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Review

Progress in Multi-Soil-Layering Systems for Wastewater Treatment

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Abstract: The use of decentralized wastewater treatment technologies is a reasonable solution for rural areas. As a decentralized treatment technology, the multi-soil-layering (MSL) system has recently drawn an increasing amount of attention owing to its merits, such as a high hydraulic load rate, small land area occupation, low probability of clogging, low investment, and low operation cost. This review summarizes the progress in MSL systems in the past decade, focusing on the directions of efforts for system optimization, the latest applications of MSL systems to various wastewater treatments, and the integration of MSL with other technologies. The great application potential of MSL systems is illustrated, and future research directions regarding better application of MSL systems are provided.

Keywords: multi-soil-layering systems; decentralized treatment technology; removal performance and mechanism; material composition; integrated system; wastewater treatment

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Citation: Ma, T.-F.; Wu, J.; Feng, L.; Chen, X.-P.; He, J. Progress in Multi-Soil-Layering Systems for Wastewater Treatment. *Sustainability* **2024**, *16*, 3330. https://doi.org/10.3390/su16083330

Academic Editor: Alberto Ferraro

Received: 29 February 2024
Revised: 7 April 2024
Accepted: 15 April 2024
Published: 16 April 2024

1. Introduction

It is reported that about half of the world's population is living in rural areas, with most of them still facing the problems of unsound or inefficient sanitation systems [1]. For most low-income countries, the use of centralized treatment systems in rural areas or peri-urban cities causes a debt burden to local residents or governments [2]. Therefore, the use of decentralized treatment technologies has become a reasonable solution.

Septic tanks are a common traditional decentralized treatment technology. Although they are simple and safe, the concentrations of suspended solids (SS), total nitrogen (TN), total phosphate (TP), and biological oxygen demand (BOD) in septic tank effluent cannot meet local emission standards [3]. Ecological technologies that rely on the filtration, adsorption, and degradation of soil, plants, and microorganisms to purify wastewater are suitable for remote rural areas due to their very low energy requirements and specialized operations [4,5]. Common decentralized treatment technologies primarily include constructed wetlands (CWs), soil filtration systems, and oxidation ponds. These technologies also have their own limitations. For example, slow soil filtration systems and oxidation ponds require large land areas [4,6], while CWs require large investments, as well as having high operation and maintenance costs [7].

The multi-soil-layering (MSL) system has gained increasing interest in recent years as a promising alternative to common centralized and decentralized technologies. The MSL system was first proposed by Japanese researchers in the 1990s. MSL systems are an improvement based on soil treatment systems. An MSL system primarily consists of soil mixture blocks (SMBs) and permeable layers (PLs) that are constructed in the form of stacked bricks to form a modularized soil treatment system [8]. MSL systems avoid the disadvantages of traditional soil filtration processes, such as occupying large amounts of land, clogging problems, poor nitrogen removal performance, and a low treatment load. They have the advantages of requiring a low investment and low operation costs [4]. Many studies on MSL systems have been conducted since the first MSL system was proposed. It was initially proposed for treating domestic wastewater, but the applications of MSL systems have been extended to treating landfill leachate, polluted river water, aquaculture wastewater, industrial wastewater, antibiotics wastewater, and other aquatic areas [9–13]. Moreover, to further enhance MSL system performance, many studies have examined optimizations of MSL systems and pollutant removal mechanisms [14–18].

In this review, the progress in MSL systems during the past decade is summarized with a focus on the direction of system optimization efforts, the latest applications of MSL systems to different wastewater treatments, and the integration of MSL systems with other technologies. This review provides a deeper understanding of MSL systems and facilitates their further application.

2. Primary Mechanisms

Different from traditional soil treatment systems, an MSL system is a modularized soil ecosystem that contains aerobic areas and anaerobic areas, where SMBs constitute anaerobic areas and PLs form aerobic areas (Figure 1) [19]. SMBs are generally mixtures of soil, sawdust, charcoal, rice straw, and iron particles and are placed in the system in the form of bricks. PLs fill the gaps around SMBs, and the materials used as PLs are usually gravel, pumice, or zeolite. Although it is optional, an aeration system is utilized in most cases; the reason for this will be discussed later.

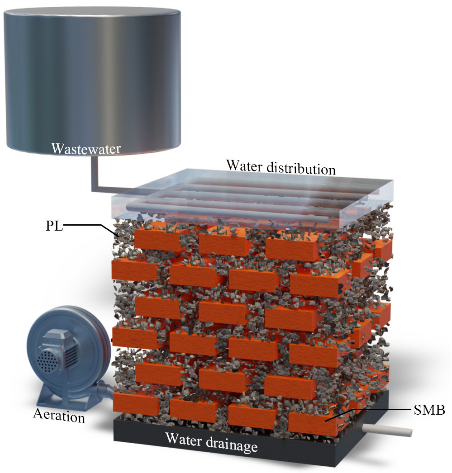

Figure 1. Schematic diagram of an MSL system. The figure is redrawn according to An et al. [4]. MSL, multi-soil-layering; SMB, soil mixture block; PL, permeable layer.

Detailed pollutant removal mechanisms can be found in previous literatures [4,19,20]. The pollutant removal mechanisms of an MSL system are shown in Figure 2. Briefly, the soil in the SMBs acts as a filter and provides places for microbes to attach and reproduce. Organic matter (OM) in SMBs, such as sawdust and rice straw, provides carbon sources for the denitrification process in SMBs. Charcoal can adsorb a variety of wastewater pollutants. Moreover, iron particles can release iron ions and promote the removal of phosphorus in water through adsorption or co-precipitation after the formation of hydrated iron oxide in SMBs or PLs [21]. Gravel, pumice, and zeolite are commonly used as PL materials. PLs fill the gaps around SMBs and, owing to their large particle size and high porosity, PLs can promote the diffusion of water flow and lower the possibility of clogging. In addition, the porous materials in PLs can also adsorb pollutants and facilitate the growth of microorganisms.

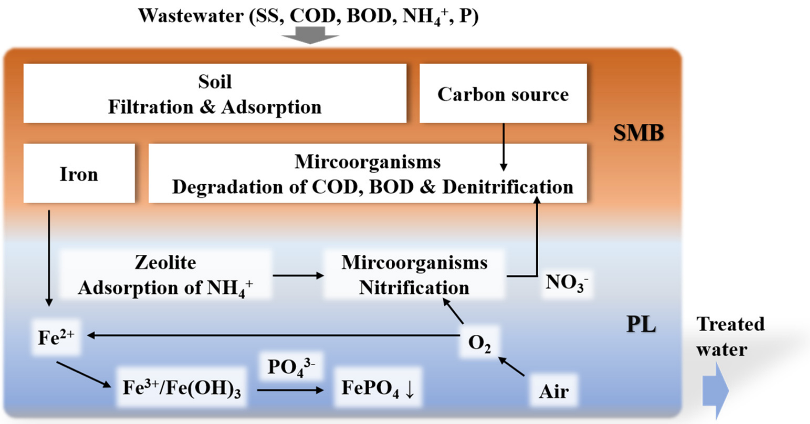

Figure 2. Primary removal mechanisms of an MSL system. The figure is redrawn according to An et al., 2016, and Guan et al., 2015 [4,19]. SS, suspended solids; COD, chemical oxygen demand; BOD, biological oxygen demand; P, phosphate; SMB, soil mixture block; PL, permeable layer.

3. Comparative Assessment of MSL and Alternative Techniques

This section provides a comparative evaluation of the MSL system and other alternative treatment techniques based on a comprehensive literature analysis. According to the reviewed literature, the MSL system demonstrates good performance in the removal of traditional pollutants from various types of wastewaters, such as SS, BOD, COD (chemical oxygen demand), NH_4^+, TN, and TP. As shown in Table 1, compared to common alternative techniques such as CWs, stabilization ponds, and sand filtration, the MSL system has advantages, such as a small land occupation, low maintenance frequency, and no odor or insect production. Indeed, these advantages are the reasons why the MSL system is considered to have great potential for application in rural wastewater treatment. However, the MSL system also has certain shortcomings, such as the risk of clogging under high hydraulic loading rates (HLRs) and the need for improved effluent sanitation efficiency. Substantial research is needed in the future to overcome the deficiencies of the MSL system and to further enhance its performance.

Table 1. Comparative assessment of MSL and alternative techniques.

Technique	Principle	Removal Performances	Advantages	Disadvantages	References
CWs	Combining the adsorption and filtration effects of soil and artificial fillers, as well as the adsorption and degradation effects of plants and microorganisms.	COD $\leq$ 75.7% BOD$\leq$ 91% SS $\leq$ 91% NH_4^+ $\leq$ 72.1% TN $\leq$ 63.4% TP $\leq$ 71.8%	Low cost Low energy Simple operation	Large land occupation Treatment efficiency fluctuates with seasonal variations High maintenance frequency Odor and insects	[20,22,23]
Stabilization ponds	Combination of the adsorption and degradation effects of microorganisms, algae, and aquatic plants.	BOD $\leq$ 91% COD $\leq$ 76% SS $\leq$ 91% NH_4^+ $\leq$ 56% TN $\leq$ 30% TP $\leq$ 21%	Low cost Low energy Simple operation	Large land occupation Treatment efficiency fluctuates with sunlight and climate variations Long hydraulic retention time High water loss by evaporation Odor, insects, and rodents	[24–26]
Sand filtration	Pollutants are intercepted by the sand and then decomposed by microorganisms.	COD $\leq$ 78% SS $\leq$ 95% NH_4^+ $\leq$ 88% TN $\leq$ 85% TP $\leq$ 50%	Low cost Small land occupation Simple operation and maintenance	Risk of clogging Odor and insects	[27–29]
MSL	Soil and filter media perform filtration and adsorption, while microorganisms carry out biodegradation.	$\geq$90% for SS, BOD, COD, NH_4^+, TN, and TP, in most cases	Small land occupation Low cost Low energy No odors, no insects Simple operation and maintenance	Risk of clogging Moderate sanitary efficiency	[20,30,31]

4. Applications to Different Wastewater Types

4.1. Domestic Wastewater

Owing to the advantages of only requiring a small land area, low construction cost, and simple operation and maintenance, MSL systems have been widely used for small-scale domestic wastewater treatment, especially for rural decentralized domestic wastewater.

In the beginning, MSL systems were originally developed to treat domestic wastewater, and showed good removal performance for SS, BOD, COD, TN, and TP in early studies [32–34]. In recent years, domestic wastewater treatment has remained one of the most important research and application directions of MSL systems. With the continuous optimization of MSL system design and operating parameters, satisfactory removal efficiencies of C, N, and P in domestic wastewater by MSL systems were obtained. Normally, the removal efficiencies of MSL systems for COD, ammonia nitrogen, TN, and TP can reach greater than 80%, 85%, 60%, and 80%, respectively [14,35,36]. For instance, Latrach et al. [37] utilized an MSL system with shape-modified SMBs to treat secondary effluent from a domestic wastewater treatment plant and found that the MSL system accomplished good removal efficiencies for COD, ammonia nitrogen, TN, and TP at 81%, 89%, 92%, and 98%, respectively. To treat rural domestic wastewater, Wang et al. [17] added sludge-based biochar materials into SMBs, which strengthened the MSL system performance, and the removal efficiencies of COD, ammonia nitrogen, TN, and TP were 80%, 90%, 65%, and 92%, respectively. Zidan et al. [38] employed a hybrid MSL system treating domestic wastewater. The hybrid MSL system is composed of a vertical flow MSL unit in series with a subsurface horizontal flow MSL unit. The hybrid MSL system showed good removal performance for the septic tank output with a concentration of total SS (TSS), COD, TP, and TN ranging from 6.70–118.70 mg/L, 40.83–210.09 mg/L, 0.64–4.77 mg/L, 16.67–41.69 mg/L, respectively, and the removal efficiencies of TSS, COD, TP, and TN were 97%, 79%, 76%, and 27%, respectively.

Hence, MSL systems are generally efficacious in removing conventional nutrients (C, N, and P) in domestic wastewater. With the growing global concern with regard to water resources, water ecological health, and drinking water safety, the standards for the effluent water quality of wastewater treatment systems have been elevated. Therefore, apart from the removal of conventional nutrients, the removal of pathogens and emerging pollutants in wastewater, such as microplastics, antibiotics, endocrine disruptors, and persistent organic pollutants, has become a new research focus in the wastewater treatment field. Nevertheless, there is currently a paucity of research on the removal performance of pathogens and emerging pollutants in domestic wastewater by MSL systems. Therefore, it is imperative to conduct research in these areas in the future.

Additionally, it is important to note that many regions around the world face water scarcity issues, and reusing treated domestic wastewater can help to alleviate this problem. The MSL system for treating domestic wastewater has been reported as being used for irrigation and can be a valuable source of plant nutrients and soil fertilizers [39,40]. Therefore, from the perspective of the fertility of irrigation water, future studies may not have to pursue a high removal efficiency of nutrients but should pay more attention to the biological safety of the effluent.

4.2. High-Strength Wastewater

4.2.1. Livestock Wastewater

In addition to domestic wastewater, MSL systems have also been used for livestock wastewater (LW) treatment in recent years. Unlike domestic wastewater, LW typically contains high concentrations of OM, nitrogen, phosphorous, and pathogenic bacteria [41]. Moreover, antibiotics, parasiticide, heavy metals, and steroid hormones are also presences in LW [42]. Therefore, if LW is discharged without proper treatment, surface water and groundwater will be contaminated. Although some conventional biological technologies have been utilized for the decentralized LW treatment, such as anaerobic sludge beds [43], and CWs [44], and have shown positive results, the problems such as the large area occupied, high operation and maintenance costs, and relatively poor performance still exist. Therefore, researchers have tried to utilize MSL systems for LW treatment. The performances of MSL systems in removing conventional contaminants in LW have been relatively satisfactory. For example, Liu et al. [18] reported that when treating LW with a microcurrent-assisted MSL system, when the concentrations of COD, TP, and TN are 1200, 10, and 120 mg/L, respectively, the removal efficiencies of COD and TP reached 95.45% and 92.0%, respectively, and the removal efficiency of TN was at 60–75%. When treating anaerobically digested swine wastewater with initial concentrations of ammonia nitrogen, TN, and TP, these appeared as 682.6, 761.8, and 22.8 mg/L, respectively, and the highest removal efficiencies of ammonia nitrogen, TN, and TP were 94.2%, 94.4%, and 92.5%, respectively [45]. Guo et al. [46] found that when dosing 0.1 critical micelle concentration of biosurfactant into the MSL system for treating anaerobically digested swine wastewater, under the condition of hydraulic loading rate (HLR) being 120 $L/m^2/d$ and the inflow ammonia nitrogen being 1000 mg/L, the ammonia removal performance of the MSL system was enhanced, and a maximum ammonia removal efficiency of 93% was reached. These reports confirm that MSL systems have good potential to be used for removal of conventional nutrients in LW treatment. However, there are few reports on the removal efficiencies of MSL systems for antibiotics, pathogens, steroids, and heavy metals in LW. Future research should focus on these aspects to ensure the health and safety of MSL effluent for water and soil environments.

4.2.2. Food Industry Wastewater

In addition to LW, many food industry wastewaters also have the characteristics of high nitrogen, phosphorus, OM, and pathogen contents, such as dairy industry wastewater [47] and rice noodle industry wastewater [48]. Regarding the wastewater produced in extensive centralized food industrial parks, it is common to have centralized sewage treatment facilities for processing. However, when it comes to the wastewater generated by small-scale handcrafted food production in remote and underdeveloped villages [49], as well as dairy wastewater on islands [47], centralized sewage treatment systems and bioremediation technologies based on soil and plants are not feasible due to their high operating and maintenance costs and large land area requirements. In such situations, the MSL system has demonstrated encouraging potential for application. When treating dairy industry wastewater (TN: 5.39–44.6 mg/L, TP: 17.76–21.39 mg/L) with an MSL system, it was found that the MSL system, using Leilehua soil as an aerobic layer, removed the inorganic nitrogen and phosphate by 22–93% and 64–99%, respectively, and by utilizing a constant aeration rate and sucrose addition, the nitrogen and phosphorus removal performances can be further improved [47]. In the scenario of utilizing an MSL system to treat rice noodle wastewater in a handicraft village, the COD, ammonia nitrogen, phosphorus (PO_4-P), and TSS removal efficiencies were 67.42%, 53.1%, 44.73%, and 80%, respectively, with the initial influent concentrations of these pollutants appearing as 197.50–766.25, 24.55–135.35, 8.56–24.20, and 37.60–132.00 mg/L, respectively [49]. Compared to the vertical flow constructed wetland (VFCW) used for rice noodle wastewater treatment, the MSL system showed similar removal efficiencies for COD, ammonia nitrogen, and TSS, and a higher phosphorus removal efficiency [49]. Generally, the MSL system can meet the local effluent requirements for OM and suspended matter, but nutrient removals require further strengthening. Therefore, it is promising that, after optimization according to food industry wastewater characteristics, MSL systems can be employed as an alternative technology for food industry wastewater treatment.

4.2.3. Landfill Leachate

Landfill leachate is also a typical type of high-strength wastewater, which has low biodegradability and a high content of COD, nitrogen, and toxic substances [50,51]. Although biological processes are commonly employed for landfill leachate treatment, the refractory organic matters and high ammonia contents are still challenging [51]. The integration of advanced oxidation technology and other technologies as post-treatment methods has good application possibilities in the field of leachate treatment [52–54], but the treatment cost is typically high [55]. Guan et al. [9] utilized an MSL system to treat rural unsanitary landfill leachate (ammonia nitrogen: 59.9 ± 22.0 mg/L, COD: 218.4 ± 133.8 mg/L, TP: 1.3 ± 1.1 mg/L) and studied the influence of different HLRs and intermittent aeration on the MSL system performance. For 184 days running, results indicated that under an HLR of 200 L/m^2/d and no aeration, the effluent ammonia nitrogen, COD, and TP met the local emission standards. Moreover, a higher HLR and intermittent aeration can not only solve the clogging problem of MSL systems, but also strengthen the nitrification process of the systems. These results suggested that the MSL system is suitable for nutrient removal in landfill leachate. However, to the best of our knowledge, there is currently no research on the performance of MSL systems in the removal of toxic substances in landfill leachate, such as xenobiotics and heavy metals. In order to practically apply MSL systems to the treatment of landfill leachate, a significant amount of research in this direction needs to be conducted in the future.

4.3. Water/Wastewater Containing Special Pollutants

In addition to the aforementioned wastewater, researchers are expanding the application of MSL systems to the treatment of water/wastewaters containing special pollutants.

4.3.1. Microcystins

Water bodies contaminated with microcystins (MCs) are potential risks to the environment and human health [56]. Among the MCs, the most toxic is MC-LR [57]. Although slow sand filtration [58] and CW technologies [59] have shown good performance for MC-LR treatment, more studies are required for large-scale applications of slow sand filtration. In addition, CWs require large land areas. In order to address the limitations of the aforementioned technologies, Aba et al. [60] endeavored to employ an MSL system for the treatment of simulated surface water contaminated by MC-LR. The MSL system utilized pozzolan as the PL material, and local sand, iron chips, charcoal, and sawdust as the SMB materials. Following a five-week operation, the MSL system successfully achieved a removal rate of over 95% for the MC-LR. This study serves as a testament to the potential of MSL systems for the treatment of water bodies contaminated by MCs. It should be noted that, in this study, distilled water containing MCs was used, whereas actual water bodies may have more complex compositions, such as the presence of algae and various organics. The existence of these substances may affect the performance of the MSL system in removing MCs. To date, this is the only study that has been conducted on the application of the MSL system to water bodies contaminated by MC. Further research on MSL systems' removal performance for MC-LR and other MCs in real-world water bodies is needed.

4.3.2. Trace OMs

Trace OMs, such as drug-active compounds and endocrine disruptors, will produce disinfection by-products during drinking water treatment, thus posing a threat to public health [61]. However, the removal of these trace OMs has not been given full attention in wastewater treatment plants (WWTPs) [62]. To efficiently remove trace OMs from wastewater, Maeng et al. [63] employed an MSL system as a tertiary wastewater treatment technology. The results showed that the MSL system can remove greater than 80% of pentoxifylline, caffeine, 17-acetinyl estradiol, estradiol, and 17-estradiol in wastewater. These results demonstrated that the MSL system is promising as a trace OM removal technology in tertiary wastewater treatment. It should be noted that the trace OMs were spiked into the influent of the MSL system at 2 μg/L, which is relatively higher than the concentration of those found in the aquatic environment [63]. Future studies based on the real concentrations of trace OMs or real water/wastewater should be carried out. Meanwhile, more research on the removal efficiencies and mechanisms of MSL systems for other types of trace organic contaminants are still needed.

4.3.3. Residual Antibiotics

Residual antibiotics in the environment pose a threat to water safety and environmental microbial diversity [64,65]. The existing soil-based treatment technology has good performance for antibiotic removal [66], but there are also problems such as instability [67], large land area occupation, a low load rate, and clogging. To solve the above problems, Song et al. [10] utilized an MSL system to treat poultry wastewater containing sulfamethoxazole (SMX). It was found that the SMX removal efficiencies were stabilized above 91% after 40 days of operation with a low influent SMX concentration (1 mg/L) and pH (pH = 3), as well as medical stone used as a PL material. The findings of this study suggest that the MSL system holds promise for the treatment of wastewater that contains antibiotics. However, there is currently a lack of research on the removal effects and mechanisms of other types of antibiotics by the MSL system. Moreover, in the real world, there may be kinds of residual antibiotics existing in water/wastewater. Therefore, the removal performance of the MSL system for multiple residual antibiotics, and the adaptation mechanisms of microbes in the MSL system to multiple residual antibiotics are still unclear. Therefore, a significant amount of research is still needed in these areas in the future.

5. Strategies for System Optimization

5.1. SMBs

5.1.1. Size and Shape

A good reactor structure design is critical to improve reactor performance and reduce construction costs. For MSL systems, the design of SMBs has an important impact on MSL system performance. In most lab-scale studies, the size of the SMB is closer to a square brick [9,15,16,60,68], but it should be noted that narrower and thinner SMBs benefit MSL systems more.

Narrower SMBs increase the number of PLs between SMBs, which is more conducive to good water flow distribution, thus improving system performance and lowering the risk of clogging [69,70]. Additionally, narrower SMBs increase the overall side surface area of SMBs in MSL systems, which is conducive to contact between wastewater and the SMBs, thus improving the removal performance of MSL systems [71,72]. Thinner SMBs are conducive to allowing the full play of the decontamination function of SMBs because the upper portion of thicker SMBs is more prone to clogging [71]. Moreover, thinner SMBs can increase the SMB layers in the MSL system, thereby increasing the total surface area of SMBs, which is conducive to improving the removal performances of COD, BOD_5, SS, and TP [72]. Therefore, the sizes of SMBs with dimensions within the range of (4.5–9) cm × (4.5–9) cm × (2–4) cm (length × width × height) may potentially enhance system performance.

Additionally, MSL system performance can be further optimized by improving the SMB shape. Although there is only one report on the optimization of SMB shape so far, it is quite encouraging and inspiring. The U-shape SMBs (Figure 3a) that Latrach et al. [37] designed create a meandering pattern of water flow through the system, allowing water to flow more evenly throughout the system and reducing the dead zone area, compared with a standard MSL system (Figure 3b). In addition, this design also increases the hydraulic retention time and the effective volume of the system, which further enhances the removal effects of the MSL system for coliforms and nutrients. When treating secondary effluent from a domestic wastewater treatment plant, the removal efficiencies of the MSL system with U-shaped SMBs for ammonia, TN, and TP all increased by more than 10% compared with the MSL system with common SMBs. Therefore, the design of the SMB shape also plays a critical role in improving MSL system performance, and in future studies more attention should be paid to this direction.

Based on the aforementioned studies on the optimization of SMBs, future works on optimizing both the shape and size of SMBs simultaneously might achieve unexpectedly good results.

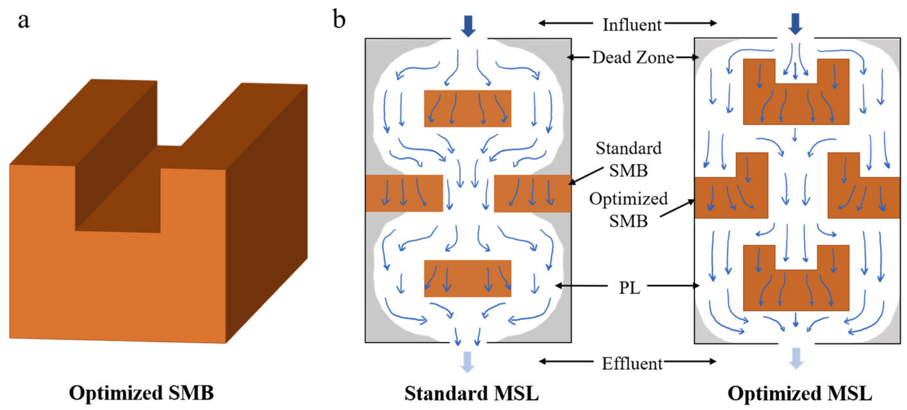

Figure 3. Optimized SMB (**a**), and an illustration of the dead zone in a standard MSL and an optimized MSL (**b**). The figure is redrawn according to Latrach et al., 2018b [37]. SMB, soil mixture block; PL, permeable layer.

5.1.2. Material Components

In earlier studies [9,35,36], SMBs primarily consisted of sawdust, rice straw, iron powder/iron slag, charcoal, and soil. These components play different roles in contamination removal, and there is room for improvement of SMB components to optimize MSL system performance.

- Optimization of carbon sources

The traditional natural carbon sources, such as sawdust and rice straw, although cheap and accessible, often also have high nitrogen content and risk of clogging [73]. Therefore, optimization of carbon sources in SMBs is necessary. In recent years, solid OM substrates, acting as carbon sources and biofilm carriers, have shown good application prospects in low carbon-to-nitrogen ratio wastewater treatment due to a good carbon release effect and significantly improved denitrification efficiency [74–76]. This provides a new perspective for optimizing the material composition of SMBs. It has been proven that addition of a new carbon source in SMBs benefits denitrification processes in MSL systems. For example, Zhou et al. [77] developed a new blended carbon source, polyhydroxybutyrate (PHBV) sawdust, and mixed it with melon stone, blast furnace washing slag, and humus soil in SMBs. The PHBV sawdust contributed to the increase in denitrification gene abundance, the enhancement of energy metabolic processes, and the stimulation of enzymatic activity of histidine kinase, glycogen phosphorylase, and ATP enzymes. As a result, the denitrification performance of the MSL system was strengthened. Song et al. [10] added poly-(bubulosuccinate) (PBS) into SMBs, and it was mixed with surface soil, sawdust, and iron powder at a ratio of 1:7:1:1, thus achieving improvement of the denitrification efficiency. However, Hong et al. [73] found that when mixing PBS, soil, slag, and iron powder into SMBs, the removal of ammonia nitrogen and TN was negatively influenced. This was possibly due to the carbon released by the PBS in SMBs in the upper layers, which facilitated the growth and activity of heterotrophic bacteria, and this inhibited the activity of nitrification bacteria. This resulted in a reduction in the ammonia nitrogen removal efficiencies, further weakening the denitrification process and eventually leading to a decline in the TN removal efficiency.

It is important to note that the above studies did not explore the cost issue of the new carbon sources. Since these new carbon sources are all prepared through artificial synthesis, it is likely that their cost is higher than that of traditional carbon sources such as sawdust and straw. Considering that relatively low construction and operation costs are also one of the advantages of the system, future studies should give full consideration to the cost issue of new carbon sources while taking into account the improvement of system performance brought about by carbon sources.

In addition, the current research has not addressed the issue of the lifespan of the carbon source used within SMBs. When the carbon source is depleted over the MSL system's service life, it is likely to have an adverse effect on the MSL system's denitrification performance. How should we efficiently and cost-effectively replenish it? There are no answers yet.

- Change of iron source

As previously mentioned, iron particles can release iron ions and promote the removal of phosphorus in water, and thus play an important role in the phosphorus removal performance of MSL systems. Using cost-effective and accessible iron sources, such as local natural materials or solid waste containing iron, to replace iron particles is promising. Chen et al. [78] attempted to utilize local common red clay containing iron oxide instead of iron particles to add into SMBs. Although the TP removal efficiencies of the MSL system that utilized local red clay were lower than that of the MSL system using iron particles, using local red clay showed a higher removal efficiency of P per gram Fe, and effectively reduced the overall cost of the MSL system. Waste steelmaking slags have also been employed to replace iron particles in SMBs, and a more than 85% phosphorus removal efficiency has been reported [79]. However, the aforementioned studies focused on the influence of iron source on the removal of phosphorus; whether the change in iron source affected the community structure in the SMBs and thus impacted the removal performance of other pollutants, such as COD, BOD, and TN, was not mentioned. Since iron affects bacterial activity and microbial community structures [80–82], which are related to the system's performance in removing various pollutants, future research should fully consider the impact of iron sources on the overall performance of MSL systems, rather than focusing solely on the removal of phosphorus.

- Addition of functional microorganisms

The adsorption and degradation functions of microorganisms play critical roles in the pollutant removal performance of MSL systems. Optimizing MSL system performance through the addition of functional microorganisms is worthwhile. Currently, no studies that have examined the direct addition of functional microbial agents have been reported. This may be due to the high cost of microbial agents, which mean that the use of them is not an economic option. Activated sludge from WWTP contains a large number of functional microorganisms and is cheap and easy to obtain, and can be an alternative to functional microbial agents. Song et al. [11] added activated sludge into SMBs with dry weight ratios of 0%, 5%, 10%, and 20% in an MSL system for turtle aquaculture wastewater treatment. It was found that a 20% addition of activated sludge into the SMBs significantly increased the ammonia nitrogen removal efficiencies of the MSL system. A denaturing gradient gel electrophoresis analysis implied that by adding activated sludge into the SMBs, the nitrification bacteria in the MSL system were enriched, and the stability of the nitrification process was also improved. This study demonstrated the good prospect of the addition of exogenous functional microorganisms to improve the MSL system's performance. However, the functional bacteria in the activated sludge adapted to the MSL system. In another case, Hong et al. [73] reported that an activated sludge addition adversely influenced the COD removal efficiencies owing to the death of some bacteria that could not survive the anaerobic environment in SMBs. In addition, the indigenous microorganisms in the soil and influent played a key role in the removal of nitrogen and phosphate, while the exogenous microorganisms in the SMBs did not play a primary role. This study implied that whether the introduced exogenous microorganisms could adapt to the environment of an MSL system and maintain a certain advantage in competition with the indigenous microorganisms in an MSL system was the key to determining whether the exogenous microorganism addition could improve MSL system performance. Therefore, when adding exogenous microorganisms, the operating conditions of MSL systems and the characteristics of exogenous microorganisms should be fully considered to maximize the effect of the addition of exogenous microorganisms.

In addition, it is worth noting that with the rapid development of materials science, a variety of new environmental materials for pollution control are continuously being researched and developed. For example, various metal-based and carbon-based nanomaterials [83–85], as well as other composite catalysts [86–89], have been widely used in wastewater treatment. These materials have shown good performance in pollutant removal. Therefore, new environmental functional materials may also be added into SMBs to improve MSL system performance. However, there have been no reports yet on the use of the aforementioned materials in SMB. Relevant research should be carried out in the future. Nevertheless, it cannot be ignored that these materials may have adverse effects on microorganisms [90,91] and plants [92] in soil and water bodies, and sufficient attention to this should also be given in future research.

5.2. PLs

Compared to the large amounts of studies on SMB optimization, the numbers of studies on PL optimization have been fewer. Zeolite has been used as the PL layer packing in most MSL systems because its high cation exchange capacity benefits the adsorption of ammonia nitrogen. Studies have been conducted to discover whether there are better packing materials for PLs instead of zeolite. Ho et al. [93] tried to replace zeolite with different materials for wastewater treatment. The results indicated that under a lower hydraulic load (below 0.5 $m^3/m^2/d$), expanded clay grain material, oyster shells, and used granular activated carbon could be utilized to replace the zeolite to achieve effective wastewater treatment and reduce the construction cost. Song et al. [10] first used anthracite and medical stone as PL fillers when treating poultry wastewater containing antibiotics with an MSL system, and it was found that a medical stone performed better for SMX removal because of its large surface area. However, because there are no control MSL systems with commonly used materials as PLs, nor similar studies that have been performed, it remains unclear whether using medical stone for the PLs can optimize the MSL performance for SMX removal. In the future, screening for high-efficiency and low-cost materials remains the direction for PL optimization.

5.3. HLR

The HLR has an important influence on wastewater treatment system performance. Generally, under a too-low HLR, poor pollutant removal performance may be witnessed, as the death of some microorganisms in the MSL system will occur due to insufficient nutrition, which consequently leads to a fluctuation in the effluent quality; in most cases (as shown in Table 2), low or moderate HLRs are chosen for better pollutant removal performances [9,17,45,94]. If the HLR goes too high, the pollutant removal efficiencies will decline [95].

Moreover, it should be noted that in filters and biofilters, a higher HLR may cause clogging. This is because a higher HLR commonly results in a massive accumulation of SS and an increase in biofilms, which lower the porosity and thus lead to clogging [96,97]. Although MSL systems are characterized by high water permeability due to the brick-layer-like pattern of PLs and SMBs, clogging was reported when the HLRs became too high. Guan et al. [9] witnessed clogging of the two MSL systems running under HLRs of 800 and 1600 $L/m^2/d$ when treating leachate from rural unsanitary landfills with MSL systems, while the other two MSL systems operated stably under the HLRs of 200 and 400 $L/m^2/d$. Masunaga et al. also reported that the MSL system was clogged when treating domestic wastewater containing high contaminations with HLRs of 1250, 1500, and 2000 $L/m^2/d$ [98]. Although the removal of SS in wastewater by pretreatments is recommended and generally accepted to reduce the clogging risk of CW [99], it seems appropriate aeration and proper operation cycle are enough to solve the clogging problem of MSL systems [9,98], and this will be discussed in later sections.

Table 2. Summary of studies on the influences of HLR on MSL systems.

Type of Wastewater	HLRs Applied ($L/m^2/d$)	Optimal HLR ($L/m^2/d$)	Removal Efficiencies under Optimal HLR (%)	Other Primary Parameters of MSL Systems	Reference
Domestic wastewater	500, 1000, 1250, 1500, and 2000	500	For low concentration wastewater: COD: 89; ammonia nitrogen: 98; TN: 44; TP: 73 For high concentration wastewater: COD: 94; ammonia nitrogen: 98; TN: 45; TP: 89	MSL size(cm): 50 × 10 × 139 (L × W × H) SMB size(cm): 20 × 10 × 10 (L × W × H) SMB composition: andisols, sawdust, and granular iron metal in volume ratio of 6:2:2 PL: zeolite (0.1–0.3 cm in diameter) Aeration: not used	[98]
Rural domestic wastewater	400, 800, 1200, 1500, and 2000	800~1200	MSL system with SMBs containing sludge-based biochar: COD: 90; ammonia nitrogen: 90.01; TN: 67.75; TP: 90.98 MSL system with SMBs containing wood chips: COD: 87; ammonia nitrogen: 84.01; TN: 57.74; TP: 87.45 MSL system with SMBs containing charcoal: COD: 82; ammonia nitrogen: 79.81; TN: 50.98; TP: 83.14	Effective volume of MSL system: 28 L (130 cm in height) SMB composition: local soil, iron chips, and sludge-based biochar/wood chips/charcoal in dry mass ratio of 7:1:2 PL: zeolite Aeration: not used	[17]
Domestic wastewater	200, 300, and 500	200	COD: 92.46; ammonia nitrogen: 98.53; TP: 97.84; TN: 22.19	MSL size (L × W × H, cm): 50 × 50 × 70 SMB size (L × W × H, cm): 12.5 × 50 × 5, and 15 × 50 × 5 SMB composition: local soil, cinder, and bio-ceramic in weight ratio of 6:3:1 PLs: zeolite (0.1–0.3 cm in diameter) Aeration: not used	[94]
Synthetic domestic wastewater	300, 400, and 500	400	COD: 93.4; ammonia nitrogen: 94.9; TN: 80.4; TP: 94.7	MSL size (cm): 45 × 25 × 70 (L × W × H) SMB size (cm): 22 × 11 × 8 (L × W × H) SMB composition: clay soil and sawdust in dry weight ratio of 85:15 PLs: zeolite (0.3–0.5 cm in diameter) Aeration: not used	[95]
Anaerobically digested swine wastewater	80, 120, 160, and 200	160	ammonia nitrogen: 94.2; TN: 92.5; TP: 94.4	MSL size (cm): 45 × 25 × 70 (L × W × H) SMB size (cm): 22 × 11 × 8 (L × W × H) SMB composition: clay soil and sawdust in dry weight ratio of 85:15 PLs: zeolite Aeration: not used	[45]
Leachate from rural unsanitary landfills	200, 400, 800, and 1600	200	COD: 72.0; ammonia nitrogen: 97.4; TN: 66.5; TP: 96.2	MSL size (cm): 50 × 10 × 75 (L × W × H) SMB size (cm): 10 × 9 × 3.8 (L × W × H) SMB composition: soil, sawdust, iron, and charcoal in dry weight ratio of 5:3:1:1 PLs: zeolite (0.3–0.5 cm in diameter) Aeration: only the MSL systems with high HLR (800 and 1600 $L/m^2/d$) were executed when necessary	[9]

Note: HLR, hydraulic loading rate; MSL, multi-soil-layering; SS, suspended solids; TN, total nitrogen; TP, total phosphate; COD, chemical oxygen demand; SMB, soil mixture block; PL, permeable layer.

Since a higher HLR could reduce the required construction materials and land occupation of MSL systems, and consequently lower their cost, MSL systems with higher HLRs will always be welcomed on the premise of guaranteeing pollutant removal performance. To find the optimum HLR of an MSL system, it is necessary to fully conduct a lab-scale test and a pilot test so as to provide references for scale-up research in practical applications. In addition, the use of computer simulation analysis to predict the optimal HLR is a direction worthy of further study. Some studies have used machine learning and neural networks to predict the pollutant removal performance of MSL systems [100–102]. In the future, perhaps the optimal HLR can be predicted using a simulation according to the primary parameters of an MSL system.

5.4. Aeration

Operation with proper aeration also helps to improve the performance of an MSL system.

Studies in earlier years have shown that whether there is aeration or not in the MSL system has a large impact on the pollution removal performance [33,34,103,104]. In studies of domestic wastewater treatment, it was found that the removal efficiencies of BOD_5, COD, TN, and TP were notably increased after aeration was introduced into an MSL system that had not previously been aerated [104]. A longer aeration period was found to improve the BOD_5 and SS removal performances in an MSL system using intermittent aeration [34]. When an MSL system that had been operating under continuous aeration stopped being aerated, the TN removal efficiencies in the MSL system displayed a trend of increasing for a month, and then a trend of decreasing after that [33]. The increasing trend of TN removal was likely caused by the enhanced denitrification process favored by the anaerobic environment, and the decreasing one was likely due to the inhibition of the nitrification process. Recent studies have also confirmed the positive effects of aeration for the removal of OM, TN, ammonia nitrogen [105,106], and phosphate [47].

In MSL systems with continuous aeration, aeration intensity has been found to contribute greatly to improvements in the MSL system performance. After the aeration intensity was doubled (from 1000 L/min to 2000 L/min) in a study treating LW with an MSL system, the removal efficiencies of the colored substances and COD increased by 9.0–14.6% and 13.9–23.7%, respectively [28]. Similarly, in a study treating polluted river water with an MSL system, ammonia nitrogen was totally removed when the aeration intensity was increased to 16,000 L/m^2/h [13].

Proper intermittent aeration also creates improvements in MSL system performance. Luanmanee et al. [107] reported that under a local tropical climate with high temperatures all year round, the MSL system performed best in the alternating operation mode of aeration for three days (20,000 L/m^3/d) and then stopping aeration for two months. When Guan et al. [9] used MSL systems to treat leachate from rural landfills, it was found that three periods of intermittent aeration during the middle stage of the experiment not only helped to solve the clogging caused by a high HLR (800 and 1600 L/m^2/d), but also enhanced the nitrification process and COD removal.

5.5. Operation Cycles

Due to the accumulation and clogging of pollutants during the operation of an MSL system, a reasonable operation cycle is conducive to MSL system performance. Masunaga et al. [98] found that under an HLR of 2000 L/m^2/d, the highest net removal rates (g/m^2/d) were obtained by running for 4 months and then resting 2 months for high-concentration domestic wastewater treatment, and running for 7 months and then resting 2 months for low-concentration domestic wastewater treatment. In addition, clogging was alleviated. However, there are no more studies on the operation cycles of MSL systems. More studies in this direction should be conducted to discover the optimal operation cycles that can both better the MSL system performance and lower the operation and maintenance costs.

5.6. Proper Pretreatment

To ensure the stable performance of the MSL system and its application to a wider range of wastewater scenarios, it is essential to carry out appropriate pretreatment based on the characteristics of the wastewater, especially when the wastewater contains potential limiting factors such as heavy metals and pharmaceutical components. Researchers have confirmed that heavy metals and pharmaceutical components can adversely affect the performance of biological treatment processes in wastewater treatment systems [108–111]. Pretreatment through physicochemical methods, such as adsorption, precipitation, oxidation, and dilution of pollutant concentrations, can help to mitigate the impact of limiting factors on the biological processes within the system [112,113]. Currently, there are no related studies on MSL systems; it is necessary to conduct research on pretreatment targeting limiting factors in wastewater on the basis of controlling overall costs of MSL systems in the future.

6. Integration with Other Technologies

Apart from optimizations of MSL system parameters, integration of an MSL system with other treatment technologies can also improve wastewater treatment performance because the integration can enhance the advantages of various technologies while making up for the shortcomings of the technology itself.

6.1. Integration with CWs

In current studies, it is most common to integrate MSL systems with CWs (Figure 4). For example, Koottatep et al. [114] hybridized an MSL system with a VFCW to construct an MSL-VFCW system (Figure 4a) to process septic tank effluent. The MSL-VFCW system was planted with Chrysopogon zizanioides, the SMBs consisted of laterite soil, sawdust, and powdered charcoal, and the PL layer was composed of zeolite. The MSL-VFCW system showed a good pollutant removal performance during a three-month operation. Under the same conditions, the average ammonia nitrogen removal efficiency of the integrated system was 95.86%, while that of the CW system was only 61.76%. Moreover, under optimized conditions, greater than 84% of the total COD and BOD_5 were removed, and a remarkable ammonia nitrogen removal efficiency of 96.77% was achieved.

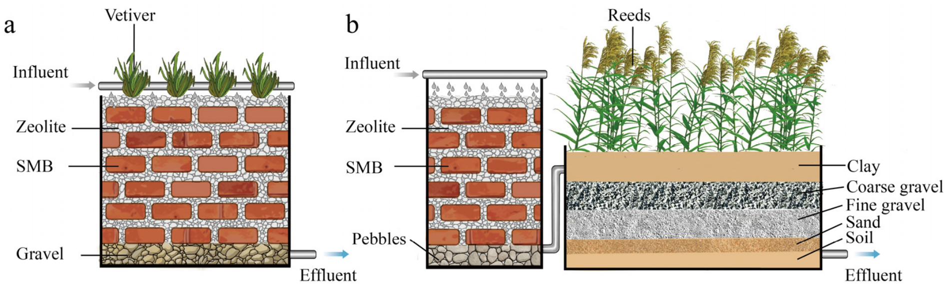

Figure 4. The integration of MSL systems with CWs. (**a**) The hybridization of an MSL system with a VFCW (the figure is redrawn according to Koottatep et al., 2018 [114]). (**b**) The combination of an MSL system with a subsurface flow CW (the figure is redrawn according to Song et al., 2019 [106]). MSL, multi-soil-layering; SMB, soil mixture block; CW, constructed wetland; VFCW, vertical flow constructed wetland.

Similarly, a pilot-scale MSL-CW system planted with Canna was utilized to treat septic tank wastewater [115]. During its four years of operation, the average removal efficiencies of the total COD, soluble COD, and total BOD were approximately 71%, 65%, and 80%, respectively, and the average removal efficiencies of ammonia nitrogen and TP were both greater than 75%. In addition, the total coliform and E. coli counts in the effluent were all less than 100 MPN/100 mL. Although the MSL-CW system showed no significant difference in contaminant removal when compared to a CW under the same operating conditions, the clogging problem in the MSL-CW system was lessened.

The integration of MSL systems into CWs has also been applied for rice noodle wastewater treatment, and this system showed a better removal performance for COD, phosphorus, and total coliform than the MSL systems and CW systems [49].

The aforementioned studies suggest that the integration of MSL systems and CW systems can not only maintain or improve the pollution removal performance of CW systems, but also effectively reduce the clogging risk and the land occupation of CW systems.

In addition to the hybridization of MSL systems and CWs, MSL systems can be directly connected with CWs (Figure 4b). In a study by Song et al. [106], the MSL system was employed as the core treatment unit. In addition, a multifunctional anaerobic tank was established at the front of the MSL system, and a subsurface flow CW was installed behind the MSL system. The removal efficiencies of COD, BOD_5, TP, ammonia nitrogen, and TN were 92%, 93%, 92%, 86%, and 76%, respectively. In addition, the greenhouse gas emissions were lower than that of centralized wastewater treatment. Hence, this integrated system is promising for use as an environmentally friendly rural wastewater treatment technology. Therefore, in areas where CW systems have already been used for decentralized wastewater treatment, connecting MSL systems in series with CWs will be a recommended option for upgrading decentralized wastewater treatment systems.

6.2. Integration with Filters

Some scholars have integrated MSL systems with filters to reduce the operating cost and improve pollutant removal performance.

The integration of an MSL system with a filter helps to reduce the operation costs. An integrated system consisting of a coarse zeolite trickling filter and an MSL system (Figure 5a) using intermittent wastewater feeding can operate stably without aeration, and this effectively reduces the operation cost of the integrated system [116]. In a trickling filter, the use of coarse zeolite with a low packing density and a size of 35 mm, as well as an intermittent wastewater feeding mode, favored the air flow in the filter and ensured that there was no shortage of oxygen in the trickling filter. Therefore, the use of a trickling filter can achieve an initial degradation of OMs and ammonia nitrogen without aeration. The concentration of OM and ammonia nitrogen in the wastewater entering the MSL system after being treated by the filter is greatly reduced. As a result, the MSL system can effectively remove pollutants without aeration and clogging.

Additionally, the integration of an MSL system with a filter benefits pollutant removal performance. The TN removal performance requirements of MSL systems have not been satisfied in several studies [17,35,45,102]. To enhance the TN removal performance, Zhang et al. [117] developed an integrated system by connecting a horizontal flow MSL system (HFMSL) to a vertical flow trickling filter (VFTF) to treat rural septic tank wastewater. The VFTF was primarily used for OM degradation and nitrification, while the HFMSL system was used as an anaerobic treatment unit for denitrification. By optimizing the operation parameters, the VFTF-HFMSL system reached a TN removal efficiency as high as 92.8%, and the removal efficiencies of COD, ammonia nitrogen, and TP were also greater than 92%.

In addition, because PLs are characterized by relatively high porosity and large pores, an MSL system has limited capacity for removing bacterial indicators in wastewater [33,35,107]. Sand filters are satisfactory for removing bacteria [118,119], therefore, Latrach et al. [120] connected an MSL system and a sand filter in sequence to treat domestic wastewater (Figure 5b). The results showed that when the HLR was 100 $L/m^2/d$, the system showed a high removal performance for bacterial indicators (the Log 10 removals for total coliforms, fecal coliforms, and fecal streptococci were 4.46, 4.47, and 4.13 Log units, respectively) and achieved 100% removal of parasitic eggs. In addition, the removal efficiencies of conventional contaminants, such as SS, BOD_5, COD, TN, and TP, reached more than 92%.

These studies suggest that the effective integration of MSL systems with filtration systems can further enhance their wastewater treatment performance.

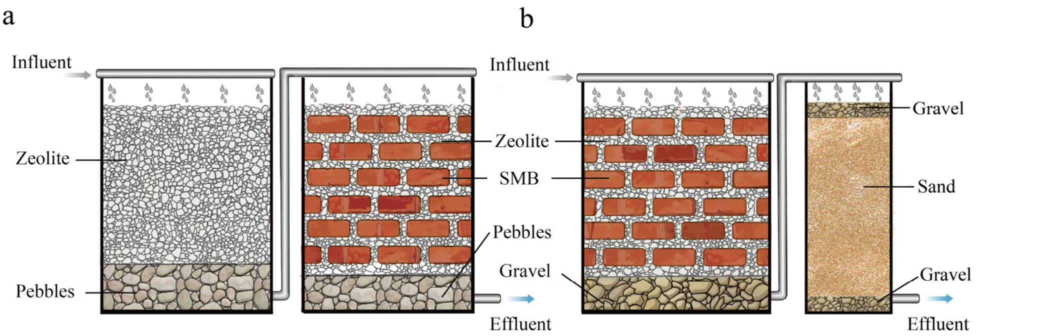

Figure 5. The integration of MSL systems with filters. (**a**) The integration of a trickling filter with an MSL system (the figure is redrawn according to Luo et al., 2014 [116]). (**b**) The combination of an MSL system with a sand filter (the figure is redrawn according to Latrach et al., 2016 [120]). MSL, multi-soil-layering; SMB, soil mixture block.

7. Conclusions

An MSL system is a wastewater treatment system with low energy consumption, low construction cost, and small land area occupation. Its applications have been expanded from the initial rural dispersed domestic wastewater to LW, food industry wastewater, antibiotic wastewater, and other types of wastewaters, and good application prospects have been demonstrated. Effluent quality can be further improved by integrating with technologies such as constructed wetlands and filters. To better promote the application of MSL systems, future research should be conducted according to the following aspects:

(1) The performance of MSL systems to remove unconventional pollutants. Previous studies have paid much attention to the removal of conventional pollutants by MSL systems, such as the COD, BOD, SS, nitrogen, phosphorus, and fecal coliform content. There is only one report regarding antibiotic removal, and there exist no relevant studies on new pollutants such as persistent organic pollutants, microplastics, and endocrine disruptors. The harmfulness and wide-source nature of the above-mentioned unconventional pollutants have attracted an increasing amount of attention in society. In addition, some new pollutants have been detected in rural domestic wastewater. As a potential treatment technology of rural domestic wastewater, the MSL system should improve its performance for the removal of these new pollutants.

(2) The decontamination mechanism of MSL systems. Computer analyses and simulations should be utilized to reveal the removal process of complex pollutants inside MSL systems, to screen the key parameters of MSL system operation, and to predict the performance of MSL systems. This will help researchers to attain a deeper understanding of the operational mechanism of MSL systems and provide references for optimizing system operations. Future research should pay more attention to these issues so as to provide strong references for the engineering applications of MSL systems.

(3) Further reduction of operating costs. Aeration equipment operation and maintenance are the primary sources of cost in an MSL system. In the future, to minimize the cost, MSL systems should be combined with other technologies, such as trickling filters. This will allow for stable operation and the achievement of good treatment results without aeration. In addition, green energy, such as wind and solar energy, can be utilized to power aeration.

Author Contributions: Conceptualization, T.-F.M. and J.H.; resources, J.W. and J.H.; writing—original draft preparation, T.-F.M. and J.W.; writing—review and editing, L.F. and X.-P.C.; project administration, L.F.; funding acquisition, T.-F.M., J.W. and J.H. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Chongqing Science and Technology Bureau (cstc2019jxjl20007), the China Postdoctoral Science Foundation (Grant No. 2020M683218), and the Doctoral Fund of Southwest University of Science and Technology (20zx7109).

Data Availability Statement: No new data were created or analyzed in this study. Data sharing is not applicable to this article.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

SS	Suspended solids
TSS	Total SS
TN	Total nitrogen
TP	Total phosphate
BOD	Biological oxygen demand
COD	Chemical oxygen demand
CWs	Constructed wetlands
MSL	Multi-soil-layering
SMBs	Soil mixture blocks
PLs	Permeable layers
OM	Organic matter
LW	Livestock wastewater
HLR	Hydraulic loading rate
VFCW	Vertical flow constructed wetland
MCs	Microcystins
MC-LR	Microcystin-LR, L stands for leucine and R for arginine
WWTPs	Wastewater treatment plants
SMX	Sulfamethoxazole
PHBV	Polyhydroxybutyrate
PBS	Poly-(bubulosuccinate)
ATP	Adenosine Triphosphate
HFMSL	Horizontal flow MSL
VFTF	Vertical flow trickling filter

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