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# Effect of Nitrogen Supplement on Methanotrophic Activities in Sandy Loam and Organic Compost-based Landfill Cover Soil

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**Abstract:** The use of sandy loam and compost as final landfill cover material promote methanotrophic activities and reduce methane emission. In this study, the effect of nutrient supplement in the form of ammonium and nitrate to sandy loam and organic compost-based landfill cover soil was investigated in batch experiment. The results revealed the addition of ammonium inhibited methane oxidation in sandy loam; however, some stimulatory effect was found in the compost case. The influence of nitrate addition was not observed. Methanotrophic activity in compost-amended material was found five times higher that in sandy loam. The effects of ammonium nitrogen were also confirmed in soil column experiment operated with leachate irrigation.

**Key words:** Methanotrophic activity, methane oxidation, nutrients, landfill cover, compost.

## Introduction

Methane ($CH_4$) emission from landfill contributes up to 10% of global $CH_4$ emission to the atmosphere (IPCC, 2001). However, biological sink of atmospheric $CH_4$ by bacteria in upland soil is responsible for about 6% of global $CH_4$ emission (Mer and Roger, 2001). Landfills are regarded as an important source and sink of atmospheric $CH_4$. The aerobic soil cover materials on top of landfill can control the emission of $CH_4$ via microbial $CH_4$ oxidation by $CH_4$ oxidizing bacteria or methanotrophs which utilize $CH_4$ as a sole carbon and energy source in their metabolism pathways under aerobic condition (Hanson and Hanson, 1996). Enhancement of landfill cover soil potential in reducing $CH_4$ emission through methanotrophic activity is one attractive practice for landfill management in developing countries.

Many prior experiments have studied the performance of landfill cover soil in reducing $CH_4$ emissions. Sandy loam was generally used in those studies due to its loose texture and coarser grain size than other fine grained soils and clays which affected oxygen diffusion into landfill lower soil and also $CH_4$ consumption. Recently, compost was introduced as final cover material due to high porosity, water retention capacity and organic content which directly support adequate oxygen, moisture content and supplemental organic matter for methanotrophic activity (Christophersen et al., 2001; Humer and Lechner, 2001; Streese and Stegmann, 2003; Wilshusen et al., 2004; Mor et al., 2006). Furthermore, leachate irrigation was also reported to benefit $CH_4$ oxidation in vegetated landfill cover soil (Maurice et al., 1999). This could be attributed to an increased amount of roots in vegetated cover soil which might provide a better environment for methanotroph besides available oxygen and contribute to a greater $CH_4$ oxidation capacity. However, effect of nutrient supplement on methanotrophic activity in different landfill cover soil material has not been reported. The objective of this study was to determine the effects of nutrient nitrogen (ammonium—$NH_4^+$ and nitrate—

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$NO_3^-$) supply on $CH_4$ consumption in both sandy loam and compost in column and batch experiments.

## Materials and Methods

### Landfill Column Experiment

The column experiment was performed in acrylic columns with 15 cm diameter and 100 cm height to simulate landfill cover system. Two different types of materials (sandy loam and compost) were filled into the columns with each 60 cm depth and operated under simulated landfill condition by purging at the bottoms with artificial landfill gas, 60% $CH_4$ and 40% $CO_2$, at flow rate of 4 mL/min (equivalent to $CH_4$ flux of 14 mol $CH_4/m^3$.d) and intermittent irrigating with leachate or rainwater. The properties of sampled cover materials and leachate characteristics are summarized in Tables 1 and 2.

**Table 1: Characteristics of cover materials—sandy loam and compost**

| *Properties* | *Unit* | *Sandy Loam* | *Compost* |
|---|---|---|---|
| pH (1 : 2.5) | - | 7.30 | 6.36 |
| EC (1 : 2.5) | dS/m | 0.24 | 7.15 |
| CEC | c $mol_c$/kg | 4.8 | 22.3 |
| Moisture content | %d.w. | 14.48 | 51.73 |
| Bulk density | $kg/m^3$ | 1,250 | 566 |
| Total organic carbon | % | 0.61 | 12.22 |
| TN | mg/kg | 1,880 | 19,645 |
| $NH_4^+$ - N | mg/kg | 13.26 | 34.06 |
| $NO_3^-$ - N | mg/kg | 16.93 | 120.41 |
| Available P | mg/kg | 8 | 5,845 |
| Available K | mg/kg | 39 | 5,780 |

**Table 2: Characteristics of leachate**

| *Parameters* | *Unit* | *Value* |
|---|---|---|
| pH | - | 8.01 ± 0.43 |
| EC | mS/cm | 11.3 ± 3.6 |
| Temperature | °C | 28.4 ± 1.9 |
| BOD | mg/L | 31.3 ± 6.1 |
| COD | mg/L | 494 ± 40 |
| TKN | mg/L | 647 ± 38 |
| $NH_4^+$ - N | mg/L | 584 ± 44 |
| $NO_2^-$ - N | mg/L | n.d. |
| $NO_3^-$ - N | mg/L | 0.22 ± 0.02 |
| TP, mg/L | mg/L | 4.08 ± 1.01 |
| BOD/COD | - | 0.07 ± 0.02 |
| COD/TKN | - | 0.76 ± 0.11 |

### Determination of Methanotrophic Activities

Batch study of methanotrophic activity was conducted to evaluate the capacity of sandy loam and compost as landfill cover soil. These materials were collected from the surface layer (0-15 cm) of simulated landfill cover columns in column experiment. Ten grams of each cover material was transferred into 188 mL serum bottles sealed with rubber septum and aluminum ring, incubated at an initial headspace $CH_4$ concentration of 9%, and $CH_4$ concentration was monitored with time by gas chromatography (GC) on daily basis. All incubation experiments were performed in duplicate.

Methanotrophic activity was defined as maximum specific $CH_4$ consumption rate which was determined from the reduction of $CH_4$ concentration in the bottle headspace according to incubation time and weight of incubated soil.

### Effect of Nitrogen Supplement on Methanotrophic Activity

To investigate the effect of $NH_4^+$ and $NO_3^-$ amendment on methanotrophic activity, $NH_4Cl$ and $KNO_3$ were added to each cover material (sandy loam and compost) at the contents of 10, 30, 50 and 100 mg N/kg soil. The control incubation was prepared by adding distilled water instead of nitrogen nutrient solution. All amended cover materials were incubated in serum bottles at an initial concentration of 9% $CH_4$ as previously mentioned.

## Results and Discussion

### $CH_4$ Consumption Rate in Soil Column Experiment

The study of methanotrophic activity in simulated soil columns was conducted using two different types of materials, i.e. sandy loam and compost. The columns were operated with leachate irrigation in order to control moisture content and supply nutrients to the materials. Methanotrophic activity in both landfill covers was expressed in terms of $CH_4$ consumption rate. During steady active period (Figure 1), it was found that $CH_4$ consumption rate in compost was higher than in sandy loam, especially in case of leachate irrigation. Compost could maintain higher $CH_4$ consumption rate of about 12 mol $CH_4/m^3$.d over 240 days, while sandy loam provided lower rate of 9 mol $CH_4/m^3$.d during the first 120 days and 6 mol $CH_4/m^3$.d afterwards (Figure 1b). However, in rainwater case, shorter steady active period (100-120 days) was observed in both compost and sandy loam (Figure 1a). High porosity and water-holding capacity characteristics of compost directly supported

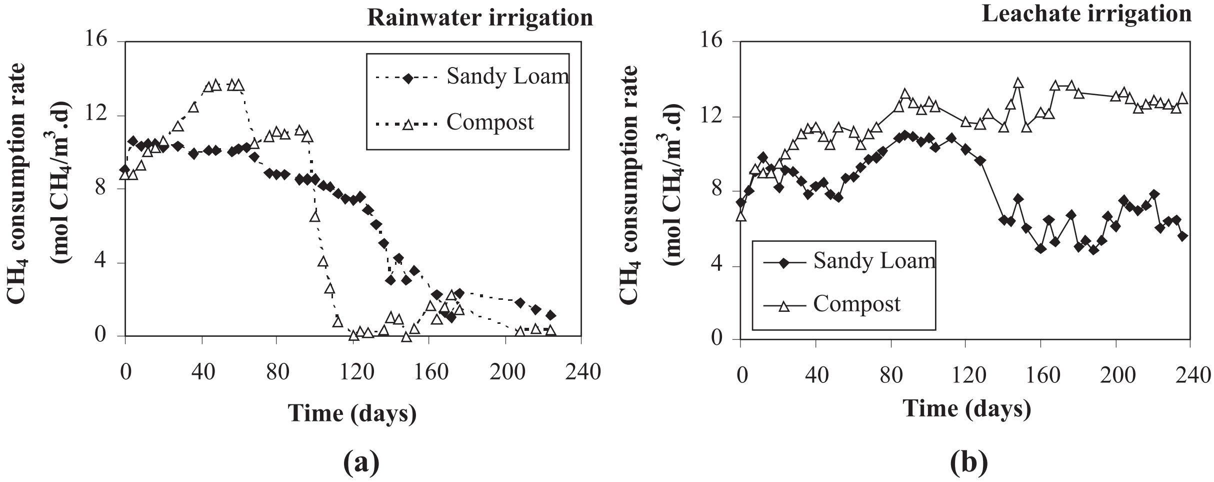


**Figure 1: $CH_4$ consumption rate in column experiment of sandy loam and compost with (a) rainwater and (b) leachate irrigation.**

adequate oxygen and moisture content for methanotrophic activity (Streese and Stegmann, 2003; Wilshusen et al., 2004). Besides the beneficial physical properties, high organic matter content also considerably influenced the methanotrophic activity. Several studies (Christophersen et al., 2001; Humer and Lechner, 2001) reported high methanotrophic activity in high organic material such as compost. Moreover, Seghers et al. (2005) demonstrated that abundance of methanotrophs (especially Type II) was significantly higher in the organically fertilized soil which was correlated with the enhancement of $CH_4$ consumption. They also suggested that compost application directly affected the presence of Type II methanotrophs in compost. The application of compost as cover material could alter the methanotrophic bacteria community by providing organic matter and subsequently increased methanotrophic activity.

Furthermore, comparison of $CH_4$ consumption rate in leachate application with rainwater application is shown in Table 3. In sandy loam, $CH_4$ consumption rate in the former active period (day 0-120) was not different between leachate and rainwater irrigation (about 9 mol $CH_4/m^3$.d). Subsequently, capacity in rainwater case was dropped to zero (Figure 1a) while that of leachate case was continued in the later active period (day 120-240) at lower capacity of 6 mol $CH_4/m^3$.d. Contradictory observation was found in compost material. Capacity of $CH_4$ consumption in the later active period (day 100-240) of leachate application (13 mol $CH_4/m^3$.d) was higher than that of former period (day 0-100) in both leachate and rainwater applications (11 mol $CH_4/m^3$.d). From these column results, leachate irrigation affected the methanotrophic activity in sandy loam and organic compost in different manner. Inhibitory effect was found in sandy loam whereas stimulatory effect was shown in compost material. Therefore, effect of leachate on methanotrophic activity in sandy loam and organic compost was investigated in terms of nutrient nitrogen ($NH_4^+$ and $NO_3^-$) using batch experiment.

**Table 3: Methane consumption in column experiment of sandy loam and compost with rainwater and leachate irrigation**

| *Landfill covers* | | *Active period (days)* | *Methane consumption rate* (mol $CH_4/m^3$.d)* |
|---|---|---|---|
| Sandy loam | Rainwater | 0-120 | 9.7 ± 0.8 |
| | Leachate | 0-120 | 9.3 ± 1.1 |
| | | 120-240 | 6.3 ± 0.9 |
| Compost | Rainwater | 0-100 | 10.9 ± 1.4 |
| | Leachate | 0-100 | 10.8 ± 1.2 |
| | | 100-240 | 12.5 ± 0.8 |

*All data are the average and standard deviation of methane consumption in the steady active period.

### Effect of Nitrogen Supplement on Methanotrophic Activity

Figure 2 presents the $CH_4$ consumption curve of batch experiment with $NH_4^+$ and $NO_3^-$ amendment in landfill covers. The $CH_4$ consumption or methanotrophic activity was assumed to be zero order reaction with constant rate as shown in Table 4. It was noticed that methanotrophic activity in compost without nitrogen addition was five times higher than in sandy loam. Compost clearly showed high capacity of methanotrophic activity.

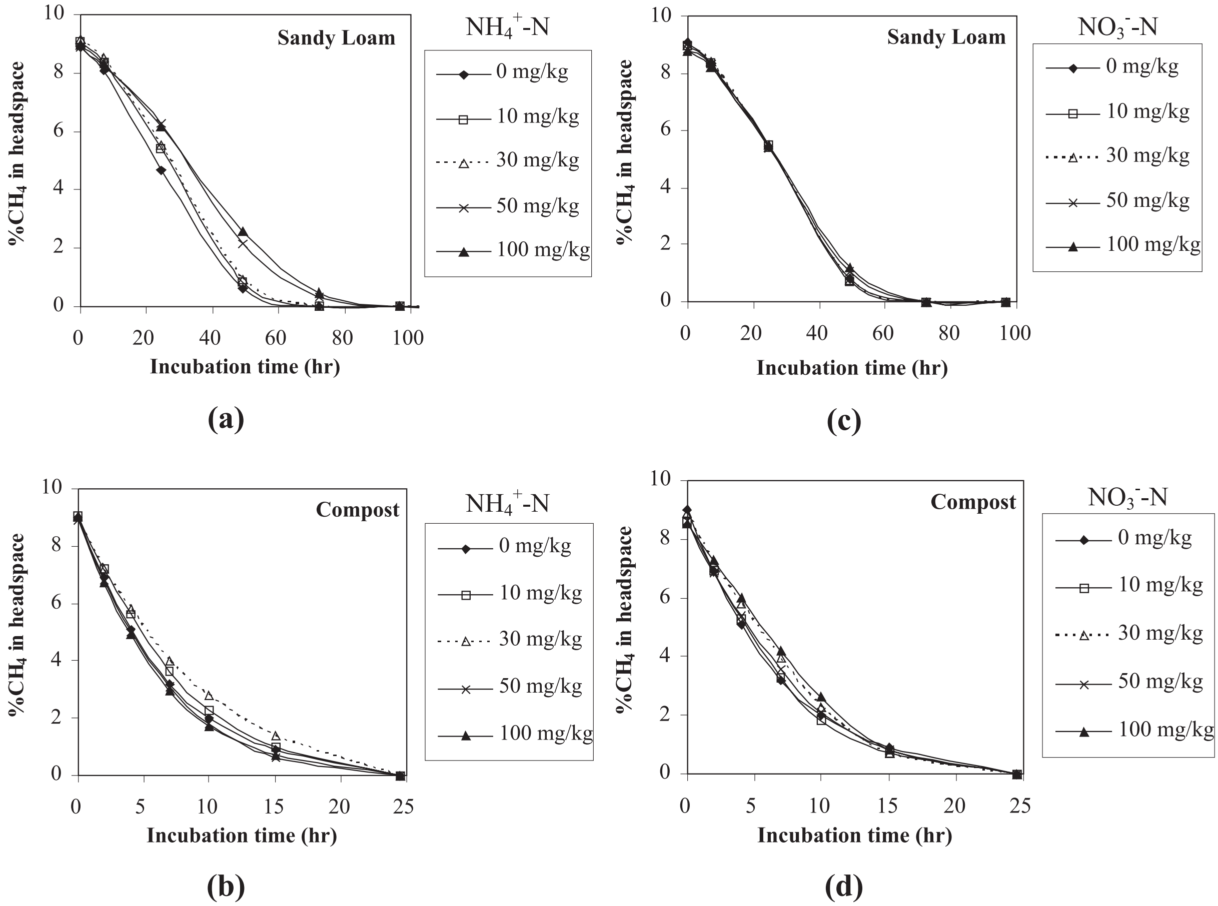


**Figure 2: Methane concentration in batch experiments with $NH_4^+$ amendment in (a) sandy loam and (b) compost; and $NO_3^-$ amendment in (c) sandy loam and (d) compost landfill cover.**

*Effect of $NH_4^+$ Addition*

The addition of $NH_4^+$ had slight inhibitory effect on methanotrophic activity especially in sandy loam with high amount of $NH_4^+$ addition (Table 4). In sandy loam, low addition of $NH_4^+$ (10 and 30 mg N/kg soil) did not significantly affect the methanotrophic activity, whereas high amount of $NH_4^+$ addition (50 and 100 mg N/kg soil) provided the inhibition of about 30%. Same inhibitory effect of $NH_4^+$ addition (50 and 100 mg N/kg soil) in landfill cover soil was also confirmed by Visscher et al. (2001). They proposed that the inhibitory effect of $NH_4^+$ was increased with the exposure time of soil to the high $CH_4$ according to the dominant microorganism at that time. Different observations of $NH_4^+$ were reported by Kightley et al. (1995), Hutsch (1998), Cai and Yan (1999) and Whalen (2000) showing strong inhibitory effect during incubation studies of landfill cover, arable, paddy and forest soil, respectively. They explained that the suppression of methanotrophic activity was due to competitive metabolism of methane monooxygenase (MMO) in methanotroph by $NH_4^+$.

**Table 4: Effect of $NH_4^+$ and $NO_3^-$ amendment on methanotrophic activity in landfill cover soil**

| *Landfill covers* | *Added $NH_4^+$ or $NO_3^-$ content (mg N/kg soil)* | *Methanotrophic activity ($\mu$mol $CH_4$/kg dry soil)* | |
|---|---|---|---|
| | | *$NH_4^+$ amendment* | *$NO_3^-$ amendment* |
| Sandy loam | 0 | 0.40 | 0.40 |
| | 10 | 0.40 | 0.39 |
| | 30 | 0.39 | 0.39 |
| | 50 | 0.29 | 0.38 |
| | 100 | 0.28 | 0.38 |
| Compost | 0 | 1.89 | 1.89 |
| | 10 | 1.86 | 1.87 |
| | 30 | 1.75 | 1.79 |
| | 50 | 1.94 | 1.78 |
| | 100 | 1.99 | 1.75 |

In case of compost, $NH_4^+$ addition at low content did not significantly affect the capacity of methanotrophic activity while high content of $NH_4^+$ (50 and 100 mg N/ kg soil) led to the slight stimulation of methanotrophic activity. The effect of $NH_4^+$ addition could be explained by following assumption as reported in various researches. Firstly, less or no effect of $NH_4^+$ addition at low content was probably due to the changes in bacteria community, i.e. possible shift between $NH_4^+$-tolerant and $NH_4^+$-intolerant methanotrophs or a relative increase of $NH_4^+$ oxidizers consuming $CH_4$ (Hutsch, 1998; Sitaula et al., 2000), and the capacity of soil to adsorb $NH_4^+$ in soil matrix (referred as cation exchange capacity; CEC) which could reduce the competitive inhibition of $NH_4^+$ for methanotrophic activity (Dunfield and Knowles, 1995; Gulledge et al., 1997). Secondly, stimulatory effect of $NH_4^+$ addition at high content was suggested by Cai and Mosier (2000) and Visscher et al. (2001). They proposed that the amount and activity of $NH_4^+$ oxidizers was increased at high $NH_4^+$ content and thus $CH_4$ consumption was also increased. Other explanation to the stimulatory effect was correlated to $NH_4^+$-N limitation of methanotrophic growth which was mitigated by $NH_4^+$ addition and subsequently increased methanotrophic activity (Visscher et al., 1999; Papen et al., 2001; Visscher and Cleemput, 2003).

From these results, it was found that compost application may affect the soil microorganism community and their ecosystem as compared to sandy loam. Thus, addition of $NH_4^+$ increased $CH_4$ consumption in compost material, but not in sandy loam. This batch experiment confirmed the observation of inhibitory effect in sandy loam and stimulatory effect in compost column studies as the $NH_4^+$ content in soil during column experiment would be increased with time. At the end of column experiment, $NH_4^+$ content in sandy loam and compost was increased to 100 and 150 mg N/kg soil, respectively.

*Effect of* $NO_3^-$ *Addition*

As shown in Table 4, the effect of $NO_3^-$ addition on methanotrophic activity was not significant in both sandy loam and compost. In sandy loam soil, influence of $NO_3^-$ was not observed even at high concentration. This result was confirmed with the studies in arable and landfill cover soil by Hutsch (1998), Hilger et al. (2000), Visscher et al. (2001) and Park et al. (2002). Additionally, Hilger et al. (2000) reported that landfill cover soil collected from the lysimeter operated after long exposure to $CH_4$ had no effect of $NO_3^-$ addition on methanotrophic activity.

In compost material, slight inhibitory effect of $NO_3^-$ was obtained only at high concentration. Nevertheless, other studies revealed stronger inhibitory effect of $NO_3^-$ (Chiemchaisri et al., 2001; Wang and Ineson, 2003) as caused by nitrite ($NO_2^-$) accumulation. $NO_3^-$ addition might restrict $NO_2^-$ oxidation through nitrification and thus accumulated $NO_2^-$ caused a decrease in methanotrophic activity (Chiemchaisri et al., 2001). Meanwhile, Wang and Ineson (2003) proposed that the toxic $NO_2^-$ which inhibited methanotrophic activity was probably produced from $NO_3^-$ reduction through denitrification. Hence, the less inhibitory effect of $NO_3^-$ found in our study was possibly due to low $NH_4^+$ content in compost (Table 1) and no $NO_2^-$ accumulation via nitrification. Furthermore, because aerobic condition was maintained throughout the batch experiment, $NO_2^-$ production via denitrification was negligible.

From these batch results, it was found that $NH_4^+$ but not $NO_3^-$ affects the methanotrophic activity. Therefore, the effect of leachate irrigation in column study could be attributed by its $NH_4^+$ concentration.

## Conclusions

In soil column experiment, $CH_4$ consumption rate of 8 and 12 mol $CH_4/m^3$.d was obtained in sandy loam and compost-based cover soil during leachate irrigation for 240 days. The effect of leachate irrigation on methanotrophic activity was found. Batch experimental results also confirmed the effect of $NH_4^+$ on inhibiting and stimulating $CH_4$ consumption rate in sandy loam and compost operated with leachate application in long-term as the accumulation of $NH_4^+$ took place in landfill cover soil.

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