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Removal of Ammonia Nitrogen (NH_4-N) from Landfill Leachate by Chemical Treatment

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Abstract

Objectives: The paper studies the removal efficiency of Ammonia-nitrogen from leachate by a chemical method named "Struvite Precipitation" as the concentration of Nitrogen (major causes of eutrophication), is several times higher in leachate than in municipal wastewater. **Sampling:** The leachate composition and sampling was done for leachate derived from a dump type landfill named 'Jawahar Nagar Dump Yard', Hyderabad and was characterized as an old leachate with pH = 8.0. **Methodology:** The treatment process tested is a batch type lab-scale experiment for precipitation of ammonia nitrogen in the form of Magnesium-Ammonium-Phosphate (MAP). **Results:** With MAP (Struvite) precipitation at the optimal Mg:N:P ratio (1.2:1:1), 78% removal of inorganic nitrogen was achieved. Maximum Ammonium removal achieved is about 95% at pH of 9.0 with Mg:N:P molar ratio 1.5:1:1.5. **Conclusions:** Comparing the results for different Mg:N:P ratios, conclusions are drawn that it is possible to achieve effluent nitrogen requirements (50 mg N/l) by treatment of leachate with MAP precipitation. Detailed description is given in the article.

Keywords: Ammonium Removal, Leachate, Molar Ratio, Magnesium-Ammonium-Phosphate, Struvite

1. Introduction

Landfill leachate is defined as any liquid that percolates through or drains from solid waste and extracts dissolved, suspended and/or microbial contaminants from it[1]. Though landfilling of organic waste is prohibited, existing landfills will continue to produce leachates contaminated with organic substances. In addition, nitrogen and toxic substances are also present in significant amounts in leachate[2]. Altogether, this will constitute an important challenge for environmental protection over the next decades. To maintain sustainability, Leachate is to be treated in an engineered pathway to prevent intense environmental and ecological degradation.

Landfill leachate is today treated mainly at municipal Wastewater Treatment Plants (WWTPs) together with household domestic wastewater. On-site pre-treatment should be carried out before discharge to the WWTP[3]. However, municipal WWTPs often refuse accepting leachate from landfill sites, due to several reasons.

First of all, leachate components might, in rare cases, have a negative impact on the treatment processes. Moreover, there is a financial interest for landfill operators to introduce their own cost-efficient on-site leachate treatment systems, as WWTPs charge them both for the volume of incoming leachate and nutrients concentrations. Hence, design and evaluation of methods for on-site leachate treatment are required.

The main leachate pollutants that have to be removed are organic compounds, nutrients, heavy metals and suspended solids[4]. Nitrogen, being one of the most important causes of eutrophication, has to be reduced foremost among the nutrients. Its concentration is several times higher in leachate than in municipal wastewater. High nitrogen concentrations and leachate toxicity create limitations for leachate treatment by conventional activated sludge process in a continuous-flow reactor.

Reviews on leachate treatment methods mention a number of treatment processes successfully applied for nitrogen removal from leachate – combined nitrification-

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de nitrification in a sequencing batch reactor, chemical precipitation as Magnesium Ammonium Phosphate (MAP) and ammonia air-stripping[5,6].

Less sophisticated methods, such as irrigation, infiltration and recirculation are also used. However they may cause groundwater pollution and other environmental problems[7]. Novel nitrogen removal technologies, such as ammonium removal over nitrite (Sharon) and combination of nitritation and anoxic ammonium oxidation (Anammox), can be also applied for leachate treatment[8].

Combined nitrification-denitrification in a Sequencing Batch Reactor (SBR) is widely applied for leachate treatment. The first step of this process is oxidation of ammonia to nitrate under aerobic conditions and the second step involves nitrate reduction to nitrogen gas in anoxic conditions[7]. Aeration (in the first phase) and addition of external carbon source (in the second phase) constitute the main expenses[8].

Another method used for nitrogen elimination from leachate is Ammonia Air Stripping which requires significant capital investment and maintenance costs for Construction of humongous towers, blowing/sucking of large quantities of air, pH adjustment before and after the process and treatment of ammonia-polluted air. Furthermore, these counter current spray towers cannot operate in cold conditions or climates because as air temperature approaches freezing, a drastic reduction in efficiency is observed[9].

Nitrogen removal as chloramines using break point chlorination requires a huge quantity of liquid/gaseous chlorine. When induced into wastewater, chlorine reacts with the organic matter leading to formation of Tri-Halo-Methane's (THMs). Moreover, break point chlorination can be used, to remove nitrogen, only when its concentration is very low. Also, due to disinfectant nature of chloramines, most of the useful microorganisms which help in further wastewater treatment are exterminated.

The fourth and final method available for nitrogen removal is by precipitation of a crystal named Magnesium-Ammonium-Phosphate (MAP Crystal - $MgNH_4PO_4{\cdot}6H_2O$), which is also known as struvite. The magnesium compound ($Mg(OH)_2$, MgO, $MgCl_2$ etc.) and phosphoric acid (H_3PO_4) or other salts have to be dosed as Mg- and P-containing substances usually occur in leachate in low concentrations, in comparison to the amount of ammonium, which has to be removed[10].

Leachate treatment technologies add additional complexities to the system design due to the higher concentrations of pollutants, their lower degradability and higher toxicity compared to municipal wastewater. For evaluation of toxicity effects, lab-scale trials should always precede the design step when treatment is considered. The aim of the study, presented in this article, was to evaluate efficiency of nitrogen removal from landfill leachate, by chemical precipitation i.e., by crystallization of struvite.

In spite of high process costs and necessity of phosphorous addition to the wastewater, this alternative has many advantages, for example struvite can be used as a valuable/profitable fertilizer because of its slow nutrient release characteristics. Struvite is more likely to form in a high pH and alkaline environment, where there is lower temperature, higher conductivity, and higher concentrations of magnesium, ammonia and phosphate[11].

In the context of leachate treatment, the constraints of the process application include need for a higher degree of carefulness, maintaining processing conditions like pH, temperature etc. Furthermore, additional experience about the process is required, as its extensive research has been carried out mainly during the last decade.

2. Leachate Composition and Sampling

The leachate, which has been treated in the experiment, was delivered from eastern leachate pond of Jawahar Nagar Dump Yard (Hyderabad, Telangana). The landfill with an area of 339 acres has a pond treatment system for leachate pretreatment before sending it to the municipal WWTP (see Figure 1). Flow equalization in a buffer pond is recommended in a leachate treatment system because of significant variations in leachate volumetric flow and composition. Hence the samples for this study were taken between the buffer pond and the aerated pond. The composition of leachate from landfill, based on the measurements in 11 different samples, taken over a three-month post-monsoon period, is presented in Table 1.

3. Methodology

A glass reactor with a total volume of 1 liter was filled with 0.5 liter of leachate, which was continuously stirred with a magnetic stirrer. $MgCl_2{\cdot}6H_2O$ and $NaH_2PO_4{\cdot}H_2O$ were added. This causes a pH drop, thus, pH adjustment with 1 M NaOH was started directly after the addition of

chemicals to maintain the pH at the selected level (7.5 to 9.5, depending on the chosen conditions). The mixture was stirred during 2 minutes at 400 rpm (fast mixing) and then during 10 minutes at 100 rpm (slow mixing).

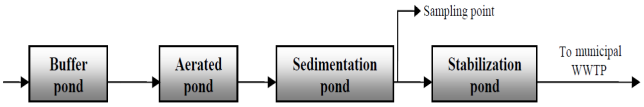

Figure 1. Current leachate treatment system at landfill.

Table 1. Composition of leachate from landfill

Sl. No.	Parameter	Value
1.	Total Nitrogen (unfiltered), mg/l	1056±21
2.	Total Nitrogen (filtered), mg/l	1007±17.3
3.	NH_4-N, mg/l	923±15.4
4.	NO_3-N, mg/l	50.7±1.4
5.	NO_2-N, mg/l	21.27±0.09
6.	Total phosphorous (unfiltered), mg/l	43.7±0.8
7.	Total phosphorous (filtered), mg/l	39.3±0.5
8.	PO_4-P, mg/l	31.8±0.5
9.	COD (unfiltered), mg/l	31.8±0.5
10.	COD (filtered), mg/l	5452±55.7
11.	SS, mg/l	391±7.3
12.	VSS, mg/l	287±6.9
13.	pH	8.0±0.1
14.	Alkalinity, mmol HCO_3^-/l	24.8±2.6

The formed struvite was then allowed to settle during 10 minutes where after samples for NH_4-N and PO_4-P measurements were taken from the upper part of the reactor. The pump for NaOH addition was run at a constant speed of 0.1 ml/s. Stopwatches were used to measure the operating time of the pump for calculation of lime consumption.

4. Results

Struvite precipitation was carried out at different Mg:N:P ratios and different pH. The increase of magnesium and phosphorous molar concentrations above the equilibrium level (1:1:1) at pH 9.0 improves nitrogen removal efficiency up to 95% with the lowest ammonium nitrogen concentration in the effluent equal to 46 mg/l at the Mg:N:P molar ratio 1.5:1:1.5 (see Figure 2). In order to reach 95% efficiency upto 245 mg/l of 1 M NaOH was required to maintain the pH on the desired level. Bars on the diagrams indicate ammonium removal (left scale), while dots indicate lime consumption (right scale).

High consumption of chemicals as well as high phosphorous concentrations in the effluent may seem to look like the utilization of high Mg and P molar concentrations economically inefficient, but if the leachate from landfill is combined with phosphorous rich municipal wastewater at WWTP, the cost for supplying P can be significantly reduced (in some cases upto 48-55%).

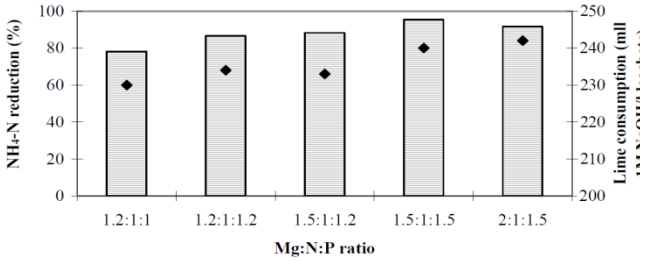

Figure 2. MAP/Struvite precipitation: Reduction of NH_4-N (%) and lime consumption (ml 1M NaOH/l leachate) depending on different Mg:N:P ratios at pH 9.0.

Nitrogen removal significantly improves with increasing pH from 7.5 to 8.5. In the pH range 8.5-9.5 the changes in nitrogen removal efficiency are insignificant, but lime consumption for pH adjustment increases with 25% (Figure 3). The most feasible conditions for struvite precipitation for Mg:N:P ratio 1.2:1:1, were found at pH 8.5.

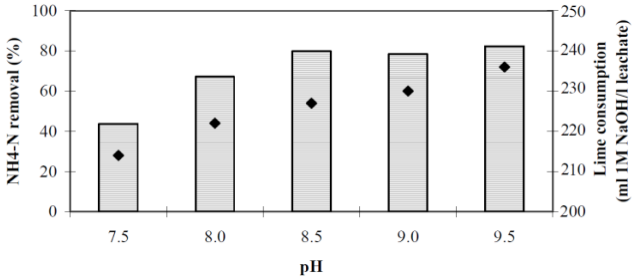

Figure 3. MAP precipitation: Reduction of NH4-N (%) and lime consumption (ml 1M NaOH/l leachate) depending on different pH at Mg:N:P ratio 1.2:1:1.

Addition of phosphorous for nitrogen precipitation resulted in a significant increase of phosphorous concentrations in the effluent with a concentration 190.8 mg PO_4-P/l after MAP precipitation at the applied Mg:N:P ratio 1.2:1:1 and pH 9.0. Efficiency of Organic removal was fairly low (20–25 %).

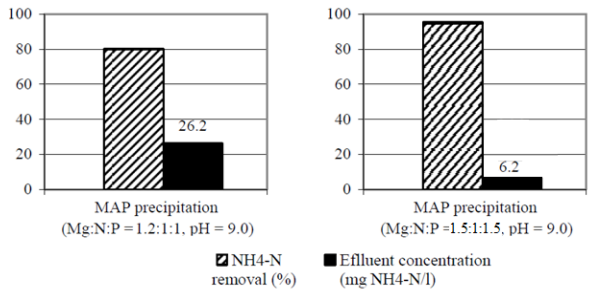

Figure 4. Ammonium nitrogen removal and effluent ammonium nitrogen concentration.

As shown in Figure 4, Struvite precipitation at Mg:N:P molar ratio 1.2:1:1 and pH 9.0 resulted in lower ammonium removal (78% with 215 mg NH_4-N/l in the effluent) and precipitation at Mg:N:P molar ratio 1.5:1:1.5 and pH 9.0 resulted in maximum ammonium removal (95% with 46 mg NH_4-N/l in the effluent).

5. Conclusion

The choice of the right process for a full-scale wastewater treatment facility is always a decision which involves a compromise between the process costs and treatment efficiency. Struvite itself might cause clogging of pipes and equipment failures but, the positive feature of struvite is the possibility of its utilization as a valuable fertilizer, thus, selling it to the farmers may partly reimburse costs for chemicals.

Another positive feature of the process is its small footprint, as the installation can be quickly mounted on site and occupies only a few square meters. Struvite precipitation is effective and economical, when compared to biological processes which require big reactor volumes, high expenses for aeration and high power/electricity consumption.

The nitrogen removal process discussed in this study is considered to be the only possible treatment strategy for the complete nitrogen removal from leachate at Jawahar Nagar landfill. The results can also be taken into account by other landfill operators, who aim at upgrading of leachate treatment systems. Study reveals that struvite precipitation allows upto 95% ammonium nitrogen removal (or 46 mg NH4-N/l in effluent).

With a Mg:N:P molar ratio of 1.2:1:1, which was found to be the optimal one in literature, magnesium salt with a concentration of 1715 mg Mg^{2+}/l and phosphorous salt with a concentration of 2330 mg P/l have to be added to the leachate. The latter also led to an increase of the phosphorous concentration in the effluent up to 400–500 % of the initial concentration of phosphorous in the leachate. Struvite precipitation requires pH adjustment throughout the process. 230 mg/l of 1M NaOH was required to maintain the pH on the desired level.

This lab-scale evaluation of struvite chemical precipitation revealed that it is perhaps very advantageous for Ammonia-nitrogen removal at higher nitrogen concentrations in leachate, especially if a small footprint of the plant is required.

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