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Potential of Aquatic Plant as Phytoremediator for Treatment of Petrochemical Wastewater in Gebeng Area, Kuantan.

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ABSTRACT

Malaysia is a vast developing country that partly relies on its oil and gas sector industries for economic growth. However, rapid industrialization has resulted in production of massive amount of wastewater daily that may contain heavy metals and other contaminants. Phytoremediation is a promising technology and reliable method that uses plants to degrade, assimilate, metabolize, or detoxify contaminant. The objectives of this study are to identify the potential of aquatic plant and the percentage of contaminant removal in treating Petrochemical wastewater via phytoremediation. Three aquatic plants were use in this study; *Eichhornia crasspies sp.*, *Pistia statiotes sp.* and *Landoltai punctata sp*. Sampling method was carried out once for every 3 days and continues for 7 weeks period. Overall, 12 parameters are used to measure the effectiveness of phytoremediation process which are; BOD, COD, DO, pH, Turbidity, Cadmium, Zinc, Iron, Cooper, Lead, Nitrate and Chromium. Analysis of data was performed by using 1-way ANOVA .The significant ANOVA ($p<0.05$) studies shown the difference in values of the monitored 12 parameters which indicate the data obtained is accurate. The percentage removal of organic and inorganic pollutant in wastewater is also successfully determined; all three plants have 100% potential removal of Nitrate, Lead, Chromium, and Cadmium metal. Dotted Duckweed has 96.98% potential in removal of Iron metal, and 98.90% iron removal for Water Hyacinth. Water Lettuce proven high percentage of removal for BOD, 57.80% and decrement of pH value of 41.93%. In the end of this research; it is proven that all three plants are potential aquatic plant in treating Petrochemical wastewater through phytoremediation process. Consequently, the result of this study is being used to establish the basic element for designing a suitable wastewater treatment to promote sustainable management and reduce water pollution.

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INTRODUCTION

In this new globalization era, water pollution has become one of the most serious problem that faced by many countries. Water pollution is the contamination of water bodies such as lakes, rivers, oceans, and groundwater caused by human activities, which can be harmful to organisms and plants that live in these water bodies. Basically, water pollution is divided into two categories; point source pollution and non-point source pollution. Point source pollution refers to contaminants that enter a waterway through a discrete conveyance, such as a pipe or ditch. Examples of sources in this category include discharges from agro-based industries, manufacturing industries, sewage and sewerage works, sullage, or pig farming. While non-point sources refers to diffuse contamination that does not originate from a single discrete source such as agricultural runoff, forestry runoff, and urban runoff [8].

Water pollution is the discharge of some undesirable materials into water in high amounts. In the other words, water pollution is an issue that comes true by artificial effects, that constraint or prevents and that destroys ecological balance [15].

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Although it is undeniable that Malaysia is rich with water resources and able to fulfill the demands of waters for its citizen to continue daily activity. However, process of globalizations that has brought rapid development to country will eventually causes environmental pollution. This problem will also leads to deterioration of river basins that is used to supply water to citizen. Although, the sources of water does not extinct, but the quantity of raw water that are safe to use is decreasing due to these water resources pollution [8]. Thus, new technologies based on environmental friendly and economical are urgently required.

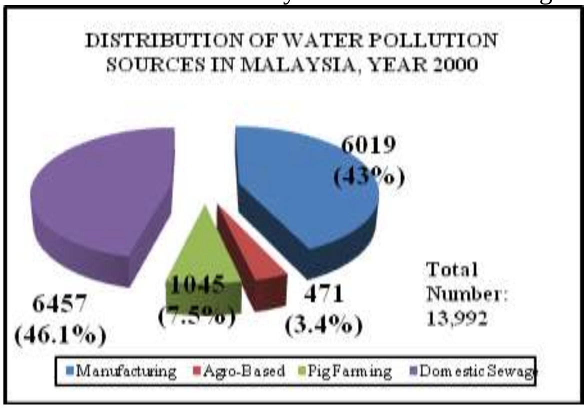

Fig. 1: Chart of Distribution of Water Pollution Sources in Malaysia, 2000 [9].

For these reasons, this research project was proposed with the aim of fulfilling this deficiency, by way of identifying the potential of aquatic plant in treating Petrochemical wastewater via phytoremediation and also with the characteristics that permit wastewater treatment to a level that meet up green technology, thus saving important amounts of reclaimed water, promoting sustainable management and reducing water pollution.

MATERIAL AND METHODS

Methodology part in this study came up with the design and selection of plant in remediating petrochemical wastewater. Malaysia has the world's 13th largest natural gas reserves, 24th largest crude oil reserves and largest production facility at a single location of liquefied natural gas. Through harnessing of its oil and gas reserves, it will generally in other way contribute to production of massive wastewater daily. Therefore, the source of wastewater for this study is located at Gebeng, Pahang. Gebeng is another Petrochemical cluster located 25km from Kuantan Town and 5 km from Kuantan airport.

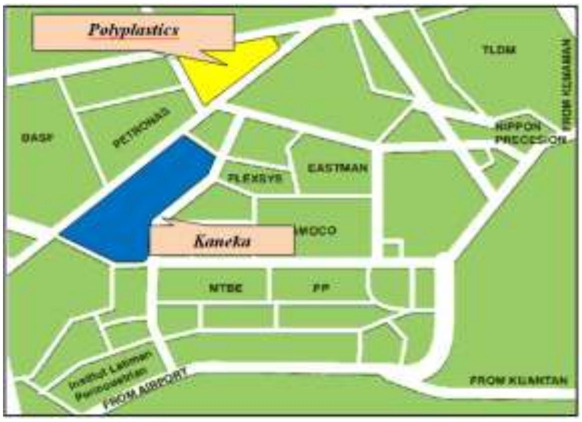

Fig. 2: Gebeng industrial area plan, [20].

In this research, it is conducted in two different places; which is in-situ and laboratory experiment. In-situ experiment is located at block Z area, in University Malaysia Pahang. This is the place where the design setup tank is settled and plantation of aquatic plant on wastewater held under supervision daily. In this method the aquatic plant is ensure to have sufficient oxygen and its growth is observed. Wastewater sample is also taken from here for the 12 parameter characteristic testing purposes.

Laboratory experiment or testing is done in (Fakulti Kejuruteraan Awam & Sumber Alam) FKASA Environmental Laboratory of University Malaysia Pahang. In this laboratory the water sample taken from in-situ in tested physically, chemically and biologically to determine its characteristics based on 12 parameters.

The wastewater qualities and plant growth especially the rhizobium part will be monitored. Apart from that, other parameters such as Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Dissolved Oxygen (DO), pH, Turbidity, Cadmium (Cd), Zinc (Zn), Iron (Fe), Cooper (Cu), Lead (Pb), Nitrate (NO_3) and Chromium (Cr). All these 12 parameters are referred according to American Public Health Associate (APHA).

Before conducting the experiment, firstly it is very important to make sure that the site for in-situ experiment is well prepared. All listed material is checked according to specification ordered and tools needed to construct experiment site are also ready. These are done so that the construction of site for experiment is done perfectly and material is assembled accordingly to plan. The dimension is properly measured, and all the assembled system is checked before the experiment begins. Water Hyacinth, Water Lettuce and Dotted Duckweed with approximately the same size, were used for the removal of organic and inorganic contaminant. Water Hyacinth and Water Lettuce can be found easily in lake as it known to grow profusely in polluted water bodies, and eutrophic lake. Both of these aquatic plants is obtained from the lake near Tasik Gambang, meanwhile the Dotted Duckweed plant is bought from nursery due insufficient time in looking and harvesting the plant. These aquatic plants were washed thoroughly with tap water followed by de-ionized water prior to the experimentation.

Industrial effluent or wastewater is taken from two selected petrochemical plant which are; Polyplastics Asia Pacific Sdn Bhd. and Kaneka Malaysia Sdn Bhd. Both of the effluent will be mixed inside the centralized tank. Water samples are often obtained by filling a container held just beneath the surface of the water, commonly referred to as a dip or grab sample. Through the use of special depth samplers (such as a Van Dorn bottle), grab samples can also be obtained from deep waters. This is important as distinct thermal and chemical differences can occur throughout the water column.

Frequency of experiment sampling is decided to be done once in three days. Each of the samples taken will be tested for the selected 12 parameters. This process will continue for 7 weeks, and the total samples will be 48 samples. Overall, there will be 12 set of sampling for 4 different containers. The measurement of selected 12 parameters for wastewater samples effluent will be carried out by using Spectrometer DR/2500 based on the SMEWW [2], as shown in **Table 3**.

Table 1: Parameters standard testing method.

No	Parameter	Unit	Testing Method	Standard Method Code
1	BOD_5@20°C	mg/L	Azide Modification Method	APHA 5210B#
2	COD	mg/L	Reactor Digestion Method	HACH 8000#
3	DO	mg/L	Membrane Electrode Method	APHA 4500.O(G)#
4	pH	-	Electrometric Method	APHA 4500-H^+-B(C)#
5	Turbidity	NTU	Nephelometric Method	APHA 2130 B#
6	Cadmium	mg/L	Dithizone Method	APHA 3500-Cd#
7	Zinc	mg/L	Zincon Method	APHA 3500-Zn(B)#
8	Iron	mg/L	FerroZine Method	HACH 8147*
9	Cooper	mg/L	Bicinchorinate Method	HACH 8506*
10	Lead	mg/L	Dithizone Method	APHA 3500-Pb(B)#
11	Nitrate	mg/L	Cadmium Reduction Method	HACH -8192
12	Chromium	mg/L	1,5-Diphenylcarbonhydrazide Method	APHA 3500-Cr(B)#

RESULTS AND DISCUSSION

The statistics performed by using an analysis-of-variance (ANOVA) table perform using Microsoft Excel. Generally, 1-way ANOVA is been using in analysing the data; removal of contaminants is taken as one factor. Meanwhile, significant of all statistical analysis accepted at ($p<0.05$). The ability and the endurance of the three selected plants to live in the industrial wastewater are determined by the condition and physical appearance of plants during the period of experiment and end of experiment.

Referring to **Table 2** Percentage of Removal of Contaminant by 12 parameters; each set of data was arranged execute to 1-factor of ANOVA ($p<0.05$) as follows: different types of plants were taken as one factor. The significant studies shown the difference in values of the monitored 12 parameters which indicate the data obtained is accurate.

Table 2: Percentage Removal of Contaminant.

Percentage of Removal of Contaminant (%)			
Parameter	Water Hyacinth	Water Lettuce	Dotted Duckweed
BOD	52.29	57.80	46.15
DO	39.44	37.19	31.24
COD	62.20	57.95	48.65
pH	34.12	41.93	29.10
Turbidity	-95.73	-96.68	-91.87
NO_3	100.00	100.00	100.00

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Fe	96.90	91.72	96.98
Zn	71.43	77.78	92.86
Cu	88.89	100.00	100.00
Pb	100.00	100.00	100.00
Cr	100.00	100.00	100.00
Cd	100.00	100.00	100.00

The results indicated that the BOD of effluent remediate by Water Hyacinth plants has decreased from 13.08 mg/L to 6.24 mg/L. As for Water Lettuce the value of BOD also decreased from 15.64 mg/L to 6.60 mg/L and for Dotted Duckweed it decreased from 10.92mg/L to 5.88 mg/L after seven weeks period. The potential removal of BOD by plants is Water Lettuce > Water Hyacinth > Dotted Duckweed. The summary of ANOVA showed significant differences with $P<0.05$ where P obtained is 0.000012. All BOD results prove negative correlation coefficients with time. The variance summary concluded that the ability of BOD removal of Water Lettuce plants is highest, followed by Water Hyacinth, Dotted Duckweed and control sample.

Meanwhile, the control sample has shown an almost flat line in the graph, which means that there is a very small reduction of Chemical Oxygen Demand. The value decreased is measured from 68 mg/L to 50 mg/L. A higher value of COD shows a higher sign of the measured effluent is polluted. The potential removal of COD by plants are Water Hyacinth > Water Lettuce > Dotted Duckweed. The summary of ANOVA showed significant differences with $P>0.05$. The obtained P value from data analysis for COD parameter was 0.21. However, all COD results prove negative correlation coefficients with time; which indicate a significant reduction of COD value along the experiment.

Water Hyacinth has shown the highest decreasing rate of Dissolved Oxygen concentration that is from 7.91 mg/L to 4.79 mg/L. Followed by Water Lettuce, where the value of Dissolved Oxygen decreased from 8.12 mg/L to 5.10 mg/L. On the other hand, Dotted Duckweed reduced the concentration of Dissolved Oxygen from 8.45 mg/L to 5.81 mg/L. The summary of ANOVA showed significant differences with $P<0.05$. The significant of P value obtained from the 1-way ANOVA anaylsis is 0.02. All DO results prove negative correlation coefficients with time. Meanwhile, by referring to the variance value; Water Hyacinth has the highest value, followed by Water Lettuce, Dotted Duckweed, and control sample.

Water Lettuce has a stepper line, which indicates that it has the highest ability in reducing the pH value from alkali of 10.16 to 5.90. These then, followed by Dotted Duckweed pH reduction from 9.76 to 6.92. As for Water Hyacinth, it reduces the value of initial effluent pH from 8.09 to 5.33. Obviously, from the reduction of pH value through remediation of the above three plants the end value of pH obtained were in the range promoted by [24] as in paragraph above. The summary of ANOVA showed significant differences with $P<0.05$. All pH results prove negative correlation coefficients with time. P-value obtained is 0.0002.

Water Hyacinth significantly increased the turbidity from 2.04 NTU to 47.80 NTU, as for Water Lettuce the turbidity significantly increased from 1.46 NTU to 44.01 NTU. Meanwhile, Dotted Duckweed increased the turbidity of the wastewater sample along the experiment from 1.38 NTU to 8.03 NTU. The summary of ANOVA showed significant differences with $P<0.005$. Based on 1-way ANOVA analysis, the obtained significant value of P is 0.003; this small value of P indicates that the smaller the p-value, the more certainty of the group means are different and significant.

The most potential plant in reducing the value of Nitrate is Water Lettuce, whereby it decreased the value of Nitrate from 0.80 mg/L to 0.00 mg/L. The second potential is Water Hyacinth, where it decreased the value Nitrate from 0.80 mg/L to 0.00 mg/L. As for Dotted Duckweed, the value decreased is from 0.80 mg/L to 0.00 mg/L. Based on the concentration reduction of Nitrate by these three plants, it is proven that these three plants have 100% potential of Nitrate ion removal. The summary of 1-way ANOVA for Nitrate parameter showed significant differences with $P<0.05$; the P-value obtained is 0.0002. This significantly small value of P indicates that the data obtained is accurate.

Dotted Duckweed has the highest potential in removing iron metal, where the removals range from initially 0.600mg/L to 0.013mg/L, followed by Water Hyacinth with the decreased of iron value from 0.420mg/L to 0.013mg/L. Water Lettuce reduced the value of iron metal from 0.290mg/L to 0.024mg/L.

The summary of ANOVA showed significant differences with $P>0.05$. Based on the 1-way ANOVA analysis the P-value obtained is 0.27. Although the significant P-value is more than 0.05 but from the data its shown that all plants results to prove negative Iron concentration correlation coefficients with time. This indicates that, there are still reductions of iron concentration proven in this experiment; whereby Water Hyacinth has the highest variance value followed by, Water Lettuce, Dotted Duckweed, and control sample.

Water Hyacinth has the highest potential in removing zinc metal, where the removals range from initially 0.21 mg/L to 0.06 mg/L, followed by Water Lettuce with the decreased of iron value from 0.18 mg/L to 0.04 mg/L. Dotted Duckweed reduced the value of iron metal from 0.14 mg/L to 0.01 mg/L. The control samples also show a very small reduction of iron value that is initially from 0.08 mg/L to 0.05 mg/L. The removal of heavy metal zinc by Water Hyacinth (*Eichorrnia crassipes sp.)* in present study is in agreement with [25]. The summary of ANOVA showed significant differences with $P<0.05$. The P value obtained from the 1-way

ANOVA analysis is 0.002. All plants results proved negative zinc concentration correlation coefficients with time; which indicate a significant reduction of zinc concentration in wastewater sample over time.

All the three plants have the potential in reducing the value of Copper. The most potential plant in reducing the value of copper is Water Lettuce, whereby it decreased the value of cooper from 0.10 mg/L to 0.00 mg/L. The second potential is Dotted Duckweed, where it decreased the value copper from 0.09mg/L to 0.00 mg/L. As for Water Hyacinth, the value decreased is from 0.09 mg/L to 0.01 mg/L. For the ANOVA analysis, confirm that the removal of copper by these three plants was proportional with time with the significant of $P<0.05$. In fact, from the 1-way ANOVA analysis the P-value obtained is very small that is 0.0001; which indicated that the data is significantly accurate.

Water Hyacinth has the highest potential in removing lead metal, where the removals range from initially 0.029 mg/L to 0.000 mg/L, followed by Water Lettuce with the decreased of lead value from 0.027mg/L to 0.000 mg/L. Dotted Duckweed reduced the value of lead metal from 0.027 mg/L to 0.000 mg/L. The control samples also show a very small reduction lead value that is initially from 0.026 mg/L to 0.018 mg/L. The summary of ANOVA for Lead parameter showed significant differences with $P<0.05$. From the 1-way analysis of ANOVA obtained P-value is (6.69×10^{-6}); this is a very small value of P which significantly show the difference in means value thus proving that the hypothesis made is accurate whereby the all these three plants has the potential in removing lead from wastewater.

Water Hyacinth has the highest potential in removing chromium metal, where the removals range from initially 0.060 mg/L to 0.000 mg/L, followed by Water Lettuce with the decreased of chromium value from 0.059 mg/L to 0.000 mg/L. Dotted Duckweed reduced the value of chromium from 0.059 mg/L to 0.000 mg/L. The control samples also show a very small reduction chromium value that is initially from 0.051 mg/L to 0.043 mg/L. From the value of chromium concentration reduction by these three plants, all plants prove 100% of chromium removal from the wastewater sample.

All the effluent with plants shows a decreasing value of Cadmium from time to time. This shows that all the three plants have the ability to removes cadmium from a wastewater. Among these three plants, Water Hyacinth has the highest potential in removing cadmium, the value of chromium decreased from 0.007 mg/L to 0.000 mg/L. Then, followed by Water Hyacinth, the reduction range is from 0.005 mg/L to 0.000 mg/L and Dotted Duckweed from 0.004 mg/L to 0.000 mg/L. This significant difference of cadmium removal by three different plants species is positively compliance with the statement of [3] whereby the heavy metal uptake potential largely varies with plant species.

Conclusion:

The present study proved Water Hyacinth (*Eichhornia Crassipes sp.*), Water Lettuce (*Pistia Stratiotes sp.*) and Dotted Duckweed (*Landoltia Punctata sp.*) are potential aquatic plant in treating Petrochemical wastewater through phytoremediation process. All these three selected plants have the ability in accumulating contaminant and metal ion.

The percentage removal of organic and inorganic pollutant in wastewater is also successfully determined. For BOD parameter, Water Lettuce has the highest percentage of removal with 57.80%, followed by Water Hyacinth 52.29%, and Dotted Duckweed with 46.15% of contaminant removal. As for DO, Water Hyacinth has the highest percentage of removal that is 39.44%, followed by Water Lettuce 37.19%, and Dotted Duckweed 31.24%. On the hand, removal of COD is highest by using Water Hyacinth with percentage of 62.20%, and second highest removal by Water Lettuce 57.95% and Dotted Duckweed 48.65%. However, as for pH, Water Hyacinth has the highest percentage of decrement that is 41.93, then Water Hyacinth of 34.12%, and lastly by Dotted Duckweed with removal of 29.10%. Measurement on the turbidity of wastewater shown an increasing of turbidity value with highest negative turbidity removal for Water Lettuce of -96.68%, followed by Water Hyacinth, -95.73%, and Dotted Duckweed of -91.87%. The removal of Nitrate, Lead, Chromium, and Cadmium showed 100% of removal for all three plants. This indicates that, all these three plants have shown 100% removal for these four metals. Meanwhile, the removal of Iron is leading by Dotted Duckweed, with 96.98% of removal, followed by Water Hyacinth 96.90% and lastly Water Lettuce of 91.72%. For the removal of Zinc ion, Dotted Duckweed has the highest percentage of removal with 92.86%, followed by Water Lettuce and Water Hyacinth 77.78%, and 71.43%. Water Lettuce and Dotted Duckweed had shown a 100% removal of Copper, meanwhile 88.89% removal of copper by Water Hyacinth.

Based on the experiment observation of plant growth and endurance towards wastewater, the plants is find out to be still alive and green till the end of experiment; which clearly shown that all the plant used has the endurance in living in industrial wastewater. In conclusion, these Water Hyacinth, Water Lettuce and Dotted Duckweed have the potential to be commercialized as a phytoremediator for treatment of wastewater as they have the endurance to grow in wastewater. Along the observation there was no associate production of some morphological symptoms of toxicity such as yellowing of leaves, growth retardation and chlorosis. Therefore, these plants are determined to be able to grow and have endurance towards wastewater for phytoremediation.

These three plants have many features that result in a high potential for environmental cleanup. Energy costs and expenses are reduced and natural resources are conserved because plants use solar energy. Plants are adapted to a wide range of environmental conditions of the environment to some extent. The unique enzyme and protein systems of some plant species appear to be beneficial for phytoremediation. As a result of these advantages, phytoremediation has considerable potential for environmental restoration of contaminated sites.

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