marriott Tank.

S3 experiment

After selecting appropriate soil aggregate size and the thickness for setting up the hardpan layer, experiment was conducted in S3 to ensure the reproducibility of the MPLs concerning these parameters. The triplicate sieved soil

MPLs were constructed according to the procedure described in the previous section and were also left drained freely for 10 days until the percolation rate became steady at about 2 cm/day. Then, the percolation rate was regulated at 1.0 cm/day using a peristaltic pump connected to the drainage pipes at the bottom of each MPL. Mini piezometers were also installed to measure the hydraulic head profiles of the MPLs. Bromide tracer (sodium bromide) was applied to each MPL as described above for a breakthrough curve (BTC) monitoring. The applied mass of tracer was 2.925 g to achieve the initial concentration in water of 65,000 ppm. Percolated water samples were taken daily until 30 days after tracer application (DATA). The water samples were analyzed by titration method (Reber and McNabb 1937).

S4 and S5 experiments

As sieving soil is laborious and time consuming, especially for making the plow layer, it is advantageous to omit this step if the concurrent error is acceptable. Therefore, three MPLs in S4 experiment was tested with plow layer produced from un-sieved soil. The optimized hardpan layer of the sieved soil MPL was used in this experiment. Un-sieved soil was also air-dried paddy soil taken from the same farm. It was soaked with water to saturation 24 h before installation of the un-sieved soil MPL. Then, right before being transferred into the MPL, the plow soils were mixed by electric mixer (UT2203-Kaku Han, Makita Corporation, Japan) for 6, 12, 15 s, respectively, for simulating the puddling by rotary plow. In S5 experiment, three MPLs were prepared by optimized soil mixing time of 6 s to monitor the transport of bromide tracer. Mini piezometers were installed to measure the hydraulic head profiles of the un-sieved soil MPLs. A peristaltic pump was installed 10 days after soil filling to control the percolation rate at 1 cm/day. Again, sodium bromide was then applied to the surface water at the initial mass of 198 mg. Percolated water samples were collected at interval of 2 days until 30 DATA. Surface water samples were taken 0, 1, 2, 3, 5, 7, and 14 DATA. Bromide concentration in the water samples was determined by ion chromatography.

Ion chromatography

The water samples were passed through a 0.2-μm polysulfone syringe filter (Advantec, Tokyo, Japan). The filtrate was diluted with deionised water if necessary and then transferred to a 4 ml vial for analysis. The Shimadzu ion chromatography system consisted of a LC-20AD pump, an SIL-10AI autosampler, CDD-10A 3.2 μS/cmFS conductivity detector, Shim-pack IC-A3 column (4.6 mm I.D. × 150 mm L.l) with Shimpack IC-GA3 guard column. The mobile phase was a solution of 0.8 mM *p*-hydroxybenzoic acid, 3.2 mM Bis–Tris, and 50 mM Boric acid at flow rate of 1.2 ml/min in isocratic mode. Column temperature was kept at 40°C. Injection volume was 100 μl. The limit of determination was 1.0 mg/l.

HYDRUS-1D

HYDRUS-1D (Simunek et al. 2008) is a Microsoft Windows based public-domain computer program that can simulate water flow and solute transport in variably saturated porous media. The program numerically solves both the Richards equation for saturated–unsaturated water flow and the Fickian-based advection–dispersion equation for solute transport. The transport equation includes provisions for linear equilibrium adsorption and zero-order production. The governing flow and transport equations are solved numerically using the Galerkin type linear finite element scheme. The program is coupled with a non-linear least square optimization program based on Marquardt–Levenberg type algorithm (Marquardt 1963) for inverse modeling of soil hydraulic and/or solute transport parameters. A full description of the model has been given in Simunek et al. (2008). In this study, the HYDRUS-1D model was used to inversely obtain transport parameters by fitting to the BTCs of bromide concentrations in percolated water to evaluate the tracer movement in the MPL.

Darcy's law was applied at each mini piezometer installation point to calculate hydraulic conductivities K_s. As steady state water flow was established, the hydraulic head gradient was obtained from the slope of the line fitted to the observed hydraulic head values as shown in Fig. 5. Hydraulic conductivities are then obtained from Darcy's law (Hasegawa and Warkentin 2006). Since the percolation in this study is 1 cm/day, the hydraulic conductivities are equal to the slopes of the linear regression lines in Fig. 5. The upper boundary condition for water flow simulation was a constant hydraulic head that corresponds to the ponding water depth, while the lower boundary condition

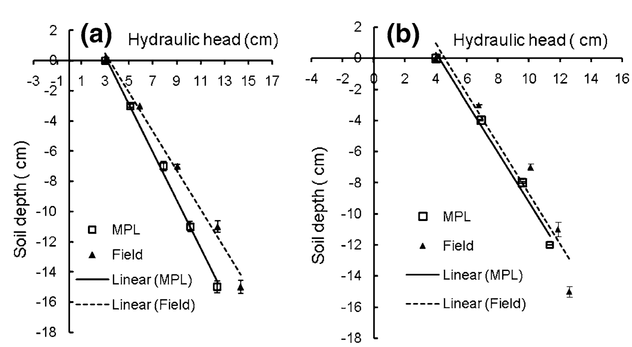

Fig. 5 Hydraulic head in sieved soil MPLs (**a**) (S3), un-sieved soil MPLs (**b**) (S5), paddy field, and their linear regression lines

for water flow simulation was a constant flux of 1 cm/day that is equal to the observed flux. The initial conditions for the hydraulic heads are based on observed data as shown in Fig. 5. The bromide concentration in surface water was used as the upper boundary condition for the solute transport, while, for the bottom boundary conditions, a second-type boundary condition where the concentration gradient at the bottom is set to zero (Simunek et al. 2008) was used. The initial bromide concentration was zero for the entire simulation domain.

MPLs and HYDRUS-1D model efficiency

In order to evaluate the goodness of fit between the measured in MPLs and simulated data using HYDRUS-1D, the statistical measures (Loague and Green 1991) were used. They are defined as follows:

Coefficient of residual mass (CRM)

$$\mathrm{CRM} = \frac{\sum_{i-1}^{n} (O_i - P_i)}{\sum_{i-1}^{n} (O_i)} \quad (1)$$

The coefficient of determination (CD)

$$\mathrm{CD} = \frac{\sum_{i=1}^{n} (O_i - O_\mathrm{m})^2}{\sum_{i=1}^{n} (P_i - O_\mathrm{m})^2} \quad (2)$$

Modeling efficiency (EF)

$$\mathrm{EF} = \frac{\sum_{i=1}^{n} (O_i - O_\mathrm{m})^2 - \sum_{i=1}^{n} (P_i - O_i)^2}{\sum_{i=1}^{n} (O_i - O_\mathrm{m})^2}, \quad (3)$$

where O_m is the mean of the measured data, O_i and P_i are the observed and predicted values in sample i. n is the number of samples.

The CRM was used to measure the tendency of the model to overestimate or underestimate the measured values. A CRM value close to zero indicates good simulation. A negative CRM indicates a tendency of the model toward overestimation. The CD is a measure of proportion of the total variance of observed data explained by the predicted data. When the value of CD reaches unity, it indicates a perfect correspondence with the measured data. The lower limit of CD can be zero. EF is a measure for assessing the accuracy of simulations. The maximum value for EF is 1, which occurs when the simulated values perfectly match the measured values. However, if EF is less than zero, the model-simulated values are worse than simply using the observed mean as the best estimate of the data points. In addition, the square of the correlation coefficient value (RSQUARE) calculated by HYDRUS-1D after each simulation was then used for the evaluation.

Results and discussion

Soil physical characteristic of paddy field and MPL

Figure 1 shows a typical soil cross section of a paddy field. In a paddy field, there is a variable ponding water depth above the top surface soil. Meanwhile, the top layer of the field soil itself comprises a low flow-resistant plow layer, followed below by a high flow-resistant hardpan layer, with a nonpuddled subsoil layer with, silty loam, or sandy loam soil type comprising the bottom (Kohno et al. 1995; Liu et al. 2001; Chen et al. 2002; Aimrun et al. 2004). Table 1 shows physical and chemical characteristics of plow layer of the paddy soil used for entire experiments (Watanabe et al. 2007).

Soil aggregate distributions of plow soils in paddy field of FM Honmachi and the sieved soil MPL are shown in Table 2. Although, the soil material was collected from the same paddy plot, physical characteristic of the soil in the paddy field and in the sieved soil MPL may have some different parameters including bulk density, soil aggregate distribution. Essentially, the distribution of soil aggregate size of the plow layer in the sieved soil MPL was almost similar to that of paddy field soil after mixing/puddling (Table 2). In contrast to the soil aggregate distribution, the bulk densities shows remarkable differences between the paddy field soil and the MPL soil as indicated in Table 3. Particularly right after puddling/mixing, the difference is obvious probably due to the soil puddling by rotary plow in the field and soil mixing by hand in the sieved soil MPL and also soil mixing by electric mixer in the un-sieved soil

Table 1 Physico-chemical properties of paddy soil (0–15 cm) in experimental plot (Watanabe et al. 2007)

Physico-chemical properties	Value
pH (H_2O)	6.5
Organic carbon content (%)	3.96
Total carbon content (%)	4.77
Total nitrogen content (%)	0.44
Cation exchange capacity ($cmol_c$/kg)	22.5
Particle density (g/cm)	2.50
Sand (%)	37.6
Silt (%)	31.8
Clay (%)	30.6
Soil texture (ISSS)	Light clay (LiC)

ISSS International Society of Soil Science

Table 2 Soil aggregate distribution of plow soil in paddy field and in sieved soil MPL after puddling/mixing

Soil aggregate size (mm)	Paddy field soil (%)	Sieved soil MPL soil (%)
$0.95 < D$	20.4	21.7
$0.475 < D \leq 0.95$	16.8	18.2
$0.20 < D \leq 0.475$	18.3	17.5
$0.10 < D < 0.20$	11.2	10.9
$0.05 < D \leq 0.10$	9.2	7.7
$0.025 < D \leq 0.05$	8.8	7.9
$0.01 < D \leq 0.025$	9.3	7.5
$D \leq 0.01$	6.1	8.5

MPL. In the paddy field, the bulk density of plow soil rapidly increased from the time of right after soil puddling to 10 days after soil puddling then it slowly increased at the time between 10 and 30 days after soil puddling. As indicated by the standard deviation the plow soil in the MPL seems to be more homogeneous than the field plow soil. In addition, these bulk density values agreed with reported bulk density of paddy soil in Japan (Kohno et al. 1995; Aimrun et al. 2004). After 30 days, the bulk densities of the MPLs and the paddy field had little difference.

The observed percolation rates at 10 different spots in the paddy plot 10 days after puddling varied greatly ranging from 0.4 to 1.3 cm/day. It is noted that during the process of puddling, soil colloid particles or small soil particles may settle into the open cracks of the hardpan layer and thus impeding the percolation capability of the hardpan layer. Meanwhile, the average value of percolation rate was 0.9 cm/day which agreed with the data from a previous study carried out in the same farm (Watanabe et al. 2007). In order to simulate the field water flow, the percolation rate in MPLs was adjusted to 1 cm/day.

Sieved soil MPLs

Hardpan layer plays important role in controlling percolation of paddy water, and significantly influences the water recharge process (Koga 1991; Wopereis et al. 1992, 1994; Tournebize et al. 2006; Aimrun and Amin 2009). In addition, plowing and compacting are two major factors that influence the formation of the hardpan layer and markedly reduce the percolation rate (Liu et al. 2005).Therefore, constructing an appropriated hardpan layer of MPL was very important in order to simulate desired percolation rates. Several studies reported that percolation rate in paddy field ranges from 0 to 3 cm/day (Chen et al. 2002; Kukal and Aggarwal 2002; Maie et al. 2004; McDonald et al. 2006). Ministry of Agriculture Fishery and Forestry (MAFF) reported that the average percolation rate in Japan's paddy field was 1–1.8 cm/day (MAFF 1996). Therefore, in this study, the hardpan layer of MPL was constructed to have adjustable percolation rate within 0–2 cm/day. In addition, because the surface soil was always in flooded condition before drainage, the plow soil and the hardpan soil were assumed to be saturated (Chen et al. 2002).

Effect of soil aggregate size and thickness of hardpan layer on percolation rate

Figure 3a illustrates the influence of the soil aggregate size of the hardpan layer on the percolation rate in S1 experiment. Both permeable capacities and changes in percolation rate with time were greatly affected by the soil aggregate size of the hardpan layer. The percolation rate, immediately after installation, was approximately five times greater for the hardpan layer constructed with the aggregates smaller than 2 mm (14.58 cm/day) as compared to that constructed with the aggregates smaller than 0.250 mm (3.04 cm/day). After 10 days, with soil aggregate smaller than 2 mm, the percolation rate decreased to 4.96 cm/day that is nearly one-third of the initial percolation rate of 14.58 cm/day. Meanwhile, when aggregates smaller than 0.250 mm were used, the percolation rate became about the half of the initial value after 10 days. In general, the decline of the percolation rate was very fast in the first 3 days, while it became moderate in the next 3 days before reaching a steady state after 8 days. The rates of decline were more profound for the hardpan layer made with larger aggregates. The soil aggregate size of 0.425 mm was chosen for the next experiments since it can generate percolation rate of about 2 cm/day after 10 days of the MPL installation (Fig. 3a). Regarding the potential to control percolation rate, the soil aggregate size of 0.250 mm was also good for the MPL. However, a lot of

Table 3 Bulk densities of plow soil in Honmachi field and MPL (g/cm^3)

	Right after mixing/puddling	After mixing/puddling 10 days	After mixing/puddling 30 days
Field	0.791 ± 0.019	0.886 ± 0.022	0.901 ± 0.002
Sieved soil MPL (S3)	0.839 ± 0.005	0.903 ± 0.006	0.915 ± 0.004
Un-sieved soil MPL (S5)	0.812 ± 0.003	0.912 ± 0.007	0.918 ± 0.003

Note. Data are presented by average value ± standard deviation value labor and time was required for sieving due to its small aggregate size. Therefore, using soil with the aggregate size of 0.425 mm was more convenient. The percolation rate can be adjustable to simulate desirable rates below 2 cm/day using flow regulator or valves.

Figure 3b shows percolation rates in four MPLs with 1.5-, 2.0-, 2.5-, and 3.5-cm thick hardpan layer, respectively, in S2 experiment in which the soil aggregates smaller than 0.425 mm were used. After 10 days, the percolation rates in three MPLs were stabilized. The percolation rate of MPLs with 1.5-, 2.0-, 2.5-, and 3.5-cm thick hardpan layer were 2.53, 2.07, 1.78, and 1.49 cm/day, respectively (Fig. 3b). The thicker the hardpan layer was, the smaller the percolation rate was, due to the increased flow-resistance in the hardpan layer. Two cm thick hardpan layer is chosen for later use since it provided percolation rate of about 2 cm/day and it required moderate time and labor for its preparation (Fig. 3b).

Evaluation of the reproducibility of soil profile among MPLs

In S3 experiment, the percolation rates of three replicated MPLs during 10 initial days were identical (Fig. 3c). The hydraulic head distribution in the plow soil of sieved soil MPL 30 days after installation is shown in Fig. 5a. At the depth of 3 cm, the hydraulic head ranges from 5.0 to 5.2 cm, while it increases to a range of 12.0–12.8 cm at the depth of 15 cm. Table 4 shows the calculated hydraulic conductivities for the plow layers of each soil profiles in MPLs and in the paddy field. The hydraulic conductivity (K_s) for each plow layer ranged from 1.289 to 1.607 cm/day. In comparison with the un-sieved soil MPL and the paddy field (with 4 cm ponding water) in Fig. 5b, their hydraulic conductivities are similar (Table 4). However, there is a difference between the sieved soil MPL and paddy field (with 3 cm ponding water) in Fig. 5a. Their hydraulic conductivities are 1.593 and 1.289 cm/day for the sieved soil MPL and paddy field, respectively (Table 4). In addition, Fig. 5 and Table 4 show that the field measurement seems to have larger variation and the r^2 values of MPL are greater than those of the paddy field.

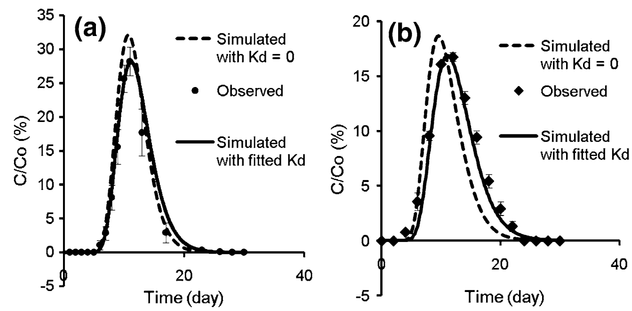

Fig. 6 Breakthrough curves of bromide tracer in sieved soil MPLs (**a**) (S3), un-sieved soil MPLs (**b**) (S5), and HYDRUS-1D simulation. *Co* is the initial concentration of bromide tracer in each set of experiment

Consequently, MPL can produce similar hydraulic conductivities as the paddy field. The hydraulic conductivity of the hardpan layer that used for this MPL experiment was 0.120 ± 0.01 cm/day in the all experiments. The hydraulic conductivity of the hardpan layer is about one-tenth of that of the plow layer because the soil aggregate sizes of the hardpan layer were smaller than those of the plow layer.

Figure 6a shows the averaged observed BTC and simulated one for bromide tracer in sieved soil MPL. Solute BTC is plotted for the relative concentration measured in the leachate versus time after tracer application. The tracer was detected about 6 DATA, while the outflow concentration reached the maximum value at 11 DATA. Considering the pore volume, the concentration peaks appeared at 1.02 pore volume. The almost symmetrical shape of the BTC also indicates that any tailing resulted from non-equilibrium flow (Simunek and Van Genuchten 2008) was not remarkable as shown in Fig. 6a. The bromide tracer mass was recovered at 90% in this experiment.

Combination of the hydraulic head distribution in Fig. 5a and the BTC of tracer in Fig. 6a indicate that flow characteristics of soil profile in three sieved soil MPLs prepared by proposed materials and methods are reproducible. In other words, the proposed approach for preparing the paddy soil profile for the MPL from dried sieved soil with prescribed conditions such as soil aggregate distribution, bulk density, and hydraulic head, can generate

Table 4 Hydraulic conductivities (K_s) in plow layers of sieved soil MPL, un-sieved soil MPL, and paddy field (cm/day)

	Ponding water depth (cm)	Linear regression equation	r^2	Hydraulic conductivity (K_s)
Field	3	$y = -1.289x + 4.342$	0.989	1.289
Sieved soil MPL (S3)	3	$y = -1.592x + 5.075$	0.997	1.593
Field	4	$y = -1.607x + 7.370$	0.934	1.607
Un-sieved soil MPL (S5)	4	$y = -1.598x + 6.720$	0.990	1.598

Percolation rate is 1 cm/day

Note: y is soil depth (cm) and x is hydraulic head (cm), r^2 is the square of correlation coefficient soil hydraulic response similar to those in the actual paddy field.

Un-sieved soil MPLs

Effect of soil mixing time on percolation rate in MPLs

In addition to the hardpan layer, characteristics of the plow layer also affect the percolation rate. In general, in cultivation, before the crop, farmers puddle their paddy fields in order to soften the soil of the plow layer, level the soil surface, and reduce percolation losses through soil surface. In paddy field, the reduction of percolation rate through puddling has been studied intensively (Singh et al. 2001; Kukal and Aggarwal 2003; Arora et al. 2006, Mousavi et al. 2009). Experimental results with well-controlled microplots (Wopereis et al. 1992) and in laboratories (Sanchez 1973; Osari 1988; Adachi 1992) show that puddling could reduce infiltration rate of 500–1,000 times, while most field studies reported a reduction of less than five times (Humphreys et al. 1992). Liu et al. (2005) reported that plowing and compaction reduced percolation rate 14 times in a simulated paddy field. In this study, in order to simplify setting up process of plow layer of MPL, un-sieved soil collected from field, was intently used. In order to simulate puddling procedure in the actual paddy field soil, the plow layer soil was mechanically mixed, just before setting up the un-sieved soil MPLs. An appropriate mixing time to achieve the designed percolation rate was investigated in this experiment. Figure 4 shows that the percolation rate decreases when the mixing time increase. As the mixing time increased from 6 to 15 s, the initial percolation rate greatly reduced from 3.8 to 2.5 cm/day and the stable percolation rate after 10 days also decreased from 2 to 1 cm/day. Similar to the puddling process in the paddy field, the mixing propeller destroys soil aggregate and soil structure, eliminates large soil pores, provokes compaction, reduces macropore, increases micropore volume and lessens the hydraulic conductivity of the plow soil. Moreover, Liu et al. (2003) reported that the settling of fine suspended clay-type can deposit on the pore via surface filtration and significantly reduce the percolation rate. In Fig. 4, increasing the mixing time increases the amount of suspended clay particles; therefore, percolation rate was reduced. Consequently, the mixing time of 6 s per total 6 kg of dried soil by prescribed device gave an appropriate percolation rate at 2 cm/day after 10 days of installation of the un-sieved soil MPLs. However, note that the percolation rate will be influenced by the specification of devices such as power and shape of mixing propellers.

In S5 experiment, hydraulic conductivities in the plow soil layer of the un-sieved soil MPL was calculated in the same way as in the sieved soil MPL above. It shows that hydraulic conductivities in MPL and the paddy field were the same as shown in Table 4. The concentrations of bromide tracer in the surface water in three un-sieved soil MPLs were almost identical during the treatment time of tracer. The data were used as the boundary condition in HYDRUS-1D. BTCs of the conservative bromide tracer are shown in Fig. 6b. The BTCs appeared to be symmetrical, with less tailing. Bromide tracer was detected in the leachate after four DATA and the relative concentration peak was measured between 10 and 12 DATA. The relative concentration peak appeared at 1.1 pore volume. The slightly delayed arrival of the moderate peak concentration instead of a quick, sharp peak in this study suggests that the preferential flow is negligible in MPL. In addition, small tailing of BTC was the indication of the slight effect of physical non-equilibrium flow on inputs of tracer. However, in general, the flow in the MPLs seems to be homogeneous. As the plow soil was well saturated and mixed before installation it supports that soil layers in MPLs are prepared homogenously without major macropore or edge flow.

Evaluation of soil hydraulic response by HYDRUS-1D simulation

In order to examine soil hydraulic condition and percolation phenomena of MPL, HYDRUS-1D model was applied to MPLs data in S3 and S5 experiments. For each MPL type, two simulations were conducted. In the first simulation, the sorption of bromide tracer on soil was not considered ($K_d = 0$). The dispersivities of both the hardpan layer and the plow layer were fitted parameters. On the contrary, in the second simulation, we considered the sorption of bromide tracer. K_d and dispersivities of both the hardpan layer and plow layer were the fitted parameters. The results showed that in the first simulation only sieved soil MPL had good fitted BTC (Fig. 6a) with EF = 0.909 (Table 5). However, un-sieved soil MPL had less well fitted BTC (Fig. 6b) with EF = 0.769 (Table 5). The fitted dispersivities in the hardpan layers of both sieved soil MPL and un-sieved soil MPL are 12.584 and 35.485, respectively (Table 6) although these hardpan layers have

Table 5 MPLs and HYDRUS-1D model efficiency

Simulation	CD	EF	CRM	RSQUARE
Sieved soil MPL (S3)				
$K_d = 0$	0.989	0.909	0.259	0.979
Fitted K_d	0.990	0.970	0.106	0.986
Un-sieved soil MPL (S5)				
$K_d = 0$	1.001	0.769	0.187	0.801
Fitted K_d	1.009	0.970	0.129	0.983

Table 6 The fitted parameters by HYDRUS-1D simulation

	Simulated with $K_d = 0$	Simulated with fitted K_d
Sieved soil MPL (S3)		
Plow layer		
DISP	0.365	0.626
K_d	0.000	0.072
R	0.000	1.105
Hardpan layer		
DISP	12.584	0.880
K_d	0.000	0.077
R	0.000	1.122
Un-sieved soil MPL (S5)		
Plow layer		
DISP	0.444	0.595
K_d	0.000	0.127
R	0.000	1.183
Hardpan layer		
DISP	35.485	1.082
K_d	0.000	0.112
R	0.000	1.178

DISP is dispersivity (cm), *R* is retardation factor, K_d is partition coefficient (l/kg)

the same preparation procedure. Meanwhile, in the second simulation, the observed data in un-sieved soil MPL and sieved soil MPL agreed well with simulated values as summarized in Table 5. CD, EF, and RSQUARE values are all close to unity and CRM values are close to zero, which indicates a good agreement between MPLs data and HYDRUS-1D simulation (Table 5). Moreover, the fitted parameters in Table 6 show that the dispersivities are small, which indicates the homogeneous state in soil profile of MPLs. The small values of K_d ranging from 0.072 to 0.127 and retardation factor ranging from 1.105 to 1.183 indicates slight interaction between bromide tracer and soil (Table 6). Bromide is commonly used as tracer. In several studies, bromide was considered as slightly reactive tracer such as in undisturbed soil in column study (Clay et al. 2004). The anion loss and retardation of bromide tracer because the soil contained the ferrihydrite ($Fe_5O_7(OH)\cdot 4H_2O$) was reported (Brooks et al. 1998). In addition, bromide anion exclusion and retardation have already been observed in different soils and minerals (Wierenga and Vangenuchten 1989; Ishiguro et al. 1992; Melamed et al. 1994; Seaman et al. 1995; Brooks et al. 1998). Bromide tracer does not always behave as an ideal tracer in Spodosols (Begin et al. 2003). In this study, the soil collected from FM Honmachi—TUAT contained ferrous about 25–48 g/kg (Tanaka et al. 2008). Slight interaction of the tracer with soil may be responsible for the slightly retardation of bromide tracer BTC appeared in un-sieved soil MPL in the first simulation and the incomplete mass balance of the tracer. The average bromide recoveries were 90 ± 2% for the un-sieved soil MPL in S5 and 90 ± 3% for the sieved soil MPL in S3. These values are similar to an average bromide recovery of 87% for undisturbed loamy sand soil (Lennartz and Kamra 1998) and 90.8 ± 0.6% for the undisturbed column of silty clay loam soil (Dousset et al. 2007). The bromide loss (10%) in MPL experiments may be resulted from the slight adsorption of bromide to the soil due to the high content of ferrous (Tanaka et al. 2008). It indicates that bromide tracer might not be the best tracer of water flow in certain soil profiles. On the other hand, bromide has become widely regarded as virtually nonreactive tracer. Several investigators, working in limestone, dolomite, allophonic Andisol, have found bromide to be nonreactive tracer (Schmotzer 1973; Jester and Uhler 1974; Tennyson and Seltergren 1980; Payne 1988; Ishiguro et al. 1992; Leap 1992). In that case, the non-equilibrium flow phenomena discussed in the recent research (Simunek and Van Genuchten 2008) could be an alternate to explain our phenomena. However, there was no available data in this study for the comparison.

Comparing sieved soil MPL and un-sieved soil MPL, the observed BTC of bromide tracer in un-sieved soil MPL is remarkably different from one in sieved soil MPL in term of BTC's shape and BTC's peak concentration (Fig. 6). However, both MPL types can generate the good fitted theoretical BTCs of bromide tracer which can be expressed by HYDRUS-1D model. The combination of MPL experiments and HYDRUS-1D simulation can be also applied for the analysis of other pollutants such as pesticides and nutrients. It is also noted that when the simulation conducted with more reactive solute such as nitrogen or pesticide, the transformation and degradation of the solute is also an important parameter affecting the movement of the solute in hardpan layer and plow layer.

Consequently, regarding the potential of simulating the soil hydraulic characteristics and the solute transport phenomena in the paddy field, both MPL types show appreciable capabilities. It is advantageous that the preparation of un-sieved soil MPL can be remarkably simplified. Therefore, it is more practical and less time consuming than sieved soil MPL. In other word, use of the un-sieved soil MPL to simulate the fate and transport of solute such as pesticide in paddy environment can be the practical option.

Conclusions

A method to construct the MPL to simulate the fate and transport of solute in paddy environment was developed. The MPL was proven to be an effective and convenient tool in assessing, predicting and simulating the fate and transport of solutes in paddy field. The MPL has similar hydraulic properties of soil profile with the paddy field in term of soil aggregate size distribution, soil bulk densities, percolation rate, hydraulic properties, and hydraulic head. The MPL used under controlled boundary conditions allows accurate quantification of transport processes for tracer through BTC analysis. There are good agreement between BTCs of bromide tracer in the sieved and un-sieved soil MPLs and BTCs fitted by HYDRUS-1D simulation model. BTCs analysis shows that the transport of tracer in MPLs demonstrated more physical equilibrium phenomena without major macropore and preferential flow effects. The MPLs exhibited small fraction of sorption, which resulted in slower transport or slight retardation of bromide tracer through the soil profile.

For using the MPL to investigate the solute transport in paddy environment, it is suggested that soil materials for constructing the plow layer and the hardpan layer should be taken from where the soil profile is to be simulated. The appropriate thickness of the hardpan layer is 2 cm, and the soil aggregate size is smaller than 0.425 mm. For the plow layer, in case the sieved soil is used, the soil aggregate distribution should be investigated in advance. When the un-sieved soil is used, the soil is recommended to be mixed at appropriate power such as 6 s/6 kg in case of this study.

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