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Study on the capability of treating Fe, Mn in the wastewater of several aquatic plant species

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ABSTRACT

Phytoremediation is a method of using plants to extract, isolate or detoxify pollutants through chemical, physical and biological processes. For years, this method has been considered as an effective method for wastewater treatment thanks to its high efficiency, low cost, stability, and simplicity of operation. Besides, this wastewater treatment technology is environmentally friendly since it increases biodiversity and contributes to the landscape of the environment and local ecosystems. This study aims to research the possibility to treat diluted Iron and Manganese from the wastewater using some aquatic plants such as Phragmites australis, Cyperus involucratus, Caladium bicolor, Chlorophytum Comosum, Dracaena sanderiana. The outcomes of this study indicate the significance of this treatment method as well as the factors that affect the efficiency of Iron and Manganese treatment in wastewater. Experiments have shown that after 96 hours the recorded efficiency of Fe treatment in wastewater in the environment 1 reaches 91.6 - 98.4% in all five aquatic plant species, with the highest value recorded in Caladium bicolor (EE1-Cal) (98.4%) and the lowest value recorded in Dracaena sanderiana (EE1-Dra) (91.6%). In environment 2, the treatment efficiency is lower, ranging from 75.4÷94.2%, with the highest and lowest values recorded in Caladium bicolor (EE2-Cal) and Dracaena sanderiana (EE2-Dra), respectively. The experiment for Mn treatment shows that recorded treatment efficiency in wastewater in environment 1 reaches 64.8÷95.8%, with the highest value recorded in Cyperus involucratus (EE1-Cyp) (95.8%) and the lowest value recorded in Dracaena sanderiana (EE1-Dra) (64.8%). In environment 2, the treatment efficiency is lower, ranging from 52.8÷80.6%, with the highest and lowest values recorded in Cyperus involucratus (EE2-Cyp) and Dracaena sanderiana (EE2-Dra), respectively.

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1. Introduction

Environmental pollution in general and heavy metal pollution, in particular, are being concerned by many scientists and management agencies. Many traditional methods have been applied to handle heavy metals, including physical and chemical methods, including burial. All of them have been proved to attain the fast speed of pollutant processing, yet they require complex technology and high cost. Therefore, removing heavy metals from polluted areas using plants with heavy metals tolerance and accumulation capability is considered as an effective, simple, environmentally-friendly, easy-to-deploy, and economically-effective method.

In the world, environmental pollution treatment and purification by plants (*Phytoremediation*) have been studied a lot in recent years based on the understanding of mechanisms of heavy metal absorption, metabolism, resistance and elimination of several plant species (Jerald Schonor, 2002; Diep, Garnier Zarli., 2007) resulting in many scientific and practical significant achievements. According to statistics, there are about 400 species of 45 plant families capable of accumulating heavy metals (Jerald, 2002; Majeti et al., 2003). Plant-based treatment methods have been developed in various ways and can be classified into a variety of mechanisms. The three mechanisms of Phytoextraction, Phytostabilization and Phytovolatilization are often applied to treat heavy metal pollution in soil, sediment and waste mud (EPA., 2000). Land after being treated and renovated can grow plants as normal. Plant growth at the treatment site also helps to reduce soil erosion due to wind and water, thereby preventing the spread of pollutants (Ademe., 1995).

In Vietnam, studies of using plants in environmental pollution treatment have also been conducted and recorded many positive results. Research by Diep Thi My Hanh et al. (2007) has shown the Pb uptake capability in all three parts of the trunk, roots and leaves of the *Lantana camara L.* in several Pb contaminated soil samples around Ho Chi Minh City, in which the root has been the most effective absorption organ compared to trunk and leaves (Diep et al., 2007).

The absorption capability of Pb and Zn of the *Pteris vittata L.* has also been studied by Tran Van Tua et al. (2011). The results of this research show that Pb and Zn accumulation capacity decreases over time, but the removal efficiency of Pb and Zn in soil increases due to increased biomass. In another research, five types of plants including *Zea mays L.*, *Typha angustifolia L.* (candle grass), *Phragmites vallatoria*, *Sesbania grandiflora L.*, and *Pennisetum purpureum* (elephant grass) have been used to study the absorption of three metals Cd, Cu and Zn in dredged sludge samples of Tan Hoa canal, Ho Chi Minh City. The results show that all five types have the capability to handle heavy metals and the *Pennisetum purpureum* has the highest absorption capacity while the *Zea mays L.* has the lowest one in all three parts of trunk, leaves and root (Dong et al., 2008).

However, studies often purely use plants so treatment of biomass after absorption is likely to cause secondary environmental pollution. The use of aquatic plants combined with some other filtering materials such as carriers and growing media has not been much researched. Limestone, which main chemical composition is CaCO_3 , is capable of neutralizing acids and is also a carrier material for microorganisms to stick and grow. Rice husks, an agricultural residue are used to provide the carbon source for growing plants through the cleavage of cellulose-decomposing microorganisms and at the same time are also filtration media and adsorbents, and media for the microorganisms involved in the removal of pollutants in wastewater (Luo et al., 2018; D.T. Hai et al., 2019). Therefore, this paper aims to study the capability of accumulating Fe and Mn content in wastewater prepared at the laboratory of five plant species including *Chlorophytum Comosum*, *Dracaena sanderiana*, *Phragmites australis*, *Cyperus involucratus* and *Caladium bicolor* to form a basis for further research at pilot scale.

2. Objects and research methods

2.1. Research objects

Samples of highly Fe and Mn polluted wastewater was prepared in the laboratory (Fe = 20 mg/l, Mn = 5 mg/l) combined with rice husks, limestone (Figure 1) and five aquatic plant species (Figure 2) including (*Chlorophytum Comosum* - a),

(*Dracaena sanderiana* - b), (*Phragmites australis* - c), (*Cyperus involucratus* - d) and (*Caladium bicolor* - e) of capable of growing and developing in water environment polluted with heavy metals and poor in nutrients.

2.2. Research methods

2.2.1. Experimental design

The experiments in this study have been designed at a pilot scale with a total experimental area of 30 m². Two experimental environment systems have been arranged as presented in Table 1 (Environment 1) and Table 2 (Environment 2).

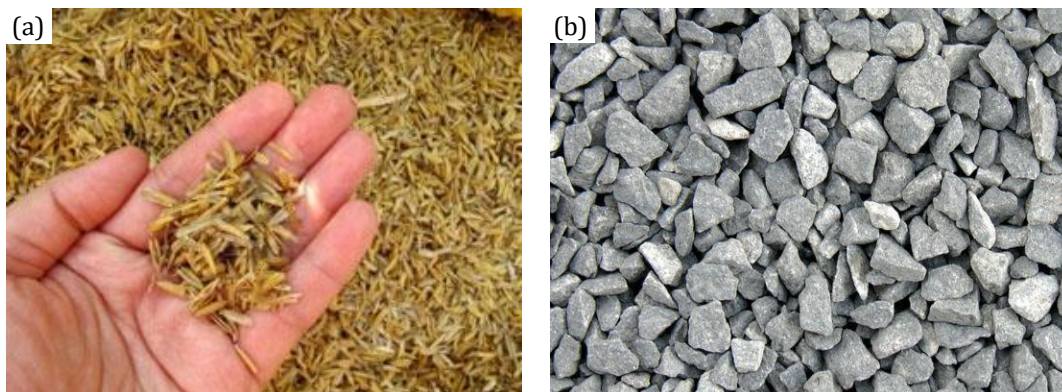


Figure 1. Rice husks (a) and limestone (b) selected to use in the research.



Figure 2. Selected five aquatic plant species.

(a) *Cyperus involucratus*; (b) *Dracaena sanderiana*; (c) *Phragmites australis*; (d) *Caladium bicolor*; (e) *Chlorophytum Comosum*.

- Experiment Environment 1 (EE1): Five types of plants (*Phragmites australis*, *Cyperus involucratus*, *Caladium bicolor*, *Chlorophytum Comosum*, *Dracaena sanderiana*) are grown in separate pots (50x35x20 cm) divided into 6 cells. The distance between cells is 15 cm. Each cell has 01 bush (3÷5 plants) and one cell contains only water (no tree planted). The average density of plants is 18 plants/m² (before conducting the experiment, the plants were grown and developed well in the experimental pots from March to June 2019). The layer of material in each pot is 20cm thick, including limestone (2x3cm size) added with hydrolysis husks. Wastewater is put into the study system with a volume of 10 liters/1 pot (containing Fe = 20 mg/l and Mn = 5 mg/l) and the initial water depth of 25 cm.

- Experiment Environment 2 (EE2): arranged similarly to the Experiment Environment 1 (EE1); however, the plants are grown directly in the wastewater without adding limestone and rice husks materials.

Sampling and analyzing wastewater in experimental pots are conducted after 3h, 6h, 9h, 12h, 18h, 24h, 30h, 36h, 48h, 60h, 72h, 96h to analyze Fe and Mn content in the wastewater for each plant, thereby assessing their Fe and Mn treatment performance.

2.2.2. Laboratory analysis method

The content of Fe and Mn in wastewater is analyzed at the Institute of Environmental Technology by the following procedure:

Mn analytical process: Draw 5 ml of the analytical sample into 50 ml glass beaker then add 1 ml H₂SO₄ (dilute). Dry the solution, cool, add distilled water to 20 ml and add 1 ml H₂SO₄ (dilute) + 0.5 ml Ag⁺ + 2.5 ml K₂S₂O₈, heat to evaporate to 10 ml. Transfer to a 25 ml volumetric flask and make up to volume with distilled water. Optical measure at 520 nm wavelength with UV-Vis spectrophotometer.

Fe analytical process: Draw 5 ml of the analytical sample into a 25 ml volumetric flask. Add 0.5 ml of hydroxylamine + 1.5 ml of acetate buffer solution. Make up to 25 ml, shake well. Draw 20 ml of the solution transformed from Fe³⁺ to Fe²⁺ into a 25 ml volumetric flask. Add 0.5 ml hydroxylamine + 2.5 ml acetate buffer solution + 0.5 ml phenanthroline reagent solution and make up to 25 ml with distilled water. Put in a dark area for 15 minutes and optical measure at 510 nm wavelengths with a UV-Vis spectrophotometer.

The treatment efficiency of Fe and Mn of each plant is assessed by the formula (Ngo Thuy Diem Trang and Hans Brix, 2012):

$$\text{Treatment efficiency (\%)} = \frac{(C_0 - C_e)100\%}{C_0}$$

Table 1. Experimental arrangement of environment 1 (with limestone and rice husks).

No.	Code	Metal concentration		Experiment Environment 1 (EE1)	Note
		Fe(mg/l)	Mn(mg/l)		
1	EE1-Cal	20	5	Planting <i>Caladium bicolor</i>	Plants were grown in pots with limestone and rice husk materials
2	EE1-Cyp	20	5	Planting <i>Cyperus involucratus</i>	
3	EE1-Dra	20	5	Planting <i>Dracaena sanderiana</i>	
4	EE1-Phr	20	5	Planting <i>Phragmites australis</i>	
5	EE1-Chl	20	5	Planting <i>Chlorophytum Comosum</i>	
6	EE1-Blank	20	5	Blank (without planting)	

Table 2. Experimental arrangement of environment 2 (without limestone and rice husks)

No.	Code	Metal concentration		Experiment Environment 2 (EE2)	Note
		Fe(mg/l)	Mn(mg/l)		
1	EE2-Cal	20	5	Planting <i>Caladium bicolor</i>	Plants purely were grown in wastewater
2	EE2-Cyp	20	5	Planting <i>Cyperus involucratus</i>	
3	EE2-Dra	20	5	Planting <i>Dracaena sanderiana</i>	
4	EE2-Phr	20	5	Planting <i>Phragmites australis</i>	
5	EE2-Chl	20	5	Planting <i>Chlorophytum Comosum</i>	
6	EE2-Blank	20	5	Blank (without planting)	

where C_0 is the initial concentration of metal ion, C_e is the concentration of metal ion at the time of the study.

2.2.3. Data processing and analyzing

All the data in the paper is analyzed and plotted with Excel 2010 with statistical significance accepted at $\rho < 0.05$.

3. Research results and discussion

3.1. Results of Fe treatment in the wastewater of aquatic plants

Results of analysis of Fe concentration in the wastewater of both 02 experimental systems after 96h experiment from 20/6/2019 to 24/6/2019 are presented in Tables 3, 4 and Figures 3, 4, 5, 6.

The results in Table 3 and Figures 3, 4 show that, the Fe concentration in wastewater in environment 1 (EE1) (planting different types of aquatic plants on limestone and rice husk materials) tends to decrease over time. The Fe concentration in the *Caladium bicolor* planting pot

(EE1) drops the most, followed by the *Phragmites australis*, *Cyperus involucratus*, *Chlorophytum Comosum* and *Dracaena sanderiana*. The treatment efficiency of Fe in wastewater reaches 91.6÷98.4%, with the highest value recorded in *Caladium bicolor* (EE1-Cal) (98.4%), followed by *Phragmites australis* (EE1-Phr) (96.65%), *Cyperus involucratus* (EE1-Cyp) (96.65%), *Chlorophytum Comosum* (EE2-Chl) (93.25%) and *Dracaena sanderiana* (EE1-Dra) (91.6%).

Similarly, the results in Table 4 and Figures 5, 6 show that the concentration of Fe in wastewater in environment 2 also tends to decrease over time but slower than environment 1. The Fe concentration in *Caladium bicolor* planting pot (EE2) drops the most, followed by the *Phragmites australis*, *Cyperus involucratus*, *Chlorophytum Comosum* and *Dracaena sanderiana*. The Fe treatment efficiency in wastewater in system 2 only reaches 75.4÷94.2%, with the highest value in the *Caladium bicolor* (EE2-Cal) (94.2%), followed by the *Phragmites australis* (EE2-Phr) (85.8%), *Cyperus involucratus* (EE2-Cyp) (82.1%), *Chlorophytum Comosum* (EE2-Chl) (79.75%), and *Dracaena sanderiana* (EE2-Dra) (75.4%).

Table 3. Changes of Fe concentration in wastewater in the environment 1 by time.

Fe concentration (mg/l)	Experimental time (hour)													QCVN40: 2011/BTNMT Column B
	0h	3h	6h	9h	12h	18h	24h	30h	36h	48h	60h	72h	96h	
EE1-Cal	20	18.16	16.31	13.92	10.57	8.62	6.51	4.95	3.28	2.34	1.35	0.72	0.32	5
EE1-Cyp	20	19.6	17.66	16.26	14.32	11.85	9.03	8.27	7.38	5.92	4.28	2.15	0.73	5
EE1-Dra	20	18.55	18.17	17.24	16.06	14.27	13.06	10.95	9.27	8.45	6.37	3.95	1.68	5
EE1-Phr	20	18.22	16.45	14.27	12.61	10.58	8.72	7.03	6.27	4.82	3.08	1.76	0.67	5
EE1-Chl	20	19.06	18.12	17.05	15.46	13.82	12.11	10.64	8.82	7.41	5.62	3.26	1.35	5
EE1-Blank	20	19.85	18.2	17.54	16.82	16.23	15.76	15.57	15.22	15.04	14.65	13.82	12.56	5

Table 4. Changes of Fe concentration in wastewater in system 2 by time.

Fe concentration (mg/l)	Experimental time (hour)													QCVN40: 2011/BTNMT Column B
	0h	3h	6h	9h	12h	18h	24h	30h	36h	48h	60h	72h	96h	
EE2-Cal	20	18.96	18.16	16.31	15.92	14.31	13.31	12.92	10.57	9.26	7.58	3.37	1.16	5
EE2-Cyp	20	19.67	19.62	18.37	17.28	16.34	15.42	14.48	12.55	10.42	8.31	6.25	3.58	5
EE2-Dra	20	19.03	18.77	18.52	17.86	16.72	16.06	15.17	13.98	12.06	9.72	7.54	4.92	5
EE2-Phr	20	18.88	18.23	17.28	16.85	15.42	14.37	13.21	11.68	9.53	7.72	4.37	2.84	5
EE2-Chl	20	19.36	19.12	18.45	17.71	16.65	15.98	14.93	13.78	11.54	9.31	6.75	4.05	5
EE2-Blank	20	19.85	19.85	19.27	18.54	18.21	18.06	17.84	17.22	16.48	16.05	15.82	15.06	5

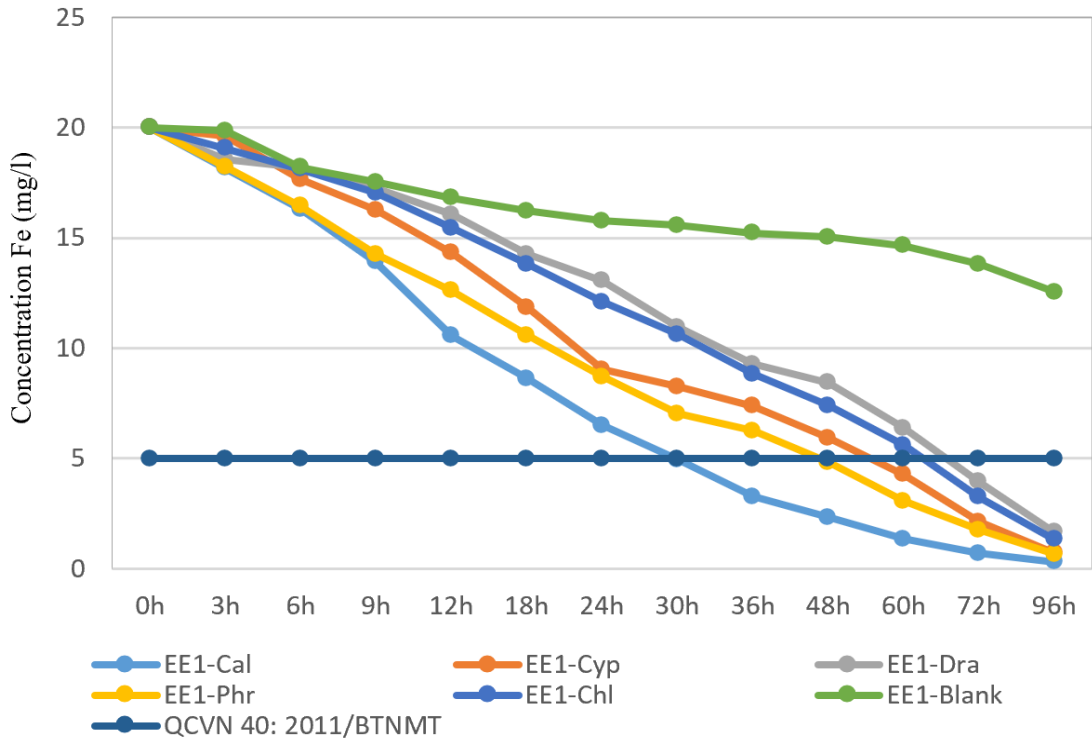


Figure 3. Change of Fe concentration (EE1).

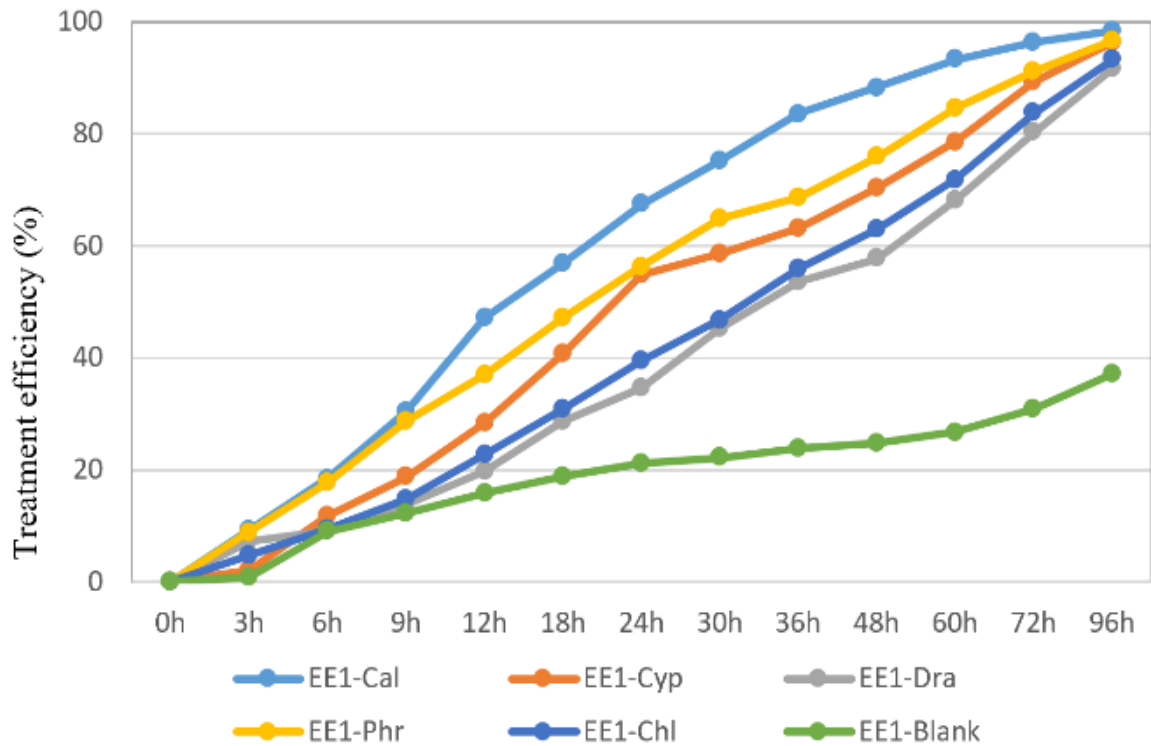


Figure 4. Treatment performance of Fe (EE1).

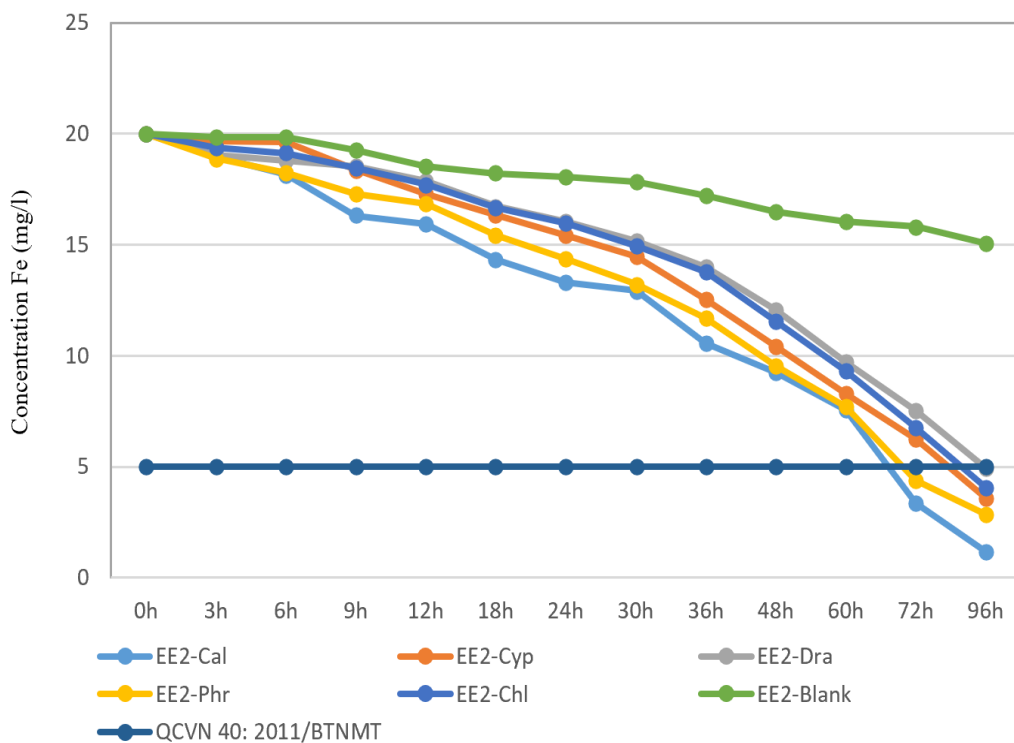


Figure 5. Changes of Fe concentration (EE 2).

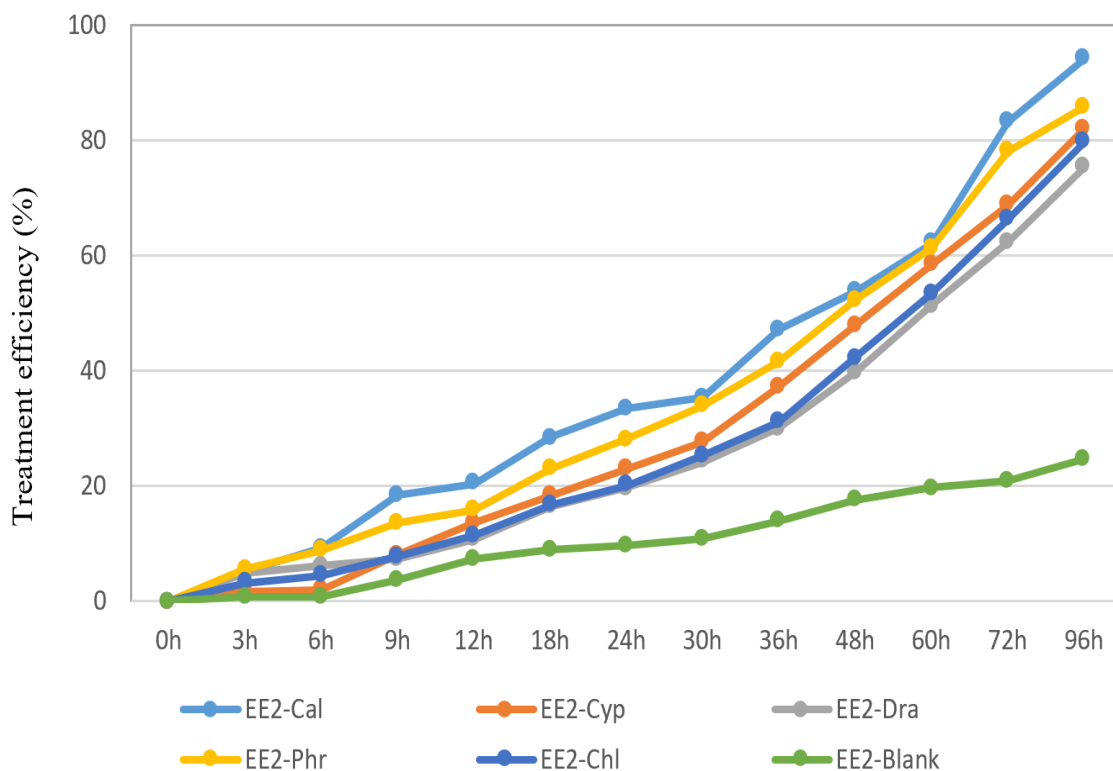


Figure 6. Treatment performance of Fe (EE 2).

In conclusion, through the above experiments and analysis, it can be seen that the *Caladium bicolor* and *Phragmites australis* are the best for the treatment of Fe in wastewater in both experimental systems.

3.2. Results of Mn treatment in the wastewater of aquatic plants

Results of analysis of Mn concentration in the wastewater of both 02 experimental systems after 96h experiment are presented in Table 5, 6 and Figure 7, 8, 9, 10.

The results in Table 5 and Figure 7 show that the Mn concentration in wastewater in environment 1 tends to decrease over time. The Mn concentration in the *Cyperus involucratus* planting pot (EE1) drops the most, followed by the *Phragmites australis*, *Caladium bicolor*, *Chlorophytum Comosum* and *Dracaena sanderiana*. The treatment efficiency of Mn in wastewater (Figure 8) tends to increase over time, ranging from 64.8÷95.8%, with the highest efficiency recorded in *Cyperus involucratus* (EE1-

Cyp) (95.8%), followed by *Phragmites australis* (EE1-Phr) (85.6%), *Caladium bicolor* (EE1-Cal) (77%), *Chlorophytum Comosum* (EE1-Chl) (69.2%) and *Dracaena sanderiana* (EE1-Dra) (64.8%).

The results in Table 6 and Figure 9 show that the concentration of Mn in wastewater in environment 2 also tends to decrease over time but slower than environment 1. The Mn concentration in the *Cyperus involucratus* planting pot (EE2) drops the most, followed by the *Phragmites australis*, *Caladium bicolor*, *Chlorophytum Comosum* and *Dracaena sanderiana*. The Mn treatment efficiency in wastewater in environment 2 (Figure 10) tends to increase over time but slower than environment 1, only reaches 52.8÷80.6%, with the highest value in the *Cyperus involucratus* (EE1-Cyp) (80.6%), followed by *Phragmites australis* (EE1-Phr) (68.4%), *Caladium bicolor* (EE1-Cal) (61.6%), *Chlorophytum Comosum* (EE1-Chl) (55%) and *Dracaena sanderiana* (EE1-Dra) (52.8%).

Table 5. Changes of Mn concentration in wastewater in system 1 by time.

Mn concentration (mg/l)	Experiment time (hour)													QCVN40: 2011/BTNMT Column B
	0h	3h	6h	9h	12h	18h	24h	30h	36h	48h	60h	72h	96h	
EE1-Cal	5.00	4.82	4.25	4.12	3.96	3.81	3.42	3.05	2.68	2.34	1.9	1.72	1.54	1
EE1-Cyp	5.00	4.76	4.08	3.51	2.32	1.92	1.68	1.31	1.07	0.95	0.72	0.33	0.21	1
EE1-Dra	5.00	4.95	4.56	4.06	3.75	3.58	3.35	2.96	2.82	2.27	2.06	1.95	1.76	1
EE1-Phr	5.00	4.78	4.37	3.94	3.52	3.29	2.48	1.92	1.77	1.56	1.15	0.94	0.72	1
EE1-Chl	5.00	4.81	4.42	4.25	3.71	3.44	3.1	2.85	2.68	1.84	1.62	1.36	1.15	1
EE1-Blank	5.00	4.96	4.95	4.91	4.88	4.8	4.73	4.69	4.66	4.61	4.57	4.5	4.45	1

Table 6. Changes of Mn concentration in wastewater in system 2 by time.

Mn concentration (mg/l)	Experiment time (hour)													QCVN40: 2011/BTNMT Column B
	0h	3h	6h	9h	12h	18h	24h	30h	36h	48h	60h	72h	96h	
EE2-Cal	5.00	4.95	4.82	4.55	4.25	4.18	4.02	3.94	3.86	3.42	3.09	2.68	2.36	1
EE2-Cyp	5.00	4.92	4.76	4.37	4.08	3.25	2.88	2.32	1.93	1.6	1.35	1.07	0.97	1
EE2-Dra	5.00	4.96	4.88	4.72	4.56	4.12	3.96	3.75	3.52	3.35	2.96	2.82	2.25	1
EE2-Phr	5.00	4.93	4.79	4.54	4.37	3.95	3.38	3.13	2.95	2.47	1.98	1.77	1.58	1
EE2-Chl	5.00	4.94	4.81	4.68	4.42	4.05	3.83	3.6	3.44	3.1	2.87	2.68	1.92	1
EE2-Blank	5.00	4.98	4.96	4.96	4.95	4.92	4.88	4.86	4.81	4.75	4.69	4.67	4.64	1

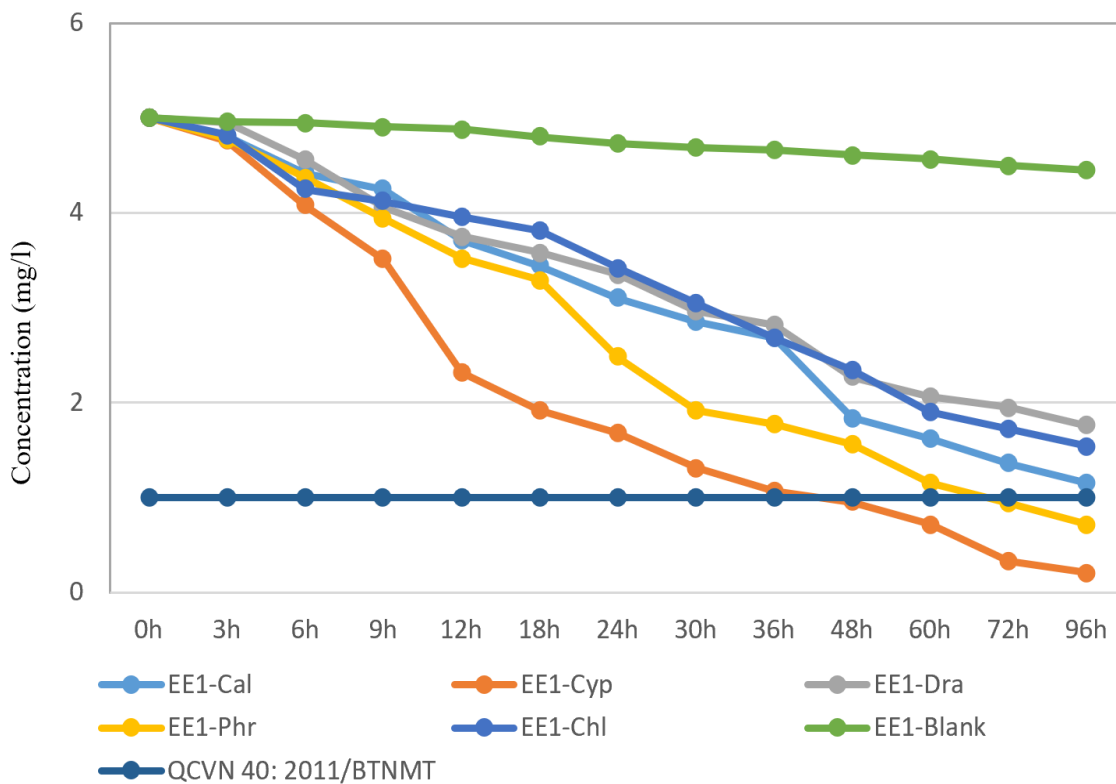


Figure 7. Changes of Mn concentration (EE1).

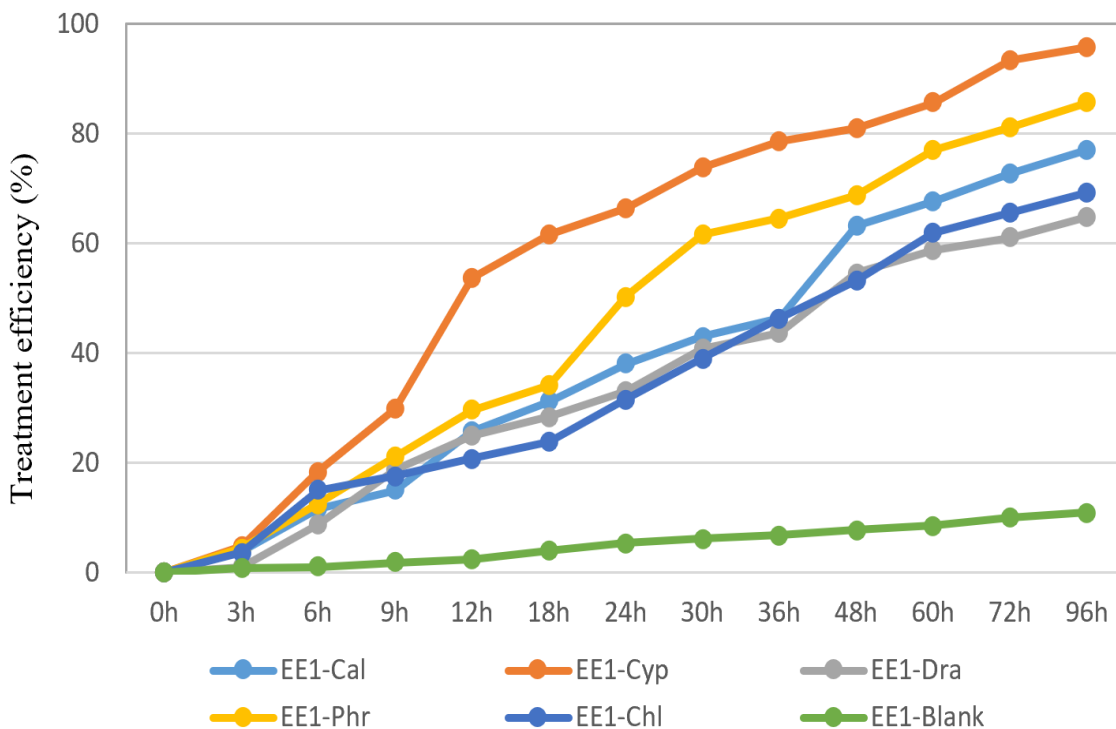


Figure 8. Treatment performance of Mn (EE1).

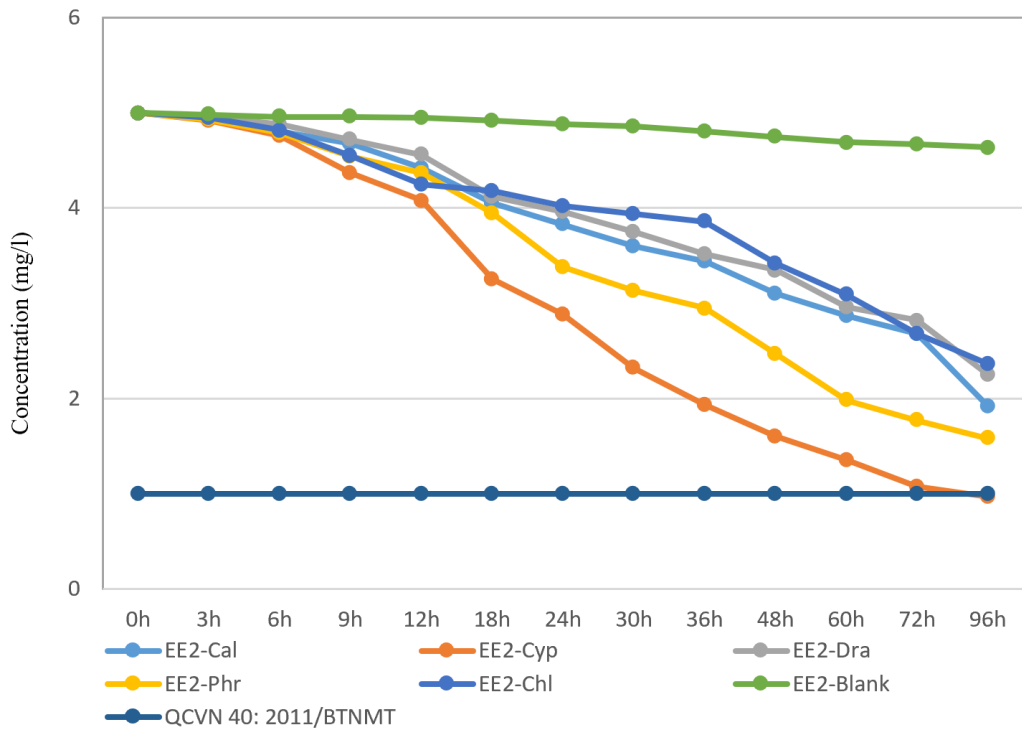


Figure 9. Changes of Mn concentration (EE2).

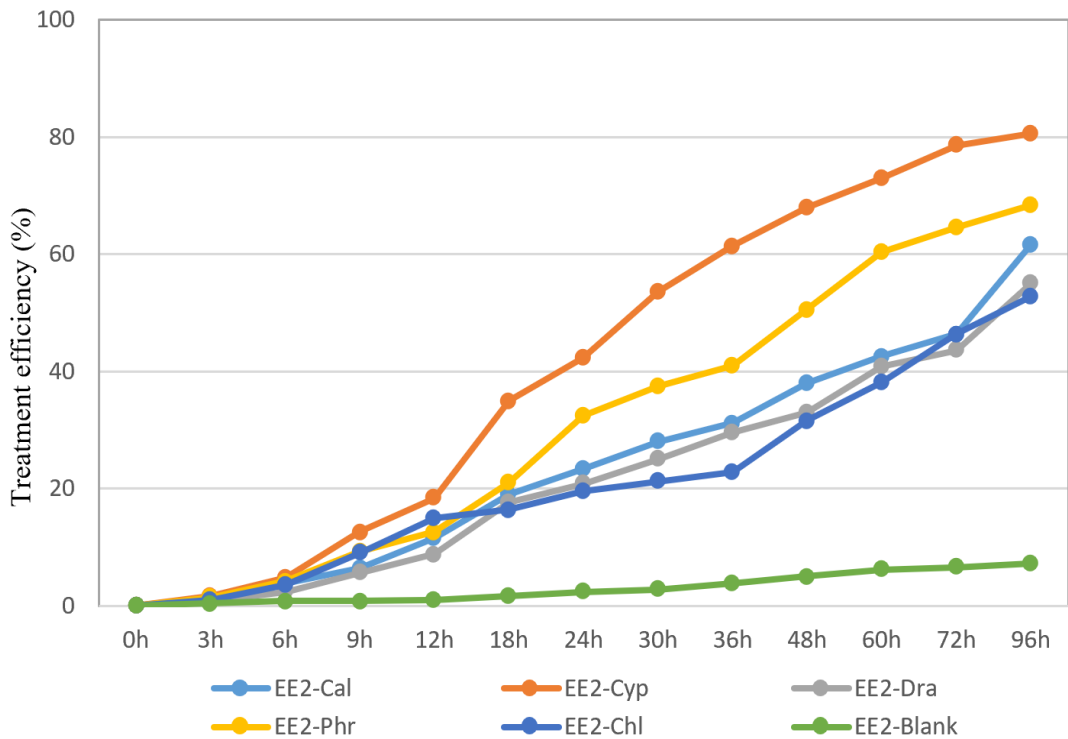


Figure 10. Treatment performance of Mn (EE2).

Through the above experiments and analysis, it can be seen that the *Cyperus involucratus*, *Phragmites australis*, and *Caladium bicolor* have the best Mn treatment performance in wastewater in both systems. However, environment 1 with added rice husks and limestone as the media has higher performance than environment 2 using purely aquatic plants. This is because limestone and rice husks themselves are also capable of filtering and purifying water (Vymazal, 2007; B.T.K. Anh et al., 2019). Filtration systems comprising aquatic plants and filtration materials capable of eliminating pollutants including organic compounds such as TSS, COD, N, P, and heavy metals have been recognized and applied in many places around the world (Vymazal, 2007; Ibekwe et al., 2016). Aquatic plants and microorganisms play an important role in removing organic compounds and heavy metals from wastewater. Meanwhile, stone, gravel, or rice husk as filter materials are capable of removing TSS and stabilizing pH to the permitted standards (Kadlec and Knight, 1996; Klomjek, 2016). The results of this study are also consistent with some published data in previous studies relating to the capability of plants to handle heavy metals. Bui Thi Kim Anh et al. (2019), when using *Phragmites australis* with limestone, gravel and rice husks to treat organic pollution in livestock wastewater from biogas tanks, indicated that pollution parameters such as TSS, ammonium, total N and P all decreased after 168 hours of the experiment. The quality of output wastewater meets QCVN 62-MT:2016/BTNMT. Luo et al. (2018) used aquatic plants in combination with rice husks, macadam, sand, through the cleavage of cellulose-decomposing microorganisms, to reduce pollution in organic-rich wastewater.

Thus, it can be seen that the filtration system using aquatic plants combined with rice husk media is capable of treating water pollution in general and heavy metal pollution in particular with short treatment time and relatively high efficiency. It is possible to apply this model to treat Fe and Mn pollution in wastewater.

4. Conclusion

Experiment for Fe treatment shows that after 96 hours the recorded treatment efficiency in

wastewater in environment 1 reaches 91.6÷98.4% in all 5 aquatic plant species, with the highest value recorded in *Caladium bicolor* (EE1-Cal) (98.4%) and the lowest value recorded in *Dracaena sanderiana* (EE1-Dra) (91.6%). In environment 2, the treatment efficiency is lower, ranging from 75.4÷94.2%, with the highest and lowest values recorded in *Caladium bicolor* (EE2-Cal) and *Dracaena sanderiana* (EE2-Dra), respectively.

Experiment for Mn treatment shows that recorded treatment efficiency in wastewater in environment 1 reaches 64.8 - 95.8%, with the highest value recorded in *Cyperus involucratus* (EE1-Cyp) (95.8%) and the lowest value recorded in *Dracaena sanderiana* (EE1-Dra) (64.8%). In environment 2, the treatment efficiency is lower, ranging from 52.8÷80.6%, with the highest and lowest values recorded in *Cyperus involucratus* (EE2-Cyp) and *Dracaena sanderiana* (EE2-Dra), respectively.

Five aquatic plant species including *Chlorophytum Comosum*, *Dracaena sanderiana*, *Phragmites australis*, *Cyperus involucratus* and *Caladium bicolor* have been selected to treat Fe, and Mn contaminated wastewater with two different experimental environments (environment 1 with added filtering material of rice husks combined with limestone; environment 2 with only aquatic plants). Experiment results show that both systems have the capability to treat Fe and Mn pollution. The concentration of Fe and Mn in wastewater in the environment 2 decreases over time but with a lower rate than environment 1 with added rice husks and limestone filter materials. Therefore, in order to effectively treat Fe and Mn in wastewater, it is necessary to combine aquatic plants with filtration materials in the same experiment system.

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