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Research article

Nutrient balancing for phytoremediation enhancement of urea manufacturing raw wastewater

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ABSTRACT

Application of urea manufacturing wastewater to teak (*Tectona grandis*) trees, a fast growing tropical timber plants, is an environmentally-friendly and cost-effective alternative for treatment of nitrogen-rich wastewater. However, the plant growth is strongly limited by lack of phosphorus (P) and potassium (K) elements when the plants are irrigated with wastewater containing high concentration of nitrogen (N). A greenhouse experiment was conducted to optimize the efficiency of teak-based remediation systems in terms of nutrient balance. Twelve test solutions consisted of 4 levels of P (95, 190, 570, 1140 mgL^{-1}) and 3 levels of K (95, 190, 570 mgL^{-1}) with a constant level of N (190 mgL^{-1}) were applied to teak seedlings every four days during the study period. Evapotranspiration rate, nutrient removal percentage, leaf surface area, dry weight and nutrient contents of experimental plants were determined and compared with those grown in control solution containing only N (N:P:K = 1:0:0). Teak seedlings grown in units with 1:0.5:1 N:P:K ratio were highly effective at nutrient removal upto 47%, 48% and 49% for N, P and K, respectively. Removal efficiency of teak plants grown in other experimental units decreased with increasing P and K concentrations in test solutions. The lowest nutrient removal and plant growth were recorded in units with 1:6:0.5 N:P:K ratio which received the highest ratio of P to K. The findings indicated that teak seedlings functioned effectively as phytoremediation plants for N-rich wastewater treatment when they were being supplied with proper concentrations of P and K.

1. Introduction

Urea fertilizer factory is one of the largest sources of nitrogen (N) pollution in water with high production rate of urea and ammonium nitrogen (Obire et al., 2008). In water and soil ecosystems, urea is quickly hydrolyzed to ammonia and ammonia is further converted to nitrate through nitrification (Islam et al., 2010). Both ammonia and nitrate are highly toxic to living organisms and causes serious damage to water bodies through eutrophication (Beutel et al., 2009; Leaković et al., 2000).

Phytoremediation, the use of natural ability of plants to absorb pollutants from wastewater, has been known as a low cost and eco-friendly remediation technique (Li et al., 2007). Excess amount of N in wastewaters can be incorporated to plant biomass and increase the plant growth (Holm and Heinsoo, 2013). A variety of plant species have been investigated for removal of N. For that purpose, plants with high transpiration capacity and commercial benefits are top candidates (Dimitriou and Rosenqvist, 2011; Holm and Heinsoo, 2013; Konnerup et al., 2009; Polomski et al., 2007). For example, Heliconia ornamental plants (*Heliconia psittacorum* L.f.) have been reported for removal of 13% of N when they were applied for treatment of domestic wastewater containing 27 mgL^{-1} N (Konnerup et al., 2009). Papyrus (*Cyperus papyrus* L.) which is used in fiber industries was able to remove 74% N from pond waters polluted with 20 mgL^{-1} N (Abe et al., 1999). *Salix* spp. that has economic potential in bioenergy industry was able to remove 41% of N from domestic wastewater containing 56 mgL^{-1} N (Kowalik and Randerson, 1994). Among available alternatives, timber plants with rapid growth, deep roots and high rates of transpiration can be applied efficiently as phytoremediators (Konnerup et al., 2009). Teak (*Tectona grandis*), a woody plant, can be considered as a suitable phytoremediator (Mary Agbogidi et al., 2007). Teak is fast growing tropical plant in *Lamiaceae* family (Krishna and Jayakumaran, 2010). This broadleaf plant produces high amounts

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of biomass and uses high rates of water and nutrients which make it suitable for nutrient phytoremediation (Anish et al., 2015). Application of teak plants as phytoremediators has advantages of attaining high nitrogen and wastewater removal (Yavari et al., 2017). However, plants need complete supply of nutrients to function properly in phytoremediation systems (Malvi, 2011). Nitrogen-enriched wastewaters with extreme levels of N and insufficient supply of other elements (mainly phosphorus, -P, and potassium, -K) cause plant nutrient deficiency and hindering plant growth and remediation efficiency (Polomski et al., 2007). Hence, it is necessary to modify nutrient availability in N-rich wastewaters to enhance productivity of plants and success of phytoremediation (Polomski et al., 2007). Several experiments signified nutrients supplied balance (N:P or N:K) in phytoremediation processes (Fujita et al., 2010; Güsewell, 2004; Lawniczak et al., 2009; Tessier and Raynal, 2003). Nevertheless, stoichiometry examinations in plant-based remediation studies have dealt only with wetland species including grasses, forbs or graminoids (Fraser et al., 2004; Lawniczak et al., 2009). Effect of N:P:K supply ratios on nutrient recovery potential of woody plants is not well understood. Therefore, this study aimed to determine the appropriate balance of P and K in high N urea manufacturing raw wastewaters to enhance remediation efficiency of teak seedling.

2. Materials and methods

2.1. Wastewater collection and analyses

Urea plant wastewater was collected from a local fertilizer plant. Wastewater samples were kept under 4 °C before execution of analysis and were analyzed for pH using a portable pH meter (EW 53013, HACHSension), total N using HACH Test'N Tube tests (HACH, Method 10072), orthophosphate (PO_4^{3-}) by PhosVer 3 Method (HACH, Method 8190) and potassium (K), calcium (Ca), magnesium (Mg), iron (Fe), zinc (Zn), copper (Cu) and manganese (Mn) by atomic absorption spectroscopy, AAS (Model AA 6800 Shimadzu). Wastewater characteristics are presented in Table 1.

2.2. Experimental units

Experimental units were set up to simulate soilless subsurface constructed wetland. Two polyethylene pots of different sizes were fitted by overlapping the rips. The smaller one (3.2 L) with drainage holes was filled with pea gravel and placed into the bigger one (8 L) filled with test solutions. In this configuration, recording the remaining volume of solution was possible with no damage to plants'roots.

2.3. Plants and experimental procedures

Seven-month old teak seedlings were obtained from Mata Ayer Research Stations, Perlis- Forest Research Institute Malaysia (FRIM). In order to acclimatize the plants to new conditions, they were removed from soil substrates and planted into the experimental units filled with 40% modified Hoagland solution one month before starting the experiment. The Hoagland solution provides all necessary nutrients for plant growth (Appendix 1) (Hoagland and Arnon, 1950).

Test solutions were prepared using tap water supply containing negligible nutrients (Table 1) and adding > 98% purified fertilizers; KH_2PO_4, $Ca(H_2PO_4)_2 \cdot H_2O$, NH_4NO_3, $Ca(NO_3)_2$ and KCl. Twelve nutrient solutions were formulated by combining 4 levels of P (95, 190, 570, 1140 mgL^{-1}) and 3 levels of K (95, 190, 570 mgL^{-1}) with a constant level of N (the average concentration of N in urea manufacturing effluent) to have a wide range of P and K ratios to N (Table 2). These concentrations were chosen based on suitable N:P:K ratios reported previously (Abod and Siddiqui, 2002; Anish et al., 2015). One additional null or control nutrient solution (with no P and K) were applied in two different kinds of experimental units; vegetated units for determining P and K effects and unvegetated units for transpiration measurements. All nutrient solutions contained other essential elements in constant non-limiting amounts (200 mgL^{-1} Ca, 48 mgL^{-1} Mg, 64 mgL^{-1} S, 1 mgL^{-1} Fe, 0.05 mgL^{-1} Zn, 0.02 mgL^{-1} Cu, 0.5 mgL^{-1} Mn, 0.01 mgL^{-1} Mo and 0.5 mgL^{-1} B). pH was maintained at 6.0–6.5 with 2 N H_2SO_4 or 10 N NaOH. The obtained fourteen nutrient solutions were replicated three times for a total of 42 experimental units. One teak seedling was planted in each experimental unit. Placement of experimental units was in completely randomized design (CRD). They were placed in an open greenhouse under a transparent rain shelter with ambient conditions. Maximum and minimum daily temperatures and relative humidity were recorded around 30.8 ± 0.3 °C, 23.7 ± 0.09 °C and 85 ± 1.6%, respectively.

At day 0, every unit was drained and flushed with tap water to remove remaining nutrients. Then, they were filled with 3800 mL of the nutrient solutions to keep a constant water level at 10 cm below the gravel surface. The experimental units were re-filled with fresh test solution every four days during the two-month experiment. Four-day of batch-loading was chosen based on a preliminarily experiment. The chosen period was maximum duration in which the plants uptake water freely without facing to any shortage of water in the containers.

2.4. Measurements

Remaining test solutions were collected from each experimental set every four days during the two-month study period. The volumes were recorded to calculate evapotranspiration rate and sampled for total N, PO_4^{3-} and K measurement using HACH Test'N Tube tests (HACH, Method 10072), PhosVer 3 Method (HACH, Method 8190) and AAS (Model AA 6800 Shimadzu), respectively. N, P and K removal percentage by plants were determined using Equation (1) (Polomski et al., 2007).

$$R = \left[\frac{\left(C_i - C_f\right)}{C_i}\right] \times 100 \quad (1)$$

where C_i and C_f are initial and final concentrations of N, P or K and R is percentage of N, P or K removal by plants.

The plants were harvested after two months of treatment with nutrient solutions. Harvested plants were transferred to laboratory and separated into leaves, stems and roots. Total leaf surface area of each plant was measured through a graph paper method described

Table 1
Composition of untreated discharge from a local fertilizer plant in Malaysia and composition of domestic water used in test solution.

Constituent	pH	Electrical Conductivity (μS cm^{-1})	Macronutrients (mgL^{-1})						Micronutrients (mgL^{-1})			
			TN	P	K	Ca	Mg	S	Fe	Zn	Cu	Mn
Urea manufacturing wastewater	9.3	163.11	190.34	1.32	1.81	29.57	3.93	6.40	0.35	0.05	0.04	0.02
Domestic water	7	65.90	0.80	0.31	0.88	5.60	8.45	1.89	0.04	0.03	0.03	0.01