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Recovering Phosphorus and Nitrogen from Leachate Collected from Chidaya Sanitary Landfill Dodoma Tanzania for Fertilizer Production

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Abstract

The demand for fertilizer is a global issue that poses a significant threat to future food security. Tanzania is among the countries which rely on agriculture for food production and contribute largely to the economy of the nation. However, farmers incur high production costs due to the utilization of expensive and environmentally insecure fertilizers. The present study investigated the potential of recovering nitrogen and phosphorus nutrients from leachate to produce fertilizer. Hanna pH meter (HI-98121) was used to determine the pH of leachate which was found to be 8.29 ± 0.33. The concentration of phosphorus and nitrogen was determined by using DR 4000 spectrophotometer and was found to be 305.24 ± 16.20 and 189.80 ± 0.165mg/L, respectively. Atomic Absorption Spectrophotometer was used in the determination of magnesium (13.13 ± 0.22 mg/L), potassium (3377 ± 123.4 mg/L), calcium (336 ± 3.65mg/L) and sodium (1270.6 ± 156.90 mg/L). The recovery of the nutrients was done through struvite precipitation, in which 74.01% of phosphorus and 57.27% of nitrogen were recovered under an optimal pH of 9.0 and magnesium-phosphorus molar ratio of 5:1. XRD patterns were analyzed by XRD Rietveld refinement method, for phase quantitative and qualitative analysis. Results show that the precipitates formed were struvite and hydroxyapatite. The elemental composition of struvite was examined by using XRF in which plant nutrients including K, P, Mg, Na and Ca were identified. Production of fertilizer is feasible by using leachate from landfills, thus, to reduce the cost of production, alternative sources of magnesium from locally available materials should be explored.

Keywords: Chidaya landfill; leachate; nitrogen; phosphorus; fertilizer

Introduction

An essential chemical substance that underpins the global food supply is fertilizer, nevertheless, the reduced access to fertilizer threatens future food security. The global fertilizer demand is currently high and is projected to increase annually (FAO, 2019, Lucero-Sorbazo et al. 2022,). The rapid growth of the human population demands and drives farmers to deliver the necessary food supplies thus, chemical fertilizers have been applied extensively to increase the growth and yield of crops in order to meet the food demand (Penuelas et al. 2023). Reports show a global increase in fertilizer demand from 107 to 110 million tonnes in 2011 to 2020, respectively. In Africa, fertilizer demand increased from 5,604 to 7,767 thousand tonnes in 2011 to 2022, respectively (FAO, 2019). Besides, Tanzania's fertilizer demand has been increasing year after year, reaching approximately 698,260 tonnes in 2021/2022 from 435,000 tonnes in 2017/2018.

Tanzania's economy depends on agriculture which accounts for more than one-quarter of the country's gross domestic product (GDP), providing 85% of exports, and about 80% of all employment (Mwigeka 2016). Despite playing a pivotal role, fertilizer availability has become a burning issue among smallholder farmers. Over 90% percent of Tanzania’s fertilizer supply is imported (Mwaijande 2019). Although Tanzania subsidizes fertilizer prices, fertilizers have become less affordable and accessible to smallholder farmers, thus, threatening to jeopardize food production and security further. Therefore, there is a need to adopt more innovative, alternative fertilizer sources, good crop practices and efficient fertilizers with minimal environmental impact.

Nutrient recovery to produce fertilizer from secondary resources has received a lot of attention in recent years (Muys et al. 2021). The increasing population in Tanzanian cities cause the production a lot of waste that is dumped in open spaces and landfills. During rainy season, water percolates in heaps of waste thus producing a liquid known as leachate. Leachate is one of the environmental pollutants that contaminate groundwater, surface waters and soil. As a result, it pollutes the environment thus negatively impacting human health. It is expensive to contain leachate and dispose of it so that it won't harm the environment or people's health. Leachates contain vital plant nutrients such as nitrogen, phosphorus, and potassium which can be recovered to produce struvite. Struvite is a hydrated compound with equimolar concentrations of magnesium, ammonium, and phosphate ($MgNH_4PO_4·6H_2O$). It is formed when magnesium is added in leachate rich in phosphorus and nitrogen at optimal Mg: NH_4^+: PO_4^{3-}molar ratio and pH. The formation of struvite is expressed as: $NH_4^+ + Mg^{2+} + PO_4^{3-} + 6H_2O \rightarrow MgNH_4PO_4 .6H_2O$. The presence of foreign ions such as K, Ca, Na, and heavy metals in leachate may affect the removal efficiency of nutrients, purity, morphology and the size of struvite (Siciliano et al. 2020).

The recovery of nitrogen and phosphorus from leachate through struvite precipitation to produce struvite is not well documented in Tanzanian. Therefore, present work provides the data on the feasibility of recovering phosphorus and nitrogen from landfill leachate found in Tanzania via struvite precipitation for fertilizer production. Results provide the way to; search for alternative source to phosphate rock which is rapidly depleting, increase fertilizer availability, and reduce environmental impacts posed by leachate, preserving natural resources and decrease food insecurity.

Materials and Methods

Materials

The raw leachates were collected from Chidaya Sanitary landfill in Dodoma Tanzania (Figure 1). It is located about 7 km west of the city centre. The landfill has been in operation since 2017. To obtain the homogeneous sample, 20L of the leachate were collected from six different points of the leachate collection pond and packed in high-density polyethylene containers. One litre (1L) of the stock sample was acidified to pH < 2 for heavy metals analysis. The samples were transported in a cool box to the Environmental Engineering laboratory at Ardhi University, Dar es Salaam. The samples were refrigerated at 4 ºC to stabilize heavy metals content and biological activities for further analysis.

Figure 1. A photograph showing the Chidaya Sanitary landfill Leachate's collection pond

Nessler reagent and ascorbic acid were used in the determination of NH_4^+ and PO_4^{3-} in leachate, respectively. Stock standard solutions of Ca, Na, Mg and K metals and such as heavy metals Fe, Pb, Cr, Cd, Ni, Cu, Zn, and Mn were used for calibration. Magnesium chloride crystals were used as the alternative sources of Magnesium. to control pH, 5M NaOH was used. All chemicals used were of an analytical grade bought from Total Med-Lab Solution and Chemical and Scientific Ltd.

Preparation of leachate sample

The collected leachate samples were filtered through watchman filter paper with a pore size of 45 µm to remove particles. For pH measurement, the sample was measured without dilution. The high concentration of PO_4^{3-}, NH_4^+ metals, and heavy metals (Table 2) necessitated diluting 0.1 mL of the leachate with distilled water to a final volume of 100 mL. This was done to lower the concentration to the instrument's detection limit and lessen interference from other substances.

Characterization of leachate

HI 2550 multiparameter analyzer (HANNA Instruments Inc., USA) was used to measure pH and ammonium- nitrogen (NH_4-N and phosphate (PO_4-P) concentration were measured by using DR 4000 spectrophotometer. Heavy metals (Fe, Pb, Cr, Cd, Ni, Cu, Zn, and Mn and metals Ca, Na, Mg and K) were determined by Atomic Absorption Spectrophotometer (AAS).

Determination of optimal conditions for struvite formation

The determination of optimal conditions for struvite formation and collection of precipitates from the reactor were carried out simultaneously. The method was used as described by Warmadewanthi et al. (2021). Twenty-five experiments were conducted in batch mode to determine the optimal pH and the desired Mg^{2+}: PO_4^{3-} molar ratio for precipitates formation. The molar ratios were selected on purpose to capture all of the PO_4^{3-} content in the raw leachate as presented in Table 1.

Table 1. Magnesium and phosphate molar ratio and pH conditions for maximum recovery efficiency of PO_4^{3+} and NH_4^+ from leachates.

Test	**Concentration of Mg $^{2+}$ (mg/L)**	**pH**	**Concentration of PO_4^{3+} and NH_4^+ (mg/L) before and after the test**				**Mole ratio Mg^{2+}:PO_4^{3+}**
			PO_4^{3+}		**NH_4^+**		
			Initial	**Final**	**Initial**	**Final**	
1	653	8.5	305	257	188	146	1:1
2		9.0	305	228	188	139	1:1
3		9.5	305	245	188	111	1:1
4		10.0	305	247	188	105	1:1
5		10.5	305	256	188	104	1:1
6	1237	8.5	289	201	175	101	2:1
7		9.0	289	143	175	93.1	2:1
8		9.5	289	145	175	87.6	2:1
9		10.0	289	220	175	108	2:1
10		10.5	289	226	175	105	2:1
11	1990	8.5	310	191	200	117	3:1
12		9.0	310	186	200	99	3:1
13		9.5	310	185	200	109	3:1
14		10.0	310	206	200	111	3:1
15		10.5	310	224	200	91	3:1
16	2953	8.5	345	254	200	152	4:1
17		9.0	345	123	191	116	4:1
18		9.5	345	186	191	108	4:1
19		10.0	345	150	191	86	4:1
20		10.5	345	148	191	83	4:1
21	2964	8.5	277	142	195	159	5:1
22		9.0	277	72	195	84	5:1
23		9.5	277	83	195	109	5:1
24		10.0	277	117	195	85	5:1
25		10.5	277	151	195	97	5:1

Recovery efficiency of NH_4^+ and PO_4^{3-} from leachate

Efficiency of phosphate and ammonium removal from landfill leachate was used as an indicator for struvite formation. It was determined by comparing the differences in concentration of phosphorus and ammonium before and after struvite precipitation as described previously by (Jabr et al. 2019) using equation 1.

$$\text{Recovery Efficiency (RE)} = \frac{C_b - C_a}{C_b} \times 100 \quad (1)$$

Where C_b is the concentration of phosphate or ammonium ions before struvite precipitation and C_a is the concentration phosphate or ammonium ions after struvite precipitation. The recovery efficiency was calculated for each pH value and mole ratio tested to determine the maximum recovery of both phosphate and ammonium in raw leachate.

Precipitate characterization

Bruker D2 Advance X-ray diffractometer was used to obtain XRD data from precipitates collected from five different Mg: PO_4^{3-} molar ratios using monochromatized Cu-Kα radiation from $2\theta = 0°$ to $70°$. Origin Pro 2019 b 64-bit and match software were used to analyze the collected data. Further, X-ray fluorescence was used to determine the elemental composition of precipitates collected precipitates.

Results and Discussion

Physicochemical properties of leachate

The results on the physicochemical properties of the leachate studied are presented in Table 2. The pH of leachate was found to be 8.62 ± 0.33 indicating that it is naturally alkaline, which is attributed to a high content of alkali and alkali earth metals. Alkaline condition favours struvite formation, thus avoiding the cost of adding commercial chemicals as pH controllers. 0.5L of the leachate required only 5-9 drops of 5M NaOH to change the pH value from 8.5 to 10.5.

Tables 2: Physicochemical properties of landfill leachate collected from Chidaya landfill

Parameter	Concentration	Acceptable ranges (TBS (2006)
NH_4-N	189.80 ± 74.44	≤30
PO_4-P	305.24 ± 16.20	≤10
Mg	13.13 ± 0.22	≤29
K	3377±123.4	NA
Na	1270.6±156.9	NA
Ca	336 ± 3.65	≤100
Cd	0.01±0.008	≤0.01
Cu	0.08±0.017	≤0.50
Zn	0.312±0.04	≤5.0
Cr	0.0191±0.03	≤0.1
Fe	2.33±0.58	≤5.0
Ni	0.02 ± 0.01	≤0.5
Pb	0.149±0.07	≤5.0

All the values are in mg/L. NA = not available

The findings indicate that the studied leachate was rich in PO_4-P and NH_4 -N, which is attributed to the breakdown of food waste materials in the landfill. The identified concentrations were enough to harvest nutrients from it compared to the sample used in the related study (Lavanya et al. 2021). The level of Mg was found to be below the optimal concentration for struvite precipitation, thus, there was a need to add it from an external source to meet the required stoichiometric ratio.

The high concentrations of Na might be primarily due to the waste being disposed of the landfill that contain high levels of common salt added to human diets. Dumping of waste that contains Ca might be the cause of the calcium content in landfill leachate. Remarkably, the high concentration of K found in leachate is attributed to the disposed remains of agricultural products in the landfill, particularly grapes, which are highly cultivated in the area where the study was conducted. Grapes are reported to contain high concentrations of K (Sousa et al. 2014). According to previous studies, the presence of high concentrations of K, Ca and Na in solutions (leachate) affects the purity of struvite formed (Johansson 2018, Moragaspitiya et al. 2019, Sakthivel et al. 2020).

The concentrations of all heavy metals analyzed in leachate were below TBS (2006) permissible limits (Table 2). The low content of heavy metals in leachate might be due to the low concentration of heavy metals in solid waste disposed in landfill. Furthermore, hazardous industrial wastes that are sorted for recycling before being disposed of in the landfills, such as electronic waste and automobile batteries, might have contributed to low concentration of heavy metals in the landfill leachate. The concentration observed suggests that, it was not enough to have an adverse impact on struvite production. In fact, if heavy metals were present in significant concentrations, co-precipitation of the metals may occur during the struvite precipitation.

Optimal conditions on recovery efficiency of phosphate and ammonium ions

The pH and Mg^{2+}:PO_4^{3-} molar ratio, influence struvite recovery from landfill leachate. These conditions were investigated in order to determine the optimal conditions to recover pure struvite from landfill leachate. The recovery efficiencies of PO_4^{3-} and NH_4^+ as indications for the formation of struvite were also examined at each Mg: P mole ratio and pH investigated. The results of the pH and Mg^{2+}:PO_4^{3-} molar ratios investigated are presented in Figures 3.

The effect of pH

The results show that, with the exception of NH_4^+, the maximum PO_4^{3-} recovery efficiency was achieved when the pH increased from 8.5 to 9.0 for each Mg^{2+}:PO_4^{3-} molar ratio investigated (1:1,2:1,3:1,4:1, and 5:1) as indicated in Figure 2. This implies that, at pH of 9, PO_4^{3-}, NH_4^+ and Mg^{2+} added from external source were present which led to formation of struvite or other phosphate compounds. At a pH 9.5 to 10.5 the recovery efficiency of PO_4^{3-} declined while for NH_4^+ the recovery efficiency declined at 9.5 and peaked at pH of 10.5. This implies that the removal of PO_4^{3-} through struvite precipitation might be reduced due to the low concentration of NH_4^+caused by the conversion of $NH_4^+$to NH_3 gas as the pH values increased. The recovery efficiency of NH_4^+at a pH of 10.5 indicates that higher pH values have a significant impact on the removal of NH_4^+ from leachate compared to PO_4^{3-}. This confirms the maximum recovery efficiency of PO_4^{3-} and NH_4^+ at pH of 9 and 10.5, respectively.

The effect of magnesium to phosphate molar ratios

The results show that as the Mg^{2+}:PO_4^{3-} molar ratio increases at a trend of 1:1,2:1,3:1,4:1 and 5:1 the trend of recovery efficiency of $PO_4^{3-}$and NH_4^+ increases as well (Figure 2). The maximum recovery of PO_4^{3-} was achieved at a pH of 9 and Mg^{2+}:PO_4^{3-} molar ratios 5:1, and that of NH_4^+ was achieved at the same molar ratios at pH 10.5. This suggests that as the Mg^{2+}: PO_4^{3-} molar ratio increased, more Mg^{2+} used to precipitate more PO_4^{3-} and NH_4^+ to produce struvite or other compounds at pH of 9.

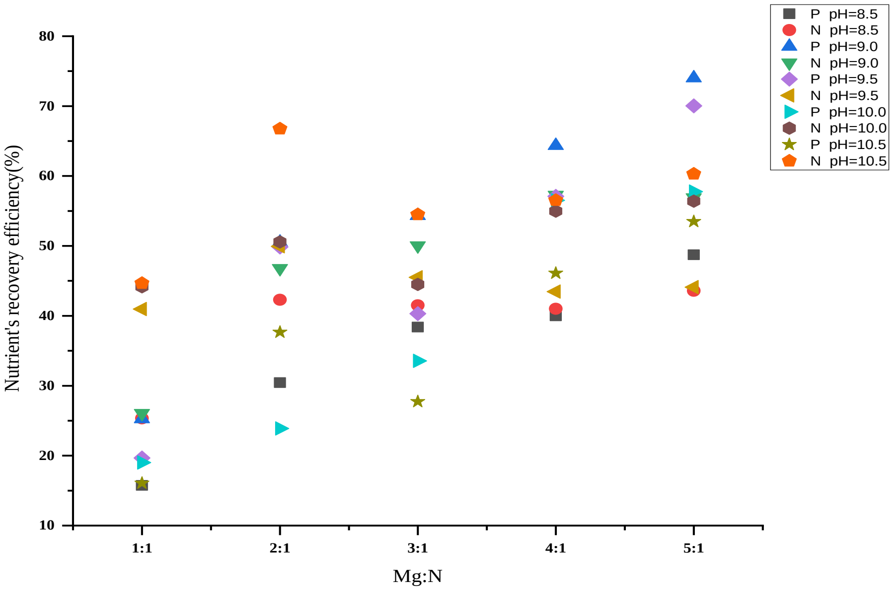

Figure 2. A combined graph for pH, Mg:N and nutrients recovery efficiency

Therefore, the optimal conditions for the maximum removal of PO_4^{3-} and NH_4^+ from leachate were determined to be a pH of 9 and Mg^{2+}: PO_4^{3-} molar ratio of 5.1. The results also demonstrate that the coupling of two factors, pH and Mg: PO_4^{3-} molar ratio influenced maximum recovery efficiency of PO_4^{3-} and NH_4^+ from the leachate rather than a single factor. This is because the maximum removal of nutrients was achieved at higher Mg: PO_4^{3-} molar ratio of 5:1 and pH of 9 compared to Mg: PO_4^{3-} molar ratio1:1 and pH of 9.

The effects of competing ions

High concentrations of K^+, Ca and Na^+ in leachate affect the removal of nutrients from wastewater (Huang et al. 2011). K^+ and Na^+ compete with NH_4^+ for Mg^{2+} and $PO_4^{3-}$to generate K-struvite and Na-struvite (Huang et al. 2019, Kabdaşlı et al. 2022). Still, Ca^{2+} compete with Mg^{2+} for PO_4^{3-} to form phosphate compounds such as hydroxyapatite ($Ca_{10}(PO4)_6(OH)_2$) (Li and Zeng 2020). The present study revealed a significant concentration of the aforementioned ions (Table 2). This suggests that they hindered the efforts to achieve the maximum recovery efficiencies of PO_4^{3-} and NH_4^+ from leachate at more than 90%.

Characterization of precipitates

Characterization of struvites was done by using XRD and XRF. XRD peak profiles of struvites in the leachate was proved to be struvite ($MgNH_4PO_4.6H_2O$), K-struvite ($MgKPO_4.6H_2O$), hazenite ($H_{28}KMg_2NaO_{22}P_2$) and hydroxyapatite ($Ca_{10}(PO4)_6(OH)_2$) crystals (Figure 4).

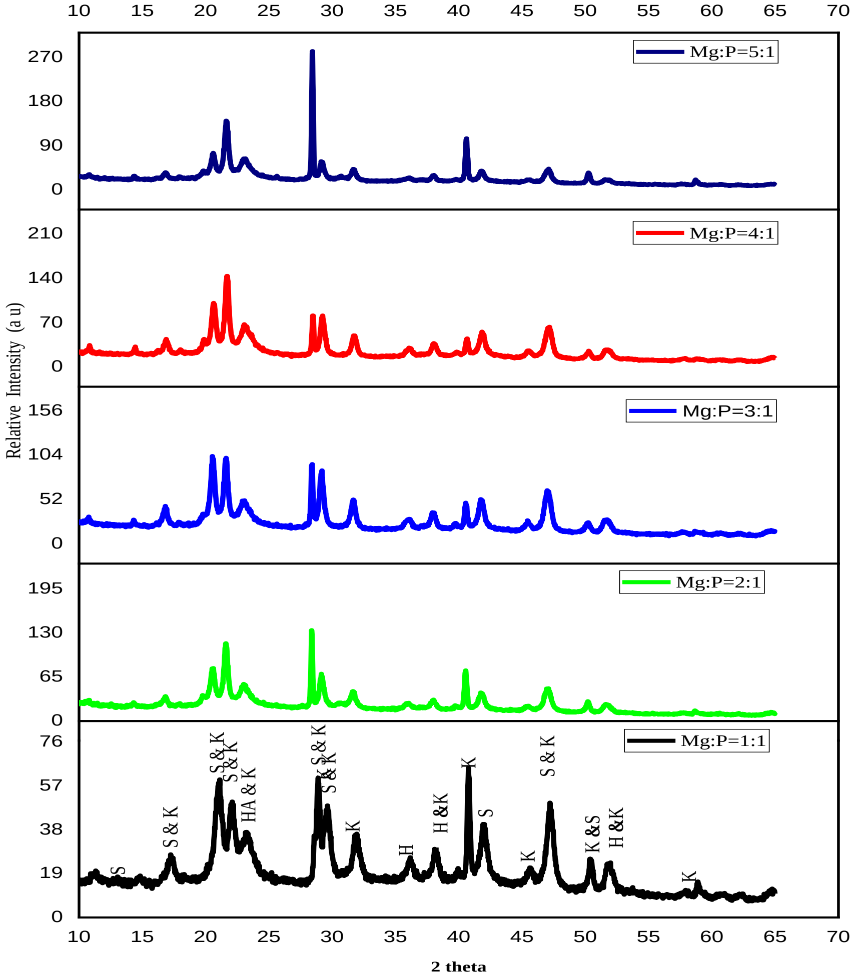

Figure 3. XRD patterns of the precipitate collected at a pH of 9 and different Mg: P molar ratios. Struvite (S), K-struvite(K), hazenite (H) and hydroxyapatite (HA). Each peak profile had been justified by the Rietveld refinement software and matched by struvite, K-struvite, hazenite and hydroxyapatite with those in the reference database model in Match software (Figure 9)

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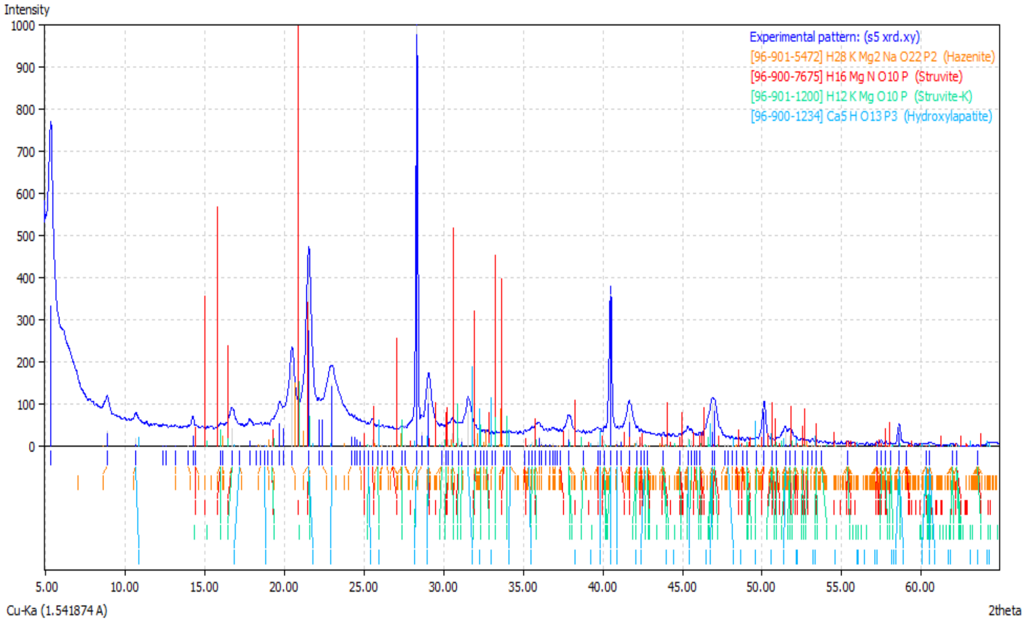

Figure 4. XRD Rietveld refinement plot of the precipitates collected from the leachate. Blue peaks are experimental, orange peaks are hazenite database, red peaks are struvite database, light green peaks are struvite k database and light blue present hydroxylapetite database

The overlapping of peaks was observed in the struvites collected from leachate (Figure 4). This implies that struvites collected from leachate contained various compounds. The quantitative analysis of the obtained struvite precipitates is presented in Table 3. Less amount of struvite was collected possibly due to the presence of K^+ and Na^+ in leachate which might be competing with NH_4^+ for Mg^{2+} and PO_4^{3-} to produce K-struvite and struvite-type phosphate mineral called hazenite. Still, the presence of Ca^{2+} ions was attributed to less amount of struvite because it might have been used to precipitate PO_4^{3-} and OH^- to produce calcium compound called hydroxyapatite. It is interesting to note that in addition to struvite, other struvites (K-struvite and hazenite) and hydroxyapatite were produced, and all of them are used as fertilizers.

Table 3. Quantitative analysis of precipitate produced by Rietveld method.

Precipitate composition	**% Composition by weight**
Struvite ($MgNH_4PO_4 .6H_2O$),	8.4
K-struvite ($MgKPO_4.6H_2O$)	29.9
Hazenite ($H_{28}KMg_2NaO_{22}P_2$)	52.7
Hydroxyapatite ($Ca_{10}(PO4)_6(OH)_2$)	9.0

Furthermore, XRF analysis confirmed the presence of plant nutrients Mg and P with exception of N which was not determined. The presence of competing ions such as K^+, Na^+ and Ca^{2+} were also confirmed in the precipitates collected from the leachate (Table 4)

Table 4: Chemical composition of precipitates determined by X-ray fluorescence

Element	% Composition by weight
K	23.30
Na	20.10
Mg	3.84
Ca	35.1
P	1.97
Cl	7.96
Si	0.73
S	1.97
Fe	0.33
Mn	ND

ND = not detected

The incorporation of K^+, Na^+, Ca^{2+} and other ions in precipitates apart from ions of interest (Mg, N and P) was due to the nature of raw leachate used (Table 2).

The effect of competing ions on struvite formation

The presence of amorphous phases in XRD diffractograms and the differences in peak positions from the reference database reveals the presence of amorphous substances and competing ions in leachate that affected the purity of struvites formed (Figure 3). In the present study, the impact of Ca^{2+} was mitigated by dosing Mg^{2+}. The amorphous phases continued to diminish as more $Mg^{2+}:PO_4^{3-}$ molar ratios increased from 1:1 to 5:1 (Figure 3). This suggests that more $Mg^{2+}:PO_4^{3-}$ molar ratio or leachate treatment were required to completely counteract the effect of Ca^{2+} on removal of plant nutrients and to obtain pure struvite. Similar results been reported elsewhere (Liu and Wang 2019, Moragaspitiya et al. 2019). Different literatures have addressed how to reduce the impact of other competing ions during struvite precipitation. The impacts of Na^+ are mitigated by increasing the $NH_4^+:Na^+$ molar ratio and applying $MgCl_2.7H_2O$ as a source of Mg^{2+} could reduce the effect of Na^+ on struvite production (Huang et al. 2011, 2019). The impact of K^+ is mitigated by increasing $NH_4^+:K^+$ (Pastor et al. 2010).

However, a specific amount of Ca, K, and Na in the precipitate generated is advantageous in addition to struvite (Mg, N, and P). Given that potassium is regarded as a key nutrient for many plants, the presence of potassium-containing compounds is particularly significant.

The effect of $Mg^{2+}:PO_4^{3-}$ molar ratio on struvite formation

The varied $Mg^{2+}:PO_4^{3-}$ molar ratios used during the struvite precipitation process had no effect on the formation of struvites, but it affected the purity of struvites formed. Impurities reduced further as $Mg^{2+}: PO_4^{3-}$ molar ratios were increased from 1:1 to 5:1 (Figure 3).

Conclusions

In the present study phosphorus and nitrogen were recovered from landfill leachate collected from Chidaya landfill for fertilizer production via struvite precipitation method. The landfill leachate contained the essential plant nutrients. The method enhanced the removal of plant nutrients from leachate in the form of struvites and hydroxyapatites. The pH of 9 and $Mg:PO_4^{3-}$ molar ratio of 5:1 were the optimal conditions to recover plant nutrients in the form of struvite. The analysis of struvite showed the presence of other forms of struvites and hydroxyapatites which are used as fertilizers. The presence of competing ions including Ca, K, and Na affected the maximum recovery efficiency of nutrients and purity of the struvite produced. Struvite formation was unaffected by the Mg^{2+} dosage in leachate, but it was affected by the purity of the struvites produced. Recovering nutrients from leachate in the form of struvites addresses the coming phosphate scarcity, lowers environmental pollution and produces fertilizer for use in agriculture to increase food production. However, further investigation is required to confirm the field applicability of the struvite produced and the availability of other locally accessible minerals such as saltwater bittern as an alternative source of Mg^{2+} to lower the cost of purchasing commercial Mg^{2+} sources. Besides, to improve the quality of the leachate produced for the recovery of nutrients, the study proposes the isolation of solid wastes before disposal.

Declaration of Competing Interest

No author claims to have any conflicts of interest related to this work.

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