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Remediation and disinfection capabilities assessment of some local materials to be applied in multi-soil-layering (MSL) ecotechnology

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ABSTRACT

The purpose of this study is to assess the improvement of the remediation effectiveness and disinfection capabilities of some local material to be applied in a multi-soil-layering (MSL) system. Different local materials such as pozzolan (Po), marble waste (Ma), and charcoal (Ch) have been tested in columns simulating MSL. These materials have been compared to materials (Br) generally used in standard MSL. To carry out this work, four columns simulating MSL systems were investigated (one standard and three others where different local materials are used as soil mixed blocks). Domestic effluent was applied to each system with a flow rate of 400 L m^{-2} d^{-1} (284.54 COD g m^{-2} d^{-1}) over 61 d. Therefore, the removal of suspended solids (SS), biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), $N–NH_4$, $N–NO_2$, total phosphorus (TP), *Escherichia coli* (*E. coli*) and *Streptococcus* from domestic wastewater was investigated. The measurement in the downstream flows of the columns shows that the Br and Po systems don't show clogging compared to the two other systems (Ch and Ma). As for the COD and BOD_5 removal, there is no significant difference between the four systems. In SS removal, however, the column (Po) remains the most efficient (93.28%); contrariwise the standard system (Br) remains the less performant (69.96%). For the TP elimination, charcoal and marble waste represents the best efficiency. Only the column-MSL with pozzolan has given convincing results in the removal of bacteria that reached 2.26 ± 0.47 CFU (colony forming unit) for *E. coli* and 2.26 ± 0.39 CFU for *Streptococcus*.

Keywords: Multi-soil-layering columns (MSL); Local materials; Treatment; Organic matter; Nutrients; Fecal indicators

1. Introduction

Wastewater discharge without treatment into natural media in rural areas is the main cause of water body pollution [1]. The absence of public sewerage systems and uncontrolled discharge of wastewater, as well as insufficient awareness of the rural population about the consequences of pollution, have led to the spread of diseases and their transmission to the surrounding population via surface and groundwater. As a result, the quality of soil and life, in general, is negatively impacted in the rural areas through water resources degradation.

Therefore, treating wastewater is a serious concern for small rural communities, with challenges of dispersed habitation and significant seasonal and even daily variation of wastewater flow and load [2], which necessitates the development of techniques more adapted to rural conditions. Hence, using a multi-soil-layer (MSL) decentralized system to treat wastewater is an opportunity to maximize the depuration efficiency at reduced costs [3].

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The MSL, a sustainable land-based system created by Wakatsuki in 1990, is easy to operate and maintain while occupying a relatively small area of land [4,5]. It is typically composed of layers of soil mixture blocks (SMB) alternating with permeable layers (PL) whose role is to prevent clogging by increasing the efficiency of wastewater infiltration rates through soil mixture blocks [6]. This system uses natural soil and zeolite to facilitate wastewater treatment through a combination of physical, biological, and biochemical processes [7]. This technology is based on the soil to improve treatment efficiency [4,7–12]. Masunaga et al. [10] and Wakatsuki et al. [4], suggested adding 10% powdered charcoal into the SMB because it attracts and adheres to organic matter as wastewater flows through the system, which increases the processing efficiency of microorganisms and ultimately achieving waste removal. A comparative study on the effect of various organic components (sawdust, rice straw, kenaf, and corncob) on the efficiency of an MSL system for domestic wastewater treatment was reported, recommending adding 10% of organic matter into the SMB [9]. Also adding iron scraps into the SMBs facilitates phosphorus adsorption considerably. In fact, Wakatsuki et al. [4] reported that mixing iron particles in the MSL system improves dramatically the phosphate fixing capacity of soil. The addition of 10 weight percentages of the iron increases the phosphate-P fixing capacity up to 5–10 g, or more, 1 kg of soil.

Compared to conventional soil systems, the MSL system could treat high hydraulic load rates (HLR) and pollutant loads thanks to alternating structures that enhance the filtration ability of soil [13].

The use of MSL system technology for wastewater has proven its efficiency worldwide. Many applications in Japan, China, Indonesia, Thailand, Hawaii, and recently in Morocco [14,15] have confirmed that MSL system purification performance could reach up to 98% reduction in biochemical oxygen demand (BOD_5), 94% of the chemical oxygen demand (COD) [10], 66.5% of total nitrogen (TN) and 96.2% of total phosphorus (TP) [16].

However, this system is characterized by moderate elimination of bacterial indicators of fecal contamination and pathogens [3], which could limit this technology when it comes to the reuse of treated wastewater for ultimate agricultural purposes. Therefore, this work main priority is to test a variety of local and cheap materials with a greater capacity to adsorb contaminants, especially fecal germs, and which could be used to improve the MSL disinfection capabilities.

The objective of this study is, therefore, to investigate the efficiency of three local materials: pozzolan, marble waste, and vegetal charcoal for use as alternative SMB materials in the MSL system in order to eliminate both fecal bacteria and organic matter from domestic wastewater, with the ultimate goal of reusing the treated water for agriculture purposes.

2. Material and methods

2.1. Structure and components of the MSL-column systems

The experimental device used in this study is composed of an upper water tank (H65 cm × L36 cm × W30 cm) from where acquired wastewater is transferred to feed the four MSL-columns with regulated flow in homogenous distribution. Subsequently, the wastewater inflow velocity is equivalent to HLR 400 L m^{-2} d^{-1} in each column.

The four MSL-column systems are packed in H30 cm × D4.4 cm polyvinyl chloride tubes which are distinguished by the nature of the granular materials used or their mixtures with different percentages as described in Table 1.

Each system is made up of two parts: a PL and SMB. The SMB has different material compositions arranged to form an alternative bricklayer as in Fig. 1. Four MSL-column were labeled (Br: with standard SMB), (Ch: charcoal), (Ma: marble), and (Po: pozzolan), all of which have particle sizes between 0.5 and 2 mm. The material constituting SMBs are described below:

Soil: the primary component of the SMB. A granulometry analysis was carried out for the used soil using a mechanical sieve analysis. The different fraction of soil texture according to the United States Department of Agriculture soil taxonomy [17] and its main characteristics are described in Table 2.

Powdered vegetal charcoal: a type of porous material with large surface areas and particle sizes of less than 2 mm. Vegetal charcoal has been utilized successfully and widely as a filter medium of the water purification because of its high cation exchange capacity and high adsorption capabilities.

Sawdust: organic matter mainly used to provide a C source for microorganisms.

Pozzolan: a natural rock made by slag (projections) volcanic basalt or similar composition. Pozzolan has both high surface area and roughness and, as such, is frequently used as a bacterial filter support [18]. However, the properties related to the area of the material grains provide higher purification capacity as quoted in [19]. The granular pozzolan material used is obtained by crushing and then sieving natural rock from quarries at Timehdit, in the region of Ifrane in Morocco.

Marble waste: the powdered marble is brought from a large private marble processing unit and is used in the experiment without any prior treatment. Its specific surface area is equal to 3.31 m^2 g^{-1}.

Table 1
Composition (% dry weight) of soil mixed blocks (SMB) for each column (Br): MSL reference, (Ch): charcoal, (Ma): marble waste and (Po): pozzolan

MSL system	Natural soil %	Charcoal %	Sawdust %	Iron %	Marble trash %	Pozzolan %
Br	70	10	10	10	0	0
Ch	70	20	0	10	0	0
Ma	70	10	0	10	10	0
Po	0	10	10	10	0	70

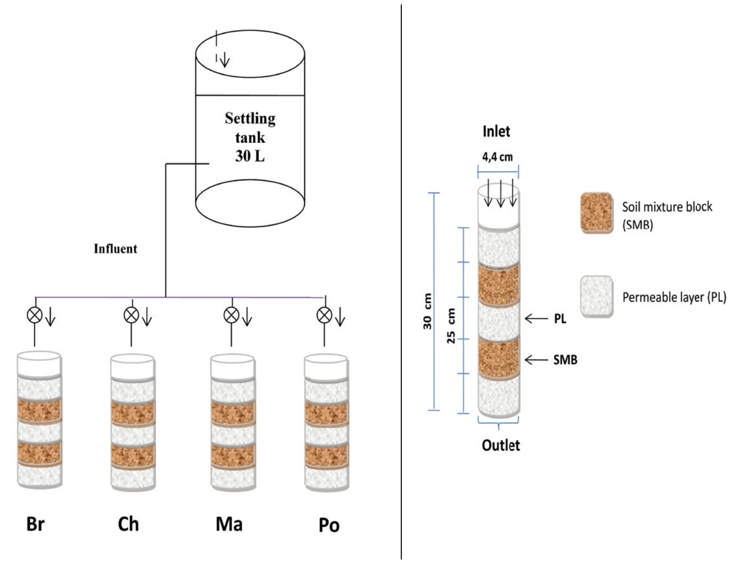

Fig. 1. Laboratory-scale MSL systems: (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

Table 2
Soil physicochemical characteristics

Parameter		Value
pH (H_2O)		7.49
TOC (%)		19.27
TKN (%)		4.75
Textural fraction (%)	Coarse sand (%)	48.42
	Fine sand (%)	19.01
	Coarse silt (%)	3.86
	Fine silt (%)	20.14
	Clay (%)	8.58

As for the PL used for all experiments, it consists of 5 cm gravel aggregate, with a diameter of 3–5 mm.

2.2. Sampling and water quality monitoring

Wastewater is weekly collected from a village (located in the Rural Commune of Tidili, Al Haouz Province), using plastic bottles for chemical essays, and sterile glass bottles for bacteriological studies, according to the French Standard Methods (AFNOR) [20] and Rodier [21]. Influent and effluent from the investigated MSL-column systems are sampled every 3 d and analyzed to determine the treatment efficiency.

The flow rate of the drained water was measured for each column. The volume of effluent is measured using a graduated flask during 1 h (the operation was duplicated three times). The flow rate was monitored during the experience after each 3 d as measurement frequency.

2.3. Analytical methods

2.3.1. Bacteriological analysis

Bacteriological analysis has focused on *Escherichia coli* (*E. coli*) and fecal streptococci. The analysis is done using the dilution method for the samples suspected to be highly contaminated [22] and filtration when they are lowly loaded. The removal of the microbiological indicators is expressed as Log units of colony-forming unit (CFU) per 1 mL (Log CFU mL^{-1}). The cultures media used are as follows: Eosin methylene blue for *E. coli* and Slanetz and Bartley medium for fecal streptococcus.

2.3.2. Physicochemical analysis

pH is measured in situ using a multi-parameter probe-type C930. The following parameters are measured according to AFNOR [20] and Rodier [21]. BOD_5 is determined by the respirometry method and the COD is analyzed according to the dichromate open reflux method [23]. Suspended solids (SS) concentration is determined by the filtration method, NH_4^+–N concentration by the indophenol method, NO_2–N concentration by the diazotization method, PO_4–P concentration by the ascorbic acid method and TP is determined as PO_4–P after potassium peroxodisulfate digestion [20].

3. Results and discussions

3.1. Characterization of used materials

A physicochemical characterization of selected materials was carried out in order to define the parameters capable of influencing physicochemical wastewater quality and biomass development.

The soil used is sandy-silt soil with a neutral pH and a high percentage of organic matter (Table 2). Marble powder is constituted mainly by carbonates (Table 3) and Pozzolan is characterized by high cation exchange capacity and very small sized particles (Table 4).

Observations by scanning electron microscopy (SEM) were carried out on the samples of powdered marble and pozzolan to characterize the surface condition of grains (Fig. 2). The morphological aspects of the two materials showed the

Table 3
Physico-chemical characteristics of marble waste

pH (H_2O)	8.91
Electrical conductivity (EC) (mS cm^{-1})	1.28
Organic C (%)	3.28
OM (%)	5.55
N (%)	0.28
$CaCO_3$ (%)	98

Table 4
Physico-chemical characteristics of pozzolan [24]

pH	8.3
Granulometry (mm)	0.5–2
Limestone (‰)	9
Cation exchange capacity (meq kg^{-1})	420
Retention capacity (%)	10.4
OM (‰)	9
EC (Mm hos cm^{-1})	0.10

presence of particle agglomerates with irregular shape and the fundamental differences in roughness at the surface. They present a very large quantity of surface micro-defects whose mean sizes are close to a few microns and therefore could favor pollutants and the biomass attachment.

The analysis of the major elements is carried out for each used material by a fluorescence X-ray diffractometer. The presence of calcium, magnesium, iron, and aluminum and chemical elements can influence the pollutants' adsorption processes on the material and also the development of biomass.

The obtained results (Table 5) show that the marble waste differs from the other materials by a rather noticeable presence of limestone. However, the pozzolan is very rich in silica (Si). Also, the soil and pozzolan are fairly rich in iron whereas the only pozzolan is rich enough in aluminum.

Table 5
Chemical composition of materials used

Element	Soil mg g^{-1}	Charcoal mg g^{-1}	Marble mg g^{-1}	Pozzolan mg g^{-1}
Ca	40.2	26.5	921.1	82.1
Mg	–	9.726	37.9	8.63
Si	4.694	2	19.3	131.7
Fe	20.7	–	4.62	15.9
Al	2.149	–	6.23	10.3
Cl	38.4	25.6	0.99	5.051
K	1.998	13.7	2.53	4.729
Ti	1.456	–	0.27	1.339
P	1.317	0.904	0.56	6.321
Mn	0.224	1.112	0.29	–
S	0.647	0.431	1.35	1.323
Zn	0.1	0.025	0.051	0.203
Zr	0.107	0.006	0.023	0.08
Cu	0.046	–	0.14	0.029
Sr	0.21	0.059	0.722	0.136
As	0.01	0.003	–	0.012
Rb	0.043	0.016		0.031
Y	0.014	0.006	–	0.022
Th	–	0.044	–	–
V	–	–	–	0.193
Cr	–	–	–	0.184
Ni	–	–	–	0.036
Ag	–	0.045	–	–
Cd	–	0.025	–	–
U	–	0.009	–	0.014
Mo	–	0.007	–	–
Pb	0.07	–	–	0.046
Te	–	–	1.9	–
Na	–	–	1.7	–
Co	–	–	0.16	–

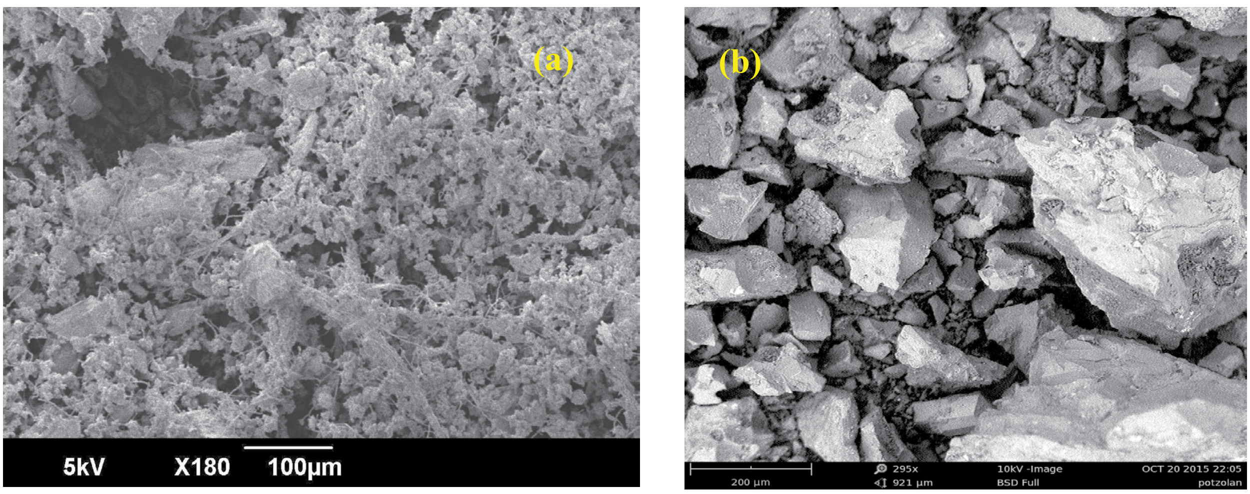

Fig. 2. SEM images of powdered marble at 100 mm (a) and pozzolan (b) at 200 mm.

3.2. Physicochemical parameters of wastewater

Several tests were performed for each MSL-column system under the same HLR conditions. The influent and effluent data were then analyzed for determining the pollution removal efficiency of each MSL-column.

Table 6 summarizes the ranges of influent and effluent concentrations and removal efficiencies during the experiment with different materials in the SMB.

3.2.1. Flow rate

Fig. 3 shows changes in the flow rate for different columns during 61 d of experimentation. In the case of (Ch) and (Ma) systems, the flow rate sharply decreases from 16 and 17.5 mL h^{-1} at the first day to approximately 3 and 5.5 mL h^{-1} at the end of the experiment respectively. On the other hand, for the system (Br) and (Po), the decrease is less important and reaches 9 and 9.75 mL h^{-1}, respectively at the end.

The decrease of the flow rate probably results from the proliferation of microorganisms and the overgrowth of biofilms in the MSL-columns. The structure of the MSL-column (Br) and (Po) systems makes possible more high-speed treatment than do (Ch) and (Ma) which cannot continue to treat wastewater because of their low permeability and probable clogging limitations. Particle size distribution of the SMB could influence clogging [11]. The difference in hydraulic behavior between the investigated columns is probably related to the difference in the particle size of the used materials. Charcoal and marble powders with small size particles could accelerate the clogging process than the other materials. Moreover, the clogging of the filter media could be explained by the retention of wastewater suspended particles in the bed pores or on the filter surface leading to an opaque aggregate which reduces water circulation. The development of microbial biomass in the filters could also contribute to the reduction of the filter bed porosity.

3.2.2. pH variation

The pH (potential of Hydrogen) measures the H^+ ion concentration of water. This parameter determines the number of physicochemical balances and depends on many factors; it is an important indication regarding the aggressiveness of water, ability to dissolve limestone.

The evolution of pH (throughout the study period) shows that wastewater is relatively neutral with an average value of 7.6 and extreme values of 8.1. For the effluent, a mean pH = 8 is observed for the four investigated systems (Fig. 4 and Table 6).

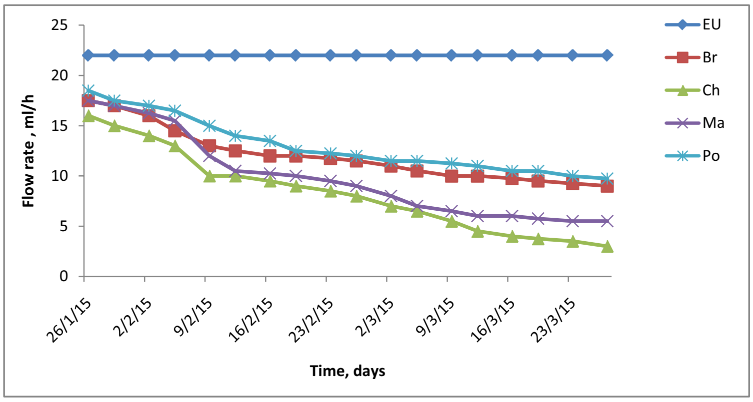

Fig. 3. Flow rate for different MSL-column systems (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

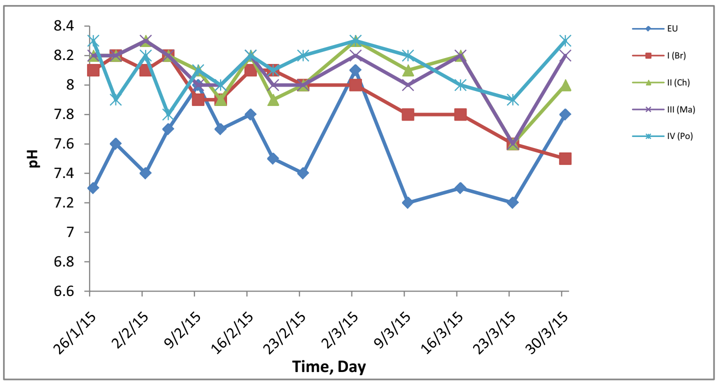

Fig. 4. Variation of the pH for different MSL-column systems (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

Table 6
Average, maximum and minimum of chemical parameters at raw wastewater (Raw WW), and the effluent of each system (Br): MSL reference, (Ch): charcoal, (Ma): marble waste and (Po): pozzolan

Parameters	Influent			Effluents											
	Raw WW			Br			Ch			Ma			Po		
	Min.	Average	Max.	Min.	Average	Max.	Min.	Average	Max.	Min.	Average	Max.	Min.	Average	Max.
Flow rate (mL h^{-1})				9	12.04 ± 0.62	17.5	3	8.38 ± 0.96	16	5.5	9.88 ± 0.98	17.5	9.7	13.04 ± 0.65	18.5
pH	7.2	7.57	8.1	7.5	7.95 ± 0.06	8.2	7.6	8.09 ± 0.05	8.3	7.6	8.09 ± 0.05	8.3	7.9	8.11 ± 0.04	8.3
EC (mS Cm^{-1})	1.84	2.07	3.11	1.84	2.09 ± 0.011	3.3	1.77	2.02 ± 0.09	2.82	1.84	2.31 ± 0.16	3.28	1.84	2.29 ± 0.14	3.07
SS (mg L^{-1})	60	106.07	160	10	30.71 ± 3.22	50	5	15.71 ± 3.47	55	5	13.57 ± 2.42	30	0	6.07 ± 1.5	20
BOD_5 (mg L^{-1})	150	209.43	391	39	53.79 ± 4.37	92	22	36.93 ± 4.23	72	23	32.21 ± 3.45	63	25	40 ± 5.13	85
COD (mg L^{-1})	425	621.34	1248	133	201.97 ± 26.77	456	122	201.03 ± 31.31	474	120	198.61 ± 29.30	446	107	208.11 ± 26.64	446
NH_4–N (mg L^{-1})	19.49	29.91	44.86	4.86	8.37 ± 0.75	17.03	5.83	8.30 ± 0.48	13.17	4.98	8.10 ± 0.46	12.63	6.22	8.25 ± 0.33	11.04
NO_2–N (mg L^{-1})	0.3	1.65	3.77	0.39	22.61 ± 7.62	82.33	0.03	4.48 ± 1.83	13.78	0.001	0.62 ± 0.33	4.66	0.13	21.05 ± 8.15	85.94
TP (mg L^{-1})	6.69	8.87	14.39	5.76	7.57 ± 0.41	10.69	3.8	5.74 ± 0.39	8.08	3.37	5.27 ± 0.34	7.02	5.57	7.26 ± 0.36	9.33

According to Luanmanee et al. [9], the pH change of the treated water by MSL is significantly related to the processes of nitrification and denitrification and the subsequent removal of TN. In addition, pH 8 could indicate that is no sign of anaerobic conditions inside the MSL-columns.

3.2.3. Evolution of the electrical conductivity

Levels of salinity, expressed in average electrical conductivity, are 2.07 mS cm^{-1} for wastewater, 2.09, 2.02, 2.31, and 2.29 mS cm^{-1} for successive systems (Br), (Ch), (Ma), and (Po) (Fig. 5). The values recorded during the analysis are important and appear not to exceed the recommended Moroccan Irrigation Standards (2.7 mS Cm^{-1}). The degradation of organic matter by bacteria contributes to the production of nutrients such as nitrogen and phosphate, which is reflected by the increase in electrical conductivity.

3.2.4. Organic matter and SS

Organic matter in wastewater is an important source of carbon for microorganisms. It is first physically and chemically adsorbed on soil and gravel surfaces and then decomposed by microorganisms [5]. The removal of the organic matter is carried out by biological degradation under oxygen conditions in the porosity of the MSL system.

During the study period, the values of BOD_5 are recorded between 150 and 391 mg L^{-1} with an average value of 209 mg L^{-1} for raw wastewater. These values are 41–94 mg L^{-1} with an average of 58 mg L^{-1} (73% of elimination) in the treated water by (Br), 24–74 mg L^{-1} with an average of 38 mg L^{-1} (82% of elimination) by (Ch), 23–63 mg L^{-1} with an average of 32 mg L^{-1} (84% of elimination) by (Ma) and from 27 to 85 mg L^{-1} with an average of 41 mg L^{-1} (81% of elimination) by (Po) (Fig. 6).

Fig. 7 shows the average COD removal efficiency for the four MSL-column systems. The COD values range is 425–1,248 mg L^{-1} with an average of 620 mg L^{-1} for wastewater and 133–456 mg L^{-1} with an average of 202 mg L^{-1} (68% of elimination) for (Br), 122–474 mg L^{-1} with an average of 201 mg L^{-1} (68% of elimination) for (Ch), 120–446 mg L^{-1} with an average of 198 mg L^{-1} (68% of elimination) for (Ma) and 107–446 mg L^{-1} with an average of 208 mg L^{-1} (66% of elimination) for (Po). No significant difference in removal rates of COD is observed between the four systems. The results show that the four systems achieved favorable efficiency with 66%–68% of COD removal.

In contrast, the removal rate of COD was usually lower than that of BOD_5 in each system, (Figs. 6 and 7, Table 6), and the results suggest that the removal of COD needed more SMB layers than that of BOD_5. This is because COD is slowly biodegradable organic matter, whereas BOD_5 represents an easily biodegradable organic matter.

Physicochemical reactions such as filtration and adsorption are probably the major treatment processes at the beginning of the four systems. It appears that the organic matter referred to as COD and $BOD_5$is easily trapped in the SMB because of the amount of pore space, large surface area and enhanced hydrophobic properties provided by the addition of charcoal. Consequently, the biological process of BOD_5 and COD removal seems to occur with time progress.

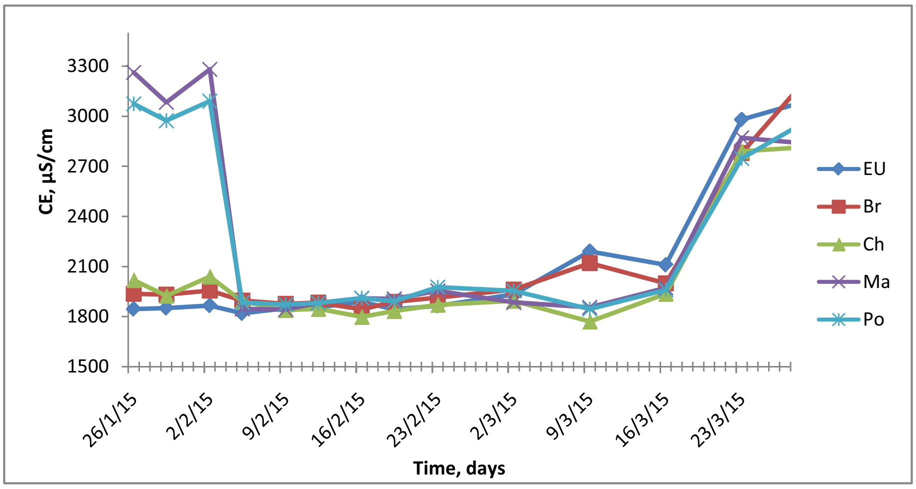

Fig. 5. Variations of electrical conductivity for different MSL-column systems (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

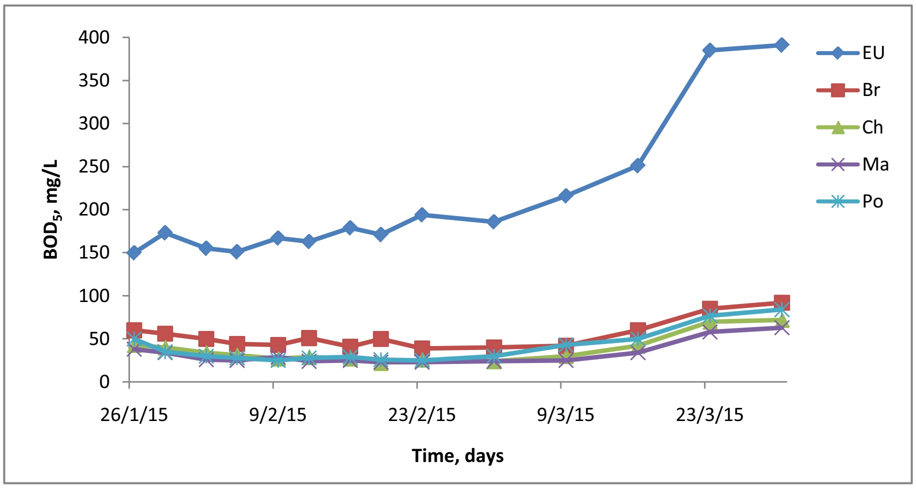

Fig. 6. Variations of BOD_5 for different MSL-column systems (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

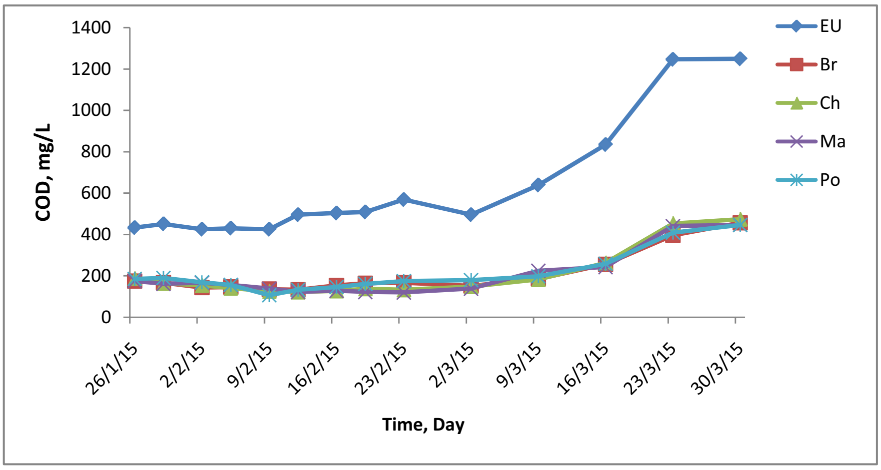

Fig. 7. Variations of COD for different MSL-column systems (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

For example, according to Sato et al. [11], the obtained removal rates in a pilot-scale study are 65.3% for COD and 75.6% for BOD_5. The reduction of organic matter is realized through biodegradation under oxygenated conditions in the porosity of the MSL reactor [5].

Fig. 8 illustrates the SS removal efficiency of the four systems. The average values are 106 mg L^{-1} for wastewater, 31 mg L^{-1} (67% of elimination) for (Br), 16 mg L^{-1} (82% of elimination) for (Ch), 13 mg L^{-1} (85% of elimination) for (Ma) and 6 mg L^{-1} (93% of elimination) for (Po). We can note that there is a notable filtration of SS from MSL-column during the study period. The results indicate that the system (Po) achieved the most favorable SS removal efficiency. Systems (Ma) and (Ch) achieved similar efficiencies, and system (Br) demonstrated the least favorable efficiency.

The best performance of the system (Po) is linked to the small-sized granular pozzolan stacked in the system, which yield the smallest pores compared with the other systems, thus increasing the filtration effect of SS. Charcoal and marble waste aggregates are used in systems (Ch) and (Ma) respectively, and therefore produce good effects. The test results indicate that the SS removal efficiency is correlated to the aggregate size and shape of the investigated materials.

3.2.5. Phosphorus removal

The TP concentration range is 6.69–14.39 mg L^{-1} with an average of 8.87 mg L^{-1} for wastewater and 5.76–10.69 mg L^{-1} with an average of 7.57 mg L^{-1} (11% of elimination) for (Br), 3.8–8.08 mg L^{-1} with an average of 5.74 mg L^{-1} (32% of elimination) for (Ch), 3.37–7.02 mg L^{-1} with an average of 5.27 mg L^{-1} (37% of elimination) for (Ma) and 5.57–9.33 mg L^{-1} with an average of 7.26 mg L^{-1} (14% of elimination) for (Po) (Fig. 9).

Filtration is a potential solution to remove TP from wastewater effectively. Filter media is the key of P removal by several mechanisms such as adsorption and precipitation within Fe, Ca or Al hydroxides in the media.

For Phosphorus removal, the comparative study shows that the four materials have neither the same capabilities nor the same efficiencies. Marble waste (37%) and charcoal (32%) seem to be the best adsorbents for phosphate. This could be attributed partly to the presence of phosphate minerals in natural materials themselves and the removal efficiency may probably result from their chemical and mineralogical composition [25].

Marble is a non-foliated metamorphic rock resulting from the metamorphism of limestone and composed mostly of calcite (a crystalline form of calcium carbonate, $CaCO_3$). As calcium ions can form stable and insoluble products with phosphate, calcium-based materials are considered to be one of the potential sorbents for phosphorus removal [26].

The addition of the marble waste in the system (Ma) leads to the precipitation of calcium dihydrogen phosphate with an optimal pH of 7 to 8 according to the following reaction [27]:

$$2H_3PO_4 + Ca(OH)_2 \rightleftarrows Ca(PO_4H)_2 + 2H_2O$$

Calcium, when present in excess simultaneously with a base (marble waste in this case), allows to achieve the

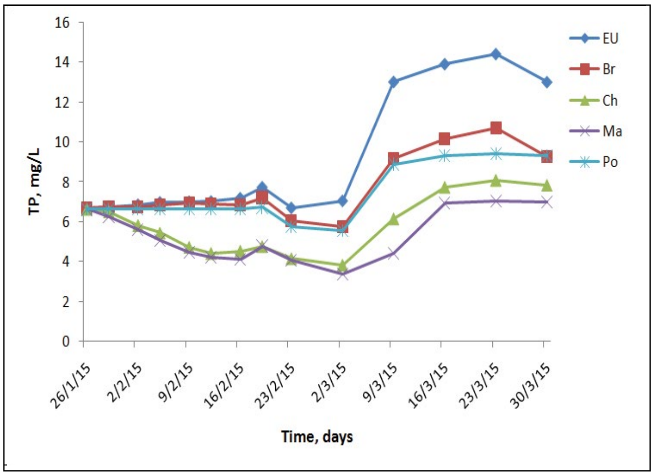

Fig. 9. Variations of total phosphorus for different MSL-column systems (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

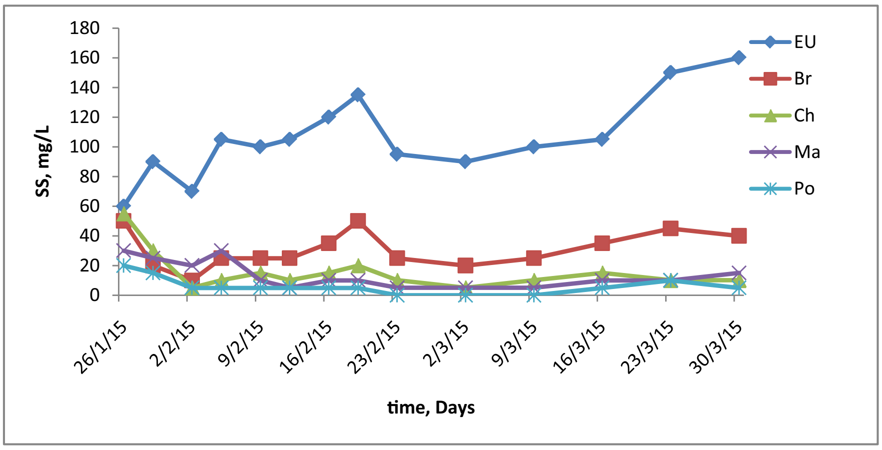

Fig. 8. Variations of SS for different MSL-column systems (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

formation of a precipitate which is sparingly soluble hydroxyapatite Ca10 $(PO_4)_6(OH)_2$. This high initial concentration of calcium ions privileges precipitation reaction [28].

In addition, the specific surface of the charcoal is very important than the other materials. The great variability of the specific area would have a great influence on the distribution of the active sites and thus the phosphate retention capacity is high as found by Ruan and Gilkes [29] in their work on synthetic aluminous goethite.

Moreover, the iron added in the SMBs also plays an important role in removing TP from wastewater [5].

3.2.6. Nitrogen

The average concentration of ammonium ion in the wastewater is 29.91 mg L^{-1} with extreme values of 19.49 and 44.86 mg L^{-1}.

NH_4–N concentrations are monitored along the study period. NH_4–N decreased notably in the first days with a maximum reduction of 62% achieved in MSL (Ma) (Fig. 10). The overall NH_4–N removal showed no significant difference between all systems (removal rate 69%, 70%, 70%, and 69% after the study period for MSL-column systems (Br), (Ch), (Ma), and (Po) respectively). However, there is no significant variation in the overall NH_4–N removal under various operating conditions. NH_4–N reduction corresponds with an increase in NO_2–N concentration in the first 18 d in systems (Br) and (Po), and after 36 d in (Ch) and (Ma) during which the latter increased from 0.49 at the influent to 7.22 and 1.07 mg L^{-1}, respectively (Fig. 11). However, throughout of study, NO_2–N concentration increased remarkably from 1.65 in raw wastewater to 22.61, and 21.05 mg L^{-1} for (Br) and (Po) systems, respectively. This concurrent NH_4–N reduction along the flow path demonstrates that (biological) nitrification is the dominant NH_4–N reduction mechanisms as evidenced by a decrease in dissolved oxygen [30].

3.3. Bacteriological parameters

The removal of *E. coli* and *Streptococcus* from the MSL system is examined for a range of soil columns mixed with different materials. Average concentrations of fecal contamination

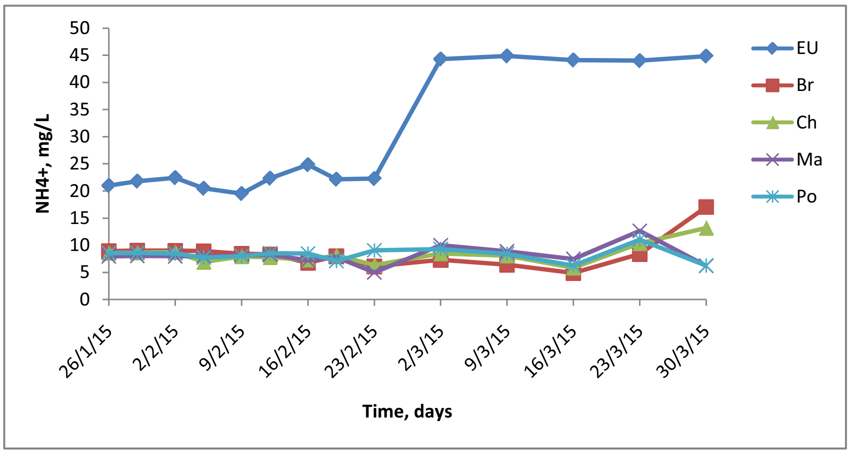

Fig. 10. Variations of NH_4–N for different MSL-column systems (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

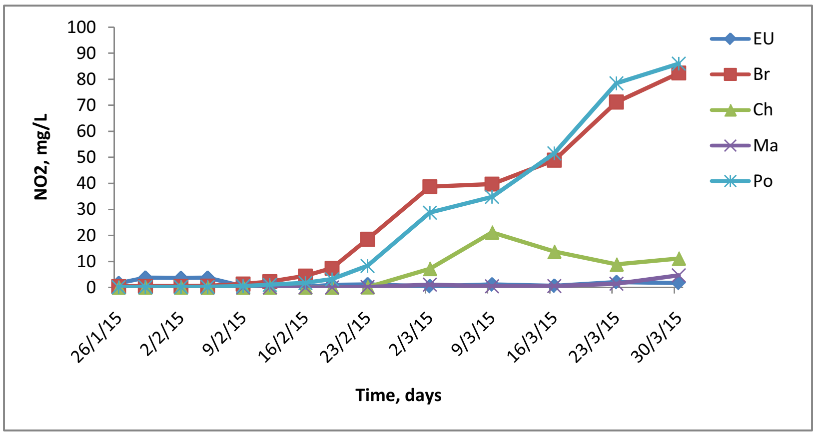

Fig. 11. Variations of NO_2–N for different MSL-column systems (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

Table 7
Summary of all the parameters (Raw WW): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste and (Po): pozzolan.

	Raw WW	Br		Ch		Ma		Po	
Flow rate (mL h^{-1})	22	9		3		5.5		9.75	
		bc		a		ab		c	
pH	7.57	7.95		8.09		8.09		8.11	
		a		a		a		a	
EC (mS Cm^{-1})	2.007	2.09		2.02		2.31		2.29	
		a		a		a		a	
		Concentration	Efficiency %	Concentration	Efficiency %	Concentration	Efficiency %	Concentration	Efficiency %
SS (mg L^{-1})	106.07	30.71	69.96%	15.71	82.37%	13.57	85.23%	6.07	93.28%
		c		b		ab		a	
BOD_5 (mg L^{-1})	209.43	53.79	73%	36.93	82%	32.21	84%	40	81%
		b		a		a		A	
COD (mg L^{-1})	621.34	201.97	68%	201.03	68%	198.61	68%	208.11	66%
		a		a		a		A	
TP (mg L^{-1})	8.87	7.57	11%	5.74	32%	5.27	37%	7.26	14%
		b		a		a		B	
NH_4–N (mg L^{-1})	29.91	8.37	69%	8.3	70%	8.10	70%	8.25	69%
		a		a	a			A	
NO_2–N (mg L^{-1})	1.65	22.61	–	4.48	–	0.62	–	21.05	–
		b		ab		a		B	
log10		Br		Ch		Ma		Po	
E. coli		1.04 ± 0.01		1.68 ± 0.23		1.25 ± 0.06		2.26 ± 0.47	
		c		a		b		A	
Fecal streptococcus		1.06 ± 0.01		1.53 ± 0.09		1.14 ± 0.05		2.26 ± 0.39	
		d		b		c		A	

a,b,c,d: grouping test results of Student–Newman–Keuls test.

indicators in MSL-column influent are 5.66×10^5 CFU mL^{-1} for *E. coli* and 1.07×10^5 CFU mL^{-1} for *Streptococcus*.

The average load of *E. coli* is 5.66×10^5 CFU mL^{-1} in wastewater; 5.22×10^4, 1.30×10^4, 3.18×10^4, and 5.84×10^3 CFU mL^{-1} respectively for (Br), (Ch), (Ma) and (Po). In terms of the bacterial load, the system (Po) effluent is notably less concentrated than the others.

For *Streptococcus*, the average values are 1.07×10^5 CFU mL^{-1} for wastewater and 9.34×10^3, 3.21×10^3, 7.72×10^3, and 8.28×10^2 CFU mL^{-1} for MSL-column systems with (Br), (Ch), (Ma), and (Po) respectively.

In general, the results show a consistent trend in the reduction of *E. coli* and *Streptococcus* for each system (Fig. 12). But, the lowest *E. coli* and *Streptococcus* performances occur in system (Br) and (Ma).

Removal efficiencies of *E. coli* are1.04 ± 0.01, 1.68 ± 0.23, 1.25 ± 0.06, and 2.26 ± 0.47 log units for (Br), (Ch), (Ma), and (Po) respectively. *Streptococcus* removals are 1.06 ± 0.01, 1.53 ± 0.09, 1.14 ± 0.05 and 2.26 ± 0.39 log units for (Br), (Ch), (Ma), and (Po) respectively. This improvement of removal rates in (Ch) and (Po) MSL-column scan be explained by favoring biomass activity in the porous medium. This is probably due to the specific large surface of charcoal and pozzolan, infiltrative surface character and adsorption phenomena. Many micropores, mesopores, and macropores are observed in the pozzolan by SEM images (Fig. 2) which show that the culture attached easily to the pozzolan owing to the abundance of pores that the microorganisms could enter.

Compared with other reports, results of bacteria removal show that there seems to be an affinity between bacteria and pozzolan. This can be justified by the high removal of the SS due to an efficient filtration by its large surface area, also the surface of solids, by its different physicochemical characteristics, can influence biomass by favoring or limiting the formation of biofilm [31]. Thus, the colonization of the support increases with the roughness of the support (the surface of contact is superior on rough surfaces as in the case of the surface of the grains of pozzolan).

The content of elements capable of acting as nutrients or electron acceptors is to be considered. Rogers et al. [32] show that microorganisms attack silicates containing phosphorus (as apatite) or iron (as oxyhydroxides) more readily.

The results in Table 7 are confirmed by the analysis of variance-Student–Newman–Keuls least significant difference

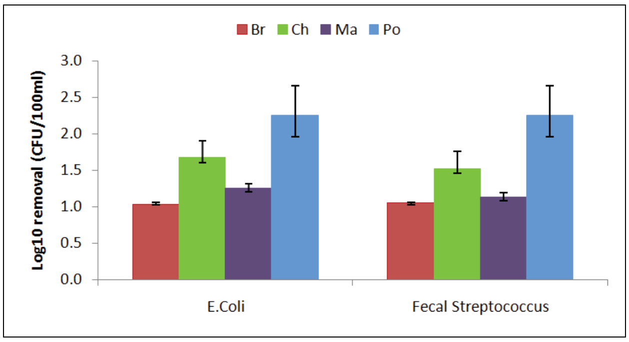

Fig. 12. Average removal efficiencies of bacterial indicators with different materials (EU): raw wastewater, (Br): MSL reference, (Ch): charcoal, (Ma): marble waste, and (Po): pozzolan.

statistical analysis. Indeed, the effect of the nature of the materials is significant ($p > 0.05$) in the case of flow rate, SS, *E. coli*, and *Streptococcus*.

4. Conclusion

Different local materials (pozzolan, marble waste, and charcoal) were investigated to improve the MSL system remediation effectiveness and disinfection capabilities to use the best of them of MSL ecotechnology for treating domestic wastewater.

Based on the results of this work, the following conclusions can be drawn:

The flow rate decreases noticeably after several days of experimentation in the case of the (Ch) and (Ma) systems. On the other hand, for (Br) and (Po) systems, the decrease was less important. This is probably due to the nature of the MSL (Br) and (Po) structures that allow faster processing than the (Ch) and (Ma) systems. The decrease of the flow rate most likely results from the proliferation of microorganisms and the overgrowth of biofilms in the MSL columns. The difference in hydraulic behavior between the different investigated columns is related to the use of materials characterized by particle size differences, especially charcoal and marble powder, that accelerate clogging processes.

The four MSL systems show no significant differences in organic removal rates which are ranked between 66%–68% for COD and 73%–82% for BOD_5. Organic matter is reduced by a microorganism's biological degradation under oxygenated conditions in the porosity of the MSL systems.

The (Po) system containing 70% pozzolan in the SMB achieved the most favourable removal efficiency of SS (93.28%), suggesting a high capacity to remove SS from concentrated domestic wastewater linked to the small-sized granular pozzolan stacked in the system which yielded the smallest pores compared with the other systems, thus increasing the filtration effect of SS. However, the (Br) system demonstrated the least favorable performance (69.96%). Statistical analysis showed that the removal efficiency of SS was significantly correlated with the size and shape of the studied materials. The elimination of SS was mainly realized by filtration on permeable gravel and soil mixture layers.

(Ch) and (Ma) MSL systems were significantly more efficient in the removal of TP than (Br) and (Po) MSL-column systems. This difference is probably due to the presence of a high quantity of Ca in marble waste and the high specific surface of the charcoal. Filter media is the key to TP removal by several mechanisms such as adsorption and precipitation within Fe, Ca or Al hydroxides in the media.

Ammonium removal showed no significant difference between all investigated systems with a removal rate of 69%, 70%, 70%, and 69% for (Br), (Ch), (Ma), and (Po) MSL-column systems respectively. A significant increase of nitrite content was observed in the outflow of (Br) and (Po) systems reaching values of 22.61 and 21.05 mg L^{-1} respectively. Probably these two systems are more aerated than the others. The co-existence of aerobic and anaerobic conditions in such a system is the most important factor controlling the efficiency of the MSL system in removing Nitrogen.

Regarding disinfection, (Po) MSL-column system showed a very significant removal which reached 2.26 ± 0.47 Log units for *E. coli* and 2.26 ± 0.39 Log units for *Streptococcus*. However, (Br) MSL-column system presented the lowest performances in removing these bacteria. The presence of many micropores, mesopores, and macropores observed in the pozzolan material by SEM images showed that the culture is easily attached to the pozzolan owing to the abundance of pores that the microorganisms could enter. However, further research regarding more optimization of the performance of such systems is needed.

References

[1] PNAR, Projet de programme national d'assainissement en milieu rurale: Assainissement liquide au Maroc: réalisations et perspectives, Programme National d'Assainissement Liquide et d'Epuration des Eaux Usées, Royaume du MAROC, 2013, p. 24.
[2] A. Kramer, L. Petta, J. Post, C. Wendland, EMWater Guide: Improving Wastewater Treatment and Reuse Practices in the Mediterranean Countries - A Practical Guide for Decision-Makers, Prepared Within the EMWater Project, InWEnt - Capacity Building International, 2007.
[3] L. Latrach, T. Masunaga, N. Ouazzani, A. Hejjaj, M. Mahi, L. Mandi, Removal of bacterial indicators and pathogens from domestic wastewater by the multi-soil-layering (MSL) system, Soil Sci. Plant Nutr., 61 (2015) 337–346.
[4] T. Wakatsuki, H. Esumi, S. Omura, High performance and N & P-removable on-site domestic waste water treatment system by multi-soil-layering method, Water Sci. Technol., 27 (1993) 31–40.
[5] X. Chen, A.C. Luo, K. Sato, T. Wakatsuki, T. Masunaga, An introduction of a multi-soil-layering system: a novel green technology for wastewater treatment in rural areas, Water Environ. J., 4 (2009) 255–262.
[6] P. Boonsook, S. Luanmanee, T. Attanandana, A. Kamidouzono, T. Masunaga, T. Wakatsuki, A comparative study of permeable layer materials and aeration regime on efficiency of multi-soil-layering system for domestic wastewater treatment in Thailand, Soil Sci. Plant Nutr., 49 (2003) 873–882.
[7] S. Luanmanee, T. Attanandana, T. Masunaga, T. Wakatsuki, The efficiency of a multi-soil-layering system on domestic wastewater treatment during the ninth and tenth years of operation, Ecol. Eng., 18 (2001) 185–199.
[8] T. Attanandana, B. Saitthiti, S. Thongpae, S. Kritapirom, S. Luanmanee, T. Wakatsuki, Multi-media-layering system for food service wastewater treatment, Ecol. Eng., 15 (2000) 133–138.
[9] S. Luanmanee, P. Boonsook, T. Attanandana, B. Saitthiti, C. Panichajakul, T. Wakatsuki, Effect of intermittent aeration regulation of a multi-soil-layering system on domestic wastewater treatment in Thailand, Ecol. Eng., 18 (2002) 415–428.
[10] T. Masunaga, K. Sato, J. Mori, M. Shirahama, H. Kudo, T. Wakatsuki, Characteristics of wastewater treatment using a multi-soil-layering system in relation to wastewater contamination levels and hydraulic loading rates, Soil Sci. Plant Nutr., 53 (2007) 215–223.
[11] K. Sato, N. Iwashima, T. Wakatsuki, T. Masunaga, Quantitative evaluation of treatment processes and mechanisms of organic matter, phosphorus, and nitrogen removal in a multi-soil-layering system, Soil Sci. Plant Nutr., 57 (2011) 475–486.
[12] R. Pattnaik, R.S. Yost, G. Porter, T. Masunaga, T. Attanandana, Improving multi-soil-layer (MSL) system remediation of dairy effluent, Ecol. Eng., 32 (2008) 1–10.
[13] Y.-D. Guan, D.-F. Xu, X. Chen, A.-C. Luo, H. Fang, Y.-Z. Song, Flow patterns of multi-soil-layering systems, Desal. Wat. Treat., 52 (2014) 4165–4169.
[14] K. Lamzouri, M. Mahi, S. Ouatar, E. Bartali, T. Masunaga, L. Latrach, L. Mandi, Application of multi-soil-layering technique for wastewater treatment in Moroccan rural areas: study of the operation process for an engineering design, J. Mater. Environ. Sci., 7 (2016) 579–585.
[15] L. Latrach, N. Ouazzani, A. Hejjaj, M. Mahi, T. Masunaga, L. Mandi, Two-stage vertical flow multi-soil-layering (MSL) technology for efficient removal of coliforms and human pathogens from domestic wastewater in rural areas under arid climate, Int. J. Hyg. Environ. Health, 221 (2018) 64–80.
[16] Y.D. Guan, X. Chen, S. Zhang, A. Luo, Performance of multi-soil-layering system (MSL) treating leachate from rural unsanitary landfills, Sci. Total Environ., 420 (2012) 183–190.
[17] Soil Survey Staff, Soil Survey Manual, USDA Agriculture Handbiiks 18, U.S. Government Printing Office, Washington, D.C., 1993.
[18] A. Seron, M.C. Dictor, B. Clozel, F. Mermillod-Blondin, Projet ANR-ECOPLUIES: Prévention et réduction des phénomènes de colmatage par modification de l'interface, Rapport. BRGM RP 56939 FR, 2008.
[19] K. Ndi, D. Dihang, P. Aimar, G.J. Kayem, Retention of bentonite in granular natural pozzolan: implications for water filtration, Sep. Sci. Technol., 43 (2008) 1621–1631.
[20] AFNOR, Recueil de norme française: eau, méthodes d'essai, 2éme édition, Paris, 1997.
[21] J. Rodier, L'analyse de l'eau: eaux naturelles, eaux résiduaires, eau de mer 7éme édition ed., Dunod, Bordas, Paris, 1996, p. 1365.
[22] Moroccan Standards, Norme Marocaine homologuée par arrêté du ministre de l'industrie, du commerce et de la mise à niveau de l'économie. Service de normalisation industrielle Marocaine (SNIMA), 2006.
[23] APHA, American Public Health Association: Standard Methods for Analysis of Waste and Waste Water, APHA, Washington, D.C., 1992.
[24] A. Aït Houssa, E. Nouga, H. Oualili, Y. Chtaibet, A. Chaddad, Fertigation de la Tomate Hors Sol dans la Zone de Douiet au Maroc, IPI Regional Workshop on Potassium and Fertigation Development in West Asia and North Africa, Rabat, Morocco, 24–28 Nov 2004.
[25] S.L. Coulibaly, Thèse: Abattement des phosphates des eaux usées par adsorption sur des géomatériaux constitués de Latérite, grès et schistes ardoisiers, 2014.
[26] C. Vohla, M. Koiv, H.J. Bavor, F. Chazarenc, U. Mander, Filter materials for phosphorus removal from wastewater in treatment wetlands—a review, Ecol. Eng., 37 (2011) 70–89.
[27] DEGREMONT, Mémento technique de l'eau, Ed. Degrémont, Paris, 1989.
[28] L. Youcef, S. Achou, Elimination des phosphates par des procedes physico-chimiques, Larhyss Journal, ISSN 1112–3680, n 04, Juin (2005), pp.129–140.
[29] H.D. Ruan, R.J. Gilkes, Kinetics of phosphate sorption and desorption by synthetic aluminous goethite before and after thermal transformation to hematite, Clay Miner., 31 (1996) 63–74.
[30] C.D.T. Abel, S.K. Sharma, S.A. Mersha, M.D. Kennedy, Influence of intermittent infiltration of primary effluent on removal of suspended solids, bulk organic matter, nitrogen and pathogens indicators in a simulated managed aquifer recharge system, Ecol. Eng., 64 (2014) 100–107.
[31] R.M. Donlan, J.W. Costerton, Biofilms: survival mechanisms of clinically relevant microorganisms, Clin. Microbiol. Rev., 15 (2002) 167–193.
[32] J.R. Rogers, P.C. Bennett, W.J. Choi, Feldspars as a source of nutrients for microorganisms, Am. Mineral., 83 (1998) 1532–1540.