

Mineralization of Urea-Formaldehyde Fertilizer and Its Availability to Oil Palm Seedling under the Tropical Environment

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Abstract

Easily dissolved fertilizers release nutrient in excess amount to be assimilated by plant roots. Some portions these fertilizers leach out from the root zone and adversely impact the environment. Controlled-release fertilizers are more favorable to reduce fertilizer loss, labor cost, and environmental impact. Urea-formaldehyde (UF) was synthesized by polymerization of urea and 40% formaldehyde solution using H_3PO_4 as a catalyst. Three mole ratios of urea:formaldehyde, namely, 1.0:1.0, 1.5:1.0 and 2.0:1.0 were synthesized. Mineralization of the UF was conducted using eight different mixtures, four different moisture, and four incubation periods. The experiment included soil alone, soil with compost, soil with UF (1.0:1.0), soil with UF (1.5:1.0), soil with UF (2.0:1.0), soil with UF (1.0:1.0) and compost, soil with UF (1.5:1.0) and compost, and soil with UF (2.0:1.0) and compost. Moisture of the mixtures was adjusted to 20%, 40% 60% and 80% water holding capacity (WHC) of the soil. The mixtures were incubated at room temperature for 1, 2, 4 and 8 weeks, the released NH_4^+ and NO_3^- were extracted by 1 M KCl and analyzed via a distillation method. Rates of mineralization increased with mole ratio of urea and moisture content of the soil. N loss increased with the moisture content. The best performance for the compromised condition was a mole ratio less than 1.5:1.0 (urea:formaldehyde) at 60% WHC. Availability of UF and serpentine-phosphate for oil palm seedlings was conducted using 10 treatments. The experiment consisted of soil without amendment; soil with Multicote; soil with cow manure; soil with cow manure and urea; soil with cow manure and UF (1.0:1.0); soil with cow manure and UF (1.5:1.0); soil with cow manure, $MgHPO_4$ and UF (1.0:1.0); soil with cow manure, $MgHPO_4$ and UF (1.5:1.0); soil with cow manure, serpentine-phosphate and UF (1.0:1.0); and soil with cow manure, serpentine-phosphate and UF (1.5:1.0). All amended soils increased vegetative growth of oil palm seedlings compared with the non-amended soils. Urea and UF increased the N content in seedling leaves, while Multicote, cow manure, $MgHPO_4$, and serpentine-phosphate increased the Mg content. The best performance was found in the combination of cow manure, serpentine-phosphate and the UF with mole ratio of 1.0:1.0.

Keywords: tropical soil, nitrogen fertilization, controlled-release fertilizer, slow release fertilizer

1. Introduction

Urea is the most commonly applied for N-source fertilizers. However, urea is substantially lost through leaching, runoff and volatilization (Yan & Zheng-yin, 2007; Jantalia, 2012; Yamamoto et al., 2016). The efficiency of N application for cereal production worldwide was estimated to be only 33% (Raun & Johnson, 1999). The N losses from agricultural lands contribute to ground water contamination, eutrophication and emission of greenhouse effect gases (Ju et al., 2009; Jantalia et al., 2012; Geng et al., 2015). Loss of urea has been manipulated by various methods such as incorporated with nitrification inhibitors or urease inhibitors, coating with synthetic or natural polymers, and chemically modified urea molecules (Trenkel, 2010; Azeem et al., 2014; Rychter et al., 2016).

Controlled-release fertilizers (CRF) are beneficial to reduce environmental pollution, cost of fertilizer application and improve the efficiency of fertilizer application. Geng et al. (2015) found that application of controlled-release urea at a rate of 70% of urea produced equal yields of rice and rape seed compared with those

of the urea. Single application of controlled-release urea as a basal fertilizer could achieve higher yield than split application of urea. Yen and Zheng-yin (2007) found that the N-use efficiency, N-agronomy efficiency and N-physiological efficiency increased by 11.4%, 8.32 kg kg⁻¹ and 5.17 kg kg⁻¹ respectively, for rice when a CRF was applied. Wada, Aragonés and Ando (1991) reported that the application of controlled-release urea with only half of ammonium sulfate can produce better yield and increase N in rice to almost an equal level. Oil palm (*Elaeis guineensis*) is an economic crop of Southeast Asian countries. It has an average potential yield of 33.2 tons fresh fruit bunches (FFB) ton⁻¹ during productive phase, and having the maximum of 40.4 tons FFB ton⁻¹ at 10 years after transplanting (Enler, Hoffmann, Fathoni, & Schwarze, 2016). However, average actual yields stagnated at around 17 to 19 tons FFB ton⁻¹. Enler, Hoffmann, Fathoni and Schwarze (2016) found that fertilization during productive phase has strongly affected on yield performance. Donough et al. (2006) reported that oil palm yields in Malaysia increased rapidly after nutrient status in the soil was improved. Therefore, fertilization is a key factor to reduce yield gap of the oil palm. General recommendation of fertilizer rate is larger than 1.2 tons ha⁻¹ to achieve high yield for oil palm having age more than 10 years after transplanting. A large amount of fertilizer loss can be expected in tropical climate, hence CRF should be introduced. Oil palm seedlings have to be cultivated in a nursery around 1 year, and fertilizers have to be applied twice a month. The CRF is also a good alternative for saving labor cost and reducing fertilizer loss. However, information on using of CRFs especially Urea-formaldehyde polymer (UF) for oil palm is still scarce.

UF has been used as a controlled release fertilizer for many decades. However, this polymer is not widely used in Southeast Asia. UF is synthesized by reacting urea (NH₂CONH₂) with formaldehyde solution by using H₃PO₄ or phosphate compounds as catalyst. Paraformaldehyde (short-chain polymerized formaldehyde) may be used instead of formaldehyde solution for reduction of water content in the reaction and product (Yamamoto et al., 2016). Typical UF products contain N between 37% and 40% (Alexander & Helm, 1990). Mineralization of UF depends on mole ratio of urea and formaldehyde, soil pH, soil moisture, temperature and microbial activities. The rate of nitrate production from UF is higher in acidic soil than in neutral soil. The released N may not sufficient to plants if the soil temperature is below 15 °C (Basaraba, 1963; Jahns & Kaltwasser, 2000; Guo, Liu, Liang & Niu, 2006). UF supplies only N, therefore other nutrients have to be supplied separately. Similar to the UF, serpentine (Mg₃Si₂O₅(OH)₄) has long been used for Mg source. However, Mg availability of this mineral is very low. The availability can be improved by an acidulation process. Reacting of Serpentine mineral with H₃PO₄ produces Serpentine-phosphate, which can be used as a source of both Mg and P (Nakamura, Yamazoe, & Kishimoto, 1956; Hanly, Loganathan, & Currie, 2005).

The objectives of this study were to examine mineralization of the UF under different mole ratios, soil moisture, and supplement with other sources of nutrients, and respond of oil palm seedling to these combinations.

2. Materials and Methods

2.1 Mineralization of UF

UF was synthesized using 1.0:1.0 (UF01), 1.5:1.0 (UF02) and 2.0:1.0 (UF03) mole ratios of urea: formaldehyde and 2 mL L⁻¹ of phosphoric acid (H₃PO₄) as catalyst. After the reaction was completed, the synthesized UFs were dried in an oven at 80 °C and ground through a 2 mm sieve. A sample of surface soil (0 to 15 cm) was taken from the Walailak University farm (latitude 8°38.145' N longitude 99°54.090' E). The soil was classified as fine, kaolinitic, isohyperthermic Typic Endoaquults. The soil sample was remixed and air-dried, gravel and debris were removed. The sample was ground and sieved through a 2 mm screen for the chemical analysis. Soil pH and electrical conductivity (EC) were measured using 1:2.5 and 1:5 of soil: water ratios, respectively. The EC at saturation point (ECe) was estimated by multiplying the EC with 6 (Shaw, 1999). Soil organic matter was analyzed using the Walkley and Black method. Exchangeable K, Ca and Mg were analyzed via atomic absorption spectrophotometry (Jones, 2001, 2003). Available P was analyzed using the molybdenum blue method (Jones, 2001, 2003). Available S was analyzed using a turbidimetric method (Jones, 2001, 2003). The microelements, namely, Fe, Mn, Zn and Cu were analyzed via atomic absorption spectrophotometry (Jones, 2001, 2003). A factorial design with 8 main-plots and 4 × 4 sub-plots was employed (Table 1). The soil (20 g) was placed into a 250 mL erlenmeyer flask, mixed with or without 2 g of chicken compost (2.32% total N, 4.86% P₂O₅, 2.90% K₂O, 3.37% Ca, 1.08% Mg), and mixed with or without 0.2 g of the UFs. Water was added into the flasks for adjusting soil moisture to 20%, 40%, 60% and 80% of the water holding capacity (WHC). The mixtures were made to stand at room temperature for 1, 2, 4 or 8 weeks. At the end of the incubation period, 50 mL of 1 M KCl was added, and the mixture was shaken for 30 minutes by a reciprocal shaker at 120 rpm. The suspension was filtered and leached with deionized water, until 100 mL of solution was obtained. The solution was analyzed for ammonium (NH₄⁺) and nitrate (NO₃⁻) by a distillation method (Mulvaney, 1996). Statistical analysis was performed using Duncan's Multiple Range test (DMRT).

Table 1. Arrangement of the mineralization experiment, each main-plot was split into 20%, 40%, 60% and 80% WHC, and incubation for 1, 2, 4 and 8 weeks

Treatments	Description
T1	Soil (20 g)
T2	Soil (20 g) + compost (2 g)
T3	Soil (20 g) + UF01 (0.2 g)
T4	Soil (20 g) + UF02 (0.2 g)
T5	Soil (20 g) + UF03 (0.2 g)
T6	Soil (20 g) + compost (2 g) + UF01 (0.2 g)
T7	Soil (20 g) + compost (2 g) + UF02 (0.2 g)
T8	Soil (20 g) + compost (2 g) + UF03 (0.2 g)

2.2 Pot Experiment

A soil sample was taken from the same place of the previous experiment (section 2.1). A completely randomized design (CRD) with 10 treatments and 5 replications was employed. A total of 8 kg of the air-dried soil was placed into a plastic pot with a diameter of 25 cm. Deionized water was added to increase the soil moisture to 65% of WHC. Fertilizers and cow manure were mixed with the soil separately to prepare 10 treatments: soil; soil with Multicote (19-10-13+1.5MgO, Haifa CRF fertilizer); soil with cow manure; soil with cow manure and urea; soil with cow manure and UF01; soil with cow manure and UF02; soil with cow manure, UF01 and MgHPO₄; soil with cow manure, UF02 and MgHPO₄; soil with cow manure, UF01 and Serpentine-phosphate; soil with cow manure, UF02 and Serpentine-phosphate (Table 2). Oil palm seedlings (90 days after germination) were transplanted into treatment pots (one seedling per pot). The experiment was carried out in a plastic-sheet covered house for 38 weeks. At the end of experiment; plant height was measured, number of fully expanded leaves were counted, and upper-ground of the seedlings were cut and their dry weight were measured. The leaf samples were collected, gently wiped with a clean cloth, and packed into marked paper bags. The samples were dried at 70 °C for 72 hours, ground and passed through a 1 mm sieve and then kept in plastic container to analyze the macronutrient and micronutrient elements. Prior to the analysis, the samples were re-dried at 70 °C for 2 hours and cooled down in a desiccator. Total-N was analyzed using the Kjeldahl method (Jones, 2001), A portion of the sample was digested by HNO₃:HClO₄ (2:1) mixture. The P concentration was analyzed using the Vanadomolybdate method, the K concentration using flame emission spectrophotometry, and Ca, Mg concentrations using atomic absorption spectrophotometry (Jones, 2001). Statistical analysis was performed using Duncan's Multiple Range test.

Table 2. Arrangement of the pot experiment, each treatment consisted of 5 replications

Treatments	Description
T1	Soil (8 kg)
T2	Soil (8 kg) + Multicote (10.3 g)
T3	Soil (8 kg) + cow manure (1 kg)
T4	Soil (8 kg) + cow manure (1 kg) + Urea (4.3 g)
T5	Soil (8 kg) + cow manure (1 kg) + UF01 (5.6 g)
T6	Soil (8 kg) + cow manure (1 kg) + UF02 (5.6 g)
T7	Soil (8 kg) + cow manure (1 kg) + UF01 (5.6 g) + MgHPO ₄ (4.6 g)
T8	Soil (8 kg) + cow manure (1 kg) + UF02 (5.6 g) + MgHPO ₄ (4.6 g)
T9	Soil (8 kg) + cow manure (1 kg) + UF01 (5.6 g) + Serpentine-P (4.6 g)
T10	Soil (8 kg) + cow manure (1 kg) + UF02 (5.6 g) + Serpentine-P (4.6 g)

3. Results and Discussion

3.1 Soil Chemical Properties

The chemical properties of the soil sample are listed in Table 3. The soil was acid and non-saline. The soil also contained low organic matter and low concentrations of P, K, Ca, Mg, S and Cu. In addition, extractable Fe and Mn were high, and Zn was in a medium range (Jones, 2001, 2003). The properties indicated that acidity and salinity of the soil do not inhibit microbial activities. However, multiple nutrients have to be amended to improve microbial activities and plant growth.

Table 3. Chemical properties and nutrient concentrations of the soil sample taken from the depth of 0 to 15 cm

Property	Unit	Value (mean±SD)
pH (1:2.5 in water)		5.42±0.01
ECe (Saturation)	mS cm ⁻¹	0.51±0.03
OM (Walkley & Black)	g kg ⁻¹	17.8±0.5
P (BrayII)	mg kg ⁻¹	11.5±0.4
S (0.01 M KH ₂ PO ₄)	mg kg ⁻¹	0.58±0.05
K (1 M NH ₄ OAc pH 7)	mg kg ⁻¹	61.7±0.1
Mg (1 M NH ₄ OAc pH 7)	mg kg ⁻¹	41.7±0.9
Ca (1 M NH ₄ OAc pH 7)	mg kg ⁻¹	423±4
Fe (DTPA extractable)	mg kg ⁻¹	85±4
Mn (DTPA extractable)	mg kg ⁻¹	30.8±0.4
Zn (DTPA extractable)	mg kg ⁻¹	1.13±0.17
Cu (DTPA extractable)	mg kg ⁻¹	0.29±0.04

3.2 Mineralization of UF

The soil sample released a small amount of N from the mineralization of the organic matter in the soil. The soil contained 17.8 g kg⁻¹ of organic matter (approximately 1 g kg⁻¹ of nitrogen), resulting in the mineralization of N. The soil nitrogen was mineralized to NH₄⁺ and then partially transformed to NO₃⁻. The total amounts of NH₄⁺ and NO₃⁻ increased with prolonged incubation periods, that is from 30±3 mg-N kg⁻¹ on the first week to 80±13 mg-N kg⁻¹ on the eighth week. NH₄⁺ was dominant on the first week, but the ratios of NH₄⁺/NO₃⁻ declined afterward. Nitrification at higher moisture content progressed faster than at lower level. On the eighth week, 49%, 60%, 88% and 93% of inorganic N presented in NO₃⁻ form at 20%, 40%, 60% and 80% WHC, respectively (Figure 1).

Soil amendment with compost released more NH₄⁺ and NO₃⁻ than the soil alone (Figure 1). However, the total amounts slightly increased with prolonged incubation periods. The results indicated that most of the mineralizable N from the compost was released on the first week. The ratios of NO₃⁻ increased with prolonged incubation period and increased moisture content. On the eighth week, NO₃⁻ was dominated for 40%, 60% and 80% WHC, whereas NH₄⁺ was dominated at 20% WHC. The results indicated that nitrification progress faster under higher moisture condition.

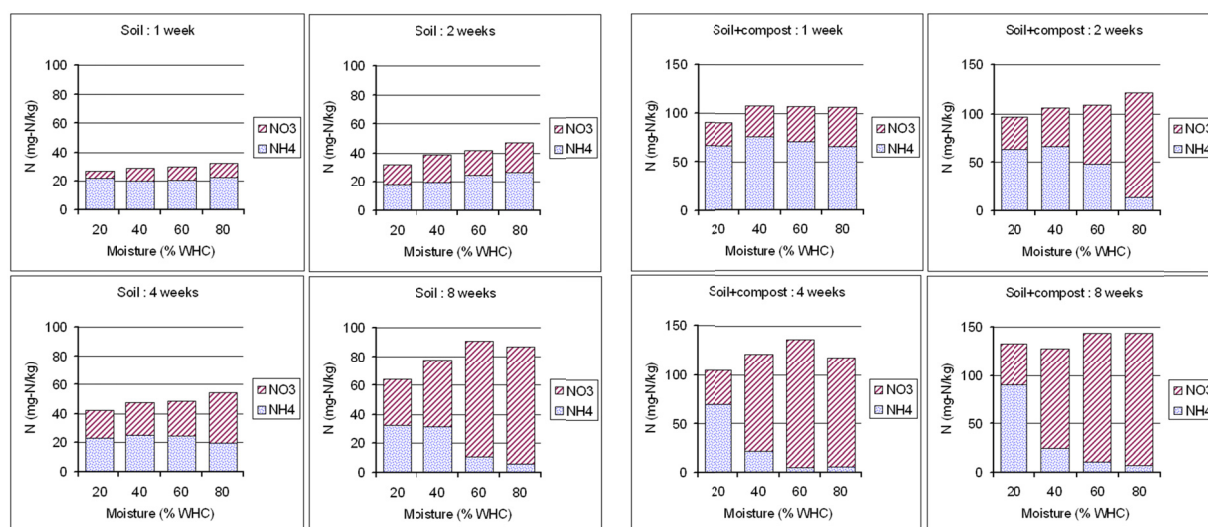


Figure 1. Releasing of NH₄⁺ and NO₃⁻ from the unamended soil (left) and soil amended with compost (right) at different moisture contents and incubation periods

The release of NH₄⁺ and NO₃⁻ from the soil amended with UF01 was slower than that of the soil with compost. The soil with UF01 released 49, 59, 54, and 74 mg-N kg⁻¹ of NH₄⁺ and NO₃⁻ at 20%, 40% 60% and 80% WHC, respectively on the first week; whereas the soil with compost released 50, 108, 107, and 106 mg-N kg⁻¹ on the

same condition (Figure 2). The amounts of NH_4^+ and NO_3^- from the UF01 gradually increased with the incubation period, but no increase was observed from the mineralization of compost after 2 weeks (Figure 1). The release of NH_4^+ and NO_3^- tended to increase with moisture content. NH_4^+ and NO_3^- were released only $92 \pm 4 \text{ mg-N kg}^{-1}$ at 20% WHC, whereas these compounds released as much as $200 \pm 2 \text{ mg-N kg}^{-1}$ at 80% WHC on the eighth week. The results indicated that mineralization rate and the nitrification progressed of UF01 was slower than that of the soil with compost. The UF01 is a source of N alone; therefore, microbial growth is limited due to lack of other nutrients.

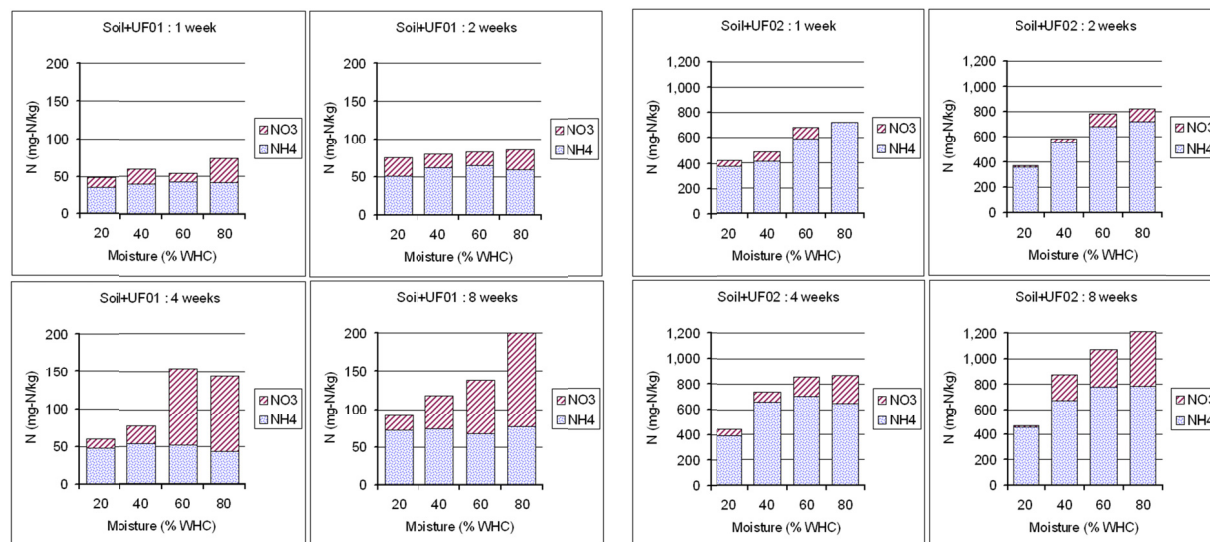


Figure 2. NH_4^+ and NO_3^- released from the soil amended with UF01 (left) and soil with UF02 (right) at different moisture contents and incubation periods

UF02 released NH_4^+ and NO_3^- faster than UF01 (Figure 2). The amount of released NH_4^+ and NO_3^- was $712 \pm 82 \text{ mg-N kg}^{-1}$ on the first week at 80% WHC, whereas $74 \pm 4 \text{ mg-N kg}^{-1}$ was released from UF01. Higher mole ratio of urea-to-formaldehyde produced UF polymer that can be more easily mineralized. The ratios of NH_4^+ were higher than those of NO_3^- for all moisture content. The results indicated that the release rate of N was faster than nitrification. Mineralization of the UF02 occurred rapidly and may be too fast to be uptake by plants.

The release of NH_4^+ and NO_3^- from the soil amendment with UF03 increased with moisture content on the first and second weeks but tended to decline afterward, especially at 80% WHC (Figure 3). The result indicated that denitrification occurred at high moisture content and extended incubation period. Most of the released N existed in NH_4^+ form for all moisture contents and incubation periods. Similar to UF02, the release rate of N was faster than nitrification. The UF03 may not suitable for using as a controlled release fertilizer.

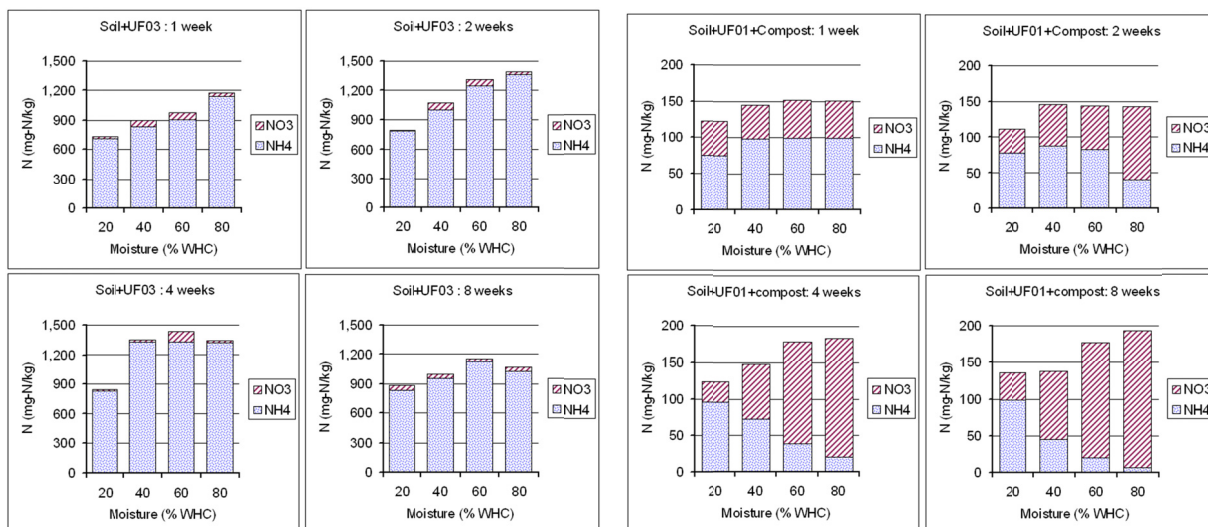


Figure 3. NH_4^+ and NO_3^- released from soil amended with UF03 (left) and soil with UF01 and compost (right) at different moisture contents and incubation periods

The mixture of UF01 and compost released a larger amount of N than UF01 alone, because a large portion of N originated from the compost. The ratios of NO_3^- increased with prolonged incubation period and moisture content. On the eighth week, the NO_3^- was released by $27.6 \pm 4.0\%$, $66.3 \pm 7.3\%$, $88.9 \pm 3.4\%$ and $96.4 \pm 0.8\%$ at 20%, 40%, 60% and 80% WHC, respectively (Figure 3). Addition of compost enhanced the nitrification in the case of UF01. The mixtures of UF02 and UF03 with compost released N similar to that of sole UF02 or sole UF03, and most of the N was in NH_4^+ form (Figure 4). Addition of compost did not significantly accelerate the nitrification in the cases of UF02 and UF03. This phenomenon may due to the very fast release of NH_4^+ and high concentration of NH_4^+ accumulated in the soil.

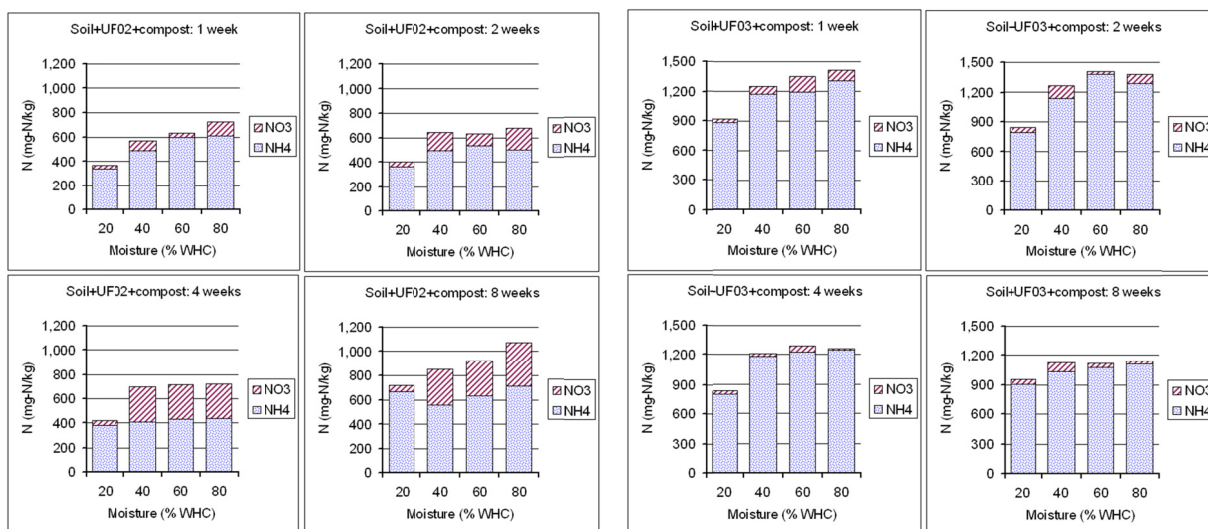


Figure 4. NH_4^+ and NO_3^- released from the soil amended with UF02 and compost (left) and soil with UF03 and compost (right) at different moisture contents and incubation periods

3.3 Pot Experiment

Growth indexes were measured at the end of experiment. Soil amended with cow manure, UF01 and serpentine-phosphate (T9) showed the highest plant height, number of leaves and shoot biomass (Figure 5). The results indicated that the oil palm seedling required additional N, P and Mg for the experimental soil. Multicote (T2) showed better performance than the soil alone. The amount of Multicote may be too little or its release rate

of nutrient may be slower than plant assimilation. Cow manure (T3) also substantially affected on the growth performance. Urea, UF01 and UF02 did not show significant differences in plant height, number of leaves and shoot biomass (T4, T5 and T6). This phenomenon may be due to the fact that the nursery period was not long. Therefore, the release rate of N did not affect the seedling growth. UF02 tended to show better results than the UF01 when the former was applied in combination with $MgHPO_4$ (T7 and T8). UF01 showed markedly better results than UF02, when the former was applied in combination with serpentine phosphate (T8 and T9).

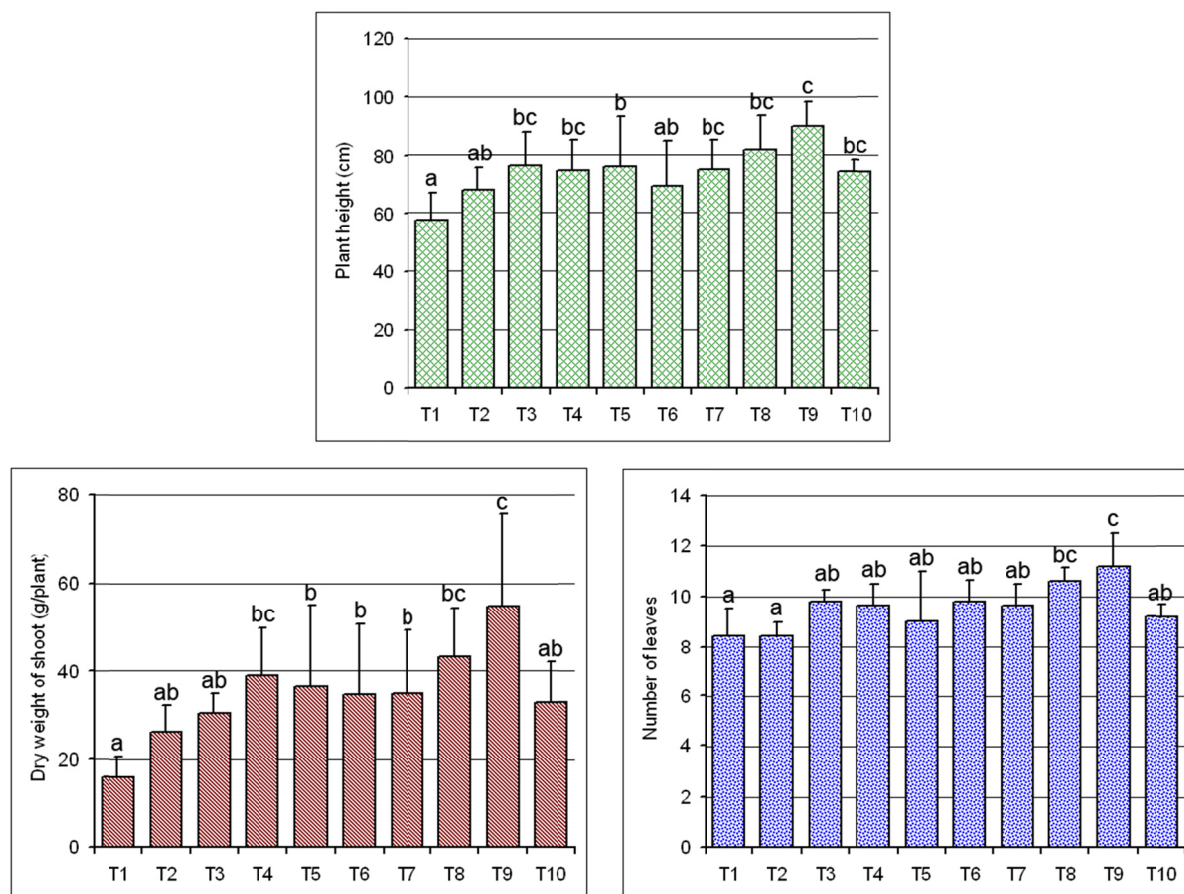


Figure 5. Plant height, number of leaves, and shoot biomass of oil palm seedling at 38 weeks after transplanting
Note. Difference letters within the columns indicate significant difference ($P \leq 0.05$).

3.4 Nutrient Concentrations in the Seedling Leaves

Leaf samples of the oil palm seedlings were collected at the end of experiment and analyzed for N, P and Mg. N in the leaves of the seedling grown in the soil without amendment (T1) was significantly lower than the those treated with N sources (Figure 6). This result indicated that N in the soil was not sufficient for seedling growth. Sufficient N concentration of 2.4% to 3.0% is suggested for oil palm (von Uexkull and Fairhurst, 1991). No significant differences in N were found in the leaves of the seedling grown in soils amended with urea, UF01 and UF02 (T4, T5 and T6). The results indicated that the rates of released N did not affect ability of plant uptake. Addition of P and Mg did not significantly affect N in the seedling leaves (T7, T8, T9 and T10). N is a translocated nutrient. Although, soil concentration declined, the sufficient level is maintained by relocation from the old leaves to the young leaves.

The concentration of available P in the soil was $11.5 \text{ mg-P kg}^{-1}$, which was generally considered as low level (Jones, 2001, 2003). Therefore, addition of P should exhibit a positive effect on plant growth. The P concentration in the leaves of the seedling grown in the soil without amendment (T1) was significantly lower than those in the other treatments (Figure 6). The result agreed well with the soil analysis. Sufficient leaf-P concentration of 0.15% to 0.19% is suggested for oil palm (von Uexkull and Fairhurst, 1991). The addition of

Multicote tended to increase P, but did not reach the sufficient level. This phenomenon may be due to the fact that the added amount was not sufficient or its release rate was too slow for the oil palm seedling. Addition of $MgHPO_4$ and serpentine-phosphate did not significantly increase the P concentrations more than that when cow manure was added. The results indicated that oil palm did not uptake P higher than the sufficient level. The highest P concentration was found in the seedling grown in the soil amended with cow manure and UF01 (T5). Slow N release of the UF01 tended to enhance P uptake.

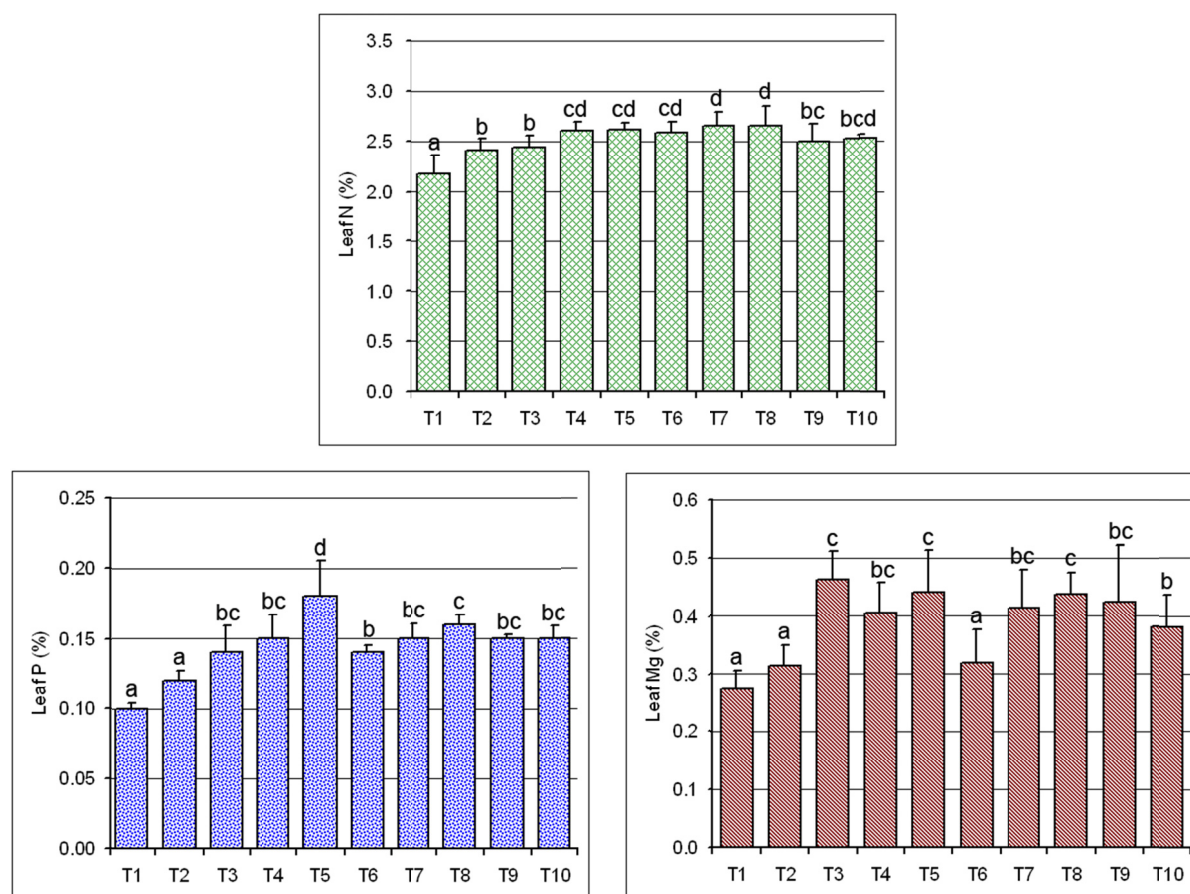


Figure 6. N, P and Mg contents in the leaves of oil palm seedling at the end of experiment

Note. Difference letters within the columns indicate significant difference ($P \leq 0.05$).

The concentration of extractable Mg in the soil was 41.7 mg kg^{-1} , which was generally considered as very low (Jones, 2001, 2003). Therefore, the amount of Mg in the soil was not sufficient for seedling growth. Plants require 1 mole of Mg for every 4 moles of N for chlorophyll synthesis. Therefore, N and Mg should be supplied in balance of mole ratio. Mg in the leaves of the seedling grown in the soil without amendment (T1) was significantly lower than those in the other treatments, except in T6 (Figure 6). Von Uexkull and Fairhurst (1991) suggested a sufficient range of 0.30% to 0.45% for Mg in oil palm leaves. Mean values of all treatments, except T1, were above the lower limit. Addition of Multicote tended to increase Mg to the lower margin. The addition of $MgHPO_4$ or serpentine-phosphate (T7, T8, T9 and T10) did not increase Mg concentration more than that with cow manure. Therefore, supplying Mg from the manure was sufficient for the seedling.

4. Conclusion

Mineralization of UF fertilizer depends on its reacted mole ratio, soil moisture and microbial substrate. A lower mole ratio of urea to formaldehyde retarded the rate of mineralization. Higher soil moisture and addition of compost enhanced the rate of mineralization. UF firstly mineralized into NH_4^+ and consequently transformed to NO_3^- . Excessive moisture in the soil enhanced the transformation rate. However, a part of N tended to loss from the soil. The mole ratio less than 1.5:1.0 (urea: formaldehyde) and soil moisture between 40 to 60% WHC are

recommended. A low fertile soil amended with cow manure, UF01 and serpentine-phosphate showed the best combination as media of oil palm seedling during nursery phase.

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