INFLUENCE OF OPERATIONAL PARAMETERS ON LEACHATE QUALITY AND GREENHOUSE GAS ABATEMENT IN THE AEROBIC-ANAEROBIC LANDFILL METHOD

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February, 2016

INFLUENCE OF OPERATIONAL PARAMETERS ON LEACHATE QUALITY AND GREENHOUSE GAS ABATEMENT IN THE AEROBIC-ANAEROBIC LANDFILL METHOD

By

NAG MITALI

A Thesis Submitted

In Partial Fulfillment of the Requirements

For the Degree of

Doctor of Engineering

Examination Committee:

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To the
DEPARTMENT OF URBAN AND ENVIRONMENTAL ENGINEERING
GRADUATE SCHOOL OF ENGINEERING
KYUSHU UNIVERSITY
Fukuoka, Japan
February, 2016

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CERTIFICATE

The undersigned hereby certify that they have read and recommended to the Graduate School of Engineering for the acceptance of this thesis entitled, "***Influence of operational parameters on leachate quality and greenhouse gas abatement in the Aerobic-Anaerobic Landfill Method*** by **Nag Mitali** in partial fulfillment of the requirements for the degree of **Doctor of Engineering**.

Dated: February, 2016

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ABSTRACT

Most of the cities in developing countries facing problems related with Municipal solid waste (MSW) disposal of high organic contents. Improper disposal of solid waste without treatment of leachate and gas lead to serious environmental issues which cause health problems as well. It is very necessary to minimize the environmental load caused by improper waste disposal including conventional landfill method. Aeration in landfills can activate microorganisms, which can execute important functions for biodegradation and accelerate landfill stabilization. In an aerated landfill bioreactor, carbon and nitrogen are effectively removed from the solid waste and leachate. However, oversupply or ineffective aeration can decrease microbial activity and increase energy consumption. A balanced and an effective utilization of both aerobic and anaerobic metabolic pathways of the microorganisms are required to accelerate landfill stabilization. The Aerobic-Anaerobic Landfill Method (AALM), originally developed from wastewater treatment technology, is considered a novel landfill method. In this method, air is injected into an anaerobic-type landfill at intervals to create aerobic zones, whereby alternative aerobic-anaerobic conditions are created in the landfill during non-aeration period. It is expected that: first, the conversion efficiency of nitrogen can be accelerated due to the existence of alternative aerobic-anaerobic conditions; second, the aerobic zones/conditions may accelerate the decomposition of organic carbon as well as reduce the emission of greenhouse gas (GHG); and third, hybrid conditions may create biostabilized landfills, thereby reducing the need for expensive perpetual landfill aftercare and reduce the energy cost as well. In order to achieve the goals mentioned above, AALM has been adopted by supplying intermittent aeration in column bioreactors to make it environmentally and economically viable.

In Chapter 1, firstly, definition of MSW, the current status of MSW, including its quantity, composition and disposal strategies, were reviewed. Then, the chronological development of various novel landfill technologies, including their advantages and disadvantages, were also introduced. Finally, the objectives and structures of the study were presented.

In Chapter 2, the AALM was evaluated by using intermittent aeration. The aeration process entailed the injection of air into plexiglass cylinders (ϕ 10 cm diameter × 200 cm height), filled with fresh organic solid waste collected from a composting plant. Different aeration routines

were applied, namely, continuous aeration (Aerobic reactor A), aeration for three days/week (Aerobic–anaerobic reactor B), aeration for 6 h/day (Aerobic–anaerobic reactor C), and no aeration (Non-aerated reactor D).The leachate quality and gas composition in each of the reactors were measured during the experimental period (408 days). It was found that Aerobic reactor A produced the best results in terms of reduction of GHGs and improvement of the leachate quality. Both reactors of B and C had significant improvement of leachate quality with Reactor A and carbon emission as CO_2 were 92, 52, 70 and 34 % in reactors A, B, C and D, respectively. Reactor C was found to be more effective than reactor B in respect of both the emission of GHGs and the leachate quality; moreover, three-fourth of energy cost can be reduced by operating Reactor C compared with Reactor A. Therefore, it is concluded that, in the AALM, air injection with an effective intermittent aeration that could create effective aerobic-anaerobic conditions, enable to reduce environmental pollutions, accelerate the stabilization of landfill waste and simultaneously reduce the energy cost.

In Chapter 3, the phenomenon occurs in transition period between aeration and non-aeration cycles was investigated by an intensive seven-day experiment conducted on the discharged leachate obtained from aerobic–anaerobic reactors B (3 days/week) and C (6 hours/day) with gas composition. It was found that aeration accelerated microbial activity and mobilization of compounds which initially increased the concentration of TOC, T-N and NH_4^+-N that led to further degradation. The transition period between the aeration and non-aeration cycles, followed the simultaneous nitrification–denitrification had a considerable effect on the leachate quality of both the reactors. The results have prospects of AALM that could reduce leachate pollutants, GHG emission and landfill aftercare as well as energy costs.

In Chapter 4, it was initiated to demonstrate the features of N_2O production rate from organic solid waste during nitrification under 20, 30, and 40 °C temperatures with 5, 10, and 20 % O_2 concentrations that of 60 % moisture content, high total organic carbon and ammonium concentrations. The experiment was carried out by batch experiment using Erlenmeyer flasks incubated in a shaking water bath for 72 hours. A duplicate experiment was carried out in parallel, with addition of 100 Pa of acetylene as a nitrification inhibitor, to investigate nitrifiers contribution on N_2O production. The production rate of N_2O in organic solid waste decomposition was in the range of 0.40-1.14 μg N/g-DM/h under experimental conditions of this study. The rate of N_2O production at 40 °C was almost double than the minimum at 30 °C and nitrification was the dominant pathway of N_2O production. However, denitrification also took place simultaneously. It was evaluated that optimization of O_2 content is one of the crucial parameters on N_2O production.

In Chapter 5, a field study was conducted to investigate the production potential of N_2O by in-situ aeration in a closed landfill site. Air was injected into the landfill site and the correlations between N_2O production and related factors are investigated. The in-situ aeration experiment was carried out by three sets of gas collection pipes along with temperature probes were installed at three different distances of one, two and three meter away from the aeration point; named points A–C, respectively. Each set of pipes consisted of three different pipes at three different depths of 0.0, 0.75 and 1.5 m from the bottom of the cover soil. Landfill gases composition was monitored weekly and gas samples were collected for analysis of N_2O concentrations. It was evaluated that temperatures within the range of 30-50 °C with 15-20 % O_2 content led up to 338 ppm of N_2O in 0.75 m depths at points A and B. O_2 below 10 % can infuse N_2O production during nitrification and above 5 % inhibit denitrification, which would affect N_2O production. The findings provide insights concerning the production potential of N_2O in an aerated landfill that may help to minimize GHG emissions with control of the operational parameters and biological reactions of N turnover.

Chapter 6 concluded all experimental results and based on the research results, the recommendations of future studies for optimization of the AALM were introduced.

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CHAPTER 1

Introduction

1.1. Definition and classification of MSW

“Waste is a left-over, a discarded product or material of marginal or limited value for the owner and which the owner wants to get rid of”.

The above definition suggests that waste includes all materials discarded from households, offices, restaurants, hotels, schools, hospitals, factories, construction, agriculture and so on, i.e., items of no material value for people or businesses. In another sense, waste refers to the material that is discarded without being resold to other persons or companies, and is costly to collect, transport and dispose of.

MSW is the solid waste collected and disposed of by or for municipalities; however, the nature of MSW varies from region to region (UNEP et al., 2005). Some countries define “MSW” as “ordinary solid waste” or “urban solid waste” managed by or for municipalities; the OECD (2010) states “municipal waste covers waste from households, including bulky waste, similar waste from commerce and trade, office buildings, institutions and small businesses, yard and garden waste, street sweepings, the contents of litter containers, and market cleansing waste”, but this definition excludes waste from municipal sewage networks and treatment, as well as from construction and demolition activities. However, the definition by the World Bank (2012) includes industrial waste, and construction and demolition waste into MSW streams. Malaysia, Vietnam and Cambodia, on the other hand, have no definition of MSW, and the Philippine definition of municipal waste refers to wastes produced from activities within local government units, including domestic, commercial, institutional and industrial wastes and street litter (Republic Act No.9003). Japan defines MSW simply as waste other than industrial waste, all of which shall be managed by or for municipalities (Waste Management and Public Cleansing Law).

The classification of urban living garbage has several ways. Such as, chemical classification method, density classification method, state classification method; It can also be classified according to the flammability or degradability, and according to the generation sources, according to the pollution on environment etc.

In chemical composition classification method, MSW can be divided into organic waste and inorganic waste. According to the density classification method it can be divided into heavy and light waste. According to the form can be divided into solid (block, granular, powder) and the form of mud; According to characteristic on flammability it can be divided into combustible and difficult to burn garbage and not burning garbage. For the degradability of MSW it can be classified as easily degradable, hardly degradable and undegradable. According to the pollution extents it can be divided into hazardous waste and general waste. Generation sources classification method divide MSW into mining, industrial, city life, agriculture and radioactive.

1.2. Generation, composition and world-wide strategy of solid waste treatment

1.2.1. MSW Generation

Current world generation of MSW levels are approximately 1.3 billion tons per year and are increase is about 2.2 billion tons per year by 2025.

Generation rate of MSW are usually influenced by various factors, such as, economic situation, the degree of industrialization, public habits and local climate. Generally, higher economic development and urbanization rate regions produce higher fraction of MSW due to the improvement of their living standards, goods consumption etc.

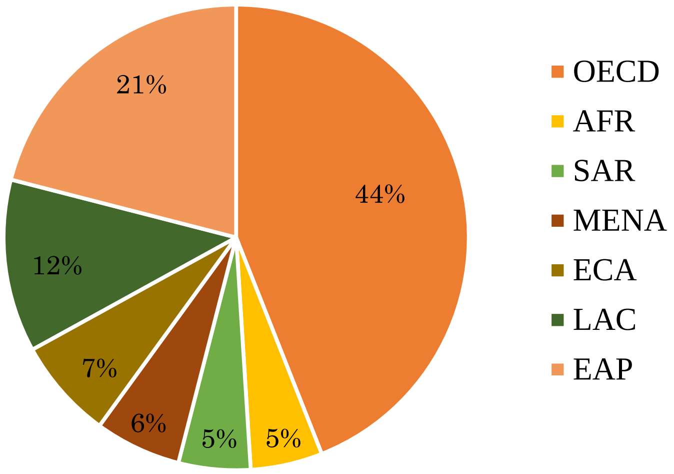


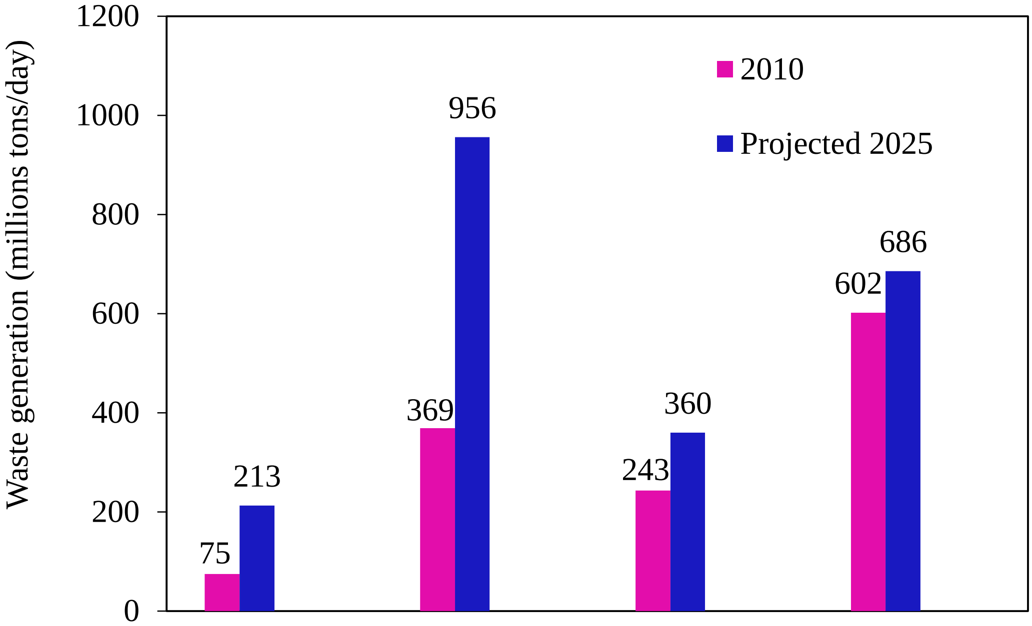
Fig.1-1 Waste Generation by Region (*Source:* World Bank, 2012)

Residents of urban area generate about twice as much waste as their rural counterparts. Fig. 1-1 illustrates world waste generation per region, where OECD countries make up almost half of the world’s waste, while Africa and South Asia produce least waste. Table 1-1 shows projected waste production for the year 2025 according to the current population growth rate in each region.

Table 1-1 Waste Generation Projections for 2025 by Region

	Available data in 2012			**Projections for 2025**			
	Total Urban Population (millions)	Urban Waste Generation		Projected population		Projected urban Waste	
Region		Per capita (kg/capita/ day)	Total (tons/day)	Total population (millions)	Urban populations (millions)	Per capita (kg/capita/ day)	Total (tons/day
AFR	260	0.65	169,119	1,152	518	0.85	441,840
EAP	777	0.95	738,958	2,124	1,229	1.5	1,865,379
ECA	227	1.1	254,389	339	239	1.5	354.810
LCR	399	1.1	437,545	681	466	1.6	728,392
MENA	162	1.1	173,545	379	257	1.43	369,320
OECD	729	2.2	1,566,286	1,031	842	2.1	1,742,417
SAR	426	0.45	192,410	1,938	734	0.77	567,545
Total	2,980	1.2	3,532,252	7,644	4,285	1.4	6,069,703

According to country income level, waste generation per capita is high in high income countries than lower income countries. Although the total generation of waste for lower middle income countries is much higher than that of upper middle and high income countries because of increasing population, income and urbanization of developing countries will generate large amount of MSW, which will be resulted in the increase of its total MSW production. Therefore, in the next decade, disposal of MSW among the developing countries, should be considered one of the concern issues.



Urban population (millions)	343 676	1293 2080	573 619	774 912
waste (kg/capita/year)	219 343	288 344	423 628	777 840
Country Income group	Lower Income	Lower Middle Income	Upper Middle Income	High Income

Fig. 1-2 Urban waste Generation by Income Level and Year

(Source: World Bank, 2012)

1.2.2. MSW Composition

The composition differs depending on the economic level of cities as well as other factors such as geographic location, energy sources, climate, living standards and cultural habits, and the sources of waste that are considered as MSW or are collected by the municipality.

Generally, low and middle income countries have a high percentage of organic matter in the urban waste stream, ranging from 40 to 85 % of the total. Paper, plastic, glass and metal fraction increase in the waste stream of middle and high income countries.

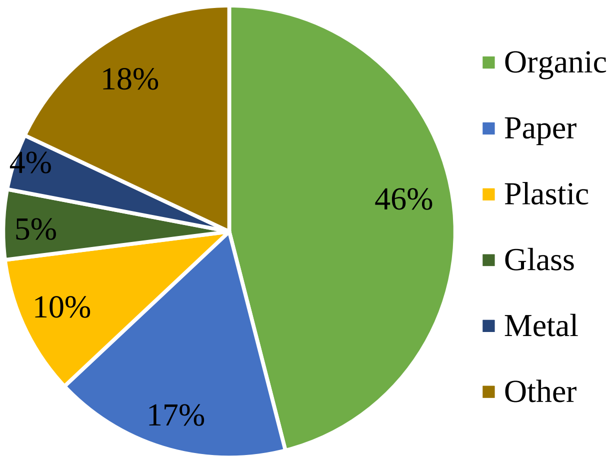


Fig. 1-3 Global Waste composition (Source: World Bank, 2012)

1.2.3. MSW Collection

Collection of MSW is a very important aspect for maintaining public health in cities around the globe. Waste collection is the collection of solid waste from point of production such as resident, industry, institute etc. to the point of treatment or disposal. MSW is collected in several ways; such as house to house, community bins, curbside pickup, and self-delivered,

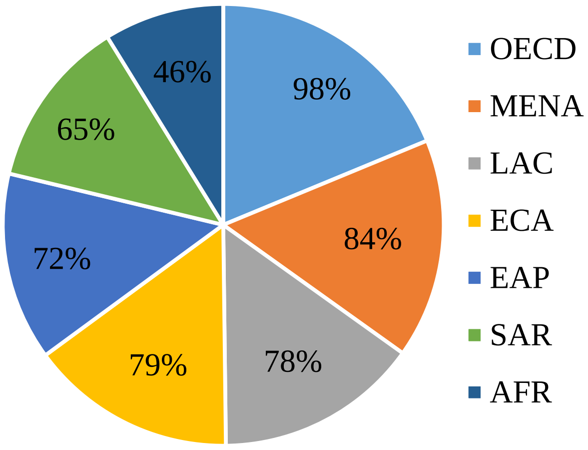


Fig. 1-4 Waste collection rates by region (Source: World Bank, 2012)

contracted or delegated service and so on. Fig. 1-4 shows the collection of MSW by rates in world regions. South Asia and Africa are the lowest with 65 % and 46 % respectively. On the other hand OECD countries have highest collection rate at 98 %.

1.2.4. Disposal strategy

Fig. 1-5 shows the approximate annual MSW disposal in the entire world reported by World Bank in 2012. Fig. 1-6 highlighted the details of MSW disposal amount and methods with variation of income levels, which has a great influence of disposal amount. Fig. 1-7 shows the difference of upper middle income and low income countries disposal methods.

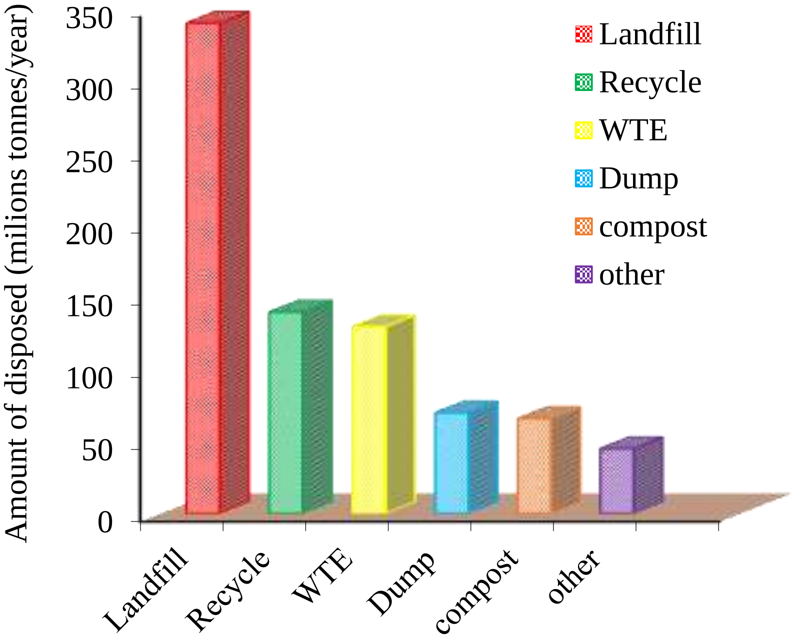


Fig. 1-5 Worldwide waste disposal strategy

(Source: Modified from World Bank, 2012)

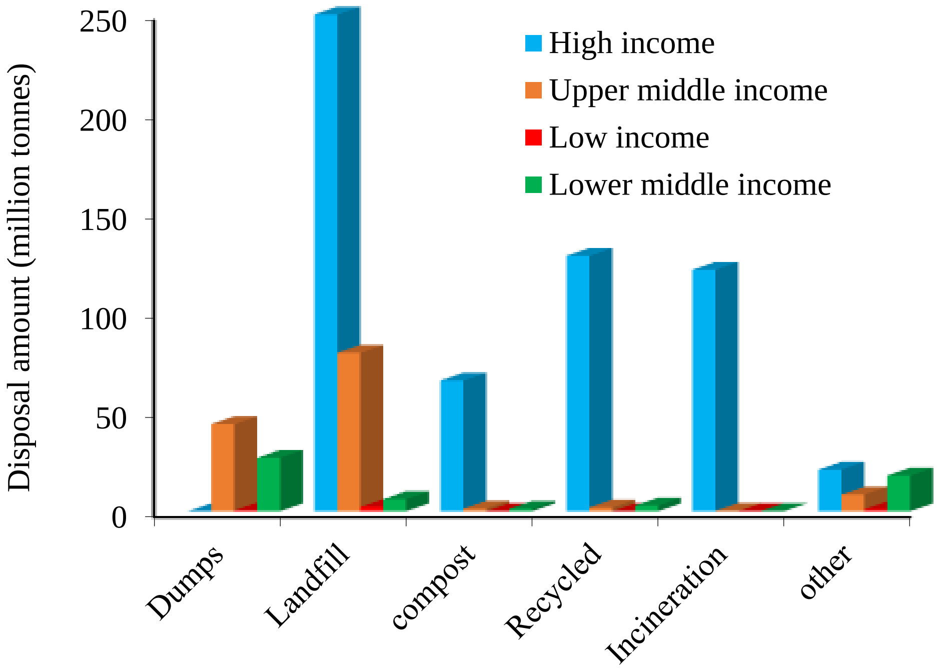


Fig.1-6 MSW disposal amount and methods with income level (Source: World Bank, 2012)

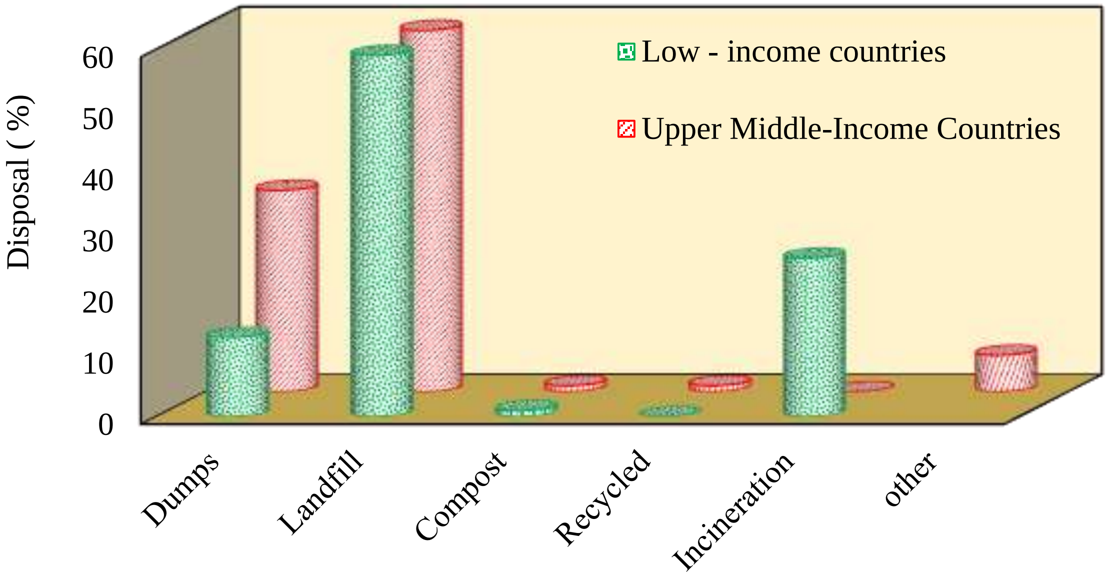


Fig.1-7 Upper and Middle income countries disposal methods (Source: Modified from World Bank, 2012)

1.3. Chronological development of concepts, technologies and management procedures for waste landfilling

It was just few decades ago uncontrolled/open dumping was the main system used in the final disposal of waste worldwide. More than half of the world’s population still apply this system as the main means of waste disposal. This waste disposal strategy is not supported by any means of modern infrastructure or environmental control and is immutable with the presence of scavengers, people who make their living from recovering marketable fractions from the deposited wastes.

In the 1960s the use of uncontrolled dumping was gradually replaced in several industrialized countries by controlled tipping, encoded by the World Health Organization in a series of guidelines used over a considerable period of time as the main technical reference point in the field.

In agreement with the principle of ”dilute and disperse” the above type of landfill provided for leachate monitoring based on attenuation throughout the unsaturated low permeability ground layers (either natural or artificial) underlying the waste. Accordingly, landfills were not sited on land characterized by highly permeable soils or at potentially vulnerable sites. Wastes were deposited in thin uncompacted layers aimed at enhancing the establishment of aerobic conditions and were covered with inert materials (preferably clay) to avoid contact between wastes and animals (dogs, birds, rodents, insects, etc.).

Provisions were laid down concerning the fencing off of landfill areas. Collection of leachate and biogas was not provided for; leachate was allowed to infiltrate into the ground, whilst biogas production was not contemplated due to the presence of aerobic conditions which were meant (optimistically) to prevent this phenomenon. In the majority of cases things did not go exactly as planned, particularly because little attention was paid to monitoring the unsaturated area and air circulation between the waste layers was extremely limited. Therefore, this type of landfill frequently resulted in noxious odors, leachate emissions from accumulation of leachate inside the landfill or in nauseating puddles around the landfill.

The repeated occurrence of similar situations and an increased public awareness of environmental issues, together with technical and scientific progress in the field, has led since the 1980s to the increasing use of "Contained landfill", featuring controlled emissions, artificial lining systems, leachate drainage and collection, waste deposition by heavy compacting, anaerobic process, biogas collection and final capping. In addition to clay as a liner, the use of synthetic membranes, initially PVC and then high density polyethylene (HDPE), became increasingly popular.

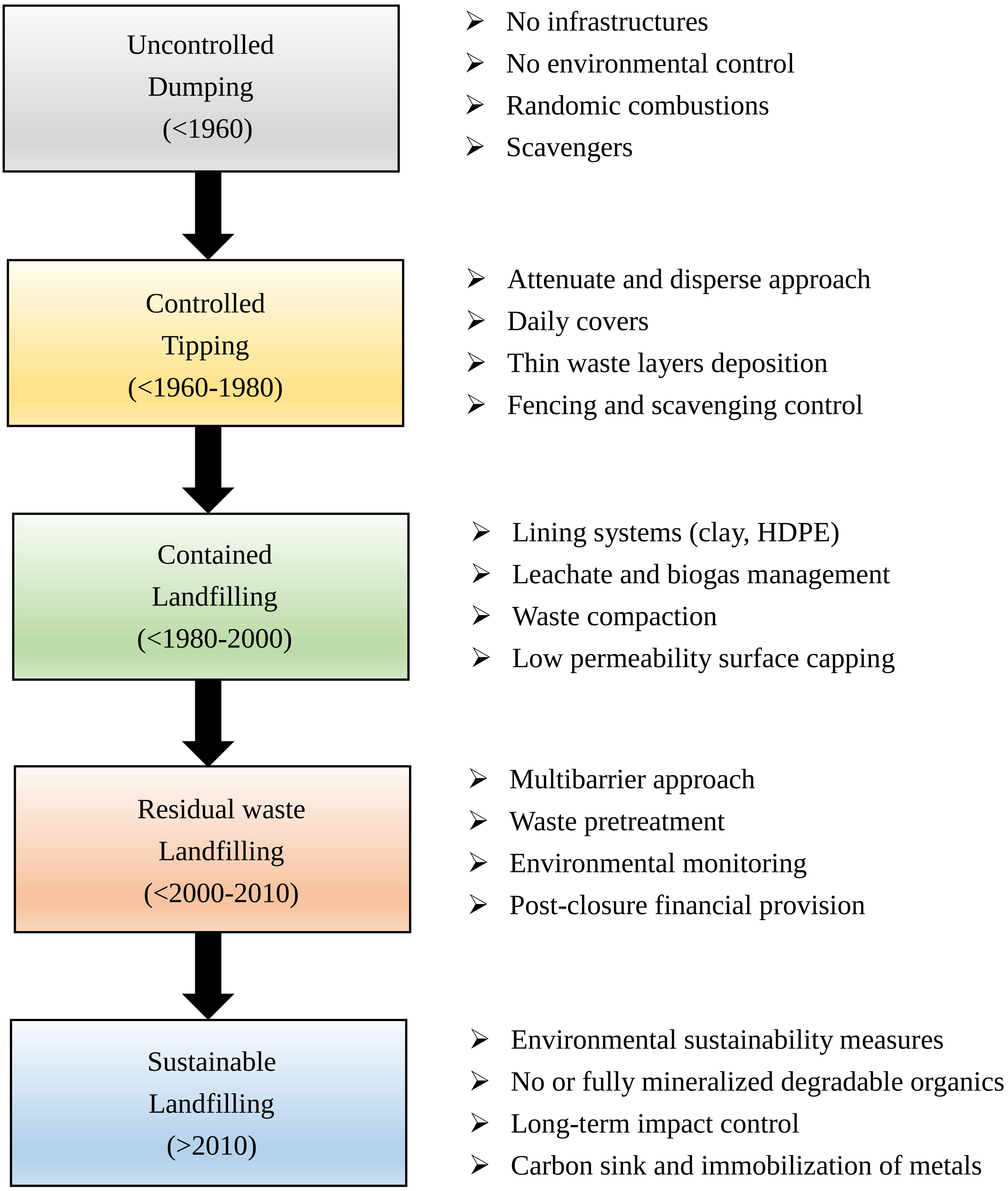
The controlled, collected emissions were naturally subjected to subsequent treatment and disposal dictating the need for specific technologies. Biogas was initially flared, although subsequently numerous recovery techniques were applied ranging from the production of thermal and electric power to upgrading of gas for domestic use and use as vehicle fuel. Increasingly complex applied treatment technologies were applied to leachate, one of the most difficult wastewaters to treat due to the inconsistency of production, high organic and ammonia content and the wide quality variation linked to landfill ageing. The simple system of aerated lagoons of the 1980s gave way to treatment options providing for high technologies of biological treatment, reverse osmosis, activated carbon absorption, chemical precipitation and oxidation, evaporation, phyto reduction, etc. (Christensen, Cossu, Stegmann, 1993).

The high management costs associated with leachate treatment have resulted in a tendency to minimize, if not to totally prevent, leachate production, thereby inhibiting the inflow of rainwater to the waste body by means of an impervious surface capping of the landfill. On the one hand the latter may solve a management issue, but on the other creates an environmental problem in view of the fact that the lack of water slows down (and even totally inhibits) biological waste degradation. This may result in a sort of waste mummification and prolonged potential for long-term impact of landfills.

Throughout the years the “Contained landfill” has displayed its numerous limitations. In particular, the “blind” acceptance of the potential efficiency of new materials has led on the one hand to an excessive use (e.g. use of multiple synthetic layers without coupling them with mineral liners), and on the other to the development of landfills on vulnerable sites on which even the least shrewd mayor of the smallest village would never have thought of siting a dump or a simple landfill. Accordingly, landfills have been developed in gravel pits, in valleys with springs, and in quarries with water seepage, etc.

The limited lasting efficacy of materials and technologies, mechanical vulnerability, inappropriate management and superficial design approach, has at times resulted in severe environmental impacts that never occurred with open dumps or old simple landfills. On referring to the reclamation of old landfills one may defame the old landfills, as major reclamation interventions are quite often performed on the “modern” contained landfills built over the last thirty years. Several examples of this can be found in Italy (i.e. Vallin dell’Aquila, La Spezia; Fossano, Cuneo, etc.)

The “Residual waste landfill” was first developed in the context of a hierarchical view of waste management and rapidly became an international reference strategy (Cossu, 2009). Subsequent to the various stages of Waste prevention, Material Reuse and Recycling, and Energy Recovery, residual wastes (ashes, non-recyclables, impurities, etc.) are formed. Although landfills continue to maintain the characteristics established for a contained landfill, they rather constitute a deposit for residual wastes. The final aim of a Residual landfill is to reduce the volumes of waste conferred, to minimize the production of greenhouse gases and generally to lower environmental impacts and risks. Moreover, the majority of industrialized countries have introduced criteria for the monitoring of landfills during the post-closure stage, proposing the



(*Source:* Cossu, R., 2015)

Fig. 1-8 Chronological development of concepts, technologies and management procedures for waste landfilling principle of financial provision and responsibility for landfill operators as long as the landfill continues to constitute a risk for the environment.

However, the evolution of the contained landfill into a residual landfill did not develop at the same pace as environmental sustainability requirements, based on avoiding leaving future generations to manage unacceptable environmental burdens.

Although present measures applied to control long-term impact (post-closure care, environmental long-term operator responsibility) are adopted by regulations, the appropriate tools for their implementation are still lacking. Indeed, termination of post-closure care is defined according to time rather than environmental performance and no criteria have been established to define acceptable conditions on which to assess operator responsibility. Furthermore, as mentioned previously, residual landfills are largely based on the technology applied for contained landfills, thus featuring the same negative characteristics ranging from the deterioration of lining and leachate drainage systems (Rollin et al., 1991; Brune et al., 1992) to the adoption of leachate minimization measures and related negative consequences.

The prevention of greenhouse gas emissions focuses mainly on the control of methane production by reducing the presence or biodegradable organic substances in waste.

Conversely, scarce importance is given to the important role of landfills in acting as “carbon sink”, permanently sequestering the non-degradable biogenic carbon present in the waste.

Lastly, the “Residual landfill”, constituting a deposit for treated wastes and process residues requires greater technical and environmental care than traditional landfills to which raw wastes are conferred. Residues may be characterized by higher concentrations of potentially polluting elements and a variation of mechanical properties that should be carefully controlled.

The modern type of landfill currently being developed on the basis of past experience, on the findings of technical and scientific research and according to ongoing environmental challenges, is the “Sustainable Landfill” incorporating the positive aspects of past landfills (e.g.

control of unsaturated background, containment and treatment of emissions, waste minimization and pretreatment) together with a coherent long-term strategy for the control of emissions and climate change issues. In particular, on the one hand the sustainable landfill aims to achieve an environmental balance over one generation (20-30 years), controlling the accumulation of mobilizable substances and uncontrolled emissions, whilst on the other hand closing the material cycle (Cossu, 2009b), thus constituting a final geological deposit for inorganic substances, and particularly for carbon associated with non-degradable or slowly degradable waste fractions such as plastics, rubber, and wood (when not most conveniently recycled) and with humic substances originating from the biological conversion of degradable. The role performed by the landfill as carbon sink is comparable to the role played by marine sediments (Bogner, 2005). Separate deposit of the high calorific fractions may also represent an intermediate storage for future energetic utilization of this material.

In order to achieve environmental sustainability in landfilling an important role is played not only by appropriate waste pre-treatment but also by in situ treatment measures such as Flushing and, when in presence of a residual biological activity, aeration. In-situ aeration, applied after Mechanical Biological Pretreatment or after intense anaerobic gas production, has proved to be an important tool in achieving Final Storage Quality, particularly when impervious top covers had been adopted previously (Stegmann and Ritzkowski, 2007).

1.4. Anaerobic Sanitary Landfill and decomposition processes

“Sanitary landfills are sites where waste is isolated from the environment until it is safe”. It is considered when it has completely degraded biologically, chemically and physically. In high-income countries, the level of isolation achieved may be high. However, such an expensive high level of isolation may not be technically necessary to protect public health. Four basic conditions should be met before a site can be regarded as a sanitary landfill. The ways of doing this should be adapted to local conditions. The immediate goal is to meet, to the best extent possible, the four stated basic sanitary landfill conditions, with a longer term goal to meet them eventually in full.

Small incremental improvements in landfill design and operation over several years are more likely to succeed than attempts to make a single, large leap in engineering expectations.

Large landfills will require more investment to improve standards than smaller sites. However, the unit cost of these improvements (measured per ton of waste landfilled or per head of population served) will decrease with increasing site size. There are financial and other benefits to sites with long operating lifetimes (ten years or more). Large regional sites serving two or more cities could be economically beneficial, providing waste transport costs are not too high.

1.4.1. Microbial Activity in Landfills

The decomposition of biowaste occurs in four stages: hydrolysis, acidogenesis, acetogenesis and methanogenesis.

Hydrolysis:

During hydrolysis, the first stage, bacteria transform the particulate organic substrate into liquefied monomers and polymers i.e. proteins, carbohydrates and fats are transformed to amino acids, monosaccharides and fatty acids respectively. Equation 1 shows an example of a hydrolysis reaction where organic waste is broken down into a simple sugar, in this case, glucose (Ostrem, 2004).

Equation 1: $C_6H_{10}O_4 + 2H_2O \rightarrow C_6H_{12}O_6 + 2H_2$

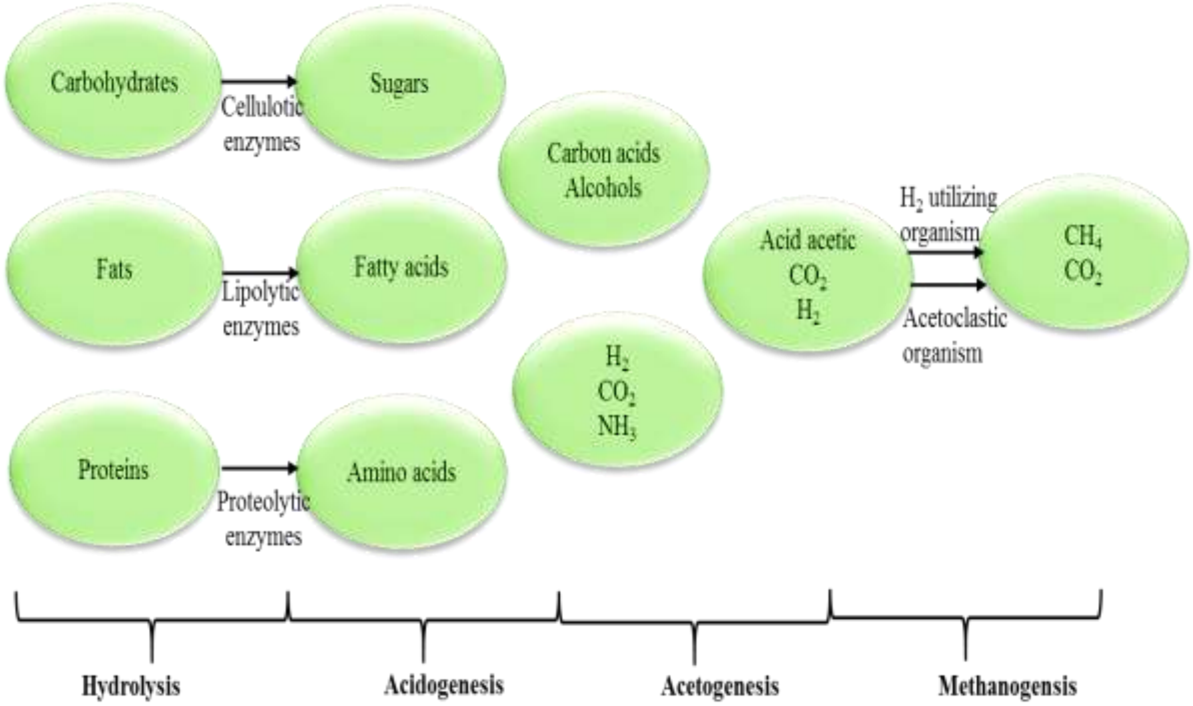


Fig. 1-9 Degradation steps in anaerobic digestion process

Acidogenesis:

In the second stage, acidogenic bacteria transform the products of the first reaction into short chain volatile acids, ketones, alcohols, hydrogen and carbon dioxide. The principal acidogenesis stage products are propionic acid (CH_3CH_2COOH), butyric acid ($CH_3CH_2CH_2COOH$), acetic acid (CH_3COOH), formic acid ($HCOOH$), lactic acid ($C_3H_6O_3$), ethanol (C_2H_5OH) and methanol (CH_3OH), among other. From these products, the hydrogen, carbon dioxide and acetic acid will skip the third stage, acetogenesis, and be utilized directly by the methanogenic bacteria in the final stage. Equations 2, 3 (Ostrem, 2004) and 4 (Bilitewski et al., 1997) represent three typical acidogenesis reactions where glucose is converted to ethanol, propionate and acetic acid, respectively.

Equation 2: $C_6H_{12}O_6 \leftrightarrow 2CH_3CH_2OH + 2CO_2$

Equation 3: $C_6H_{12}O_6 + 2H_2 \leftrightarrow 2CH_3CH_2COOH + 2H_2O$

Equation 4: $C_6H_{12}O_6 \rightarrow 3CH_3COOH$

Acetogenesis:

In the third stage, known as acetogenesis, the rest of the acidogenesis products, i.e. the propionic acid, butyric acid and alcohols are transformed by acetogenic bacteria into hydrogen, carbon dioxide and acetic acid. Hydrogen plays an important intermediary role in this process, as the reaction will only occur if the hydrogen partial pressure is low enough to thermodynamically allow the conversion of all the acids. Such lowering of the partial pressure is carried out by hydrogen scavenging bacteria, thus the hydrogen concentration of a digester is an indicator of its health (Mata-Alvarez, 2003). Equation 5 represents the conversion of propionate to acetate, only achievable at low hydrogen pressure. Glucose (Equation 6) and ethanol (Equation 7) among others are also converted to acetate during the third stage of anaerobic fermentation (Ostrem, 2004).

Equation 5: $CH_3CH_2COO- + 3H_2O \leftrightarrow CH_3COO- + H+ + HCO_3- + 3H_2$

Equation 6: $C_6H_{12}O_6 + 2H_2O \leftrightarrow 2CH_3COOH + 2CO_2 + 4H_2$

Equation 7: $CH_3CH_2OH + 2H_2O \leftrightarrow CH_3COO- + 2H_2 +H+$

Methanogenesis:

The fourth and final stage is called methanogenesis. During this stage, microorganisms convert the hydrogen and acetic acid formed by the acid formers to methane gas and carbon dioxide (Equations 8, 9 and 10) (Verma, 2002). The bacteria responsible for this conversion are called methanogens and are strict anaerobes. Waste stabilization is accomplished when methane gas and carbon dioxide are produced.

Equation 8: $CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$

Equation 9: $2C_2H_5OH + CO_2 \rightarrow CH_4 + 2CH_3COOH$

Equation 10: $CH_3COOH \rightarrow CH_4 + CO_2$

These phases are not sharply defined within a landfill, the stages tend to overlap as lifts of MSW are added. Because of the heterogeneous nature of the MSW, pockets of microbial activity form, causing the different stages of decomposition to occur side-by-side within the landfill. Bioreactor landfills attempt to increase the rate of the first four phases. This is done by providing the waste with water and nutrient rich leachate during the transition and acidogenesis phase. When the MSW in the bioreactor landfill is in the adjustment stage, the easily degradable waste such as food waste and other easily degradable organic matter serves as the carbon source for bacteria and the oxygen in the air serves as the electron acceptor resulting in the following reactions:

Degradable waste + oxygen = CO_2 + H_2O + heat + biomass + Acetic Acid + Residuals CO_2 + H_2O => H_2CO_3 (Carbonic Acid)

As oxygen is consumed, the waste gradually enters the transition phase where the complex organic matter is broken into simpler organic acids driving the MSW into the acidogenesis stage. During the acidogenesis phase volatile organic acids (VOAs) reach their peak concentration within the landfill. Water and leachate are added to the MSW when it is in the transition and acidogenesis stage. As the MSW enters an anaerobic environment the anaerobic bacteria utilize other electron acceptor like nitrates, sulfates and mixtures of other wastes to further degrade the MSW. The additional moisture provides an ideal environment for the anaerobes, and the bacteria in the leachate provide an extra seeding of bacteria to degrade the organic matter. Additionally, nitrates also serve as a sink for hydrogen as reduction to ammonia occurs within bioreactor landfills. The ammonia generated within landfills usually exceeds the microbial nutrient requirements, producing a leachate rich in ammonia requiring further treatment and stabilization (Kjeldsen et al. 2002). Through leachate quality monitoring and recirculation, a system analogous to an attached growth anaerobic process is developed within bioreactor landfills allowing microbial populations to develop and proliferate until the substrate is depleted or environmental conditions become limiting (Pohland and Kim 2000). As the alternative electron acceptors are used up, the methanogenic bacteria ferment the organic acids to methane and carbon dioxide. Methanogens start utilizing carbon dioxide as their terminal 4electron acceptor and consume the high concentration of hydrogen ions that were produced during the acidogenesis phase to produce methane, resulting in the following reactions:

$$4H_2 + CO_2 = CH_4 + 2H_2O \quad CH_3COOH = CH_4 + CO_2$$

As the available carbohydrates are consumed the MSW enters the maturation phase and is then relatively inert (Christensen and Kjeldsen 1989; Gurijala and Suflita 1993; Shearer 2001; Hater et al. 2003). Within the landfill, grass, leaves, and branches are the major contributors of refuse decomposing microbes to landfills, each contributes and average of 9.8, 6.3 and 6.5 Most Probable Number – log10 cells/dry g (MPN) respectively (Barlaz et al. 1997). A number of species belonging to the genus Clostridium have been found to be present in the leachate of 17 different landfills (Van Dyke and McCarthy 2002), this species is also common in the rumen of cow stomachs (Tajima et al. 2000). As microbial activity increases within bioreactor landfills, the temperature rises. This correlation is shown in the Van't Hoff-Arrhenius equation (Metclaf and Eddy 1991):

$$kt = k_{20} * \theta^{(T-20)}$$

Where:

k_t = degradation rate constant at a particular temperature (-)

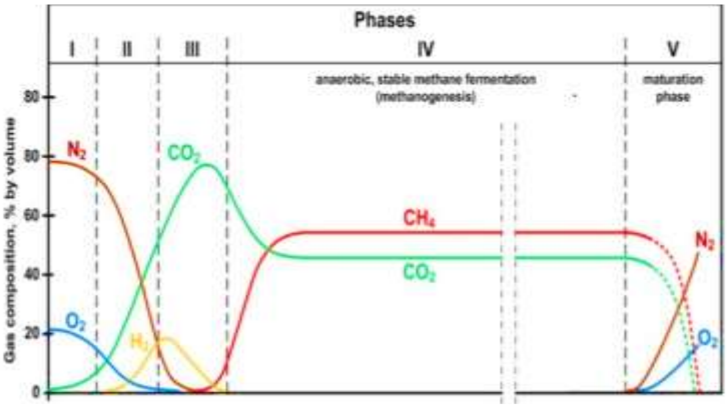
k_{20} = degradation rate constant at 20° C (0.23 is the typical value) (-)

θ = constant of 1.056 for temperatures between 20 and 30° C

T = temperature for which k is desired (° C)

With more microbial activity the temperature of the landfill naturally rises. The accumulation and distribution of moisture through leachate recirculation increases reaction rates within landfills, allowing for an increased rate of in situ waste conversion and leachate treatment. Likewise, methane generation occurs before landfill closure so the methane can be harvested and utilized for energy generation, instead of after closure where it is emitted as a green-house gas into the atmosphere. The biochemical methane potential (BMP) is an important parameter that can be used to determine MSW stability. The BMP of a sample is the amount of methanogenic degradation still possible for a sample. A high BMP indicated that the waste is still active, containing an easily available carbon source while a low BMP indicates inertness and low carbon availability (Shearer 2001).

A traditional method to determine if the landfill has entered the maturation phase is the cellulose (C) to lignin (L) ratio. A lower C/L ratio indicates stability. Factors that contribute to the slow rate of paper degradation in landfills are moisture limitation, poor shredding, low cellulose/lignin ratio (high lignin content) and the lack of inoculum (Pohland and Kim 2000). As the landfill stabilizes and reaches the maturation phase, the cellulose is consumed by the anaerobes and their concentration is minimized. When compared to the relatively inert lignin, one can determine the extent of the cellulose degradation or MSW stability. However, recent research has shown that lignin does degrade under the high temperatures and moist conditions present in bioreactors landfills. A better understanding of lignin and the anaerobic conditions



I = Initial adjustment, aerobic conditions
II = Transition phase, begin of anaerobic decomposition
III = Acid phase, hydrolysis and acidogenesis
IV = Methane fermentation phase, strictly anaerobic, methanigenesis
V = Maturation phase: air intake, methane oxidation to CO_2 and air

Fig. 1-10 Phases of landfill life

under which lignin degrades is crucial in determining the level of landfill gas production, settling rates and post closure monitoring requirements. Fig. 1-10 illustrate the phases of landfill life.

1.5. Bioreactor landfills

Bioreactor landfills are sanitary landfills that use microbiological processes purposefully to transform and stabilize the biodegradable organic waste constituents in a shorter period of time.

The Solid Waste Association of North America (SWANA), define a bioreactor landfill is "a controlled landfill or landfill cell where liquid and gas conditions are actively managed in order to accelerate or enhance biostabilization of the waste. The bioreactor landfill significantly increases the extent of organic waste decomposition, conversion rates, and process effectiveness over what would otherwise occur with the landfill."

The USEPA Clean Air Act regulations (40 CFR 63.1990, National Emissions Standards for Hazardous Air Pollutants) define a bioreactor landfill as "a MSW landfill or a portion of a MSW landfill where any liquid, other than leachate or landfill gas condensate, is added in a controlled fashion into the waste mass (often in combination with recirculating leachate) to reach a minimum average moisture content of at least 40% by weight to accelerate or enhance the anaerobic biodegradation of the waste."

Bioreactor landfills are therefore the controlled system in which moisture addition (often leachate recirculation) and/or air injection are used to create a solid waste environment capable of actively degrading the readily biodegradable organic fraction of the waste. Several researchers have documented the benefits associated with bioreactor technology (Murphy et al., 1995; Pohland et al., 1995; Reinhart et al., 1996). One advantage is that increased waste degradation rates characteristic of bioreactor landfills permit the life of a bioreactor landfill to be expanded beyond that of conventional landfills through recovery of valuable airspace. As leachate is recirculated, it is treated in situ, decreasing its organic strength and thus potential impact to the environment. In situ treatment potentially reduces the length of the postclosure care period and associated costs (Reinhart et al., 1998). Additionally, bioreactor landfills stimulate gas production; the majority of the methane is produced earlier in the life of the landfill, allowing for more efficient capture and subsequent use (Berge et al., 2005).

Although the organic strength of the leachate is significantly reduced in bioreactor landfills, ammonia-nitrogen remains an issue. The ammonia-nitrogen concentrations found in leachate from bioreactor landfill are greater than those found in leachate from conventional landfills (Onay et al., 2001). Ammonia-nitrogen tends to accumulate in both system because there is no degradation pathway for ammonia-nitrogen in anaerobic system. However, in bioreactor landfills, moisture addition and recirculating leachate increases the rate of ammonification, resulting in accumulation of higher levels of ammonia-nitrogen, even after the organic fraction of the waste is degraded (Barlaz et al., 2002).

The increased ammonia-nitrogen concentrations intensifies the toxicity of the leachate to aquatic species, potentially inhibiting the degradation processes and necessitating leachate treatment before ultimate disposal to protect receiving waters (Burton et al., 1998). It is been suggested that ammonia-nitrogen is one of the most significant long-term pollution problem in landfills, and it likely that the presence of ammonia-nitrogen will determine when the landfill is biologically stable and when post closure monitoring may end (Price et al., 2003).

Because bioreactor landfill environments are different from conventional landfills, there is a potential for a greater number of nitrogen transformation and removal processes to occur and for them to occur to a greater extent than in conventional landfills. System design of bioreactor landfills provides the flexibility in the location and duration of liquid and air injection, allowing for adjustment of pH, oxidation-reduction potential, and moisture content to create an environment conducive to microbial degradation and biological nitrogen removal (Berge et al., 2005).

Liquid addition to landfill has many advantages associated with it. Leachate recirculation involves the collection and redistribution of leachate trough a landfill. Moisture addition and movement are important factors affecting waste biodegradation, resulting in an increase in the moisture content of the waste and distribution of nutrients throughout the landfill, respectively. Optimal levels of moisture content have been found to be between 40 and 70%, on a weight basis (Barlaz et al., 1990).

Air addition has also been used as an enhancement and has been shown to enhance degradation processes in landfills at both the field and laboratory scale (Leikam et al., 1999). Adding air uniformly throughout the waste is also a challenge. Not only waste heterogeneities and compaction affect the air distribution, the presence of moisture does as well. Air will take the path of least resistance; thus, there will likely be areas of an aerobic landfill in which air does not reach, resulting in anoxic or anaerobic pockets within the waste mass (Berge et al., 2005).

Generally, bioreactor landfills undergo the same degradation processes as conventional landfills, just at faster rate and to a greater extend because of the optimization of in situ conditions. However, degradation pathways may vary depending on the operation of bioreactor landfill. Compared with conventional landfills, bioreactor landfills have shown a more rapid and complete waste conversion and stabilization process (Harper et al., 1988).

1.5.1. Anaerobic bioreactor landfill

Anaerobic bioreactor landfills is that in which moisture addition is practiced. Sources of liquid may include groundwater, storm water, infiltrating rainfall, or leachate. Adjustment of moisture content results the methane production, which has been repeatedly demonstrated in several laboratory, pilot, and field scale studies. In anaerobic bioreactors, waste degradation is enhanced and organic material is returned to the landfill via leachate recirculation, methane is generation is much faster rate. The total volume of gas produced also increases, as organics in the leachate are recycled and then biodegraded within the landfill. The majority of gas production may be confined to a few years, earlier in the life of the landfill, than traditionally occurs in conventional landfills, allowing for a more efficient capture and subsequent use (Reinhart et al., 1996).

Anaerobic bioreactor landfills are more effective at degrading the solid waste than conventional landfill. However, when compared to other types of bioreactor landfills, anaerobic system tend to have lower temperatures and slower degradation rates (Merz et al., 1970). Accumulation of ammonia-nitrogen is a disadvantage to operating the landfill as an anaerobic reactor. In anaerobic bioreactor landfills, the ammonia-nitrogen present in the leachate is continually returned to the landfill, where there is no degradation pathway for ammonia in anaerobic

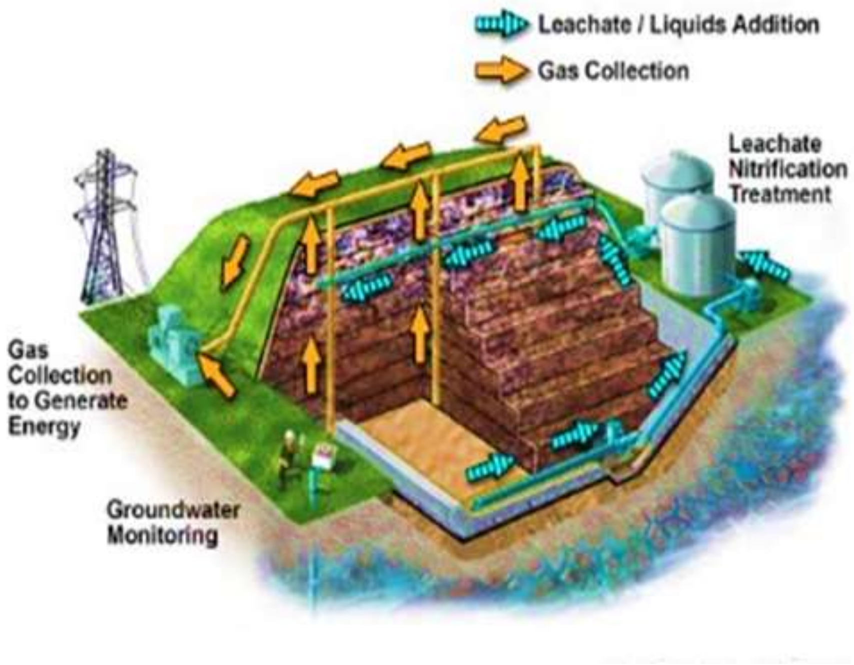


Fig. 1-11 Anaerobic bioreactor landfill

(Source: https://www.wm.com/sustainability/bioreactor-landfills/bioreactor-technologies.jsp)

environments. An advantage of operating the reactors anaerobically when compared with other bioreactor landfill types is that air is not added; therefore the operational cost are less than what would be incurred aerobically and methane can be captured and reused.

1.5.2 Semi-aerobic bioreactor landfill

The semi-aerobic landfill method was first proposed by Masataka Hanashima of Fukuoka University, Japan in 1975. In this landfill method, the pebbles and leachate collection pipes are installed at the bottom of the landfill for leachate drainage. Due to the heat generated by microorganism metabolism in landfill, the heat convection is created by the temperature difference between the internal and external. Consequently, the air can flows into the landfill

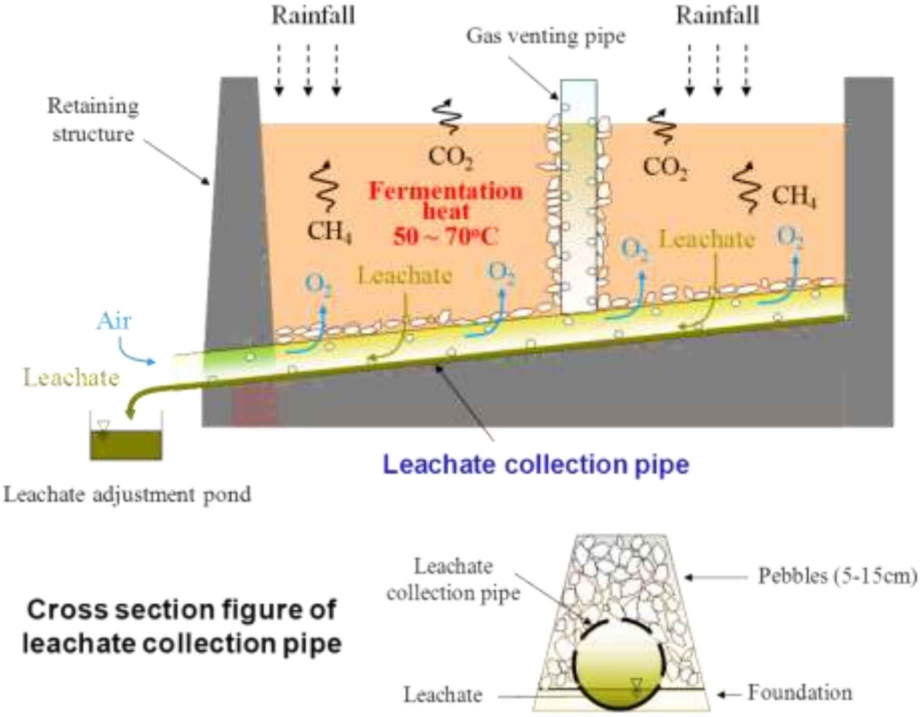


Fig. 1-12 Schematic diagram of Semi-aerobic landfill

(Source: http://nett21.gec.jp/waste/data/waste_L-1.html)

in the reverse direction of the leachate flow through leachate collection pipes. Therefore, this landfill method is easy to construct as well as operate and maintain since forced aeration facility is omitted. Moreover, the advantages that leachate collection pipes bring for the landfill system include (Shimaoka et al., 2000): firstly, it prevents leachate from stagnating in waste matrix and therefore promotes aerobic condition in the waste layer; secondly, the activity of aerobic microorganism is encouraged. And therefore it accelerates the decomposition rate of the solid waste, improves the leachate quality as well as reduces methane gas emission; thirdly, the strength of the collection pipes are complemented due to parallel utilization of the collection pipes and the pebbles; fourthly, water pressure on the liner is prevented because of the rapidly drainage of the leachate.

In semi-aerobic landfill, solid waste will be decomposed aerobically in this area where oxygen can be penetrated into. Organic carbon will be converted into carbon dioxide. However, air is hard to penetrate through the entire waste matrix. Therefore, part of the area is still under anaerobic condition. Methane will be still generated. Moreover, the anaerobic zone of semi-aerobic landfill will expand if the drainage of the leachate is insufficient or water table of leachate is too high. Besides, blocking of the aeration holes by plastic bags may also lead to the failure of this landfill scenario (Matsufuji and Tachifuji, 2007). The semi-aerobic landfill method requires appropriate design and operation in order to maintain the state of the waste layer in semi-aerobic conditions. The refuse which contains high concentration of organic matter will cause uneven settlement of the landfill, resulting in the bending and blocking of the leachate collection pipe (Jiao, 2011). Therefore, it is not suitable for semi-aerobic landfills.

Currently, recovery of the aged landfill or minimization of the post-closure management duration has drawn much attention from the all over the world. One of the most effective measures is leading air into the waste layer. The semi-aerobic landfill can meet the economic requirement due to the low operating cost. The aged landfill which operated under anaerobic condition and already installed leachate collection and gas ventilation system can be easily transformed into semi-aerobic landfill. However, for the landfill without such system, it may not be feasible and economical to install the whole pipe systems after the final soil cover work has been applied. Therefore, the semi-aerobic landfill does not suitable for open dumping site remediation.

1.5.2. Aerobic bioreactor landfill

Introducing air to landfill has been shown to enhance degradation processes in landfills, as aerobic processes tend to degrade organic compounds typically found in municipal solid waste (MSW) in shorter periods than anaerobic degradation processes (Leikam et al., 1999). Reported advantages of operating the landfill aerobically include increased settlement, decreased metal mobility, reduced ex situ leachate treatment required, lower leachate management and methane control costs, and reduced environmental liability (Read et al., 2001).

Many of the nitrogen transformation and removal process are favored by aerobic processes, including nitrification and ammonia air stripping or volatilization. Air stripping and volatilization may be favored in aerobic bioreactor landfill because of higher pH levels and temperatures that are inherent in an aerobic environment. The additional gas flow associated with air injection may also induce greater masses of ammonia-nitrogen removal (Berge et al., 2005).

During aerobic degradation of MSW, biodegradable materials are converted mostly to carbon dioxide and water. Little, if any, methane is produced, which may be viewed as either an advantage or disadvantage, depending on whether methane collection and use as an energy source is desired or required. Methane is a potent greenhouse gas; thus, if it cannot efficiently controlled and collected in anaerobic landfills, its production can be a local environment concern. Further, the solid waste environment during aerobic degradation has a fairly neutral pH, which decreases metal mobility (Hanashima et al., 1999). Volatile organic acid production is decreased in aerobic bioreactors because the anaerobic fermentation processes are limited. However, volatile acid and methane production may still occur in anaerobic pockets within the landfill (Berge et al., 2005).

The aerobic processes generate a considerable amount of heat, leading to elevated in situ temperatures as high as 66 °C (Stessel et al., 1992). The elevated temperatures increase the evaporation, which results in a significant loss of leachate. As a consequence, there is less leachate to manage. The high temperatures may limit certain biological nitrogen transformation processes from occurring, although no data regarding temperature effects are available. Additionally, the combination of the high temperatures and presence of any air may create a fire potential. However, minimizing methane production and ensuring proper moisture contents, fire potential is lessened (Berge et al., 2005).

Odor often associated with anaerobic systems, such as from hydrogen sulfide and volatile acids, are reduced in aerobic bioreactor landfills. Aerobic processes do have some odor.

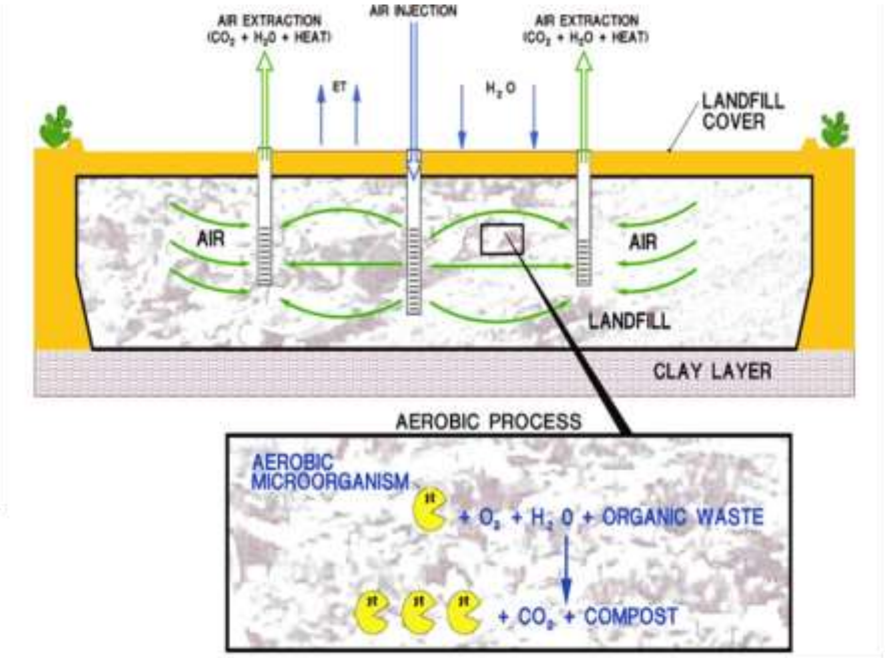


Fig. 1-13 Aerobic landfill mechanism

(Source: http://www.hgcinc.com/landfillAABE.htm)

associated with them; however, it is an earthy smell. Some odorous compounds emitted by aerobic composting include methanethiol, which has a pungent sulfide odor (Miller et al., 1992)

1.5.3.1. The basic principle of in situ aeration

The basic principle of aeration and waste gas collection is shown in Fig. 1-14. For an air supply under low excess pressure, ambient air is pressed into the landfill body by means

of aeration wells. Air resp. atmospheric oxygen is distributed by convection and diffusion processes in the landfill body. Thus, an aerobization of the whole landfill body and an accelerated degradation of the organic waste components is achieved depending on rate and duration of aeration. Waste gas is collected and treated by means of a gas collection system (gas wells with identical design). Slightly permeable / impermeable underground ground water layer aeration aggregate exhausts collection and treatment.

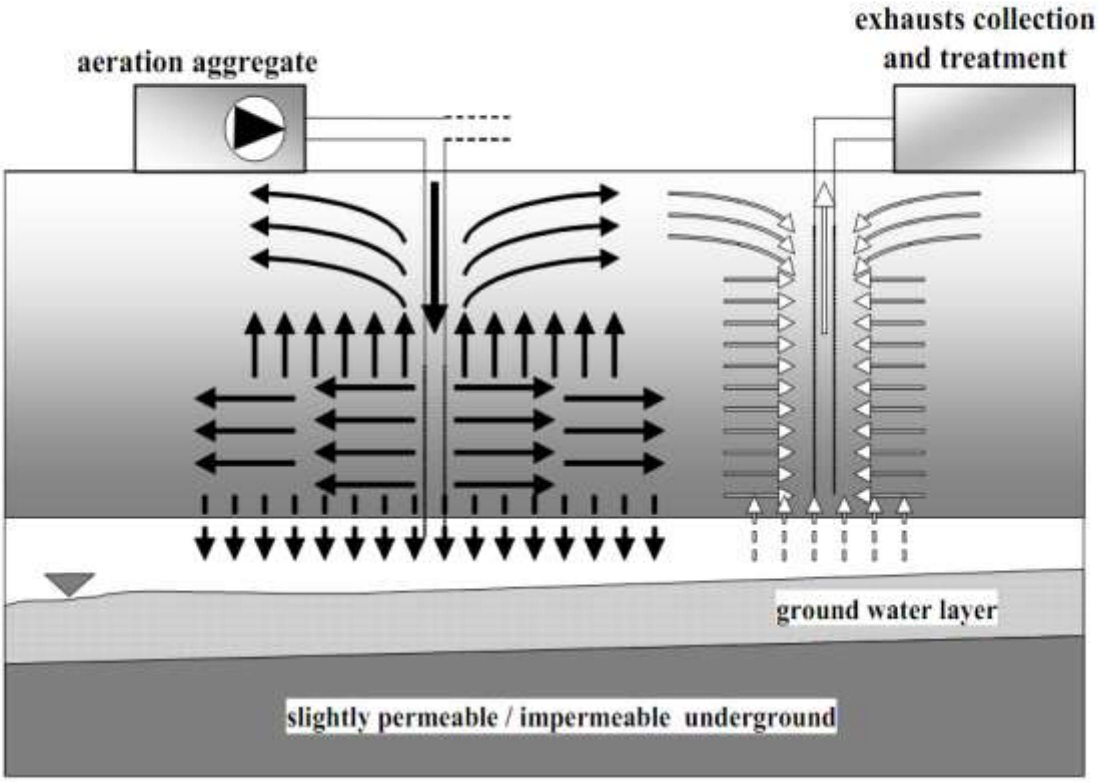


Fig. 1-14 Fundamental concept of remediation by aeration (Heyer et al; 2001)

1.5.3.2. Technical concept of low pressure aeration

The basic technical concept of the aeration of the landfill body consists of a system of gas wells, through which atmospheric oxygen is led into the landfill body via active aeration in such a way that can accelerated aerobic stabilization of deposited waste is realized. Simultaneously, the low contaminated waste gas is collected and treated in a controlled manner by means of further wells. Aeration is effected using low pressures and is continuously adjusted to meet the oxygen demand so that energy consumption is low and constantly optimized.

The landfill aeration technology has been widely used among the world in achieving different objectives (for example reduction of emission potential and aftercare period (Heyer et al., 2005), remediation of abandoned site (Cossu et al., 2003), and odor reduction (Jacobs et al., 2003)), and have found the results to be successful. However, there are still some uncertain issues or questions need to be satisfactorily addressed. The first one is related to financial cost. Although some investigation (Rich et al., 2008) shows that landfill aeration will benefit from multiple aspects such as shorten the long-term environmental costs in anaerobic landfill and landfill recover, finding a more suitable aeration concept with high oxygen utilization rate is still necessary; the second issue is about the behavior of heavy metal in leachate phase and release of non-methane organic compounds in gas phase after introducing air into a landfill. In general, the mobility of heavy metals in solid waste is high in the initial aerobic phase (Phase I) of the anaerobic landfill, but decrease as the oxygen levels decline since heavy metals tend to be retained within the waste under the reducing conditions of the anaerobic stages (Kjeldsen et al., 2002). According the research made by Kim, 2005, heavy metals, such as Al, Cu, Cr and Pb, are not immobilized in aerobic landfill as they are in the anaerobic landfill, which will lead to problems with the leaching of these toxic elements; the third issue is the rising temperature after air injection. The rising temperature as result of exothermic processes is commonly observed after air is introduced into the landfill. Temperature profile at some areas in the landfill was increased up to 70 °C (Laux et al., 2010). In order to avoid the combustion risk, landfill aeration has to stop and waits for the reduction of temperature in waste layer, or switch to a discontinuous aeration mode (Oncu et al., 2012). In this case, anaerobic degradation may be restart as indicated by the gas composition (Oncu et al., 2012).

1.5.4. Hybrid bioreactor landfill

The hybrid bioreactor landfill was operated with the combination of aerobic and anaerobic conditions, which included short-term cycling of air injection into landfill and sequencing of aerobic and anaerobic conditions. The removal of ammonia from the leachate often remained a challenge in anaerobic bioreactor landfill management. Although the organic compounds are significantly reduced in a shorter time period, the ammonia of leachate still keeps a trouble due to the lack of nitrogen degradation pathway in the anaerobic bioreactor landfill. The co-existence of aerobic, anoxic and anaerobic circumstances made in situ ammonia removal feasible in the hybrid landfill. Some researchers reported that the ammonia of leachate could be greatly reduced in the cyclic air injection type of hybrid landfill (Onay and Pohland, 1998, Price et al., 2003 and He and Shen, 2006). Furthermore, the fact that the methane production in the hybrid landfill was inhibited was also documented. However, little research has been conducted evaluating the fate of nitrogen in the hybrid landfill.

There are three types of these aerobic/anaerobic systems have been explored: short term cycling air injection landfill concept, spatial sequencing of aerobic and anaerobic landfill concept and temporal sequencing of anaerobic and aerobic landfill concept.

In this landfill, landfill is firstly operated under anaerobic condition as anaerobic bioreactor. After a certain period, depending on the composition of the MSW, methane concentration reduces to a relative level, recover of methane becomes unprofitable. The landfill system then switches into aerobic type. The late aeration of the landfill allow the anaerobic conditions in early stage to fulfill various important functions such as immobilization of heavy metals, initial degradation of certain pesticides and production of large quantities of methane for energy recovery. Therefore, some of the benefits of anaerobic phase of landfill are retained. However, the long aftercare period of anaerobic phase is reduced and the advantages of late aeration, such as ammonium removal remain.

1.5.5. Facultative bioreactor landfill

The Facultative Bioreactor combines conventional anaerobic degradation with a mechanism for controlling the high ammonia concentrations that may develop when liquids are added to the landfill. In this system, leachate containing elevated levels of ammonia is treated using the biological process of nitrification. The nitrification process converts the ammonia in the leachate to nitrate. The treated leachate is then added to the landfill. Here certain microorganisms, including the facultative bacteria, can use the nitrate in the absence of oxygen for respiration.

1.6. The Aerobic-Anaerobic Landfill Method

The Aerobic-Anaerobic Landfill Method, a novel landfill method is developed in bioreactor landfill group (Wu et al., 2013). In this method, air is injected into an anaerobic-type landfill at the appropriate depths/layers to create aerobic zones, whereby alternative aerobic-anaerobic conditions (hybrid conditions) are created vertically in the landfill. It is expected that: first, the conversion efficiency of nitrogen can be accelerated due to the existence of alternative aerobic-anaerobic conditions; second, the aerobic zones/conditions may accelerate the decomposition of organic carbon as well as reduce the emission of GHGs; and third, hybrid conditions may create biostabilized landfills, thereby reducing the need for expensive perpetual landfill aftercare.

Aerobic-Anaerobic Landfill Method have focused on aerobic and anaerobic conditions that are alternately created along the vertical direction of a landfill by applying continuous air injection. By contrast, intermittent aeration that causes a single landfill layer to alternate temporally between aerobic and anaerobic conditions has not been discussed.

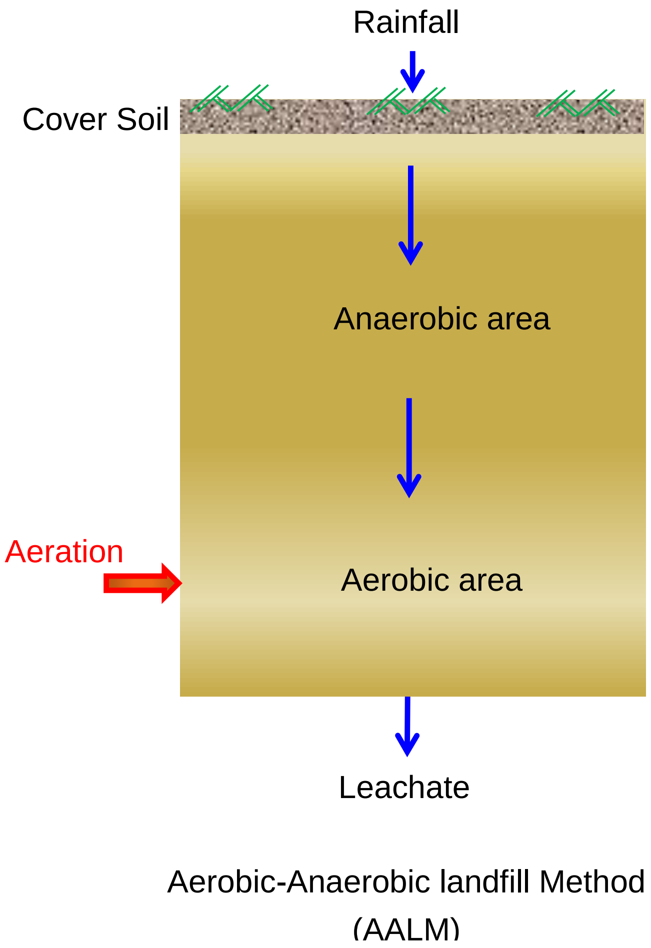


Fig. 1-15 Concept of Aerobic-Anaerobic Landfill Method

In an aerated landfill bioreactor, carbon and nitrogen were effectively removed from the solid waste and leachate (Sang et al., 2008). However, oversupply or ineffective aeration can decrease microbial activity and increase energy consumption. A balanced and effective utilization of both aerobic and anaerobic metabolic pathways of the microorganisms might be effective for accelerated stabilization.

In Aerobic-Anaerobic Landfill Method, aerobic-anaerobic conditions were alternately created in specific layers of landfill by periodical air injection as shown in Fig. 1-15. Apparently intermittent aeration is the most practical strategy to create cyclically aerobic and anaerobic conditions and to reduce energy consumption inferable to the air supply in landfill bioreactors. Rapid stabilization of solid waste is possible with intermittent aeration at various oxygen and oxidation–reduction potential levels (Mertoglu, 2007). Reportedly, an intermittent aeration strategy is favorable for separation of the acid formation phase and the methane fermentation phase, reducing the acid production time. Furthermore, intermittent aeration can stimulate nitrifiers and denitrifiers in landfill bioreactors. Some results showed that cyclic air injection system biologically stabilized the leachate in a shorter time than purely aerobic system (Berge, 2001, Pichler and Kogner-Knabner, 2000 and Reinhart et al., 2002). It was also reported that cyclic air injection system could reduce a mass of contaminants in a shorter time (Dong et al., 2007). But, there are very few studies have been done by intermittent aeration in landfill methods and almost no research has been done to investigate the transition period phenomenon during organic waste decomposition process between aeration and non-aeration periods.

1.7. Research objectives and outline of dissertation

The Aerobic-Anaerobic Landfill Method is developed for acceleration of waste degradation by altering the craftworks of the aerobic and anaerobic conditions inside the landfill. The objective of the alternative sequential aerobic-anaerobic treatment is to cause the rapid biodegradation of organic waste and other easily degradable waste in the aerobic stage in order to reduce the production of organic acids in the anaerobic stage resulting in the earlier onset of methanogenesis. In this system the inside of the landfill waste is aerated during aeration period, while the non-aeration period the aerobically decomposed waste become anaerobic condition. Therefore, the process of nitrification and dentrification occur squencencially and/or simultaneously inside the landfill. To clarify the effectiveness of application of intermittent aeration as described by the AALM, column experiments were performed in which aerobic and anaerobic conditions were alternately created in a specific layer of a column via intermittent air injection, the specific research objectives of this research are:

1) To investigate the impact of intermittent aerations on landfill leachate quality and greenhouse gas reduction;

2) To investigate the nitrification and denitrification phenomena during transition periods of aeration and non-aeration;

3) To investigate the influence of nitrification conditions on nitrous oxide production from organic solid waste;

4) To investigate the nitrous oxide production potentials with *in-situ* aeration in a closed landfill site.

The objectives of this study were met by implementing of laboratory and field study. The achievements were organized into four chapters. Besides, one chapter is used to make a brief introduction about the development of landfill technologies worldwide (Chapter 1), and one additional chapter (Chapter 6) is used to conclude the whole research work. The structure of the dissertation can be schematized as in Fig. 1-16.

Chapter 1 presents the reviewed current status of MSW including its quantity, composition and disposal strategies. Then, bioreactor landfill technologies, including their advantages and disadvantages, were also introduced. Finally, the objectives and structures of the study were presented.

Chapter 2 presents the results of lab-scale column experiments aiming at investigate the impact of intermittent aerations on leachate quality improvement and greenhouse gases abatement of organic solid waste from landfill adopted the AALM.

Chapter 3 presents the transition periods phenomena between aerobic and anaerobic conditions conducted by lab-scale column experiment in AALM.

Chapter 4 presents the results of batch experiment for the production of nitrous oxide in nitrification conditions from organic solid waste.

Chapter 5 presents a field-study results of nitrous oxide production by in-situ aeration.

Chapter 6 concluded all experimental results and few recommendations introduced.

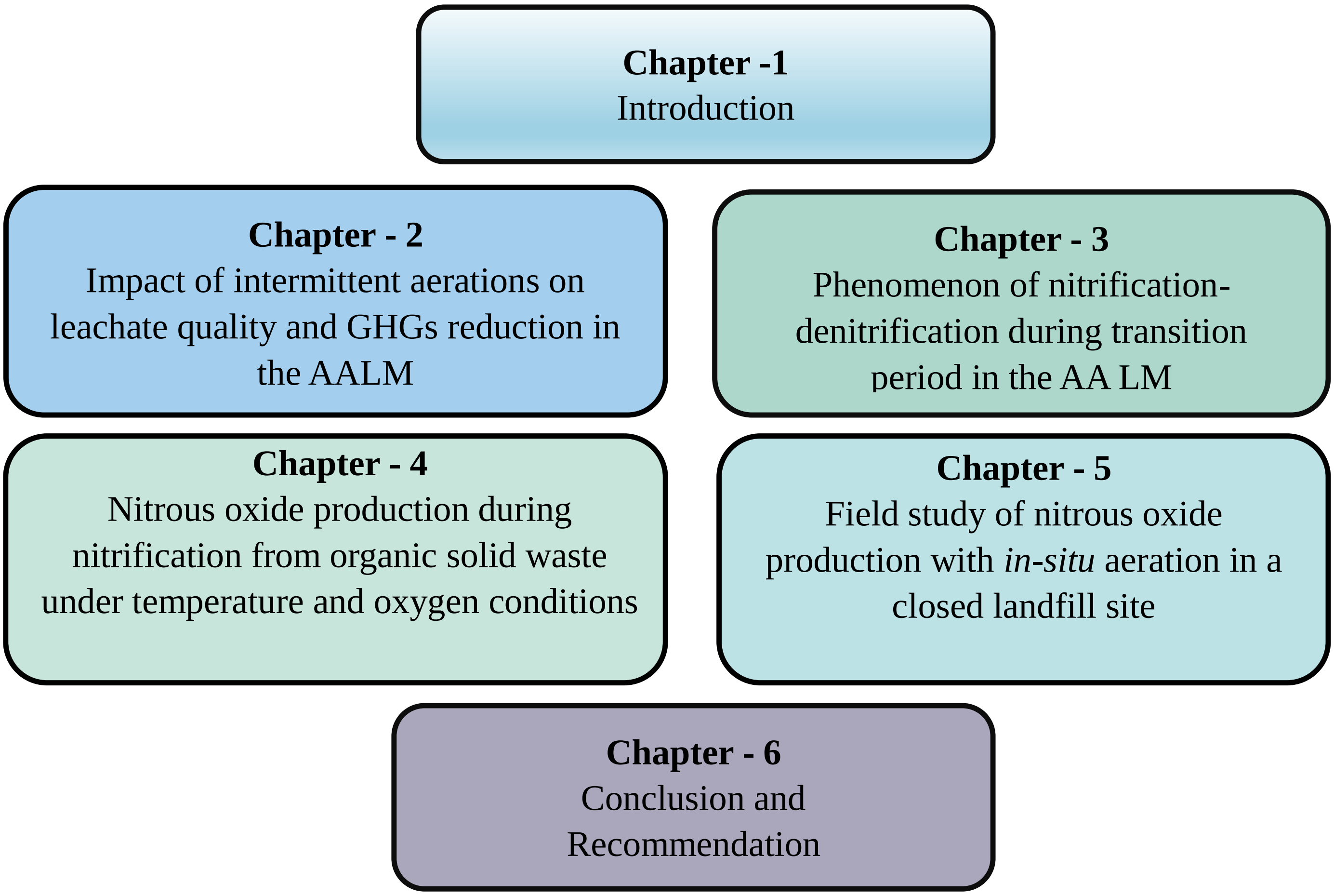


Fig. 1-16 Structure of the thesis

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CHAPTER 2

Impact of intermittent aerations on leachate quality and greenhouse gas reduction in the Aerobic–anaerobic landfill method

2.1. Introduction

The majority of developing countries have adopted the anaerobic type of landfilling as a municipal solid waste (MSW) disposal method. However, increasing attention is being given to the environmental problems associated with such landfills. These problems are high concentrations of total organic carbon (TOC) and total nitrogen (T-N) in the leachate, an extended stabilization period, and the emission of high levels of greenhouse gases (GHGs).

It is desirable that a landfilled solid-waste layer be maintained under an aerobic condition, which accelerates the decomposition of the organic matter (Hanashima et al. 1981., Hanashima et al., 1983; Mitchell, von Meien, and Krieger 2003; Mitchell et al. 2004). Moreover, relative to anaerobic decomposition, aerobic decomposition of organic matter could reduce the emission of methane gas, which has 28 times (no climate-carbon feedbacks) the global warming potential (GWP 100) of carbon dioxide (IPCC, 2013). In addition, the aerobic decomposition of organic matter could bring about the rapid stabilization of landfilled solid waste (Cossu, Raga, and Rossetti 2003; Bilgili et al. 2007). Furthermore, a worldwide considerable attention is currently being given to the reclamation of the aged landfill sites, or to reduce the period of post-closure management of the landfills. In this regard, applying the aerobic-anaerobic landfill method (AALM) of hybrid conditions may create biostabilized landfills, thereby, reducing the need for expensive perpetual landfill aftercare (Wu et al. 2014). The AALM is a novel approach to MSW management that could solve the problems associated with anaerobic landfills in both developing and in developed countries, as it combines the advantages of the aerobic type of landfill with reduced operational costs (Shimaoka et al., 2011a; Shimaoka et al., 2011b). The AALM is based on the nitrification of solid waste under aerobic conditions and denitrification under anaerobic conditions (Shimaoka et al., 2011b). The AALM could enable the development of simultaneous nitrification and denitrification processes in only one landfill cell (Berge et al. 2006), rather than in two separate anoxic and aerobic cells. Moreover, provided that the temperature is properly controlled (Raga and Cossu 2013), efficient nitrogen turnover could be achieved. In the AALM, a certain proportion of air is injected into an anaerobic landfill at a certain rate for a period of time to create a partially aerobic atmosphere in the landfill. In this way, aerobic and anaerobic atmospheres are stratified in the landfill, and the leachate from a solid waste layer is alternately exposed to aerobic and anaerobic conditions as it percolates down through the layer. Moreover, the position of the aerobic layers is controlled to achieve the optimum decomposition of the MSW by dynamically changing the air injection parameters, based on the consideration of air injection position, rate, and period, as well as the phase of decomposition of the landfilled solid waste. In this manner, the AALM could shorten the landfill aftercare period and lower the environmental loads derived from the landfill.

Based on large-scale lysimeter experiments and numerical simulations, it has been reported that air injection at a greater depth, or at the bottom layer of a landfill, was beneficial in terms of the improvement in the leachate quality, the reduction of GHGs, and the enhancement of solid waste stabilization (Shimaoka et al., 2011a; Wu et al. 2014). Previous studies have focused on aerobic and anaerobic conditions that are alternately created along the vertical direction of a landfill by applying continuous air injection. However, intermittent aeration, which results in a single landfill layer alternating temporally between aerobic and anaerobic conditions, has not been discussed. The application of intermittent aeration is also expected to reduce the operational costs, compared with continuous air injection, because surplus aeration can reduce microbial activity and increase energy consumption (Sang et al. 2009).

In this study, column experiments were conducted to determine the effectiveness of applying intermittent aeration, as is done in the AALM. The experiments, in which aerobic and anaerobic conditions were alternately created in a specific layer of a column by means of intermittent air injection, entailed examining the effect of intermittent aeration on the leachate quality and the emission of GHGs. Furthermore, the occurrence of nitrification and denitrification during the aerobic and anaerobic conditions was also investigated by means of these column experiments.

2.2. Materials and methods

2.2.1. Column experiment

The research was conducted using four laboratory-scale column reactors (A, B, C, and D). The schematic diagram of the column reactor shown in Fig. 2-1. Each column was a plexiglass cylinder (200 cm height × 10 cm diameter) in which 15 cm of gravel, 170 cm of solid waste, and 10 cm of cover soil were layered from the bottom to the top, respectively. Top 5 cm was kept as open space in the column reactors; therefore, there was possibility of the atmospheric air penetration in the reactors. An aeration pipe was installed at the bottom of the column to introduce air. Gas sampling points were set up at every 40 cm depth interval in the column reactors.

Each column reactor was filled with organic solid waste, collected from a composting plant, with a dry density of 0.32 t/m^3 and a wet density of 0.67 t/m^3 that had been shredded to pieces of less than 1 cm (1/10 of the column's internal diameter to avoid the air paths from the glass void space). The composition of the waste was primarily kitchen waste from restaurants, households, and food industries along with wood chips.

Table 2-1 presents the operating conditions of the column experiments. Column A was operated with continuous aeration (7 days/week), columns B (3 days/week) and C (6 h/day) were operated with intermittent aeration, and column D (no aeration) was operated without aeration.

The durations of air injection for columns B and C were set to three days/week (3 days continuous aeration and 4 days without aeration) and 6 h/day, respectively. The air injection rate during aeration was 7.1 l/kg-dry mass (DM) h for Aerobic (A) and Aerobic-anaerobic (B and C) columns in Stage-1 (0 to 302 days). All columns were covered with electric blankets and insulation materials to maintain the ambient temperature at approximately 30 ± 1 °C, in winter (the experimental period was Oct. 15, 2012 to Nov. 28, 2013) and in other seasons kept at 30 ± 1 °C room temperature. Temperature probes (Em50, Decagon Devices, Inc.) were placed in the middle of each column and were connected to a data logger (Em50, Decagon Devices, Inc.) to record the internal temperature. A total of 360 ml of distilled water was added weekly until 150 days of the experiment and later the amount was reduced to 280 ml to each column to simulate precipitation and generation of leachate. There was no leachate recirculation in this study.

Gas samples were collected from five-gas sampling points (25, 65, 105, 145 and 185 m depth) in each column for analysis of the O_2, CO_2, CH_4, and N_2O concentrations at 2-weeks intervals (in case of Reactor B and C, sampling was before start the aeration). Leachate was collected weekly from the bottom of the each reactor and each sample was filtered using 0.45 µm pore filter paper to analyze the pH, electric conductivity (EC), oxidation-reduction potential (ORP), TOC, T-N, NH_4^+-N, NO_3^--N, and NO_2^--N concentrations.

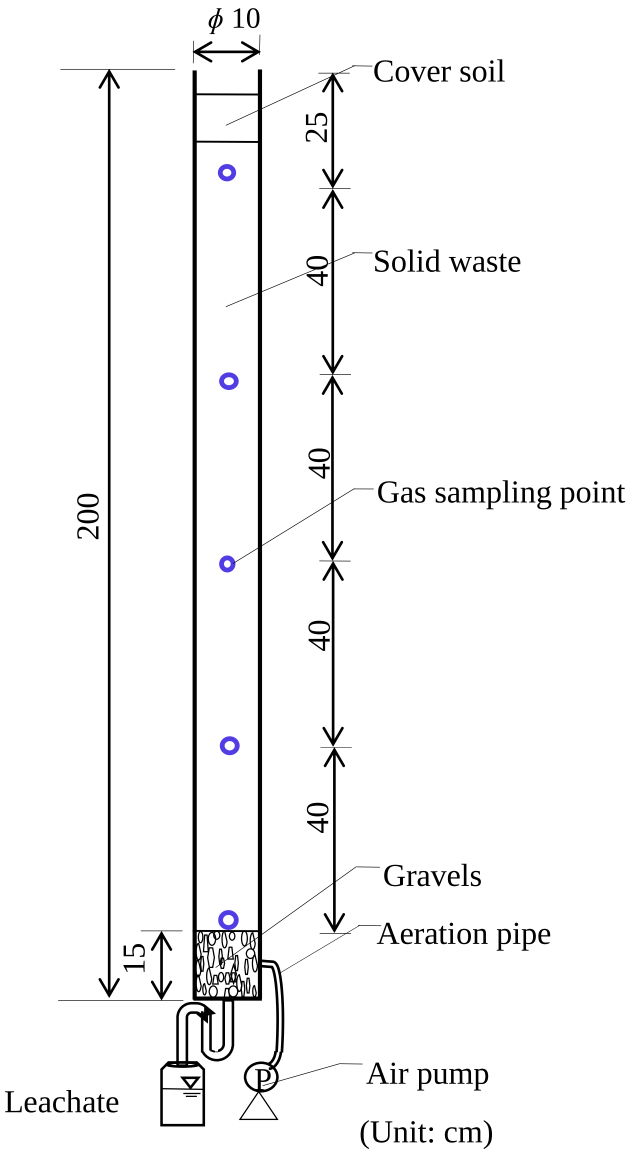


Fig. 2-1 Schematic diagram of column reactor

Table 2-1 Conditional and operational parameters of column experiment

Parameters	Reactor A	Reactor B	Reactor C	Reactor D
Moisture content (%)	50			
Packing density (t/m^3)	0.32 (Dry), 0.67(Wet)			
Volumetric ratios (m^3/m^3)	0.23 (Solid), 0.36 (Liquid), 0.41 (Gas)			
Ambient temperature (°C)	30 ± 1°(a)			
Water supply as rainfall (mL/week)	360(b) , 280(c)			
Duration of continuous air injection	7 days/week(d)	3 days/week(e)	6 h/day(f)	No aeration
Air injection rate l/kg-DM h	7.1(g) and 4.2(h)			0.0

(a)Started from 15th day of operation; (b) (0-150 days); (c) (156-408 days) ; (d) 168 hours continuous/week (5040L/week); (e) 72 hours continuous/week (2160L/week); (f) 42 hours/week (1260L/week); (g) Stage-1 (0-302 days); (h) Stage-2 (325-408 days)

It was observed that at the end of Stage-1, the ammonium concentration was exceptionally low in the discharged leachate under the high aeration rate (7.1 l/kg-DM h). Therefore, the aeration rate was subsequently reduced to 4.2 l/kg -DM h.

The dissolved oxygen (DO) concentration was measured during days 310 to 324 to determine the reduced aeration rate, i.e., from 7.1 to 4.2 l/kg-DM h, in all the aerated reactors. It was found that the DO dropped to less than 0.2 mg/l from day 325, when the aeration rate was maintained at 4.2 l/kg-DM h. Finally, during Stage-2 (days 325 to 408), which lasted for 83 days, the sampling of gas and leachate was performed once per every week. In Stage- 2, the characteristics of waste and condition of each reactor reported in Table 2-2 (waste amount was calculated based on settlement). Solid samples were collected from the backside holes of the column reactors and mixed it for analysis of waste characteristics.

2.2.2. Analysis of solid, leachate, and gas samples

TOC and T-N in solid sample were measured by TOC analyzer (SSM-5000A, Shimadzu, Japan). Loss on ignition (LOI) was measured for estimating the organic carbon content in solid phase by using a muffle furnace to heat the dried sample (at 105 °C for 24 h) to 550 °C for 4 hour. The pH, electric conductivity and oxidation-reduction potential of the collected samples were measured with a Horiba pH, EC and ORP meters. The total organic carbon and total nitrogen contents of the sampled leachate were measured using a TOC analyzer (TOC-analyzer, Shimadzu, Japan). Ion chromatography (DX-120, Dionex, Japan) was carried out to detect the concentrations of NH_4^+-N, NO_2^--N, and NO_3^--N ions in the collected leachate. The gas concentrations of O_2 and CO_2 were measured by chromatography (GC8A, Shimadzu, Japan) using Thermal Conductivity Detector (TCD). The gas concentration of CH_4 was measured by gas chromatography (GC2014, Shimadzu, Japan) using TCD in which the carrier gas was helium (He).

2.2.3. Calculation of carbon and nitrogen mass balances

In order to elucidate the reduction and conversion of carbon and nitrogen in each experimental column reactor, the mass balance of carbon and nitrogen during the experimental period of Stage-1 was calculated. The calculations accounted the mass of carbon and nitrogen in the solid, liquid, and gas phases, the discharged leachate, and the emitted gas.

Table 2-2 Characteristics of wastes in reactors in Stage-2 (at the end of Stage-1)

Parameters	Reactor A	Reactor B	Reactor C	Reactor D
Moisture content (%)	70±2	70±2	70±2	68±2
Amount of waste (kg)	6.88	7.51	7.67	8.21
Total solid (kg)	2.06	2.25	2.30	2.62
Volatile solid (%)	59.4±0.1	67.5±0.1	70.2±0.1	77.1±0.1
Waste height (cm)	130±0.0	142±0.0	145±0.0	155±0.0

The mass of initial organic carbon was measured in solid phase, whereas the mass of organic carbon at the end of Stage-1 was calculated by using the LOI data of the solid waste, under the assumption that the LOI equaled the organic matter content and that the mass of carbon was half that of the organic matter mass. The mass of carbon in the discharged leachate was the cumulative amount of the carbon discharge, calculated by using the periodical data of the leachate amount and the TOC concentration of the leachate. The mass of carbon in the emitted gas was calculated as the sum of the gas emission by advection and the diffusion during the experimental period of Stage-1. The mass of carbon emitted by gas diffusion was calculated with Fick's law, by using the gradient data of CO_2 and CH_4 concentrations (between 0.25 m and 0.05 m depth (atmospheric)), the diffusion coefficient, and the tortuosity, calculated by the Penman tortuosity model (Penman, 1940). The diffusion coefficients of CO_2 and CH_4 were 0.06 and 0.01 m^2/h (The chemical Society of Japan, 1993) and the porosity was 0.41.The advection flux of the gaseous carbon-species (J, $kg/min/m^2$) was calculated for all the aerated reactors (A, B, and C), based on the outflow rate of air from the column (Q, m^3/min), the gas concentration (C) at a depth of 0.25 m, and the horizontal cross-sectional area of the column (A, m^2), as Eq. 1. A coefficient of 0.35 was used for the adjustment of the advection flux considering the gas concentration difference between 0.25 and 0.05 m depths especially.

$$J = \frac{Q.C}{A} \qquad (1)$$

Because of the continual production of landfill gas, the increase in pressure within the landfill, together with the diffusion, caused the release of gases into the atmosphere. Therefore, gas migration could occur, attributable to pressure differential and diffusion. Based on this approach, the flux of Reactor D (non-aerated) was calculated, using Darcy's law, where the pressure gradient was calculated by the total gas pressure inside and the atmospheric pressure (assumed as 1 atm) outside and the gas concentration at 0.25 m depth. The permeability of the cover soil and the viscosity of the gas were assumed as $1x10^{-13}$ m^2 (Tanaka, N., 2000) and

1.89×10^{-5} Pa s (The chemical Society of Japan, 1993), respectively. A coefficient of 0.17 was used for the adjustment of the advection flux considering the gas concentration difference between 0.25 and 0.05 m depths and the correction of the inside total gas pressure especially.

In order to evaluate the nitrogen mass balance, the total nitrogen (T-N) was measured in the initial solid-waste sample (T-N_i). At the end of Stage-1, the final concentration of T-N (T-N_f) was calculated by assumption of the initial TOC/T-N ratio (16:1). The initial NH_4^+-N (N_{Li}), and, after Stage-1, the NH_4^+-N (N_{Lf}), was measured by leaching test (JLT-46).

The evaluation of the missing ammonia nitrogen (N_M) and the removal by gasification of the nitrogen from the reactors are intricate processes. For the evolution of ammonia transformation, it was assumed that the rate of ammonia transfer into leachate under aerobic conditions was the same as under anaerobic conditions and the conversion of ammonia mainly due to stripping (not measured in present study) and nitrification. The amount of missing ammonia nitrogen, converted by means of nitrification, calculated according to the conceptual model by Hwang and Hanaki (2000) shown in Fig. 2-2.

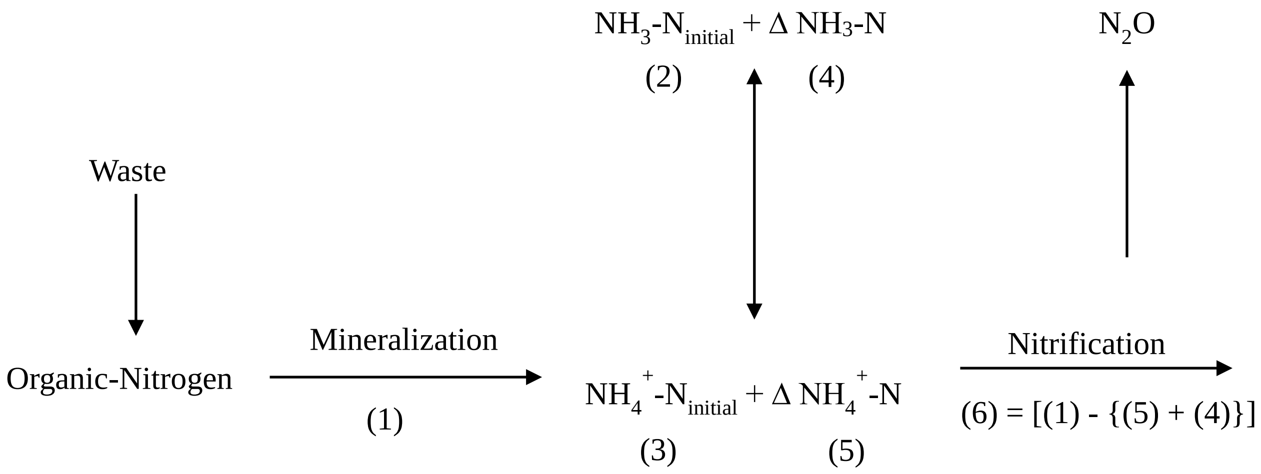


Fig. 2-2 Conceptual explanation of nitrification amount calculation

2.3. Results and discussion

2.3.1. Characterization of samples

2.3.1.1. Solid

In Table 2-3, the physico-chemical characteristics of the waste are summarized as mean values with standard deviations. In order to determine the carbon mass balance (results discussed later in this chapter), the LOI of the sample was obtained at the beginning and at the end of the first Stage (Stage-1) of measuring the organic carbon content of the bioreactors. Initial organic carbon content was 47.9 % at the beginning of the experiment, while at the end of Stage-1 it was (calculated from half of the LOI data) 29.7, 33.8, 35.1, and 38.5 % in Reactors A, B, C, and D, respectively.

The TOC and T-N values measured in the initial solid sample, obtained at the beginning of the test, were 47.9 and 3.0 %, respectively and the ratio of TOC/TN (16:1) calculated from these two values was used to determine the nitrogen mass balance in this study (results will be discussed later in this chapter).

Table 2-3 Physico-chemical chacteristics of organic solid waste with standard deviation.

Moisture content (%)	LOI (%)	pH(1) (-)	EC(1) (mS/cm)	TOC(1) (mg/kg DM)	TN(1) (mg/kg DM)	NH_4^{+}(1) (mg/kg DM)	NO_x^{-} (1) (mg/kg DM)	TOC (2) (%)	TN (2) (%)	TOC/TN (%)
53.0±2	86.7±0	5.3±0	9.4±0	83900±300	1800±100	7800±0	100±0	47.9±0.1	3.0±0.1	16:1

LOI: Loss on ignition; EC: Electric conductivity. (1) Measured by leaching test authorized by Japanese Environmental Agency, JLT-46. (2) TOC and TN measured in solid sample

2.3.1.2. Leachate

Table 2-4 shows the results for all the column reactors of the analyzed leachate samples collected at the start of the experiment, at the end of Stage-1, and at the end of the experiment. For the characterization of the leachate, the sampling started after two weeks of Stage-1.

Initially, the pH conditions in all the reactors were acidic, while at the end of Stage-1, the pH values were greater than 8.5 in all the aerated reactors, namely, A, B, and C. At the end of the experiment, the values were lower than they had been for Stage-1. In the Non-aerated reactor D, the initial pH value was approximately 5.18 and it remained almost neutral during the course of the experiment.

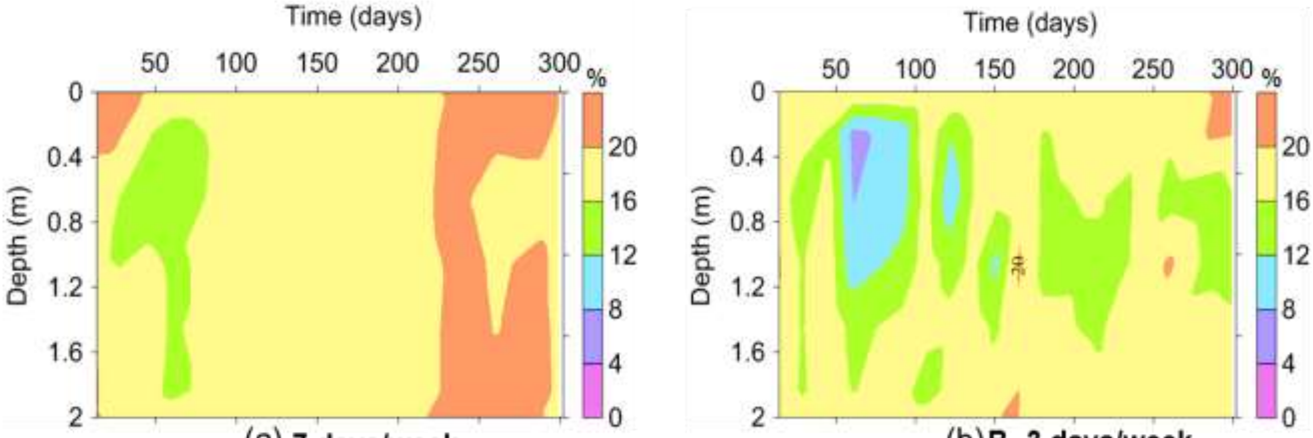
The EC had decreased from the peak 41.1, 42.4, 34.5, and 35.6 mS/cm (on day 45, data not shown in Table 2-4) to 13.7, 20.3, 18.5, and 25.3 mS/cm in reactors A, B, C, and D, respectively, at the end of Stage-1. The evaluation of organic carbon removal and nitrogen transformation in leachate has been discussed in section 3.2.

Table 2-4 Results of discharged leachate characterization from the bioreactors A-D at the start, end of Stage-1 and end of the experiment. Analysis were carried out in triplicate and results are provided as mean values. n.d.: not detectable.

	A			**B**			**C**			**D**		
Parameters	Start	End of Stage-1	End of exp.	Start	End of Stage- 1	End of exp.	Start	End of Stage- 1	End of exp.	Start	End of Stage-1	End of exp.
pH	5.3	9.3	8.6	5.2	8.8	7.4	5.1	9.3	7.6	5.2	7.3	7.2
EC (mS/cm)	11.9	13.7	4.8	16.2	20.3	6.5	19.9	18.5	2.5	17.9	25.3	10.1
ORP (mV)	166	194	212	155	105	172	169	145	149	143	-58	-1
TOC (mgC/l)	19,390	1805	435.7	14,840	2540.5	854	18,230	4011.5	201	15,590	5643	1643
T-N (mgN/l)	3571	945	552.5	2699	1558	302.6	3300	1404.5	212.6	2993	3890	2077
NH_4^+-N (mgN/l)	1695	102.4	243.1	2054.3	303.7	142.7	2650	88.7	5.2	1966	948	455
NO_2^--N (mgN/l)	n.d	337.4	53.0	n.d	321.1	25.0	n.d	102.4	31.0	n.d	0.7	n.d
NO_3^--N (mgN/l)	n.d	245.6	201.1	n.d	127.4	137.7	n.d	31.6	173.0	n.d	26.5	n.d

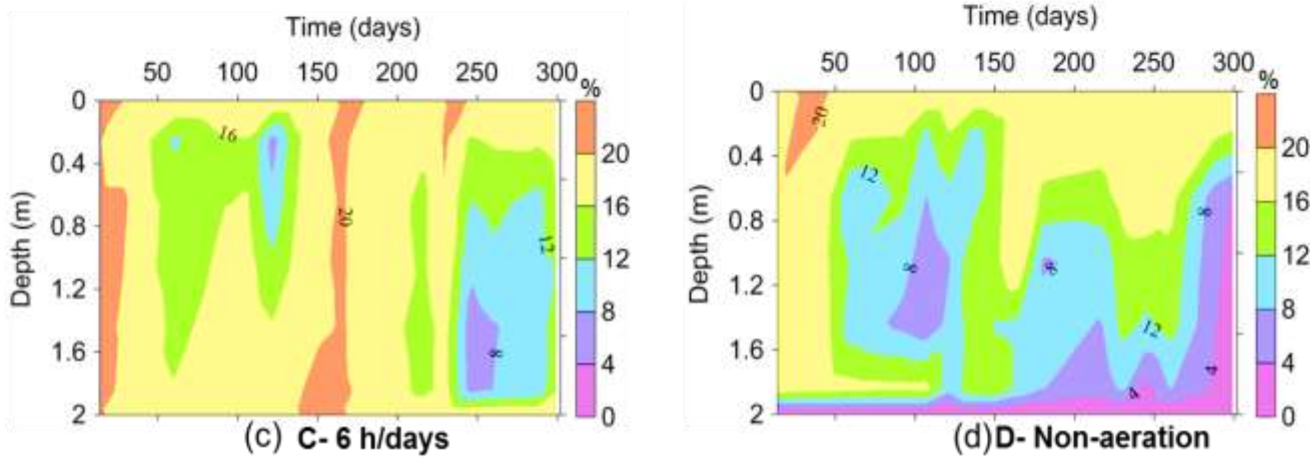
2.3.1.3. Gas

Figs. 2-3 to 2-5 illustrate the spatial variation (in percentages) during Stage-1 of O_2, CO_2, and CH_4 gas, with time and the depth, in all four of the bioreactors. The top layers of all the reactors matched the atmospheric conditions (Fig. 2-3). The oxygen concentrations were higher with longer periods of air injection, and CO_2 was generated as the consumption of O_2 started in all



(a) **7 days/week**

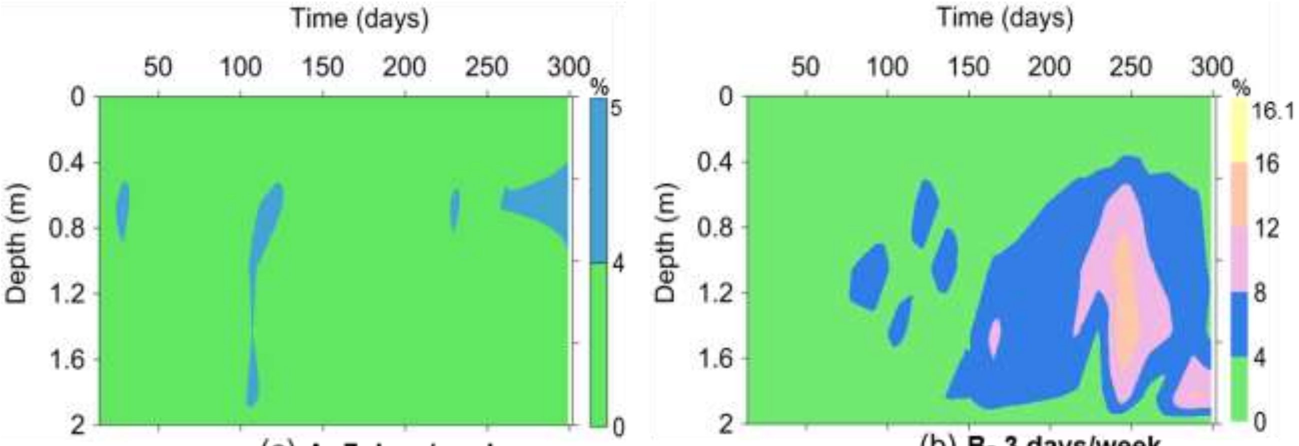
(b)**B- 3 days/week**



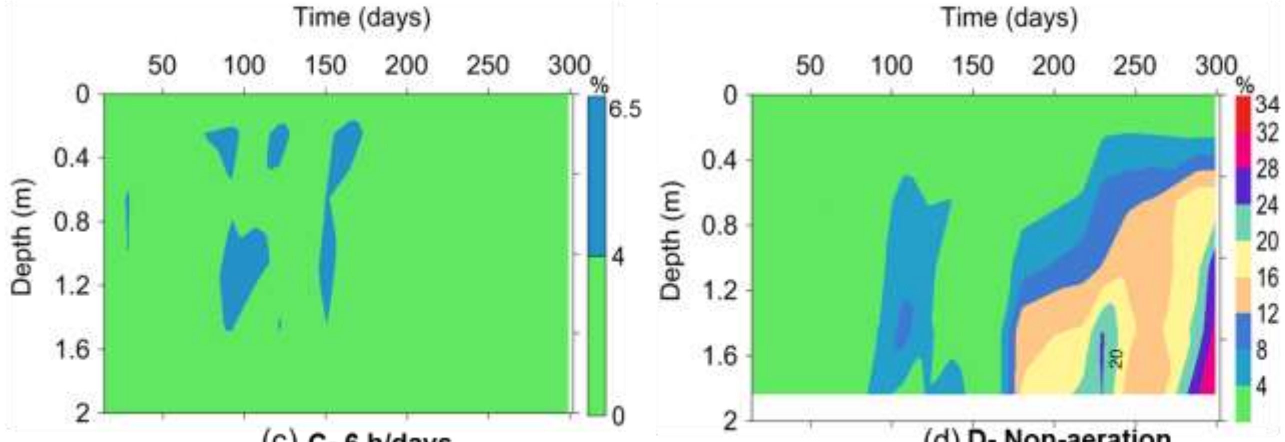
(c) **C- 6 h/days**

(d)**D- Non-aeration**

Fig. 2-3 Spatial variation of O_2 concentration in reactors with depth and time in Stage-1

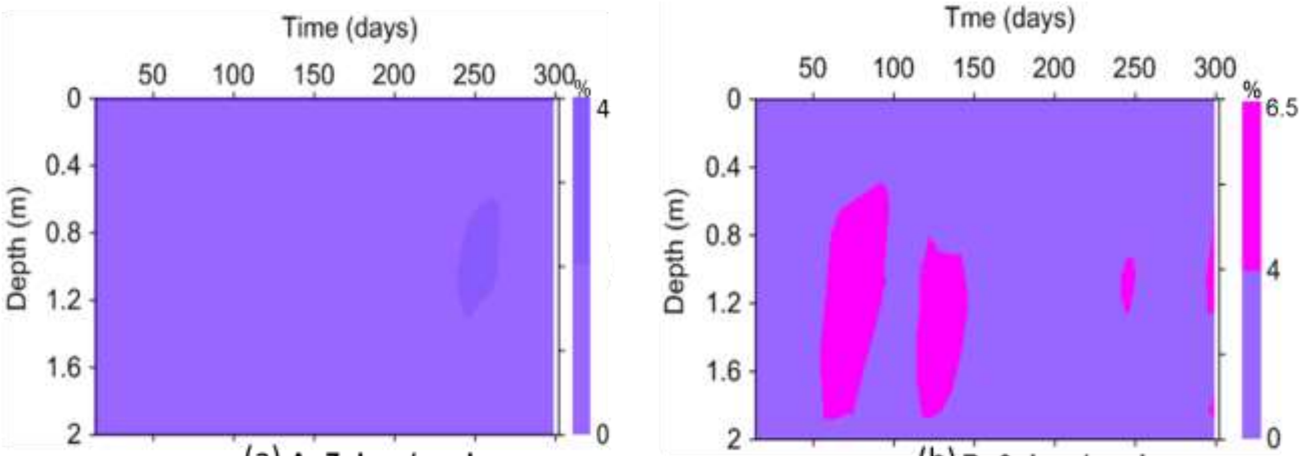


(a) **A- 7 days/week** (b) **B- 3 days/week**

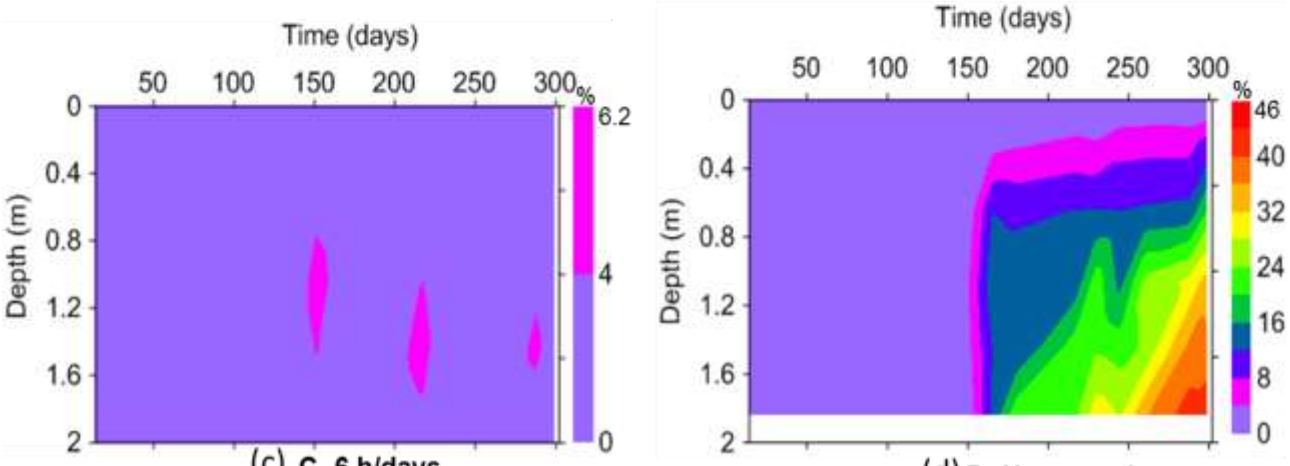


(c) **C- 6 h/days** (d) **D- Non-aeration**

Fig. 2-4 Spatial variation of CO_2 concentration in reactors with depth and time in Stage-1



(a) **A- 7 days/week** (b) **B- 3 days/week**



(c) **C- 6 h/days** (d) **D- Non-aeration**

Fig. 2-5 Spatial variation of CH_4 concentration in reactors with depth and time in Stage-1

the aerated reactors. When Reactor C was compared with Reactor B, it was found that the generation of carbon was higher in Reactor B until day 150 of the experiment. The maximum concentrations of CO_2 and CH_4 were 16, 6.5 % in Reactor B and 6.5, 6.2 % in Reactor C, respectively. In contrast, the maximum CO_2 and CH_4 concentrations in Aerobic reactor A were less than 5 and 3 %, respectively, throughout the experiment. A slight generation of CH_4 was observed in Reactors A and C from 250 to 302 days. The probable reason of this increment in

CH_4 generation was possibly due to the relatively higher inside temperature, ranged between 30 to 38 °C in aerated reactors as shown in Fig. 2-6. The reason of high temperature inside the reactors might be the passing of comparatively warm air through the air pump in summer time than winter and other seasons, in addition of energy release by decomposition of organic matter. In addition, it was observed that the generation of CH_4 had started slightly earlier (from day 66 to day 92) in Reactor B than it had in Reactor C or in the Non-aerated reactor D. However, compared with the generation of CH_4 later in the Non-aerated reactor D, the ultimate concentration was not extremely high in Reactor B.

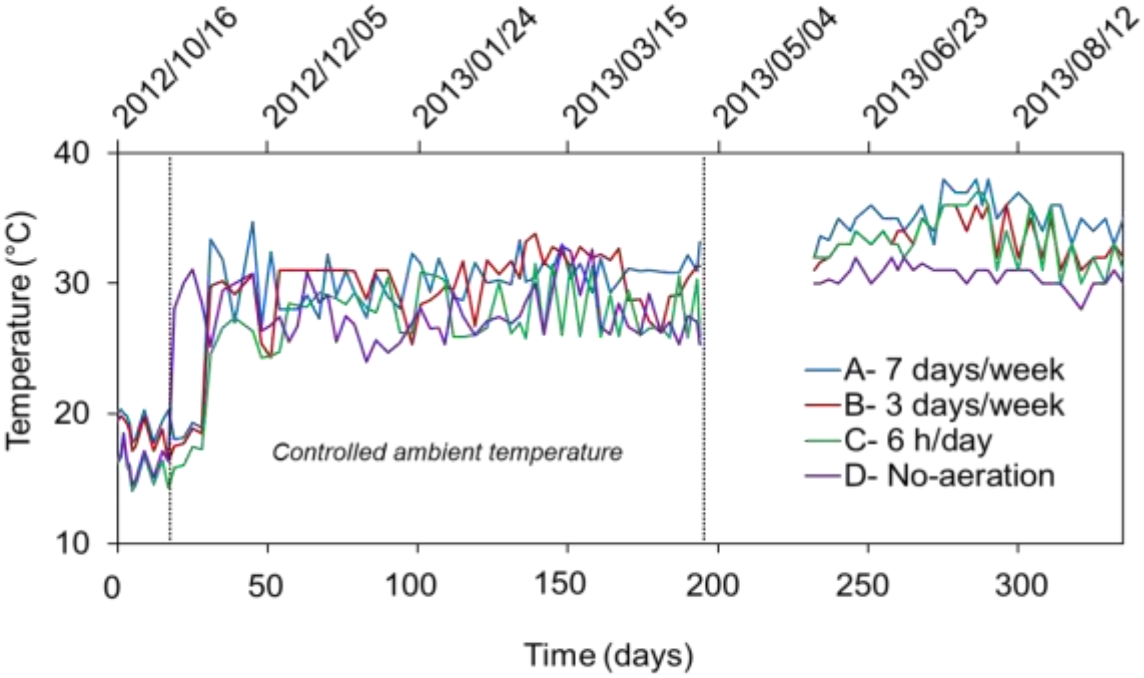


Fig. 2-6 Daily average temperature profiles inside the reactors with elapsed time

2.3.2. Evolution of aeration in bioreactors

An increase in the oxygen content in a specific layer is the most important difference found between the AALM and the anaerobic landfill methods. Oxygen concentration in the waste mass was affected by both convection, diffusion and biological activity of aerobic bacteria. In Reactor C, the air was supplied daily for six hours and the inside of the reactor remained at a nearly aerobic condition throughout the experimental period. The generation of CO_2 and CH_4 in Reactor C was markedly lower than it was in the intermittent aeration Reactor B and the Non-aerated reactor D. The impairment of oxygen diffusion and denitrification could probably be the reason for the slight generation of CH_4 in the aerated reactors, along with the production of CO_2. Moreover, the oxidation of organic matter, the nitrification or the oxidation of methane (Malgorzata P., 2014) possibly had taken place. The potential cause of the delay in generation of CH_4 in Reactor D, could be the presence of initial oxygen inside the reactor, and the addition of distilled water might flush out the anaerobic condition until middle of the experiment and later reduced water amount stimulate methanogenic phase. The ORP values in Fig. 2-7 became negative in the Non-aerated reactor D, indicating that the phase of waste degradation was shifting from the acidogenic to the methanogenic.

Furthermore, It can be noted that a high aeration rate at the bottom of the reactor was beneficial for the distribution of oxygen in the waste mass in Reactors A, B, and C, although the duration of the air supply to the reactors differed greatly. Conversely, in Reactor D (no-aeration), the consumption of O_2 was high, although it seems that the reactor was not strictly anaerobic because of the air voids in the wood chips present in the column. Because of these voids, the air in the top layer of the column was able to penetrate the surface layer inside the reactor, therefore Reactor D termed as Non-aerated reactor instead

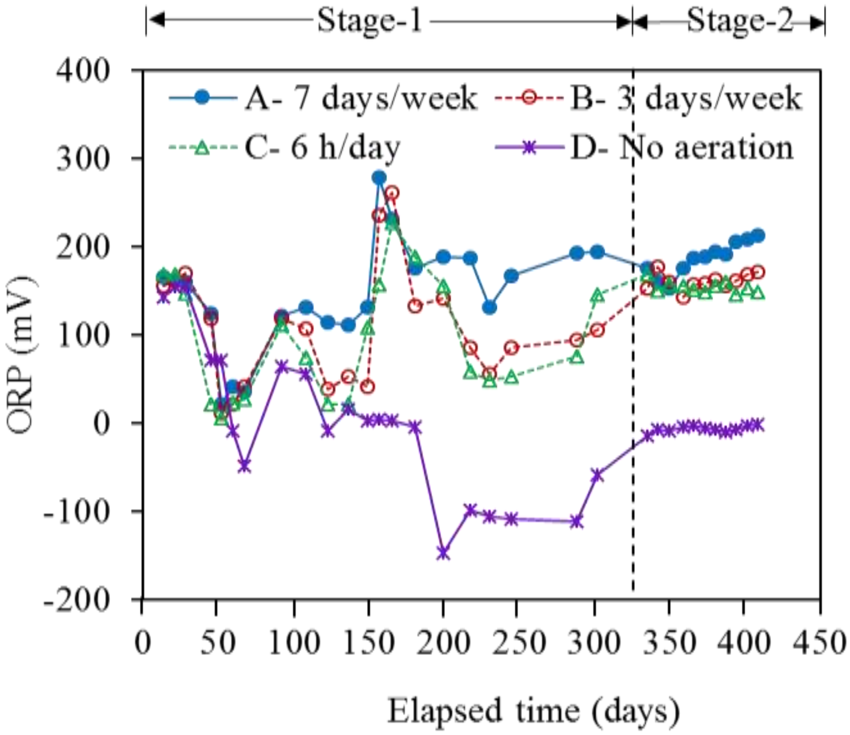


Fig. 2-7 Changes of ORP with elapsed time

of anaerobic reactor. From the middle period of the operation, the generation of CO_2 and CH_4 started under the anaerobic condition, and reached the maxima of 34 and 46 %, respectively, at approximately 288 days, when the consumption of O_2 was the highest in the bottom layers of the reactor.

Fig. 2-8 illustrates the evolution of the TOC mass changes, together with the pH; while Fig. 2-9 shows the evolution of the nitrogen mass changes for T-N, NH_4^+-N, NO_2^- -N, and NO_3^- -N in the leachate of Reactors A, B, C, and D. Based on the data on the concentrations of the leachate samples and the volume of the discharged leachate, the mass of the parameter in the liquid phase was calculated, in order to account for the effect of dilution on the leachate characteristics.

The mass of the parameters per unit of dry waste matter in the reactor was calculated by the following formula,

$$M_x = \frac{C_x . V_x}{D_x}$$

where M_x is the mass (mg/kg-DM) of parameter ‘x’ in the liquid phase in the system, C_x is the concentration (mg/l) of parameter ‘x’ in the leachate, V_x is the volume (l) of the liquid in the system, and D_x is the dry mass (kg) of initial solid waste put into the reactor.

The trends of change in the TOC mass demonstrate that when air was injected into the Aerobic and Aerobic-anaerobic reactors, the organic carbon was removed more rapidly than it was in the Non-aerated reactor. This finding indicates that the good distribution of oxygen affects the reduction of TOC. Although a sharp decrease of TOC was noted in the non-aerated reactor, the level was still almost double that of the aerated reactors at the end of Stage -1. Conversely, in Stage -2 (please refer to the TOC values shown in the secondary axis in Fig. 2-8) when the aeration rate was reduced to 4.2 l/kg-DM h, the decomposition of the organic carbon initially slowed down. However, at the end of the experiment, the TOC concentrations had gradually decreased. Significantly, the higher pH values in the leachate from the aerobic bioreactors observed by (Sang et al. 2008) during a laboratory experiment are in accordance with the findings of the current study.

The mass of the ammonium nitrogen decreased sharply from its initial high values, probably because of air stripping and the removal of the free ammonia. Increases in the temperature and the pH could promote the transformation of ion ammonium (NH_4^+) to free ammonia (NH_3) (Lei et al. 2007). The observed decrease in NH_4^+ concentrations is in accordance with the findings of a number of previous studies, showing that the hybrid conditions created by the cyclic/intermittent aeration are favorable for the reduction of total nitrogen, especially cyclic/intermittent aeration are favorable for the reduction of total

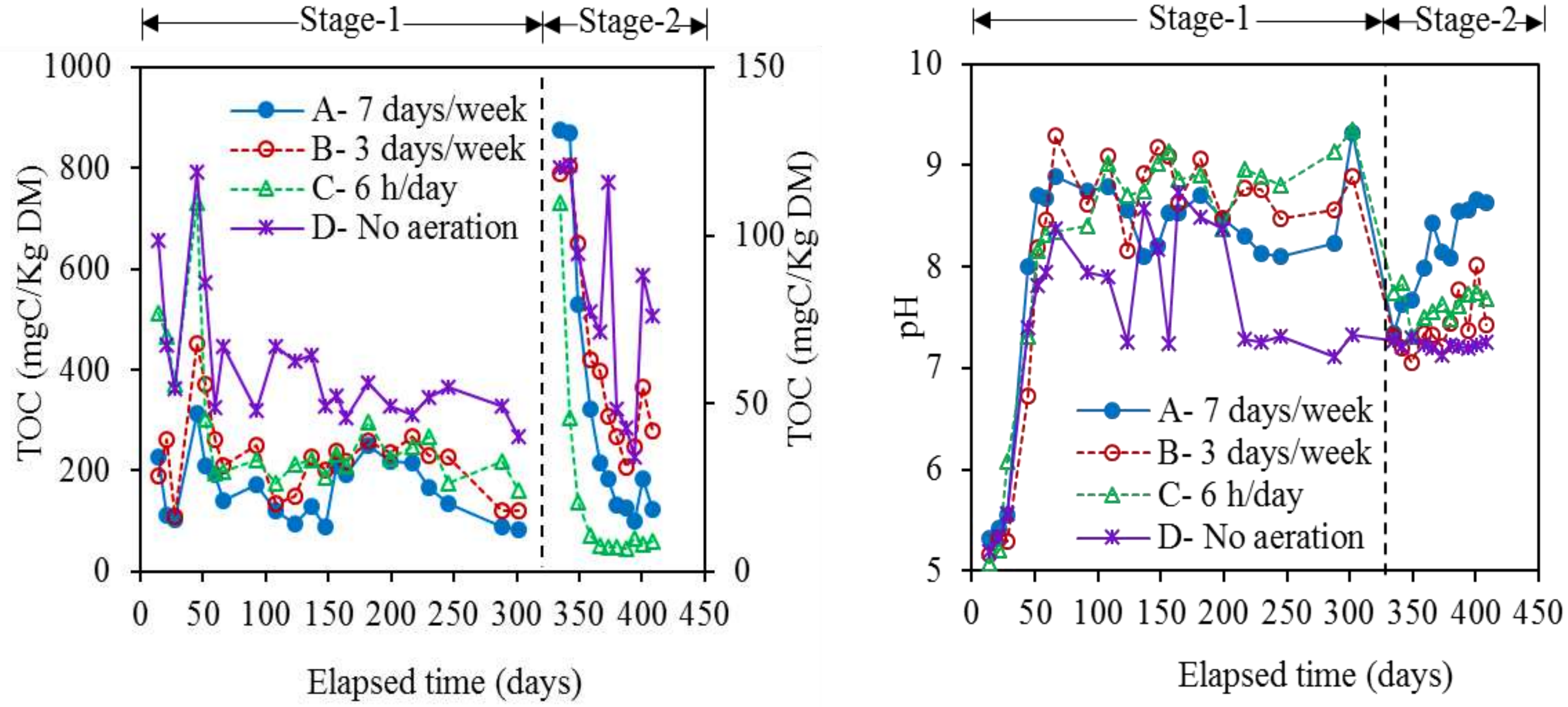


Fig. 2-8 Evolution of TOC mass changes and pH on discharged leachate with elapsed time

nitrogen, especially ammonium concentrations (Ruo He & Shen 2006)., Shao et al. 2008; Nikolaou et al. 2010). It was observed that at the end of the experiment, the removal of ammonium nitrogen in the Aerobic-anaerobic reactors (B and C) was slightly higher than it was in the Aerobic reactor.

While determining the nitrification-denitrification process in the reactors, a long lag phase (when values were not very significant) was found of nitrite/nitrate conversion from ammonium nitrogen. However, in this regard, the log-scale of nitrite and nitrate illustrates (Fig. 2-9) the difference between the Aerobic-anaerobic reactors and the Aerobic reactor.

Until day 150 of the experiment, in the lag phase, the difference between the aerobic and the intermittent aeration pertaining to nitrite and nitrate formation was observed. However, the mass changes were not at significant values, despite the rapid decrease in the NH_4^+-N concentration. Nitrite and nitrate formation was higher during intermittent aeration than it was in the Aerobic reactors, suggesting that simultaneous nitrification and denitrification occurred more readily (Sang et al. 2008) in the Aerobic reactor than in the intermittent aeration reactors.

ORP is a key index for the nitrification and denitrification process. As shown in Fig. 2-7, the ORP values in the Aerobic reactor (A) and the Aerobic-anaerobic reactors (B and C) after the initial adjustment phase were almost similar (a few ORP values in B and C could have fluctuated for the cyclic aeration). This finding indicates that simultaneous nitrification-denitrification had occurred during both the cyclic and the continuous aeration. After 200 days, the conversion of NH_4^+-N to nitrite and nitrate was obvious in all the aerated reactors and the mass changes were distinguishable according to

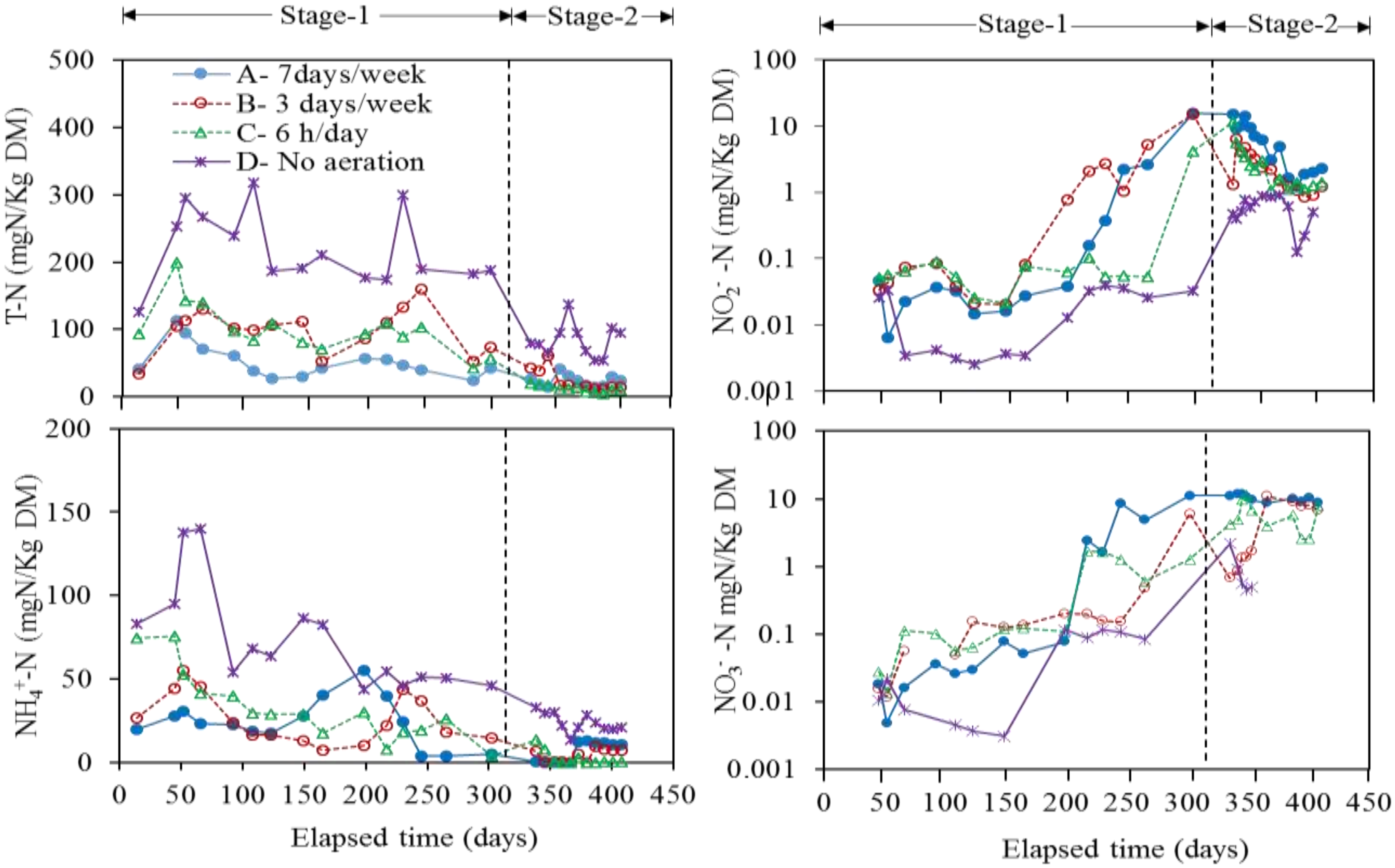


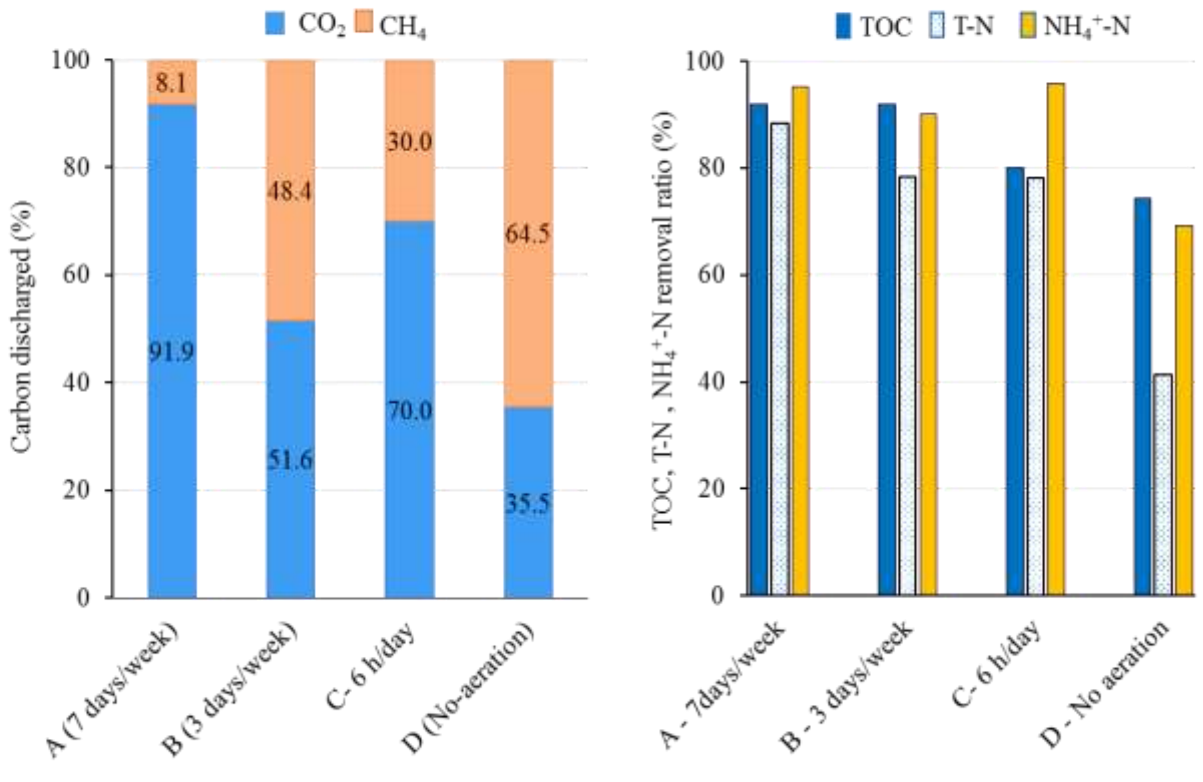
Fig. 2-9 Evolution of T-N, NH_4^+-N, NO_2^- -N and NO_3^- -N mass changes with elapsed time

the aeration status. Valencia et al. (2011) mentioned that the presence of small amounts of oxygen in the waste matrix was beneficial for the Anammox bacteria, which could also contribute to the removal of nitrogen, and could have caused the significant reduction of NH_4^+-N in Reactor D. During Stage-2, the reduced aeration rate did not show any remarkable change because the NH_4^+-N concentration in the discharged leachate had already dropped to an extremely low level at the end of Stage-1.

Lately, much attention is being focused worldwide on the emission of N_2O from aerobic landfills. The reason is that landfill aeration not only induces nitrogen removal but it could also encourage the emission of N_2O, which is 265 times (no climate-carbon feedbacks) GWP (100) of CO_2 (IPCC, 2013). In this study, all the reactors generated N_2O, producing an average of 0.0043, 0.0037, 0.0036, and 0.0048 % in Reactors A, B, C, and D, respectively, during Stage-1, when the high aeration rate was introduced. However, during Stage-2, with the lower aeration rate, the N_2O production was below the detection limit. Likely, this indicates that the generation of N_2O is high under a high rate of aeration; however, more parallel studies are required to investigate the production of nitrous oxide under different rates of aeration. Moreover, nitrification is the dominant pathway for N_2O generation with high oxygen and moisture content (Hwang and Hanaki 2000), therefore minimizing N_2O production by AALM (intermittent aeration) might be effective for limited nitrification process in landfill site. According to Berge et al. (2007) and Price, Barlaz, and Hater (2003) excess or lack of oxygen in the system contributes to the production of N_2O. However, it is also hypothesized that this could be a negligible factor in landfills because of the long residence time.

Fig. 2-10a shows the total carbon discharge as CO_2 and CH_4 (in percentage) in gas and the removal of TOC, T-N and NH_4^+-N between the aerated, intermittent, and the non-aerated reactors in leachate in Fig. 2-10b, for all bioreactors during Stage-1. It was evaluated that carbon discharge via CO_2 in Aerated reactors were higher than Non-aerated reactor. Carbon conversion via CO_2 was highest about 92 % in aerobic reactor A and 70 % in Aerobic-anaerobic reactor, C. Generation of CH_4 was higher in Non-aerated reactor D and therefore discharge amount was highest about 64.5 %.Carbon discharge via CH_4 in Aerobic-anaerobic reactor B showed higher than Aerobic-anaerobic reactor C, although the aeration duration was high. This might be the reason of long duration of non-aeration period in Reactor B (4 days/week) compared to short duration of non-aeration period in Reactor C (18 hours/day).

The removal efficiencies could indicate the difference in the removal of TOC, T-N and NH_4^+-N between the aerated, intermittent, and the non-aerated reactors. The removal efficiencies are the percentages of the differences between the peak concentration and the minimum concentration in Reactors A, B, C, and D, calculated for TOC as 91.9, 91.8, 80.1, and 74.3 %, respectively shown in Fig.2-10 (b) during Stage-1. In Aerobic reactor A and intermittent reactors B and C, the removal ratio was respectively 17.6, 17.5, and 5.8 % higher than it was in Non-aerated reactor D. The NH_4^+-N removal efficiencies in Reactors A, B, C, and D were 95.3, 90.1, 95.7, and 69.3 %, respectively. The removal ratio of NH_4^+-N in the intermittent aeration Reactors B and C was 20.8 and 26.4 % higher, respectively, than it was in the Non-aerated reactor D.



(a) (b)

Fig. 2-10 (a) Carbon discharge and (b) removal ratio of TOC, T-N and NH_4^+ -N on discharged leachate in bioreactors during Stage-1

2.3.3. Comparison of the effect of intermittent aeration

Figs. 2-10 summarize the overall removal ratios of leachate pollutants and the carbon discharge (percentage) in all four bioreactors. Fig. 2-10 clearly indicate that the performance of Aerobic reactor A (7 days/week) was the best among the tested regimens and that Non-aerated reactor D discharged the highest amount of methane gas and the lowest leachate quality. Aerobic-anaerobic reactor C (6 h/day) performed better than did Aerobic-anaerobic reactor B (3 days/week) in respect of both the leachate quality and the improvement of GHG emission. However, the duration of the air supply was 1.7 times higher in Reactor B (3 days/week) than it was in Reactor C (6 h/day). From an economic perspective, Reactor A (7 days/week) needed

168 hours of aeration per week, whereas air was supplied for only 42 hours per week in Reactor C (6 h/day), i.e., equal to one-fourth of that supplied to the Aerobic reactor. Therefore, through intermittent aeration, Reactor C (6 h/day) could reduce three-fourths of the energy costs of the Aerobic-anaerobic landfill method (AALM), compared with an aerobic landfill. Figs. 2-10 (a) and 2-10 (b) indicate the leachate pollutant removal ratios and the carbon discharge levels for Aerobic-anaerobic reactors B and C. It is suggested that an effective air supply amount at an appropriate interval is extremely important to maintain the aerobic-anaerobic conditions in a landfill in order to facilitate simultaneous nitrification-denitrification. The simultaneous nitrification-denitrification process has the advantages of speeding up the decomposition of waste, reducing GHG emissions, improving the leachate quality, and lead to early stabilization of the site. The phenomenon of pollutant reduction in Reactors B and C is described more clearly in chapter 3.

This concept of AALM by intermittent aeration holds promise for reducing leachate pollutants and could reduce the potential of greenhouse gases by accelerating the decomposition of waste under aerobic conditions. However, this method does not promise zero emissions of GHGs. Additionally, intermittent aeration could shorten the later phases of landfill aftercare (Gamperling et al., 2010) and could simultaneously reduce the energy costs.

2.3.4. Carbon and nitrogen mass balances

The mass balances of carbon and nitrogen are reported in Table 2-5 and Table 2-6, respectively. It is evident from Table 2-5 that approximately 70.0, 62.8, 60.5, and 50.5 % of the initial TOC had degraded in Reactors A, B, C, and D, respectively, during Stage-1.

The highest amount of gasification occurred in Aerobic reactor A, where the total amount of carbon measured approximately 1317 g-Cover 302 days of the experiment. In Aerobic reactor A, approximately 92 % of the carbon was released mainly as CO_2. A lowest amount of gasification observed in Non-aerated reactor D, approximately 925 g-C, where the highest amount of carbon released as CH_4. Amount of gasification from Aerobic-anaerobic reactors B and C were approximately 1133.5 and 1101.7 g-C, respectively. The Aerobic-anaerobic reactor C discharged about 69 % of the carbon as CO_2 while 50 % observed in Reactor B.

The cumulative TOC discharged via the leachate (TOC_{Le}) was shown to be higher in Reactor D than it was in the aerated Reactors A, B and C, although the concentrations in these reactors were high. The high concentration was inferred because of the evaporative effects of forced aeration, shown in Fig. 2-11, the cumulative discharge of the leachate amount. TOC_L was measured in the leachate in the system at the end of Stage-1.

Table 2-6 presents the results obtained for nitrogen mass balance. The NH_4^+-N in the solid waste at the beginning of the experiment (N_{Li}) was compared with calculated (N_f) value as the sum at the end of Stage-1 of the mass of NH_4^+-N (N_{Lf}), the cumulative value for NH_4^+-N measured in the discharged leachate (N_{Le}), in the leachate present in the system (N_L) and the amount of NH_4^+-N that have left the reactors in gas phase were designated as the missing NH_4^+-N (N_M). The concentrations of N_2O indicate that only a small amount of ammonia nitrogen had converted to N_2O. Taking into account the air stripping, the transformation of NO and N_2O as the uncounted processes for NH_4^+-N, it was assumed that 95 % of the NH_4^+-N missing from the aerated systems had been converted into nitrate. Based on the assumption made and data shown in Table 2-6, approximately 34, 27, 24 and 22 % of the initial nitrogen content had been converted by nitrification and the discharged leachate. The highest level of nitrification, approximately 17%, was observed in Aerobic reactor A. In intermittent aeration Reactors (B and C), the nitrification involved 10 and 7%, respectively of the initial total nitrogen.

Table 2-5 Carbon mass balance at the end of Stage-1

Carbon amount (g-C)	A	B	C	D
TOC_{sf} (measured)	613.22	761.16	808.33	1012.27
TOC_{Le}	14.79	20.27	25.26	32.13
C_{Gas}	1317.7	1133.5	1101.7	925.90
TOC_L	0.08	0.12	0.16	0.17
TOC_C (calculated)	1945.78	1915.09	1935.48	1970.5
TOC_{si}	2045.33	2045.33	2045.33	2045.33

TOC_{Si} and TOC_{Sf} = Initial and final TOC, respectively in solid phase; TOC_{Le}= cumulative TOC measured in extracted leachate; TOC_L = in leachate in the reactors at the end of Stage-1; C_{Gas} = sum of discharged carbon in gas phase; TOC_C = Total carbon calculated at the end of Stage-1 is the sum of TOC in solid, liquid and discharged gas.

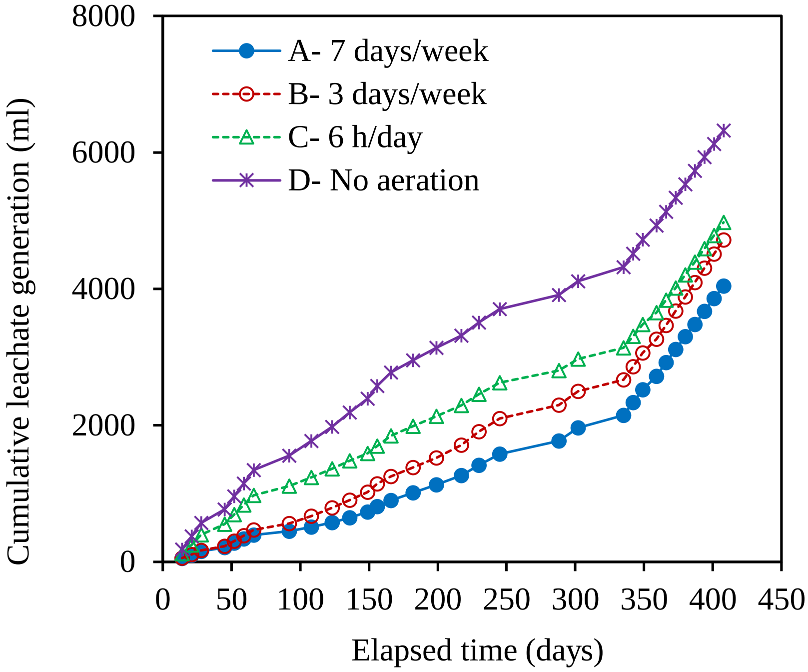


Fig. 2-11 Cumulative amount of discharged leachate

Table 2-6 Nitrogen mass balance at the end of Stage-1

Nitrogen amount (g-N)	A	B	C	D
N_{Li}	33.31	33.31	33.31	33.31
N_{Lf}	20.21	20.71	20.61	22.12
N_{Le}	1.53	1.68	2.11	4.70
N_L	0.019	0.061	0.015	0.196
N_M	22.33	12.50	8.62	2.41
NH_3-N gas	-	-	-	-
N_f (calculated)	44.1	34.96	31.35	29.43
T-N_{Si}	128.1	128.1	128.1	128.1
T-N_{sf}	38.32	47.57	50.52	63.26

N_{Li} and N_{Lf} = NH_4^+-N in the initial and after Stage-1, respectively in solid waste measured by leaching test (JLT-46); N_f (calculated)= total calculated NH_4+-N at the end of Stage-1 (sum of NH_4^+-N in solid waste (N_{Lf}), in leachate discharged during Stage-1 (N_{Le}), in leachate present inside reactors (N_L) at the end of Stage-1 and NH_4^+-N missing from the system (N_M) mainly by nitrification; T-N_{si} and T-N_{sf} = T-N contents in the solid phase at the beginning and after Stage-1. Exhaust NH_3-N gas from the reactors uncounted (not measured in this study).

2.4. Conclusions

The influence of aeration, at different intervals and controlled operational parameters, on the aerobic-anaerobic landfill method of organic solid-waste management was studied, using a laboratory-scale column experiment.

Intermittent aeration had a significant effect on the removal of carbon and nitrogen contaminants from organic solid waste when the other operational parameters, such as the ambient temperature, moisture content, and precipitation were kept at controlled levels.

Aerobic reactor A (seven days/week) had the highest removal rates of TOC, T-N, and NH_4^+-N and the lowest GHGs emissions, compared with the Aerobic-anaerobic reactors B and C and the Non-aerated reactor D.

Compared with Aerobic-anaerobic reactor B (3 days/week) and Non-aerated reactor D (no aeration), the Aerobic-anaerobic reactor C (6 h/day) exhibited improved leachate quality and more reduction of GHGs. Additionally, operating Aerobic-anaerobic reactor C was cost-effective and could save three-fourths of the energy cost of an Aerobic landfill.

The decomposition of organic carbon and the transformation of nitrogen contaminants take place rapidly during aeration periods, accelerating the degradation of organic matter as well as the removal of ammonium.

Throughout the course of the experiment, the conversion of nitrogen contaminants occurred during the different transition periods (between aeration and non-aeration), following simultaneous nitrification-denitrification.

The concept of AALM by intermittent aeration shows promise as a method for ameliorate the quality of leachate; moreover, this method could reduce the potential of emissions greenhouse gases by accelerating the decomposition of organic waste under aerobic conditions. Furthermore, intermittent aeration could shorten the later phases of landfill aftercare and could simultaneously reduce the energy costs.

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Appendix A

Greenhouse gas emission calculation

Diffusion flux:

Fick's law of diffusion: $J = -D\, \partial\phi / \partial x$

$$J = -\xi g D\, \partial\phi / \partial x$$

Where,

J = Diffusion flux (kg/m^2.s)

D= Diffusion coefficient in free air (m^2/s)

ξg= Gas tortuosity = 0.66ε (Penman tortuosity model, 1940)

ε = Waste air-filled porosity (m^3/m^3)

ϕ = Gas concentration (kg CO_2, CH_4 /m^3)

x = distance (m)

Advection flux:

Advection flux: $J = (Q.\phi)/A$

Where,

J = Advection flux (kg-gas/m^2.s)

Q= air outflow rate from column (m^3/s)

ϕ = gas concentration (kg CO_2, CH_4 /m^3)

A = horizontal cross sectional area of column (m^2)

Darcy's law:

u $=-k/\vartheta . \partial P/\partial z$

$=-k/\vartheta \quad \Delta P/\Delta z$

$=-k/\vartheta . ((P_{amb}) - (P_{amb}+\Delta P_{aeration}+\Delta P_{CH4}))/\Delta z$

$=k/\vartheta . (\Delta P_{aeration}+\Delta P_{CH4})/\Delta z$ ……………………… (1)

$Q/A = (Q_{aeration}+Q_{CH4})/A$………………… .(2)

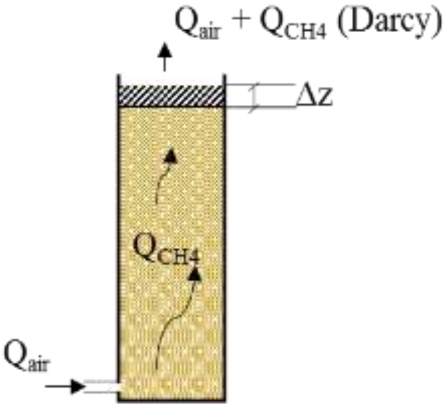


Fig. Aerated reactors

From (1) and (2),

$Q_{aeration} \quad = A. \quad k/\vartheta \ (\Delta P_{aeration})/\Delta z$

$Q_{CH4} \qquad = A. \quad k/\vartheta \ (\Delta P_{CH4})/\Delta z$

Where,

u = Darcy's avg. velocity (m/s)

k = Permeability (m2)

ϑ = viscosity (Pa s)

P = Pressure (Pa)

Z = Distance (m)

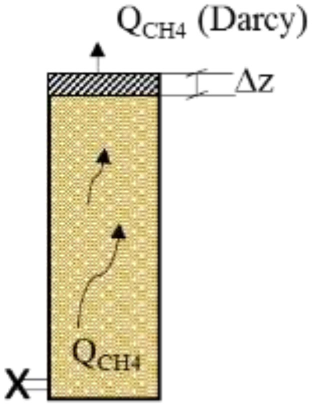


Fig. Non-aerated reactors

Table A-1: Parameters used in the carbon mass balance calculation

Parameters	*Unit*	*Values*	*Source*
Air density	kg/m^3	1.1644	
Permeability of cover soil	m^2	1E-13	(1)
Viscosity of air	pa-s	1.89E-05	(1)
Porosity	-	0.41	Measured
CO_2 diffusion co-efficient	m^2/h	0.06	(2)
CH_4 diffusion co-efficient	m^2/h	0.01	(2)
Tortuosity of CO_2	-		(3)
Tortuosity of CH_4	-		(3)

Source: (1) Tanaka, N (2000); (2) The Chemical Society of Japan (1993); (3) Penman (1940)

Table A-2: Carbon discharge amount by different emission processes

Emission process	Discharged carbon amount (g- C)			
	A	B	C	D
CO_2 – Advection	1022.74	451.70	654.17	0
CO_2 - Diffusion	62.68	28.15	39.59	56.83
CH_4 -Advection	100.97	474.16	298.05	0
CH_4 -Diffusion	1.02	4.74	3.00	10.21
CO_2- advection (pressure)	125.52	104.83	77.32	272.02
CH_4 - advection (pressure)	4.76	69.92	29.55	586.84
Total-C_{Gas}	1317.7	1133.5	1101.7	925.9

CHAPTER 3

Phenomenon of Nitrification-denitrification during Transition Period in the Aerobic-Anaerobic Landfill Method

3.1. Introduction

Most of the cities in developing countries facing problems related with solid waste disposal of high organic contents. Improper disposal of solid waste, generate leachate and gas are serious issues for environment which causing serious health problems as well. It is very necessary to minimize the environmental pollution caused by improper disposal and conventional way of landfilling for treatment of waste. Aeration induce in landfills can activated microorganisms, which can execute important functions for biodegradation and accelerate landfill stabilization (Ishigaki et al. 2003 and Sang et al. 2008). In an aerated landfill bioreactor, carbon and nitrogen were effectively removed from the solid waste and leachate (Sang et al. 2008). However, oversupply or ineffective aeration can decrease microbial activity and increase energy consumption. A balanced and effective utilization of both aerobic and anaerobic metabolic pathways of the microorganisms might be effective for accelerated stabilization.

The aerobic-anaerobic landfill method (AALM) is a novel approach to landfill that enables to reduce environmental pollution loads and accelerate stabilization. In previous studies aerobic and anaerobic conditions were alternately created in the vertical direction of a landfill by air injection (spatial AALM) (Wu et al. 2014 and Shimaoka et al. 2011). AALM can be create aerobic-anaerobic conditions alternately in specific layers of landfill by periodical air injection (temporal AALM). Apparently also intermittent aeration is the most practical strategy to create cyclically aerobic and anaerobic conditions and to reduce energy consumption inferable to the air supply in landfill bioreactors. Rapid stabilization of solid waste is possible with intermittent aeration at various oxygen and oxidation–reduction potential levels (Mertoglu et al. 2007).

Reportedly, an intermittent aeration strategy is favorable for separation of the acid formation phase and the methane fermentation phase, reducing the acid production time and furthermore, intermittent aeration can stimulate nitrifiers and denitrifiers in landfill bioreactors and therefore simultaneous nitrification and denitrification occur. But, there are very few studies have been done by intermittent aeration in landfill methods and almost no research has been done to investigate the transition period phenomenon during organic waste decomposition process between aeration and non-aeration periods.

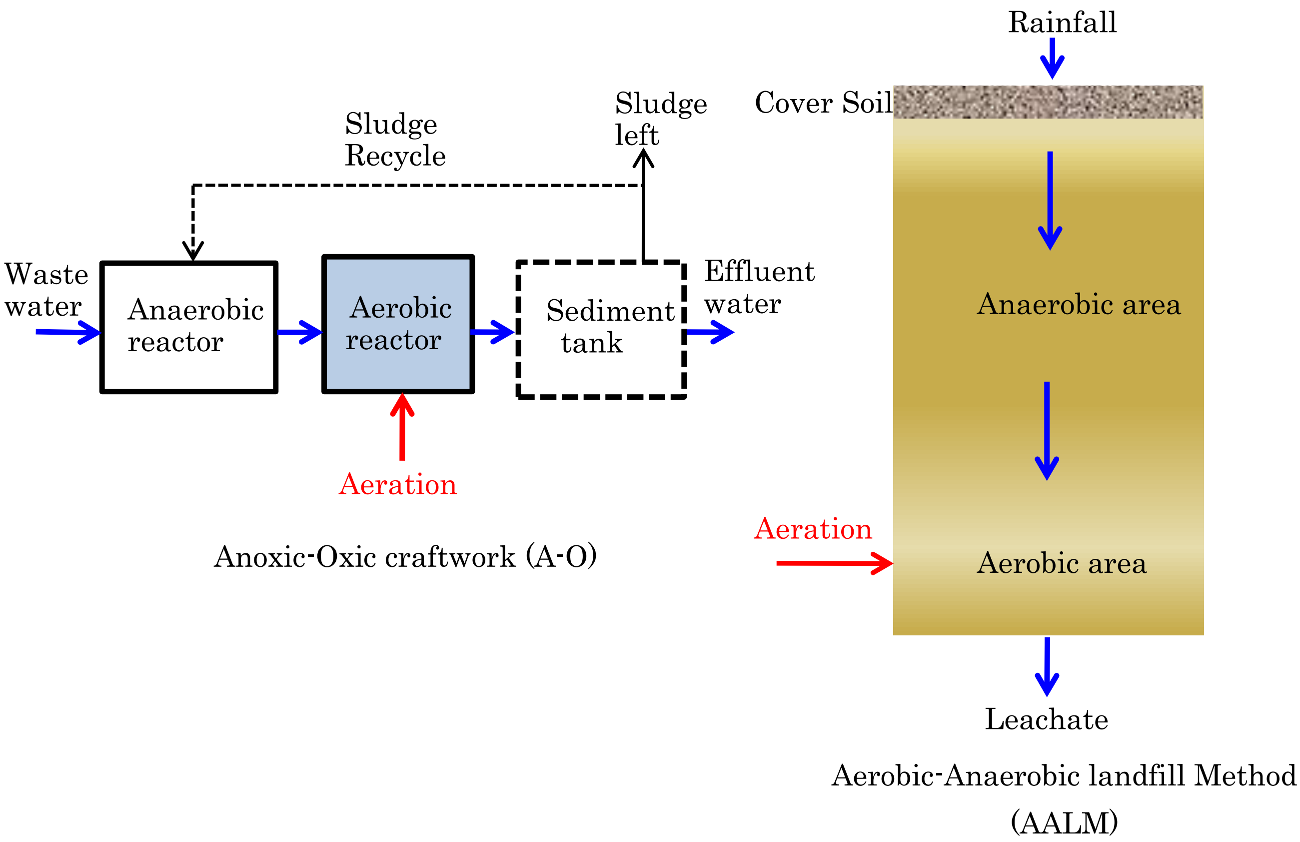
In this study, two types of intermittent aeration (B-3 days/week and C-6 hrs/day) were studied to investigate the phenomenon of nitrification-denitrification in an intensive seven-day experiment as a purpose of investigate the transition period phenomenon between aeration and non-aeration periods in terms of reducing leachate strength and GHGs emissions, which can ensure rapid stabilization.

3.2. Concept of Aerobic-Anaerobic Landfill Method

The exploration of Aerobic-Anaerobic Landfill Method (AALM) is serve with existing problems in the final disposal process of the refuse in developing countries. This idea is developed from wastewater treatment by the A/O (Anoxic/Oxic) method on as shown in Figure 3-1.

The advantage of this method is except the removal by decomposition it can also remove the nitrogen and phosphorus. It is the combined craftwork of the anaerobic decomposition method and aerobic activated sludge method. The concentration of dissolved oxygen (DO) is the key index in this method. In the stage of anoxic the DO concentration is no more than 0.2 mg/L, while in oxic stage its DO value should be more than 2 mg/L. In the anoxic period, heterotrophic hydrolysis bacteria will transform the suspended matters such as starch, fiber, carbohydrates and soluble organic pollutants in sewage into organic acid. This process degrades the large particle organic matters into little particle organic matters; transform the insoluble organic substances into soluble ones.

Thereafter, the decomposed little and soluble particles entered the oxic stage and treated under aerobic condition, the biological degradability and the efficiency of pumped oxygen can be improved. For nitrogen removal, the heterotrophic hydrolysis bacteria ammoniate the pollutants like protein and fat to release the NH_3 and NH_4^+ from the chains of organic matter or ammonia acid. Under enough oxygen condition, the nitrification autotrophic bacteria oxidize the NH_4^+ into NO_3^-. The circulation of treated water from oxic stage return to the anoxic pool and under anoxia condition, the heterotrophic bacteria can deoxidize the NO_3^- to N_2 through denitrification process, and to achieve the goal of purifying of wastewater. In the whole depth of the landfill, the anaerobic and aerobic area can be created by temporal air injection. When this alternately condition was created in the whole depth of the landfill, the downwards flowing leachate can be decomposed similar as the sewage treatment process in A/O craftwork. In order to carry on the experiment to prove the availability of the goals of AALM, laboratory scale study have been conducted by column experiment and evaluated the nitrification and denitrification processes.



Anoxic-Oxic craftwork (A-O)

Aerobic-Anaerobic landfill Method (AALM)

Fig. 3-1 Anaerobic-aerobic landfill craftwork

3.3. Materials and Methods

3.3.1. Experiment set-up

The research was conducted using two laboratory-scale column reactors (B and C). Fig.3-2 illustrates the schematic diagram of the column. Each column was a plexiglass cylinder (200 cm height × 10 cm diameter) in which 15 cm of gravel, 170 cm of solid waste, and 10 cm of cover soil were layered from the bottom to the top, respectively. Top 5 cm was kept as open space in the column reactors; therefore, the atmospheric air can penetrate the reactors from the top. An aeration pipe was installed at the bottom of the column to introduce air. Gas sampling points were set up at every 40 cm depth interval in the column reactors.

Each column reactor was filled with organic solid waste, collected from a composting plant, with a dry density of 0.32 t/m^3 and a wet density of 0.67 t/m^3 that had been shredded to pieces of less than 1 cm (1/10 of the column's internal diameter). The composition of the waste was primarily kitchen waste from restaurants, households, and food industries along with wood chips.

Table 3-1 presents the operating conditions of the column experiments. Columns B (3 days/week) and C (6 hrs/day) were operated with intermittent aeration (AALM reactors). The durations of air injection for columns B and C were set to three days/week (3 days continuous aeration and 4 days without aeration) and 6 h/day, respectively. The air injection rate during aeration was 7.1 l/kg-DM h. Temperature probes (Em50, Decagon Devices, Inc.) were placed in the middle of each column and were connected to a data logger (Em50, Decagon Devices, Inc.) to record the internal temperature. A total of 280 ml of water was added each week to each column to simulate rainfall and generate leachate.

Table 3-1 Operational condition of Aerobic-Anaerobic Landfill column

Operational Parameters	Reactor B	Reactor C
Air injection duration	3 days/week	6 h/day
Air injection rate (l/kg-DM h)	7.1	
Water supply as rainfall (mL/week)	280	
Packing density (t/m^3)	0.32 (dry), 0.67(wet)	
Moisture content (%)	50	
Ambient temperature(°C)	30 ± 1 ˚C	

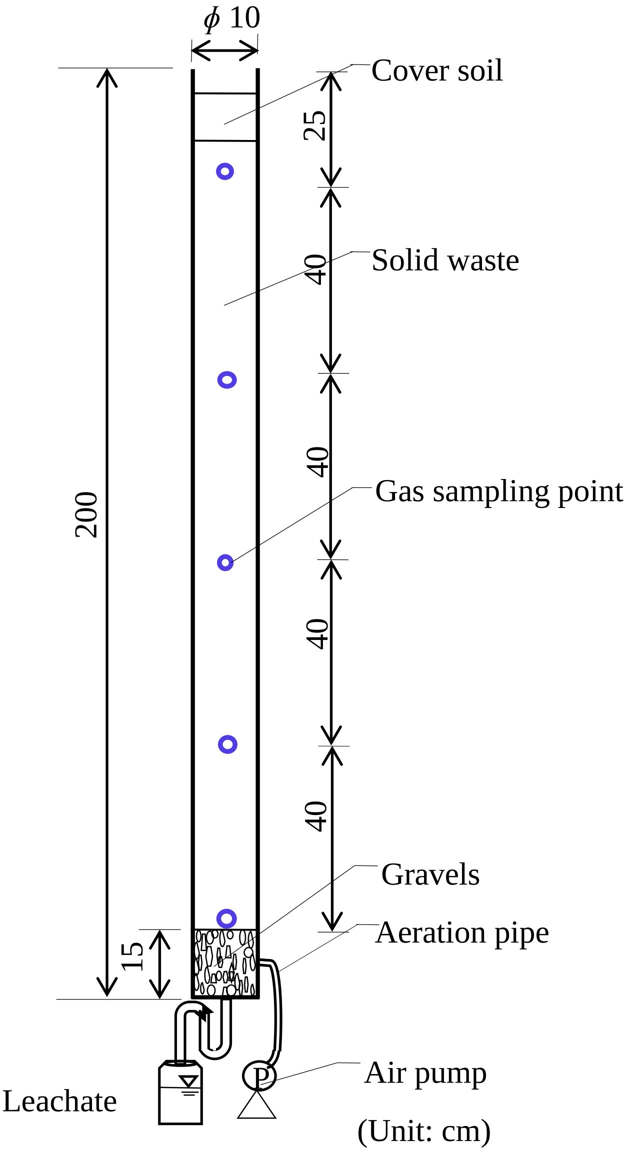


Fig. 3-2 Schematic diagram of column bioreactor

An intensive seven-day experiment was carried out from day 302 to day 309 in aerobic-anaerobic reactors B and C, with an aeration rate of 7.1 l/Kg-DM h. During this period, leachate and gas samples were collected at short intervals. The influence of the aeration and non-aeration conditions was investigated in one cycle for reactor B (3 days/week) and seven cycles for reactor C (6 h/day).

Gas samples were collected from the five gas sampling points in each column for analysis of the O_2, CO_2, CH_4, and N_2O concentrations at before aeration start, after aeration stopped and again before aeration start to complete the cycles. Leachate was collected in short intervals to monitor the pH, TOC, T-N, NH_4^+-N, NO_3^--N, and NO_2^--N concentrations. Micro-tubing pumps (Masterflex C/L, Cole-Parmer, USA) were used to sprinkle distilled water evenly to maintain the continuous leachate outflow. Leachate samples were collected by using fraction collector (CHF121SA, Cole-Parmer; USA).

3.3.2. Sampling and Analysis

The pH, electric conductivity and oxidation-reduction potential of the collected samples were measured with a Horiba pH, EC and ORP meter. The total organic carbon and total nitrogen contents of the sampled leachate were measured using a TOC analyzer (TOC-analyzer, Shimadzu). The carrier gas was pure oxygen supplied at a flow rate of 130–150 mL/min, and the oxygen gas pressure was 200 kPa. Ion chromatography (DX-120, Dionex) was carried out to detect the concentrations of NH_4^+-N, NO_2^--N, and NO_3^--N ions in the collected leachate. The columns for anions and cations were 4 × 50 mm in size, and the carrier gas was nitrogen. The gas concentrations of O_2 and CO_2 were measured by chromatography (GC8A, Shimadzu) using Total conductivity detector (TCD). The chromatographic column was a stainless steel column of 6m × 3 mm; the temperatures of the thermal conductivity were 200 °C and 40 °C for O_2 and CO_2 and 160 °C and 60 °C for N_2O. The carrier gas was helium (He), and its flow rate was 30 mL/min. The gas concentration of CH_4 was measured by gas chromatography (GC2014, Shimadzu) using TCD, in which the carrier gas was helium (He) and the temperature was 250 ^oC.

3.4. Results and discussion

3.4.1. Nitrification and dentrification phenomena in leachate characteristics

The transition period phenomenon of aerobic- anaerobic reactors was studied to assess the nitrification-denitrification process as a means of reducing the emission of GHGs and improving the leachate quality i.e., ammonia turnover, during the degradation of the organic solid waste. The representative data of leachate characteristics for nitrogen turnover for both reactors are presented in Figs 3-3 and 3-4.

As shown in Fig. 3-3, the TOC concentrations varied markedly during the transition period between the aeration and the non-aeration periods. In reactor B (3 days/week), when an aeration period started, a sudden drop was observed in the TOC. Later, values started to increase and, subsequently, gradually decreased again. The TOC decreased slightly when the aeration stopped and was subsequently shown to be almost stable. It is evident that aeration had a rapid effect and that it speeded up the decomposition of the organic carbon. Under aerobic conditions, the biodegradable materials are converted into carbon and water; therefore, compared with the anaerobic condition, the production of methane and volatile organic acid was reduced.

In reactor C (6 hrs/day), the TOC values were found to have increased rapidly in the discharged leachate samples immediately after the start of the aeration. However, after six hours, after the air supply had stopped, the TOC concentrations immediately exhibited a slow decreasing trend, which was observed over all seven days. This indicates that the transition period between aeration and non-aeration significantly influences the rate of decomposition by stabilizing or slowing down the process of decomposition during the non-aeration period, lasting 18 hours.

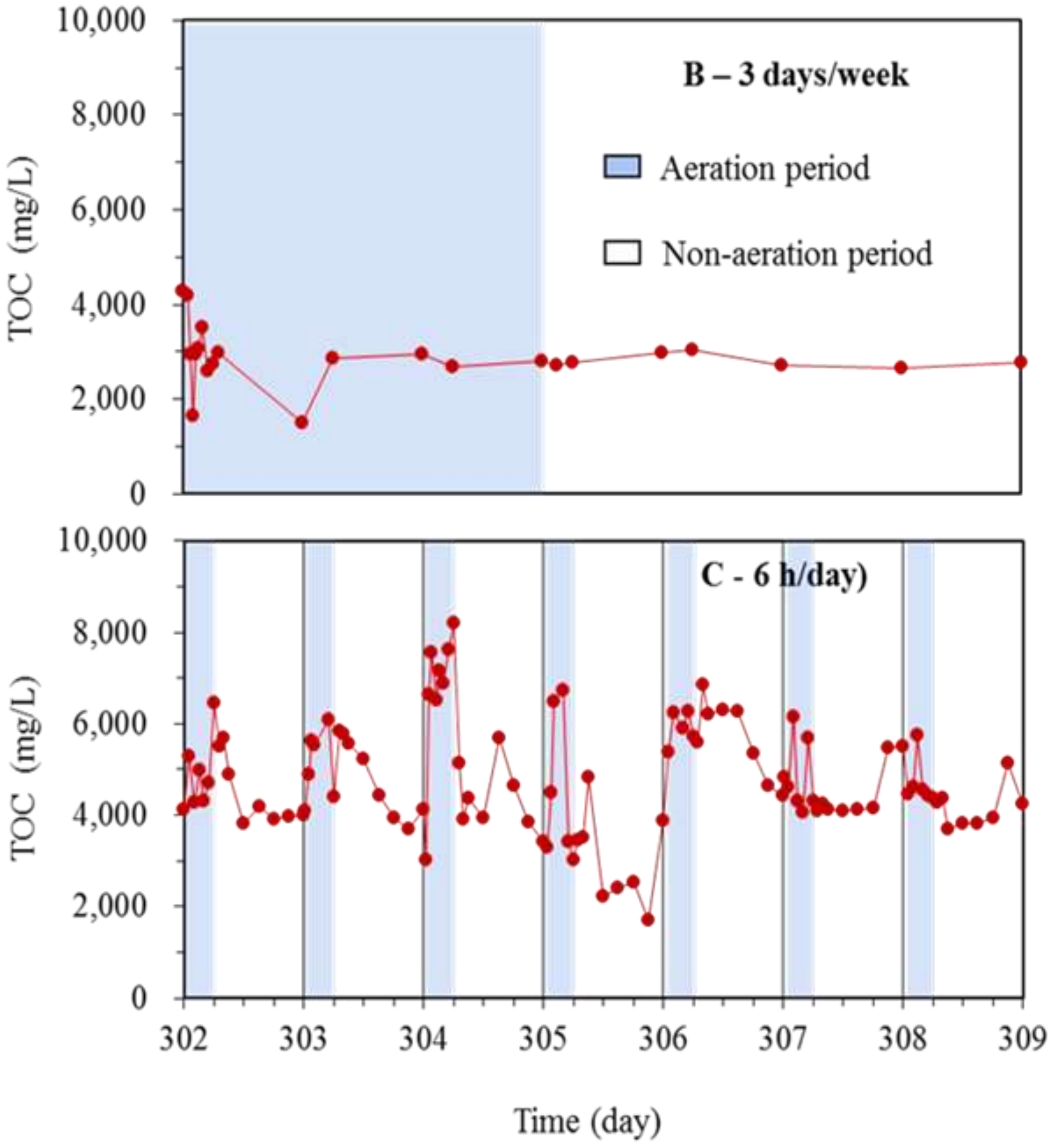


Fig. 3-3 Phenomenon of TOC in transition of aeration and non-aeration periods

Fig. 3-4 illustrates a phenomenon, similar to the change in the TOC concentration during both the aeration and the non-aeration periods that was also observed for the transformation of nitrogen compounds in reactors B and C. Immediately after the start of aeration, T-N and NH_4^+-

N showed increasing trends in both reactors B and C (few fluctuations were noted). Whereas, when the aeration stopped, both T-N and NH_4^+-N immediately continued their declining trends during all the cycles and in both reactors.

The oxidation of ammonium to nitrite and nitrate was successfully completed as a nitrification process during the aeration periods in both the reactors. Ammonium was oxidized into nitrite and nitrate during the aeration period and with no air supply. The complete and stable conversion of ammonium into nitrite to nitrate was obtained in both the reactors. According to Ritzkowski and Stegmann (2005), nitrite/nitrate is an indication of the nitrification process. If nitrite/nitrate does not remain at an elevated level, it is an indication of denitrification, which was observed during the non-aeration period of this study, as is shown in Fig.3-4. The nitrification process involves two biochemical reactions, which are (1) the oxidation of ammonium to nitrite by ammonia-oxidation bacteria, and (2) the oxidation of nitrite to nitrate by nitrite-oxidizing bacteria. It is evident that both reactions occurred during the aeration periods in both of the bioreactors; therefore, the nitrite and nitrate values increased.

During the aeration period, the concentrations of T-N, NH_4^+-N, NO_2^- -N, and NO_3^- -N increased, whereas, during the non-aeration period, there were no considerable changes. However, the formation of nitrite was confirmed during the denitrification process, which could stimulate the conversion of NO_3^- -N to N_2 gas under conditions of insufficient oxygen.

The initial increased changes of leachate quality with aeration indicate accelerated microbiological activity and the mobilization of compounds in the waste material, which leads to further degradation. In this regard, the findings of a study by (Öncü, Reiser, and Kranert 2012) are in accordance with the findings of the present study.

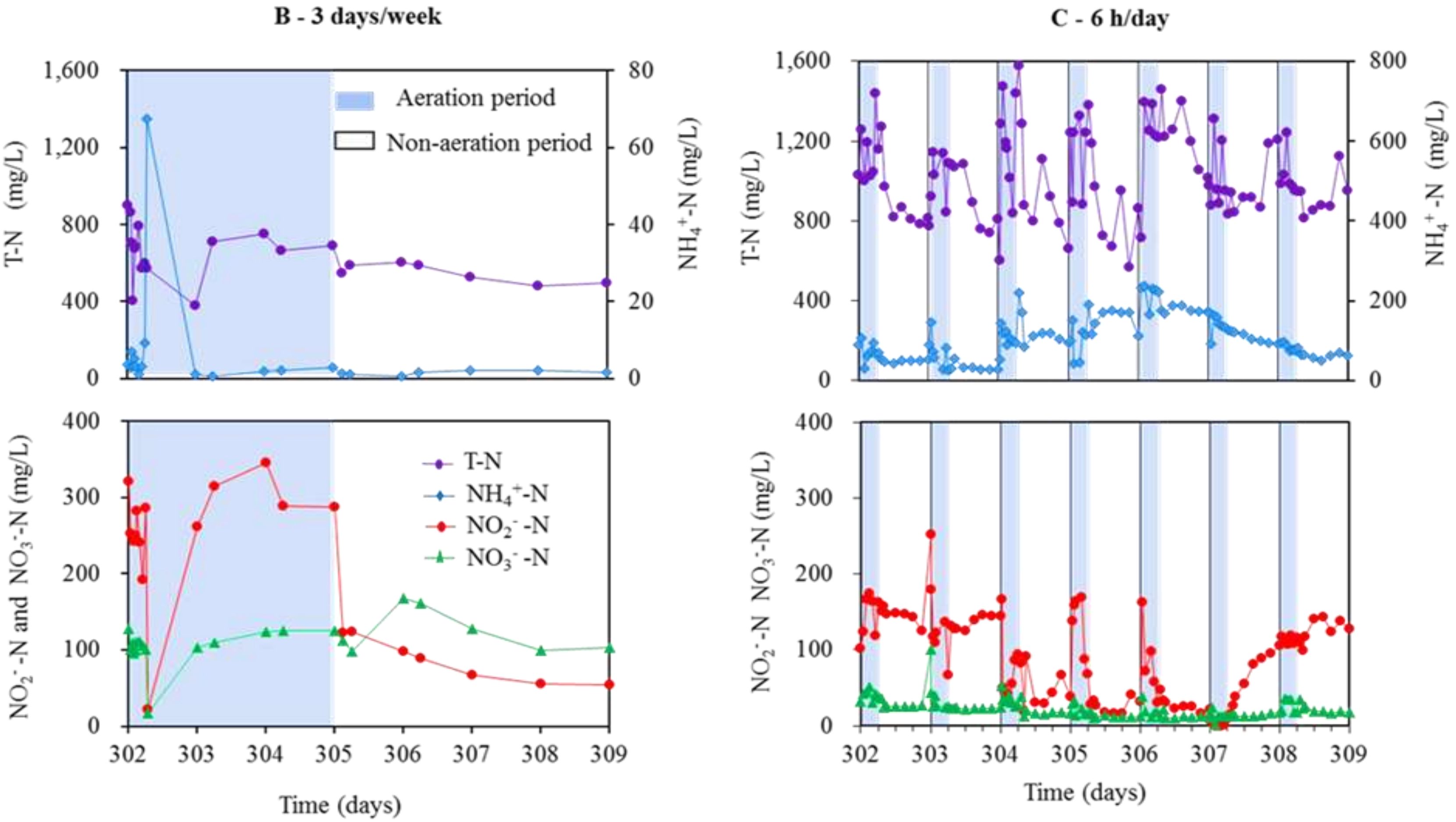


Fig. 3-4 Phenomenon of nitrogen conversion in transition of aeration and non-aeration Periods

3.4.2. Gas composition

The results pertaining to the transition period phenomena reveal the simultaneous processes of nitrification and denitrification in the aerobic-anaerobic reactors, with the presence of sufficient oxygen during the aeration periods, and with the lack of oxygen during the non-aeration periods. This is shown in Fig. 3-5 and Fig. 3-6 by the change in the gas profiles with depth and time for reactor B and C, respectively. In reactors B (3 days/week) and C (6 hrs/day), data on the gas composition during the aerated period were collected for one cycle and for seven cycles, respectively. It was found that the O_2 concentrations were high after aeration and gradually decreased after aeration had stopped. During both the aeration and the non-aeration periods, the oxygen concentration was sufficient for the simultaneous nitrification-denitrification process. The concentration of CO_2 was observed to be lower after the aeration period and higher during the non-aeration period. This is indicate that during aeration the biodegradation of organic waste accelerated the carbon conversion was mainly as CO_2. Whereas, CH_4 and N_2O were below detection limit on the entire seven-day experimental period.

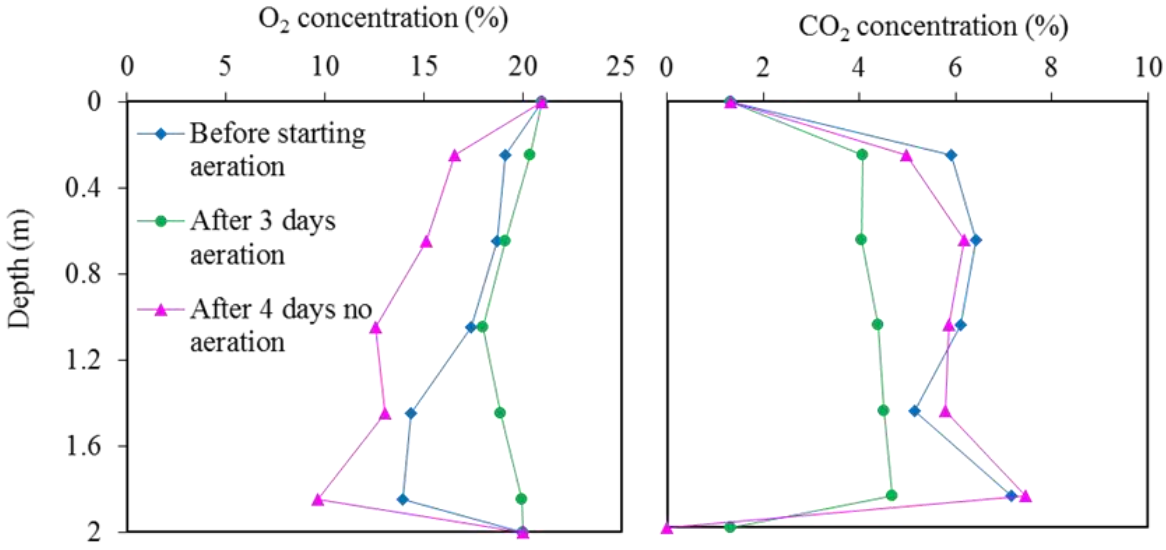


Fig. 3-5 Profile of O_2 and CO_2 concentration in reactor B during aeration and non-aeration periods (one cycle)

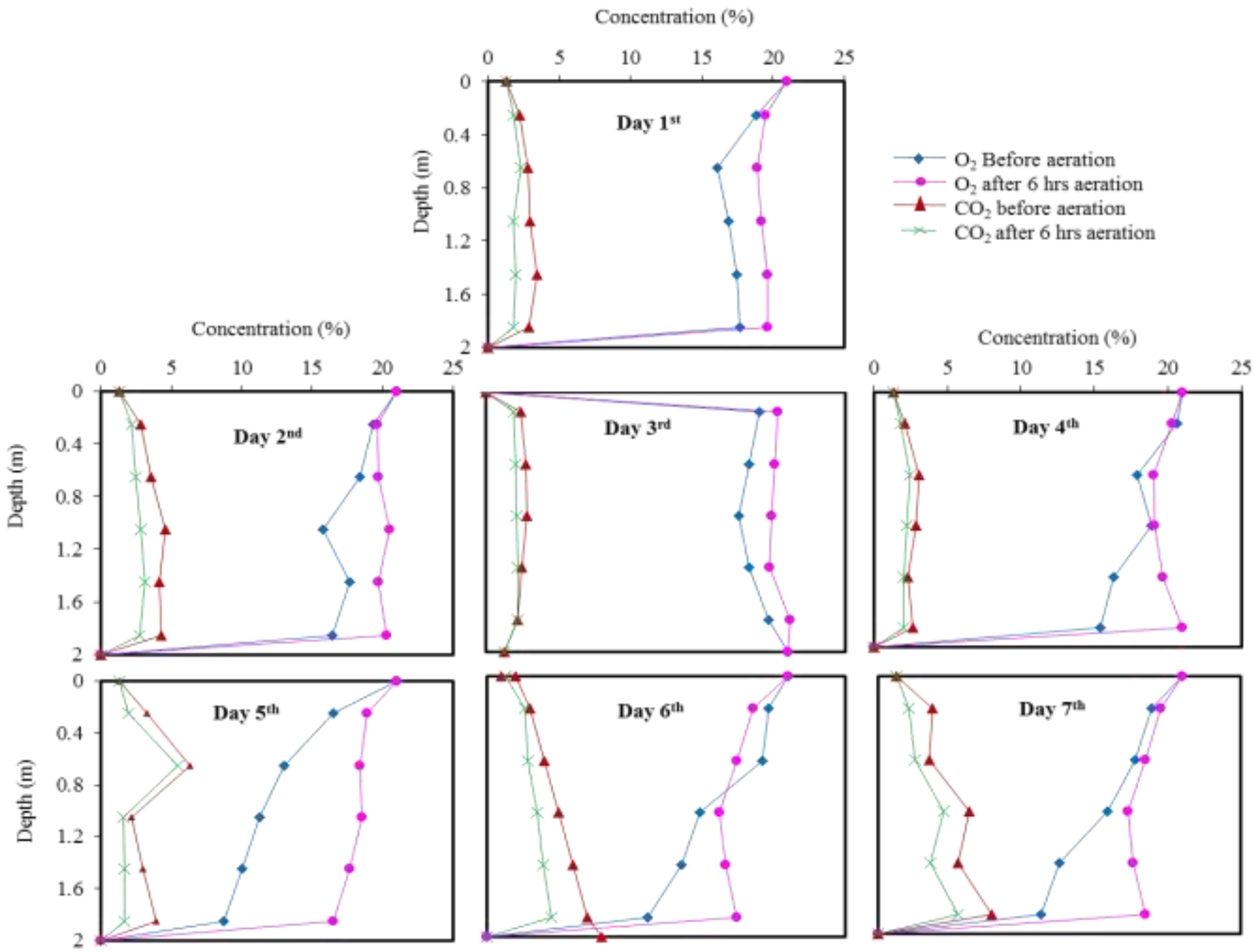


Fig. 3-6 Profile of O_2 and CO_2 concentration in reactor C during aeration and non-aeration periods (seven cycles)

3.5. Conclusion

Transition period phenomenon in this study reveal that during aeration period the decomposition of organic solid waste is accelerated and therefore initially increase the concentration of TOC, T-N and NH_4^+-N. Nitrogen transformation was following nitrification in aeration period and denitrification was occurred in non-aeration periods.

Both simultaneous nitrification and denitrification were observed during the transition periods between aeration and non-aeration periods with sufficient oxygen concentration inside the bioreactors.

The transition period phenomenon indicates that the aerobic-anaerobic landfill method by intermittent aeration is an efficient system for the removal of nitrogen contaminant i.e., ammonia transformation from the anaerobic system. Moreover, the nitrification and denitrification processes occurred simultaneously. These findings could be used in a pilot plant and a full-scale intermittent aeration landfill for the removal of nitrogen from anaerobic landfills.

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CHAPTER 4

Nitrous oxide production during nitrification from organic solid waste under temperature and oxygen conditions

4.1. Introduction

Nitrous oxide (N_2O) is a strong greenhouse gas with global warming potential of approximately 298 times that of carbon dioxide (CO_2) over a 100-year period (IPCC 2013). It has received great attention because it accounts for about 7.9 % of the overall greenhouse effect and has destructive potential for the stratospheric ozone layer (IPCC 2007; Ravishankara, Daniel, and Portmann 2009). Therefore, even low amounts of N_2O emission are unwanted.

It is estimated that agricultural soils contributed about 60% of the atmospheric N_2O emissions. (Mosier et al. 1998; Kroeze, Mosier, and Bouwman 1999). Another important N_2O emission source is wastewater treatment reported by the Environmental Protection Agency of the United States (2006) and IPCC 2007. Solid waste is one of the anthropogenic sources of greenhouse gas and landfill is a potentially high emitters of methane (CH_4) and N_2O. The environmental conditions in the landfills are favourable for the microbial production of N_2O as landfills containing organic waste with high nitrogen (N) level, and have alternating aerobic and anaerobic sites in the landfill volume.

The combination of aeration and leachate recirculation aeration to landfill may be an effective way for in-situ N removal. However, this can produce large amounts of $N_2O$and air addition to landfill layers can develop a favourable environment for simultaneous nitrification and denitrification, thereby allowing N removal and waste degradation (Berge, Reinhart, and Batarseh 2007; Shao, He, and Li 2008). Prantl et al. (2006) reported that aeration caused a rapid and complete reduction of ammonium in well-decomposed refuse. Other studies in different scales have been conducted (Jun et al. 2007., Onay and Pohland 1998., Price et al. 2003., Ritzkowski et al. 2006), most of which focused on the efficiency of nitrification or denitrification or both following aeration of landfills.

N-containing organic compounds in solid waste are degraded into ammonium, which is then converted into nitrate by ammonia- and nitrite-oxidizing nitrifiers. Nitrate, which is the final product of nitrification, is gasified to nitrogen gas in anoxic condition by the metabolism of several nitrate-, nitrite- and nitrous oxide-reducing denitrifiers. During these two biological processes, formation of N_2O as by-product from nitrification, or as an intermediate in denitrification occurs. Fig. 4-1 shows the N_2O generation pathway during nitrification and denitrification.

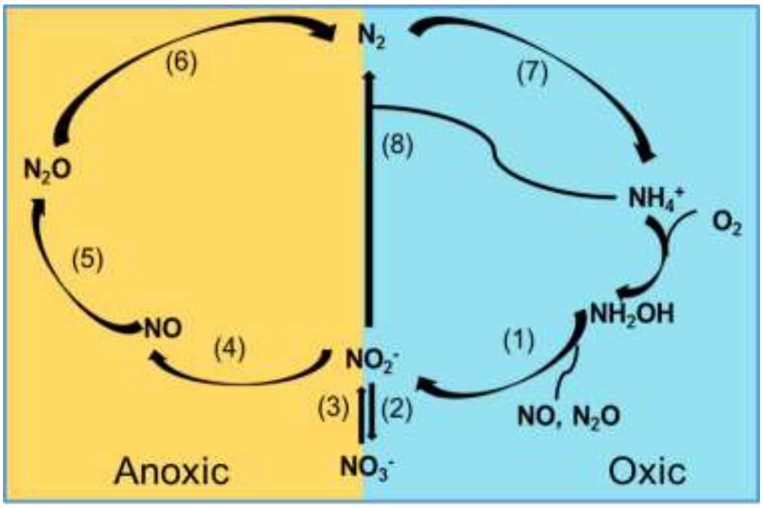

Fig. 4- 1: N_2O generation pathway; (1) Aerobic ammonium oxidation (AOB); (2). Aerobic nitrite oxidation (NOB); (3). Nitratereduction to nitrite (DEN); (4). Nitrite reduction to nitric oxide (AOB and DEN); (5). Nitric oxide reduction to nitrous oxide (AOB and DEN); (6). Nitrous oxide reduction to nitrogen gas (DEN); (7). Nitrogen fixation; (8). Ammonium oxidation with nitrite to nitrogen gas (Anammox). (Kampschreur et al. 2009).

However, the fraction of N_2O produced in each of these reactions varies considerably (Firestone et al. 1989., Bleakley and Tiedje 1982). Nitrification is carried out by autotrophs in aerobic conditions, whereas denitrification is accomplished by heterotrophs in anoxic conditions. During field scale closed landfill aeration, Powell et al. (2006) inferred that N_2O production was promoted on the basis of the observation that increasing gas flow did not lead to the reduction of N_2O concentration. In lab-scale landfill column experiment, He et al. (2011) observed that N_2O production was positively correlated with the prolonged aerobic time, and negatively correlated with the C/N ratio in the recycled leachate.

The mechanisms of nitrification or denitrification in solid waste mass were more intricate than in the case of wastewater treatment. Physical and chemical characteristics of solid particles and aerobic status of the bulk space among the particles may cause various unexpected results during a biological reaction process (Hwang and Hanaki 2000).

Previous studies (Skopp, Jawson, and Doran 1990., Linn and Doran, 1984) showed that the rate of nitrification was relatively high when the moisture content of the soil was 50–60 %. Moreover, N_2O production could increase with the increase in moisture content (Blackmer, Bremner, and Schmidt 1980., Yoshida and Alexander 1970). BREMNER and BLACKMER (1978) reported that the release of N_2O was completely stopped by addition of nitrification inhibitors like acetylene (C_2H_2) or nitrapyrin. C_2H_2 was used to investigate the contribution of nitrification and denitrification to nitrous oxide emission. Commonly, a low partial pressure of acetylene (l-100 Pa) was used to inhibit nitrification and, consequently, its contribution to the nitrous oxide production (Bedard and Knowles 1989.,Berg, Klemedtsson, and Rosswall 1982)). The difference between N_2O production in a control without C_2H_2 and in the C_2H_2 treatment was ascribed to nitrification. The remaining N_2O production in the C_2H_2 treatment was assigned to denitrification.

Intensive studies have been done to investigate the N_2O emissions in various ecosystems, such as, agricultural fields, wetlands, forests, and grasslands (Chen et al. 2000., Kiese & Butterbach-Bahl 2002., Ghosh et al. 2003., Erisman et al. 2008) whereas only a few investigations have been conducted about N_2O emissions in municipal solid waste (MSW) landfills. It is estimated that in developing countries more than 60 % of landfilled waste is organic with high moisture content. Therefore during in-situ aeration in a landfill site it is significant to investigate the forcing factors of N_2O generation under different O_2 concentration with temperature variations, i.e. nitrification conditions (O_2 range of 5-20 % and temperature of 20-40 °C).

This study mainly focused on the production of N_2O emission potential from organic solid waste of landfill remediation with air injection at different oxygen concentrations. First, batch incubation was carried out to investigate the N_2O production potential through nitrification for different temperature and aerobic status with high moisture (60 %) and substrates concentrations, i.e. total organic carbon (TOC) and NH_4^+-N. Second, the reactions responsible for biological nitrification and denitrification were identified through applying C_2H_2 as an inhibitor of nitrification; and third, N mass balance was calculated.

4.2. Materials and methods

4.2.1. Materials

The organic solid waste, with dominant composition of kitchen waste that from restaurants, households and food industry, was collected from a composting plant, Japan. Table 1 shows the physico-chemical characteristics of the solid waste.

Table 4-1 Physico-chemical characteristics of the solid waste with standard deviations

MC (%)	pH* (-)	EC* (mS/cm)	ORP* (mV)	TOC* (mg C/g-DM)	TN* (mg N/g-DM)	NH_4^+-N* (mg N/g-DM)	NO_x^- -N* (mg N/g-DM)
44.50 ±1.4	4.97±0.3	9.58±0.6	-3.6±1.3	36.2±1.3	12.1±0.3	2.5±0.1	0.09±0.0

MC: Moisture Content, EC: Electric Conductivity; ORP: Oxidation-Reduction Potential

* Measured by Japanese Leaching Test, JLT-46.

4.2.2. Incubation and sampling

In this experiment, three Erlenmeyer flasks (325±0.5 ml) were filled with 100 g of the above mentioned organic solid waste and capped with septum-fitted rubber stoppers. Flasks were purged with helium gas at 0.1 Mpa for about 15 min and then oxygen was injected in the flasks till the oxygen concentration reached the designated concentrations: 5, 10 and 20 %. Moisture content was maintained at 60 % by adding desired amount of distilled water in each flask. A duplicate experiment was carried out in parallel, but with addition of 100 Pa of C_2H_2 as a nitrification inhibitor. All flasks were incubated in a shaking water bath at 180 rpm using three different temperatures of 20, 30 and 40 ^{o}C. All flasks were pre-incubated for 1-2 d at three designated temperatures to recover nitrification activity of the solid samples before starting the experiment. The experiment lasted for 72 h and gas sampled from the head space of the flask after 1, 18, 24, 42, 48, 66 and 72 h by withdrawing 1 ml of gas sample with 1-ml analytical syringe. Oxygen and nitrogen concentrations inside the flask head space were analyzed at least twice a day. Pure O_2 was added in each flask to maintained desired concentration. The amount was decided by pre-experiment (*data not shown*).

4.2.3. Analytical Methods

Initial pH, Electric conductivity (EC), Oxidation reduction potential (ORP), nitrate, nitrite, ammonium and TOC, and Total nitrogen (T-N) measurement were carried by a leaching test authorised by Japanese Environmental Agency, JLT-46. After 72 h of the incubation experiment, the leachate of the solid waste was extracted using a centrifuge machine (H-200nRS, Kokusan, Japan) which was operated for 10 min at 100 rpm. Finally the collected leachate was filtered through a 0.45 µm filter unit for analysis of leachate characteristics: pH, NH_4^+-N, NO_2^--N, NO_3^--N, TOC, T-N. TOC and T-N were measured by a TOC analyser (Shimadzu, Japan). The carrier gas was pure oxygen supplied at a flow rate of 130–150 mL/min and oxygen gas pressure was 200 kPa. Ion chromatography (DX-120, DIONEX, Japan) was used to quantify the concentrations of NH_4^+-N, NO_2^--N, and NO_3^--N ions from collected leachate. The columns for anion and cation were 4 × 50 mm in size and the carrier gas was nitrogen. The sampled gas was analysed using a gas chromatograph (GC-8A, Shimadzu, Japan) equipped with a Porapak Q column and a Thermal Conductivity Detector (TCD). Oxygen and nitrogen concentrations inside the flask were analysed by gas chromatograph (GC-8A, Shimadzu, Japan) with TCD. Each of the experiment done in duplicate and each analysis above was conducted in triplicate. The results are provided as mean values.

4.2.4. Calculation of N_2O production rate

The N_2O production rate was expressed as mass of nitrogen in N_2O per dry mass (DM) of solid waste. The mass of N_2O was comprised by two parts, N_2O in liquid and gas phase. The amount of N_2O dissolved in liquid phase was recovered by the Bunsen adsorption coefficient (Wilhelm, Battino, and Wilcock 1977). The amount of N_2O in gas phase, was calculated by conversion of the N_2O gas concentration in the flask head space, quantified by GC-8A, into the N_2O gas concentration under standard condition (1 atm, 273.15 K) and multiplied the converted concentration by the head space of the flask.

4.2.5. Calculation of Nitrification rate

Two sets of leachate samples were used to measure the nitrification rate. One sample had 100 Pa of C_2H_2 added as an inhibitor to completely block nitrification, and the other one was identical with the former except C_2H_2 was added. Change between initial (0-time) and after 72 h of incubation (t-time) of NO_3^--N concentration per unit time was estimated as net nitrification rates in two samples. The difference in net nitrification rates between the two samples equalled the gross nitrification rate.

4.2.6. Calculation of nitrogen mass balance

A mass balance calculation was done for the nitrogenous compounds to verify the nitrification and denitrification process, based on the conceptual model as shown in Fig. 4- 2, of calculating the amount of nitrification and denitrification. The amount of organic nitrogen was calculated as the difference between T-N and inorganic nitrogen. The amount of organic nitrogen mineralised to ammonia [(i) in Fig. 4-2] represent the inflow to the ammonia balance, whereas the outflow from the ammonia represents the amount of nitrification. Since the increase in ammonia [(iv) + (v) in Fig. 4-2] was the difference between these two, the amount of nitrification was estimated from this balance. Nitrogen gas was calculated from the balance reduced total nitrogen and increased ammonia and N_2O. Free ammonia nitrogen (NH_3-N) concentration was assumed to be in aerobic condition and pH was considered 8.5 (average pH in aerobic condition). The free ammonia fraction was calculated using Eq. (1).

$$[NH_3 - N] = [NH_4^+ - N] \cdot 10^{pH} / [\left(\frac{K_a}{K_w}\right) + 10^{pH})] \qquad (1)$$

Where NH_3-N is the free ammonia as N (mg/L); NH_4^+-N is the total ammonium as N (mg/L) and K_a and K_w are the dissociation constants of ammonia and water. The ratio of K_a/K_w is the related to temperature in Eq. (2).

$$\frac{k_a}{k_w} = \exp[\frac{6344}{273 + °C}] \qquad (2)$$

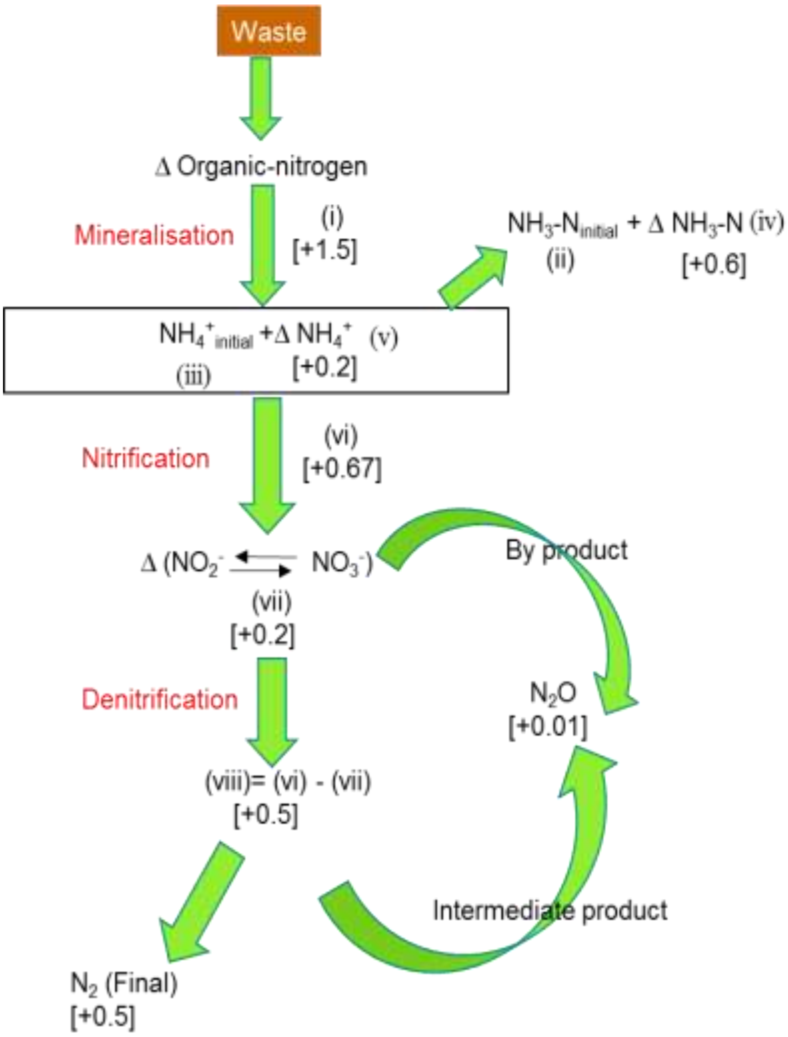

Fig 4.2 Conceptual model to calculate the amount of nitrification and denitrification (Results when O_2 concentration 10% at 30°C, values in brackets are avg. mass changes in each step as mg N/g-DM

4.3. Results and discussion

Fig. 4-3 illustrates the N_2O production rate with and without C_2H_2 treatment in different temperatures. Without C_2H_2 treatment (controlled conditions), N_2O production rate ranged from 0.40 to 1.14 μg N/g-DM/h, and with C_2H_2 ranged from 0.09 to 0.67 μg N/g-DM/h. It was observed that in the control condition, N_2O production rate was high under higher O_2 concentration. N_2O production rate was increased from 0.41 to 0.82, 0.40 to 0.79, and 0.68 to

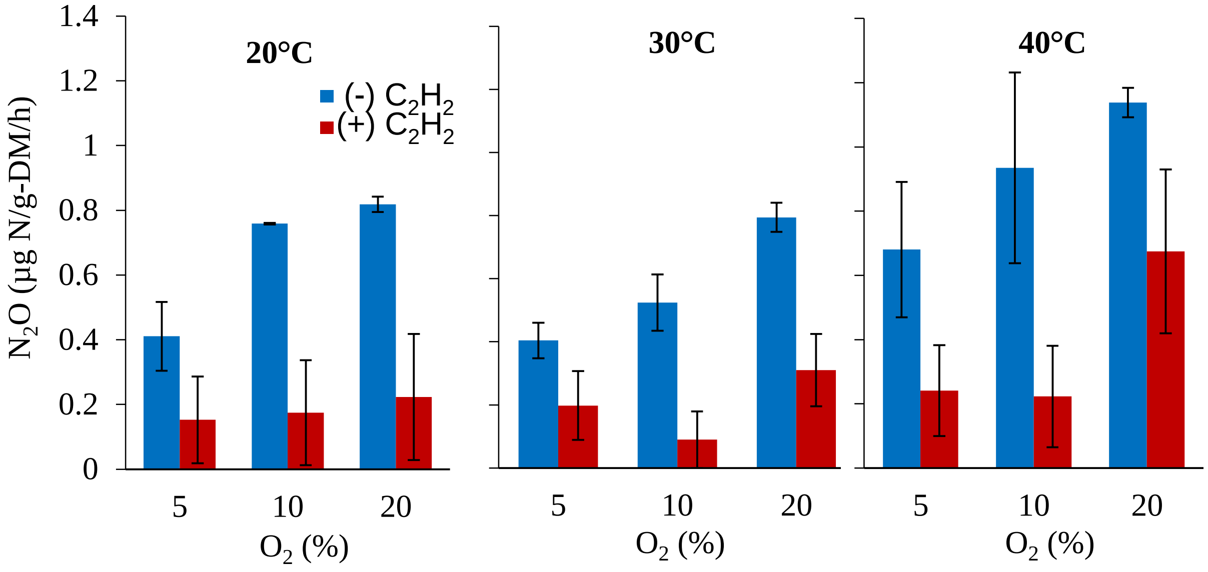

Fig 4- 3 N_2O production rate with temperatures and O_2 concentration

1.14 µg N/g-DM/h as the concentration of O_2 increased from 5 to 20 % in 20, 30 and 40 °C temperatures, respectively. In all cases, C_2H_2 treatment inhibited the generation of N_2O gas to a large extent. C_2H_2 inhibits ammonia oxidation and therefore inhibits N_2O production by both nitrification and nitrifier denitrification, and O_2 suppresses the reduction pathway of nitrifier denitrification. Since the difference between the production of N_2O in a control without C_2H_2 and in the C_2H_2 treatment was ascribed to nitrification and the remaining N_2O production in the C_2H_2 treatment was assigned to denitrification, we presume that nitrifier denitrification was restrained under this condition (with C_2H_2). Hence, it was evaluated that about 63, 77 and 73 % of N_2O was produced by nitrification at 20 °C; 50, 82, and 61 % at 30 °C and 65, 76, 41 % at 40 °C with 5, 10, and 20 % O_2 concentration, respectively. Therefore, assessing the contribution of nitrification and denitrification with C_2H_2 treatment to N_2O production during organic solid waste decomposition is a suitable method. However, production of N_2O at 30 and 40 °C was observed to be a little higher in the presence of C_2H_2 at 20 % O_2 concentration (Fig. 4-3). This might be due to the availability of O_2, redox potential, and the availability of substrates, which are the regulators of some nitrifiers that also able to denitrify and thus cause significant N_2O release (Loecke, 2009; Burford, 1975; Ellis S, 1998).

Moreover, low content of O_2 might have inhibited the activity of nitrifiers and enhanced the denitrifiers. Therefore, more information on different ammonia oxidizers and their sensitivity to C_2H_2 is needed to clarify N_2O production during solid waste decomposition. Temperature also had a strong influence on N_2O production. Production rates of N_2O at all designated O_2 concentrations in the bulk spaces in controlled condition were higher at 20 and 40°C than at 30 °C. However, the inhibitors did not always cause the expected reduction in the production of N_2O compared to the controls (Wrage et al. 2004) which accords with this study. The important point is that N_2O was not necessarily generated only by nitrification, but also by denitrification. The availability of Carbon (C) and N typically affect the rate of denitrification; however, some other crucial parameters can also regulate N_2O production by dentrification, such as C and nitrate ratio, electron donors and electron acceptors ratio (Senbayram et al. 2009). Moreover, Beck-Friis et al. (2015) reported that many different bacteria can denitrify and some of them can survive in thermophilic and mesophilic temperatures.

The gross nitrification rate was maximum at 30 °C and the minimum at 40 °C, illustrated as shown in Fig. 4-4. The gross nitrification rate increased gradually with increase of O_2 concentration and the trends were similar for all designated temperatures. These results show that at optimum temperature, N_2O production rate is affected by nitrification process. On the other hand, nitrifiers might inhibited at 40 °C because they have an optimal temperature range of 30–35 °C (Willers et al. 1998) and consequently denitrification might affect N_2O production at the same time as nitrification is occurring (in presence of O_2).

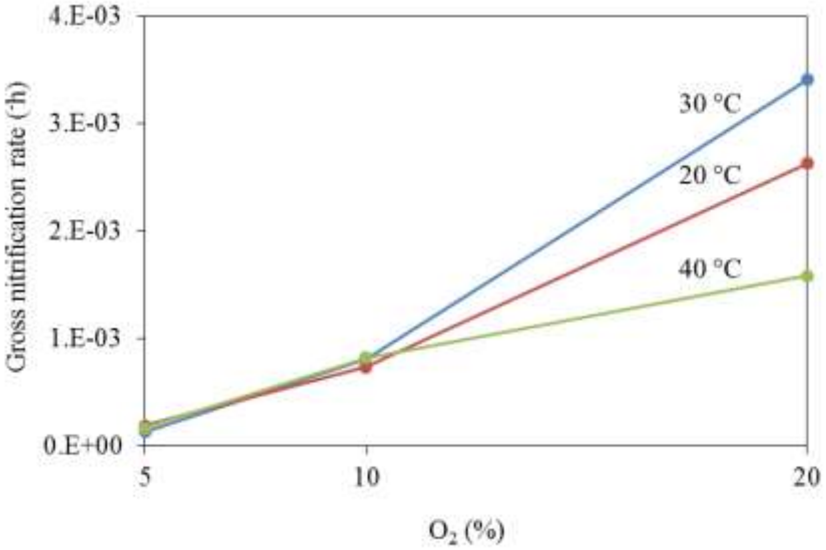

Fig. 4- 4 Variation of gross nitrification rate with temperatures and O_2 concentrations

High O_2 concentration accelerated the decomposition of solid waste and therefore also accelerated N transformation as shown in Fig. 4-5. It was observed that conversion of NH_4^+-N and NO_x^--N increased with gradual increase of O_2 concentration in the bulk space. Highest NO_x^--N formation was observed in 20 and 30 °C temperature as 0.46 and 0.54 mg N/g-DM, respectively, at 20 % O_2 content. On the other hand, 40 °C confined the accumulation of NO_x^--N than 20 and 30 °C. Moreover, low formation of NO_x^--N at 5 % O_2 content indicated that nitrification was limited by a low content of O_2.These results infer that nitrification is the dominant pathway of N_2O production as a by-product during spontaneous decomposition of intermediates of ammonia oxidation, e.g. hydroxylamine (NH_2OH). Furthermore, incomplete oxidation of NH_2OH might lead to the development of N_2O during nitrification (Hooper and Terry 1979).

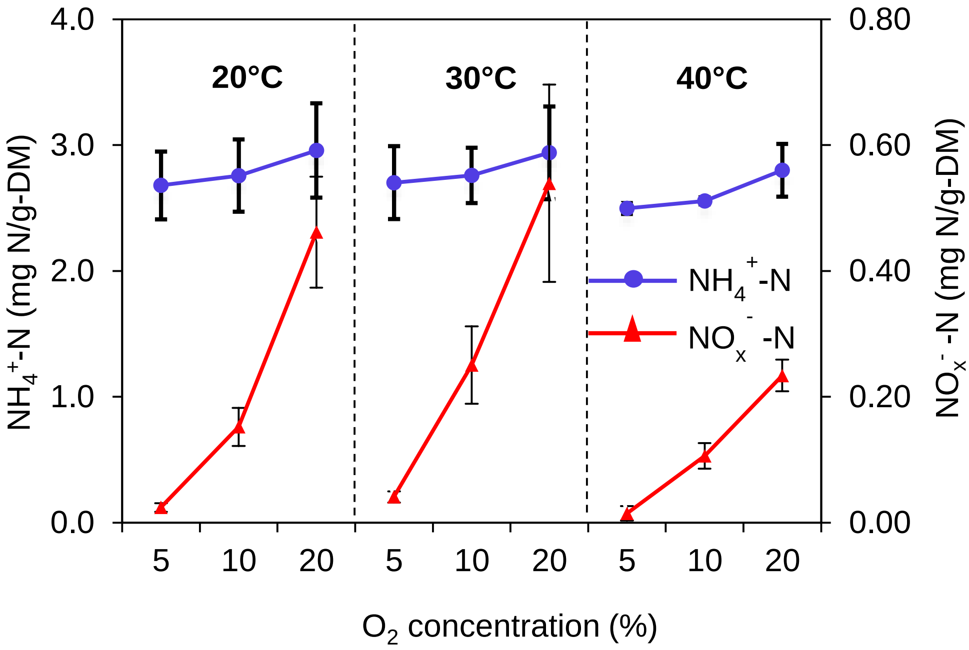

Fig. 4- 5 Nitrogen conversion with temperatures and O_2 concentrations

Table 4-2 summarises the nitrogenous mass balance and estimated values with occurence of nitrification and denitrification in all designated temperatures with O_2 concentrations of 5, 10 and 20 % in the bulk spaces. Fig. 4-6 illustrates that the amount of nitrification (nitrified ammonium) increased with increased O_2 , while denitrification diminution occurred with increased O_2 concentrations. However, temperature has a potential influence on nitrification in organic waste decomposition. The highest amount of nitrification was 0.57, 0.67 and 0.74 mg N/g-DM at 5, 10 and 20 % O_2, respectively, at 30 °C. In contrast, amounts of nitrificaton were about half at 40 °C compared with 20 and 30 °C, showing that the nitrification process is inhibited in higher temperature even with high O_2 content. In aerobic conditions, a combination of nitrification and denitrification occurs regardless of the efficiency of the aeration system, but on a micro scale (Ritzkowski, Heyer, and Stegmann 2006).However these pathways appear after ammonia volatilization due to the low growth rate of autotrophic nitrifiers and potential competition with heterotrophs when organic substrate is abundant (Berge, Reinhart, and

Townsend 2005), which is in accord with the results of this study. Hollocher et al. (1981) found that under aerobic conditions the biotic process of chemoautotrophs using ammonia as an energy source produced N_2O by several pathways: nitrifier nitrification, nitrifier denitrification, and nitrification-coupled denitrification.

The results of the current study show that the optimization of O_2 concentration is one of the crucial factors in N_2O production because aeration in landfills accelerate the decomposition of organic matter and therefore release energy that increase temperature, reduce CH_4 production and promote simultaneous nitrification and denitrification processes. Increasing O_2 content enhanced the nitrification process, which was also affected by temperature, with N_2O produced as a by-product. Therefore, investigating the effects of environmental and operational conditions such as temperature, moisture content, and O_2 content are significant and valuable for the consideration of N_2O production from solid waste landfill where decomposition of organic solid waste is a long-term process. The findings of this study will help understanding of the N and temperature dynamics of N_2O production during aeration in large-scale solid waste landfill remediation with appropriate control of environmental and operational parameters.

Table 4-2 Initial and final values for N-mass balance during 72 h, at different temperature and O_2 concentration (analysis were done in triplicate and values are provide as mean values.

Parameters (mg N/g DM)	**Initial values**	**Final values**								
		20°C			30°C			40°C		
		5[a]	10[a]	20[a]	5[a]	10[a]	20[a]	5[a]	10[a]	20[a]
Total -nitrogen	12.1	11.2	11.3	11.6	11.0	11.1	11.2	11.0	11.0	11.0
Organic-nitrogen	9.5	8.5	8.4	8.2	8.2	8.0	7.8	8.5	8.3	8.0
Inorganic-nitrogen	2.6	2.7	2.9	3.4	2.7	3.0	3.5	2.5	2.7	3.0
Nitrate	0.088	0.02	0.06	0.19	0.01	0.06	0.25	0.01	0.06	0.12
Nitrite	0.0	0.01	0.1	0.27	0.03	0.19	0.29	0.0	0.05	0.12
Nitrous oxide[b]	0.0	0.02	0.02	0.02	0.01	0.01	0.02	0.03	0.01	0.02
Nitrogen gas	0.0	0.6	0.5	0.2	0.6	0.5	0.3	0.3	0.26	0.17
Ammonia	0.0	0.3	0.3	0.3	0.5	0.6	0.6	0.8	0.9	0.9
Ammonium	2.5	2.68	2.76	2.95	2.71	2.85	3.35	2.85	2.90	2.99
Amount of nitrification	-	0.52	0.55	0.59	0.57	0.67	0.74	0.22	0.28	0.32
Amount of denitrification	-	0.59	0.49	0.22	0.62	0.51	0.29	0.29	0.27	0.17
TOC (mg C/ g DM)	36.2	54.74	55.6	56.05	54.65	47.32	49.01	54.79	55.21	55.20

a: O_2 concentration (%); b: Mean calculated value

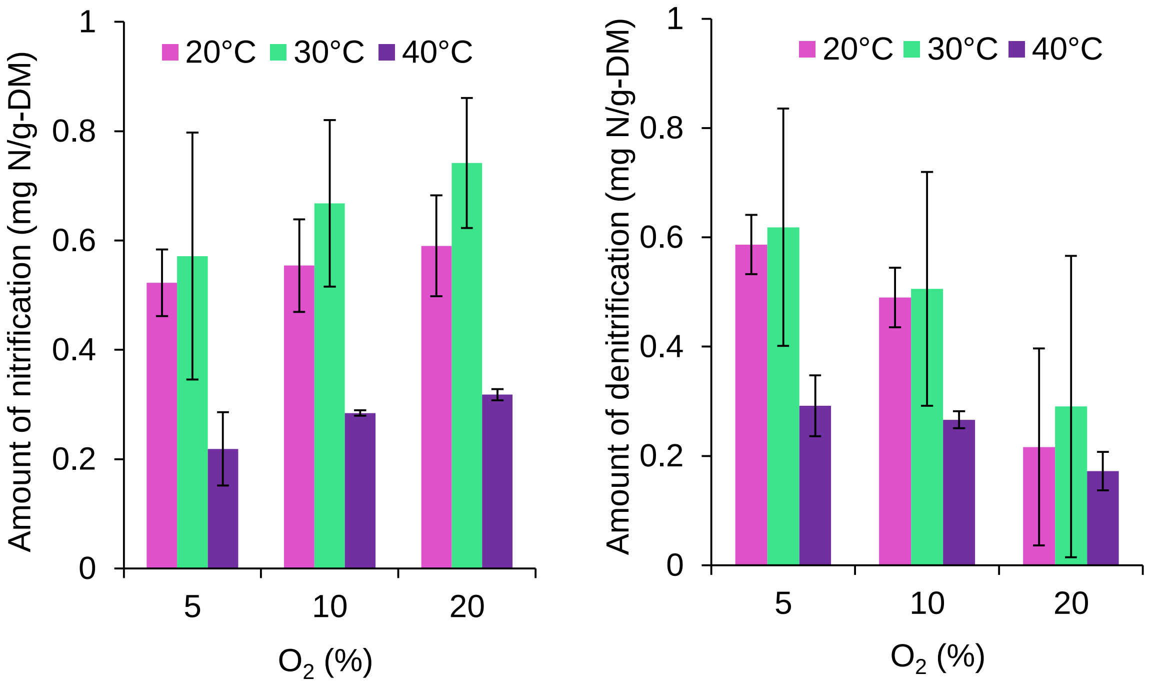

Fig. 4-6 Amount of nitrification and denitrification with temperatures and O_2 concentrations

4.4. Conclusion

This study was carried out to clarify the potential of N_2O production during nitrification process in organic solid waste decomposition. The batch incubation experiments of organic solid waste under different oxygen concentrations and temperatures produced the following specific findings:

1. Nitrification, denitrification and N_2O production depended on O_2 concentration and temperature.
2. The production rate of N_2O in organic solid waste decomposition was in the range from 0.40 to1.14 μg N/g-DM/h.
3. Nitrification was the dominant pathway of N_2O production; however, some N_2O production was due to denitrification because both nitrification and denitrification took place simultaneously.

4. Treatment with C_2H_2 is a suitable method for assessing the nitrifier contribution to the N_2O emission from organic solid waste. However, further study is needed to clarify the sensitivity of ammonia oxidizer with respect to N_2O production.

This study will help understanding of N and temperature dynamics for N_2O production in large-scale landfills. The results provide insights into N_2O production potential during aeration that may help to minimize greenhouse gas emissions with appropriate control of the environmental or operational parameters and N turnover. Further microbiological study is needed to verify the potential of N_2O production during both nitrification and denitrification and to clarify the influence of different parameters and substrate concentrations such as moisture content, TOC, and NH_4^+-N.

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CHAPTER 5

Field Study of Nitrous Oxide Production with *In-situ* Aeration in a closed Landfill Site

5.1. Introduction

Nitrous oxide (N_2O) is a trace gas that has gained considerable attention, as it has an important contribution to global warming and participates in the depletion of stratospheric ozone layer. Therefore, even a very low amount of N_2O is even undesirable. Around the world, landfilling continues as a primary method to manage municipal solid waste (MSW), which is one of the high emitters of greenhouse gases such as CH_4 and N_2O during the biodegradation of solid waste. In recent years, landfill aeration with or without leachate recirculation has attracted increasing attention worldwide for fast, controlled and sustainable conversion of landfills into a biological stabilized condition. In traditional anaerobic landfill, ammonia, the long-term pollutant, can be dramatically removed in aerobic landfill since the introduced air can create a favorable environment for simultaneous nitrification and denitrification (Price, Barlaz, and Hater 2003; Prantl et al. 2006; Ritzkowski, Heyer, and Stegmann 2006;Berge, Reinhart, and Batarseh 2007). N_2O originates from the biodegradation, or combustion of nitrogen (N)-containing solid waste during microbial nitrification and denitrification. During these two processes, formation of N_2O as a by-product from nitrification, or as an intermediate product of denitrification (Wrage et al. 2001). However, the fraction of N_2O produced in each of these reactions varies considerably (Firestone and Davidson, 1989; Bleakley and Tiedje 1982).Nitrification is carried out by autotrophs in aerobic conditions, whereas denitrification is accomplished by heterotrophs in anoxic conditions. Therefore, landfilling and landfill leachate treatment may have huge contribution to release of N_2O into the atmosphere.

Recently, N_2O emission from aerobic landfill started to draw much attention from the world since landfill aeration induced not only N removal, but can also impel N_2O emission. During field scale closed landfill aeration, Powell et al. (2006) inferred that N_2O production was promoted on the basis of the observation that increasing gas flow did not lead to the reduction of N_2O concentration. In lab-scale landfill column experiment, He et al. (2011) observed that N_2O production was positively correlated with the prolonged aerobic time, while negative related with the C/N ratio in the recycled leachate.

N_2O emissions have been investigated intensively in various ecosystems, such as, agricultural fields, wetlands, forests and grasslands (Chen et al. 2000; Kiese and Butterbach-Bahl 2002; Ghosh, Majumdar, and Jain 2003 and Erisman et al. 2008), whereas there have been few investigations of N_2O emissions in municipal solid waste (MSW) landfills. However, fluxes of N_2O from MSW landfills are comparatively higher than those from other systems such as agricultural and forest soils (Rinne et al. 2005). Some studies have examined the effects of cover soils and leachate surface irrigation on N_2O emissions from MSW landfill to determine several biochemical reactions which may responsible for N_2O emission (Zhang, He, and Shao 2008; Zhang, He, and Shao 2010). However, to our knowledge no study has investigated the production potential of N_2O inside the aerated MSW landfill, where landfill aeration technology may induce N_2O production in presence of oxygen.

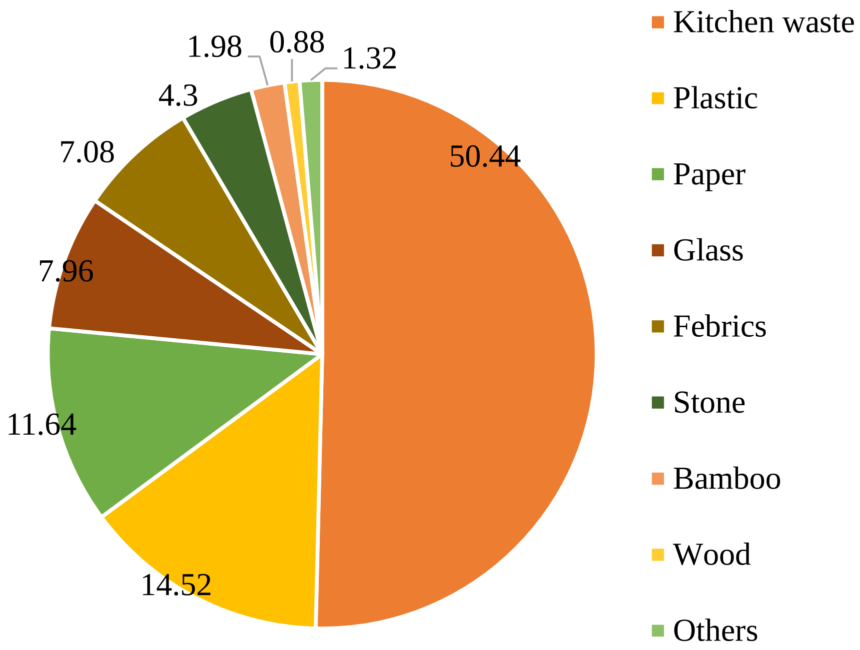
The main purpose of this study was to investigate the production potential of N_2O inside the aerated landfill. In this study, air was injected into a closed landfill site and the major N_2O generation factors analyzed with landfill gases. Correlative analysis of temperature in the landfill cell has been conducted to understand the effects on N_2O production as well. The findings provide insights concerning the production potential of N_2O in an aerated landfill that may help to minimize greenhouse gas emissions with appropriate control of operational parameters and biological reactions of N turnover.

5.2. Materials and methods

5.2.1. Characteristics of Landfill Waste

About 50.44 % of the waste deposited in the Chongming landfill site, China,was kitchen/organic waste in original composition and about 15 and 12 % were plastic and paper, respectively shown in Fig.5-1(a). Before starting the in-situ aeration experiment kitchen/organic waste was about 20 % (wt.) and 19 and 10 % were measured plastic and paper, respectively, shown in Fig. 5-1 (b). The deposited landfill solid waste was collected from the experimental site after removing the cover soil and measured the composition on a weight basis.

(a)



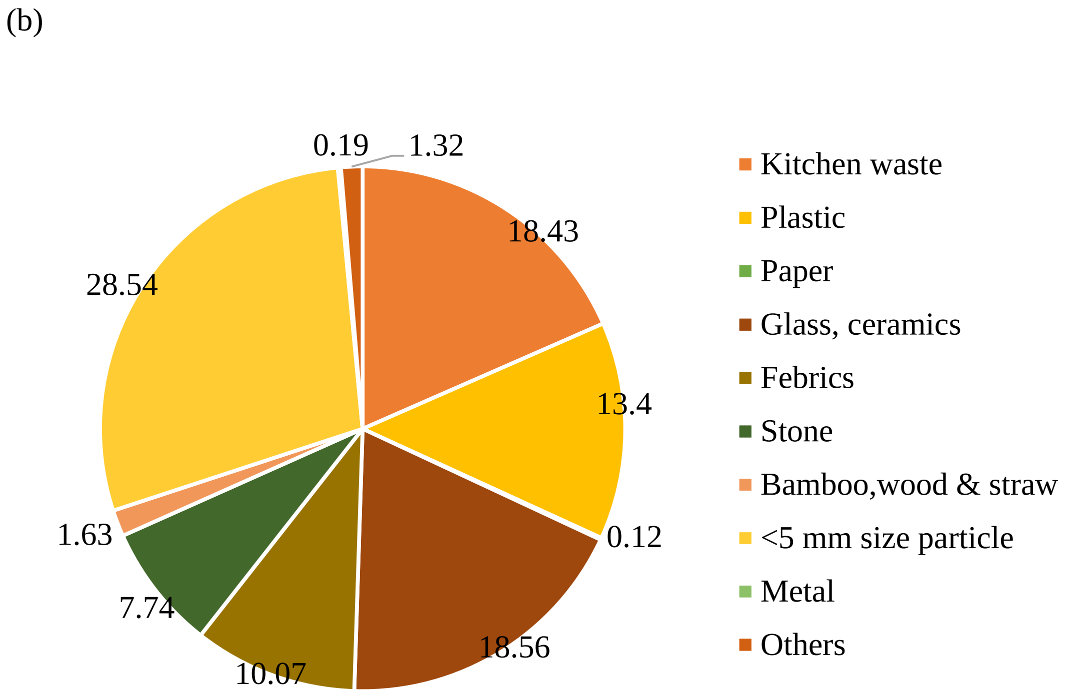


Fig. 5-1 (a) Original composition of Chongming landfill (b) Composition before start the in-situ aeration

5.2.2. Site Description

Chongming MSW landfill started in operation in 2006 and closed in 2013. The landfill site is located north-west of Chongming Island at 31°27′00″ N, 121°09′30″ E and 3.5~4.5 m above sea level. In this area, the mean annual precipitation is 1003.7 mm and the mean annual temperature is 15.3 °C (“Baidu Encyclopedia -the world's largest Chinese encyclopedia”. 2015). The total landfill site has an area of 3.3 ha, where 1,250,000 tons (fresh matter) of untreated municipal and commercial waste were deposited. The depth of disposed solid waste was between 3 and 8 m and covered with a thin layer of soil material and a compost/soil mixture. The area selected for the in-situ aeration project has a surface of approximately 125 m^2 (25 m x 5 m) and was situated at the center of the Chongming landfill site (Fig. 5-2).

Fig. 5-2 Experiment site at Chongming landfill, Shanghai, China

5.2.3. Experimental Set-up

The outline of the experiment to observe and verify the in-situ aeration effect on N_2O generation in the closed landfill is shown as Fig. 5-3. The aeration system consisted of air compressor (0.8 MPa and capacity 42 L/min, BEBICON, Hitachi), gas regulator, flow meter, air hoses, and air injection pipe. The air injection pipe was installed at the experimental site and the air injection point which is the tip of the air injection pipe was located at 3.8 m depth from landfill surface. First, cover soil was removed from the experimental site and second the landfilled solid waste was dug out and the aeration pipe and three sets of gas collection pipes along with temperature probes were installed. Three sets of gas collection pipes were installed at three different distances of 1.0, 2.0 and 3.0 m away from the point where the aeration pipe was installed; named points A, B and C, respectively. After installing all pipes, landfilled waste and the covered soil were replaced. Each set of gas collection pipes consisted of three pipes different in length, and their tips were set at three different depths of 0.0, 0.75 and 1.5 m from the bottom of the cover soil, respectively. Names of monitoring points different in depth are called as the depth from cover soil bottom in this study. The cover soil thickness was 2.3 m

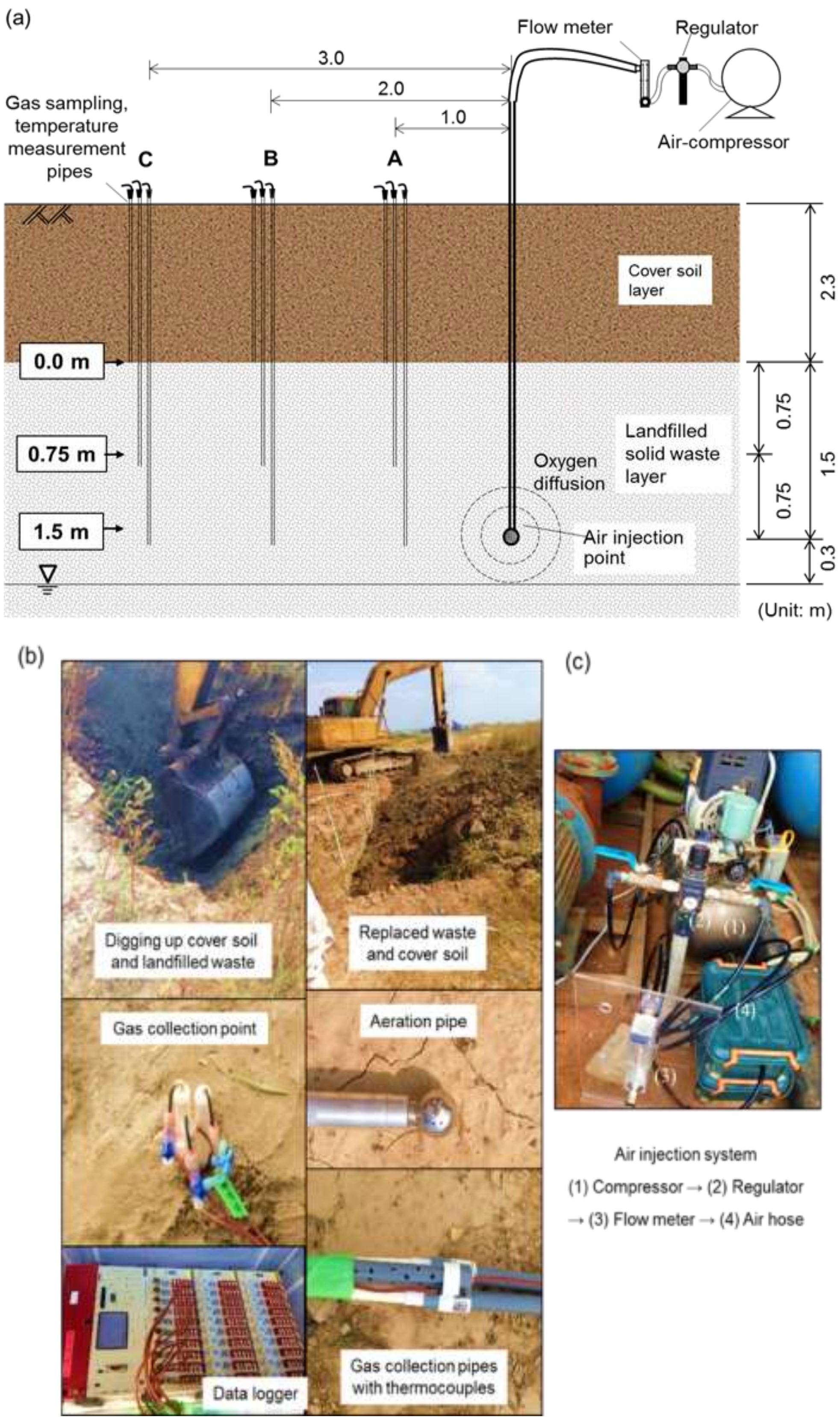


Fig. 5-3 (a) Outline of the in-situ aeration experiment (b) Measurement method and parts (c) Aeration system from the surface. In each gas monitoring pipe, concentrations of O_2, CO_2, and CH_4 were monitored in every week using a portable landfill gas analyzer (GA 5000). Temperature was measured at each monitoring depth by thermocouples connected to the portable dada logger (TDS-150 and FSW-10, Tokyo Sokki, Kenkyujo Co., Ltd., Japan) at 15 min intervals. During monitoring of landfill gas (LFG), water came out from 1.5 m monitoring depth as the water table was very close to the monitoring depth of 1.5 m. Therefore gas samples could be collected and monitored only from 0.0 and 0.75 m depths. Air flow rate was controlled 1.0 l/min until 24 day and from 24 day the air flow rate was maintained at 5.0 l/min. The in-situ aeration experiment lasted for 107 day.

5.2.4. Analytical Methods

Initial solid waste characteristics were measured by Japanese Leaching Test (JLT-46, a method to test the conformity of a material to soil environmental standard and also the environmental safety of a material that a solid waste is recycled in Japan) for analyzing pH, Electric conductivity (EC), total organic carbon (TOC), ammonium-N (NH_4^+-N), nitrite –N (NO_2^- -N), and nitrate-N (NO_X^- -N). pH and EC measured by Horiba pH and EC meters. Loss on ignition (LOI) was measured to estimate the organic carbon content in the solid phase using a muffle furnace to heat the dried sample (at 105 °C for 24 h) to 550 °C for 4 hours. The total organic carbon and total N contents of the sampled leachate were measured using a TOC analyzer with Total Nitrogen Measuring Unit (TNM-1) (Shimadzu, Japan). Ion chromatography (DX-120, Dionex, Japan) was carried out to detect the concentrations of NH_4^+-N, NO_2^--N, and NO_3^--N ions in the collected leachate. Composition of O_2, CO_2, and CH_4 were measured using a portable gas analyzer (Geotech GA-5000, UK). The N_2O concentrations in the gas samples were analyzed using a gas chromatograph (GC-Agilent 7820, USA) equipped with an electron capture detector (ECD). The GC was equipped with a pre-column (Porapak Q80/100 mesh, 1m x 2 mm) and a main column (Porapak Q80/100 mesh, 3 m x2 mm). A 1-ml sample loop and a 10-port valve were used to inject the gas samples. The temperatures of the columns and the ECD were 100 and 300 °C, respectively. The carrier gas used was a mixture of 91 % argon and 9 % CH_4. N_2O was quantified using the standard curves generated from certified standard gases (National Institute of Metrology, PR China).

5.3. Results and discussion

5.3.1. Preliminary Landfill Characterization

Table 5-1 shows the physico-chemical characteristics of collected solid waste before commencing aeration in the experimental site. At 24 d, a leachate sample was collected from 2 m away from aeration point at 1.5 m depth and the chemical characteristics of the leachate were analyzed, shown in Table 5-2. It is thought that the leachate percolation driven by the gravity tends to higher TOC, NH_4^+-N in landfill cells. In addition, under aerobic conditions the positive redox potential affects the mobility of organic compounds and other reactions are also affected such as, solubility and sorption properties of organic contaminants (Rich, Gronow, and Voulvoulis 2008).The initial increase was due to increased mobilization of organic and inorganic nitrogen compounds as well as release of already NH_4–N and T-N containing leachate in the pore spaces of waste. Formation of NO_x^- -N indicated that N conversion had also commenced in 2 m away from aeration point.

Table 5-1 Solid waste characteristics before started the experiment

Moisture content (%)	LOI (%)	pH (-)	TOC (mg/l)	NH_4^+-N (mg/l)	NO_x^- -N (mg/l)
47.6	31	4.91	1294.5	30391.3	0.0

Table 5-2 Leachate characteristics after 24 days of the in-situ aeration

pH (-)	EC (mS/cm)	TOC (mg/l)	NH_4^+-N (mg/l)	NO_x^- -N (mg/l)
5.1	43.9	1634.8	34289.4	10.3

5.3.2. Effects of Aeration on Temperature and the Landfill Gas Composition

Generally, landfill aeration leads to temperature in a range of 35-50°C due to intensive aerobic conversion processes (Heyer et al. 2005). Fig. 5-4 show the temperature development in the in-situ aeration landfill site with LFG composition changes with time.

Before starting the air supply, the temperature was about 24 °C; whereas after aeration started at a rate of 1 l/min, temperature had no significant change although there was a slight increase at 1.5 m depth near to the aeration point. The air flow rate was increase at 5 l/min from 24 d and the temperature profiles rapidly started to increase, especially for points A and B, and there was a gradually increasing trend during the entire aeration period as shown in Fig. 5-4 (a). In-situ aeration of waste leads to a significant change in gas composition and production when compared with anaerobic condition. Presence of oxygen accelerates the decomposition of organic matter in waste; and distribution of carbon load during aerobic decomposition is concentrated on the creation of gaseous products. Fig. 5-4 (b) represents the O_2, CO_2, CH_4 and N_2O concentrations changes with depth and time during the in-situ aeration of the experimental landfill site. Before commencing aeration (i.e. when no O_2 was available), CH_4 contents were 60 -71 % (v/v) and CO_2 contents around 35 % (v/v) at all gas wells. It was

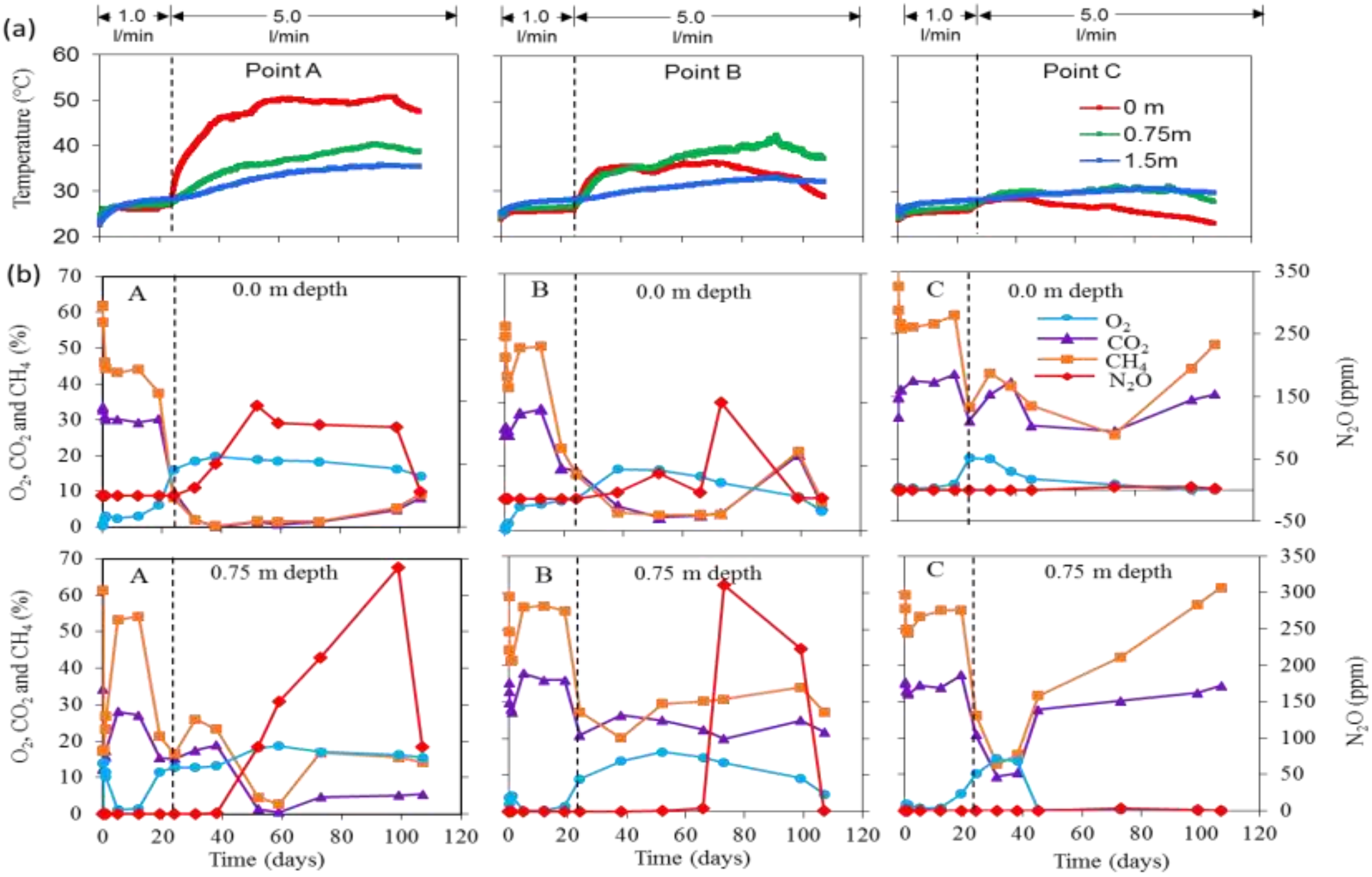


Fig. 5-4 (a) Temperature development (b) LFG compositions with time by in-situ aeration observed that during aeration rate at 1 l/min, CO_2 and CH_4 were very high due to the low O_2 concentration, although a slight decrease occurred immediately after starting the aeration. This result suggests that the aeration rate was not sufficient to enhance the microbial activity and the high organic carbon and ammonium concentration, probably made nitrification and dentrification very weak in this stage. When aeration rate was increase to 5 l/min there was a dramatic decrease in CO_2 and CH_4 were throughout the aeration periods at points A and B, with O_2 concentration within the range of 10-20 % (v/v). After starting aeration, CH_4 concentration was reduced by about 85 % within 3 months, in accordance with the study of a pilot cell in Live Oak Landfill (USA) by Read, Hudgins, and Phillips (2001). It was also noticed that the CH_4 concentration reduced from 60 to 15 % in point A at 0.75 m depth; and 60 to 9 % at 0 m depth, where CO_2 to CH_4 ratio rose from 0.55 to 0.88 at 0 m depth. Aeration enhances the carbon conversion mainly as CO_2, which complies this study at deeper depth (0.75 m) at points A and B. Heyer et al., (2005a) found that in a aerated landfill, CH_4 concentration rapidly decreased (3-15 %) and CO_2 within the range of 10-20 %, which is consistent with results of the present study. In light of Figure 5-4 (b) at points A and B, rapid decrease in CH_4 and increase CO_2 levels with O_2 consumption evidently shows the influence of the aeration measures. The rapid decrease of methane occurred by the in-situ aeration which inhibits the activity of methanogen and accelerates the activity of methane-oxidizing bacteria simultaneously through increase of oxygen content. At point C, CO_2 and CH_4 contents decreased during 24-40 d for O_2 concentration in the range of 5-15 % (v/v), while the concentration of both CO_2 and CH_4 found higher at the end approximately 35 and 60 % (v/v), respectively, under relatively lower percentages of oxygen (below 5 %).

In contrast, during the high aeration rate (5 l/min), observed N_2O generation was ranged from undetectable to 338 and 310 ppm at points A and B, respectively; whereas at point C, N_2O generation was very low (up to 5 ppm) or below detection limit as O_2 concentration was observed below 5 % (v/v). It can be inferred that the existing and adequate oxygen caused by in-situ aeration is the dominant factor on N_2O production. However, in-situ aeration provided inadequate O_2 to inhibit nitrification and promote the decomposition of immediate product, NH_2OH in nitrification and N_2O production (Davidson 1992).

After increasing the air flow rate, N_2O production began to increase; this phenomenon can be explained by the reaction rates of ammonia oxidation and nitrite oxidation studied by Tadahiro et al. (2014) on wastewater treatment. It was evaluated that due to high air flow rate, higher N_2O produced because of relatively high ammonia oxidation rate in the beginning which caused NO_2-N accumulation. On the other hand, in low air flow rate both NO_2-N and N_2O concentrations were low, probably because nitrite oxidation was relatively high. Accumulation of NO_2^- -N had good correlation with N_2O production observed He et al. (2001) in aerated composting of food waste during nitrification. Therefore, it is hypothesized that with high aeration rate, accumulation of nitrite is one of the potential factors for N_2O production. Moreover, observed NO_x^--N (Table 5-2) in this study possibly owing to the beginning of nitrification even at low air flow rate. It is assumed that with sufficient O_2 concentration due to high aeration rate prompted NO_2^- -N accumulation that also observed in previous study (Ritzkowski, Heyer, and Stegmann; 2006) in aerated landfill and laboratory-scale bioreactor study by (Nag, Shimaoka, and Komiya 2015).

In Fig. 5-4 (b), at high aeration rate, a very sharp increase of N_2O was found at point A (at both depths) and it was often observed when O_2 concentration was above 15 % (v/v). Increasing oxygen concentration from 5-20 % (v/v) led to increase N_2O production at points A and B reveal that nitrification process is the dominant pathway of N_2O formation. It has been suggested that N_2O concentration increase with decreasing O_2 concentration during nitrification; and the results of the present study from 73 d were in accordance with this suggestion, when O_2 concentration started to decrease. Robinson, Barr, and Last (1992) mentioned that the accumulation of nitrite and nitrate occur when nitrification become stronger; and denitrification become wreaker as a result of decreased organic carbon. Hence N_2O production did not accord with increase nitrite and nitrate concentration as a key influencing factor studied by Sun et al. (2013). However, at point C, N_2O generation was detected at very low level when O_2 concentration was below 5 % (v/v). It is noticeable that at point C, though O_2 concentration was ranged between 0 to 15 % at the initial period of high aeration from day 24 to 40 but N_2O generation was undetectable. This might happen for the incomplete nitrification or denitrification. Weakening of both nitrification and denitrification is thought to change the trend of N_2O production (Sun et al. 2013). These observations suggest that N_2O reduction is possible by controlling the rates of each nitrification by changing air flow rate. High production of N_2O inside the aerated landfill occurred for O_2 concentrations above 15 % v/v in this study might developed a strong nitrification process, which stimulated N_2O formation as a by-product. He et al. (2011) mentioned that prolonged aerobic conditions can result in strong production of N_2O.

5.3.3. Correlative Analysis among Landfill Gases

In order to find out the in-situ aeration effects on GHGs (CO_2, CH_4 and N_2O) and volume ratio of CO_2 and CH_4, the Pearson correlative analysis based on the collected sampling data of point A and B was conducted.

From Table 5-3, it can be found that both CH_4 and CO_2 had a significant ($P < 0.01$) negative correlation with O_2 concentration. Usually, abundant O_2 content can inhibit CH_4 production and improve CO_2 production through methane oxidation. Oxygen distribution in waste layer is a multi-stage process influenced by concentration diffusion among the pore space in waste layer, aerobic metabolism of organic carbon and nitrification process. But the N_2O formation occur during nitrification and denitrification processes. The negative but not close correlation between CH_4 and N_2O indicated that CH_4 and N_2O are formed through different biological processes. On the other hand, O_2 and N_2O had a significant positive correlation at point A (0 m depth, $P < 0.01$ and 0.75 m depth $P < 0.05$). At point B, positive correlation observed but not very close. This phenomenon elicit that higher O_2 content induce formation of higher N_2O. Therefore, during the inhibition of CH_4 production by in-situ aeration, acceleration of N2O production is not inevitable. Optimization of O_2 distribution is a key of attenuation N_2O and CH_4 production synchronously.

Table 5-3 Correlation among landfill gases: Pearson's correlation coefficients (R-value) and *P*-value

		R-value				*P*- value			
Sampling point	LFG	CO_2	CH_4	N_2O	CO_2/CH_4	CO_2	CH_4	N_2O	CO_2/CH_4
A – 0.0 m	O_2	-0.99	-0.99	0.707	0.394	< 0.01	< 0.01	< 0.01	*NS*
	CH_4	0.978	-	-0.683	-	< 0.01	-	< 0.01	-
	CO_2	-	-	-0.736	-	-	-	< 0.01	-
A- 0.75 m	O_2	-0.951	-0.98	0.525	-0.278	< 0.01	< 0.01	< 0.05	*NS*
	CH_4	0.94	-	-0.427	-	< 0.01	-	*NS*	-
	CO_2	-	-	-0.644	-	-	-	< 0.01	-
B – 0.0 m	O_2	-0.8	-0.841	0.492	0.808	< 0.01	< 0.01	*NS*	< 0.01
	CH_4	0.952	-	-0.509	-	< 0.01	-	< 0.05	-
	CO_2	-	-	-0.564	-	-	-	< 0.05	-
B – 0.75 m	O_2	-0.781	-0.876	0.388	0.583	< 0.01	< 0.01	*NS*	< 0.05
	CH_4	0.902	-	-0.295	-	< 0.01	-	*NS*	-
	CO_2	-	-	-0.401	-	-	-	N*S*	-

NS: Not significant

5.3.4. Correlative Analysis of Temperature and Landfill Gas Composition

At point A and B, oxygen concentration was mostly in the range of 15 to 20 % (v/v), therefore aerobic microbial activity accelerated the organic matter decomposition, leading to a release of energy and increased temperature. Temperature at 0 and 0.75 m depth for point A reached up to 50 °C and 40°C, respectively and at point B ranged within 30-40 °C. The raised temperature in the waste body was unequivocal indication of release thermal energy as a result of intensive aerobic conversion process with reduction of CH_4 and increase the concentration of CO_2, simultaneously. Temperatures at Point C remained below 30 °C throughout the experimental period as oxygen concentration was observed in a range between 0 to 5 % (v/v) except the initial period of high aeration rate; which indicates that release of energy from accelerated decomposition of organic matter was limited by insufficient O_2 content.

It was evaluated that during the experimental period, N_2O was below detection limit at low aeration rate as temperature ranged within 25 to 30 °C. Increased aeration rate (5 l/min) stimulated N_2O production with increased temperature up to 50 °C. These results reveal that temperature had a strong influence on N_2O production with high aeration rate, as temperature increases due to energy release from aerobic decomposition of organic matter. Increasing temperatures are the major factor determining N dynamics and the moisture losses during aeration. Temperatures above 40 °C are intensifying both hydrolysis and ammonification processes (Scheffer and Schachtschabel, 2009) while at the same time inhibiting nitrification (Robinson, Olufsen, and Last 2015). Data from the in-situ aeration in this study showed a significant ($P < 0.01$) positive correlation between temperature and N_2O generation at point A and B, within the range of 30-50 °C as shown in Table 5-4.

Table 5-4 Correlation between temperature and landfill gases: Pearson's coefficients (R-value) and *P*-value

		R-value			*P*-value		
Sampling points	Temperature	CO_2	CH_4	N_2O	CO_2	CH_4	N_2O
A – 0.0 m	30-50◦C	-0.922	-0.888	0.836	< 0.01	< 0.01	< 0.01
A – 0.75 m	30-40◦C	-0.725	-0.558	0.815	< 0.01	< 0.05	< 0.01
B – 0.0 m	35-40◦C	-0.755	-0.758	0.583	< 0.01	< 0.01	< 0.05
B – 0.75 m	35-42◦C	-0.811	-0.804	0.605	< 0.01	< 0.01	< 0.05

From Table 5-4, it was evaluated that temperature has a significant ($P < 0.01$) negative correlation with CO_2 and CH_4 indicate that optimum aerobic condition has notable impact on GHGs reduction in landfill cell. At points A and B, N_2O concentrations were higher for deeper than shallower depths (0.75 and 0.0 m, respectively) (Figure 3). When temperature increased to 30 °C, N_2O generation started to increase and gradual increase of temperatures up to 40 °C, N_2O production was increased very sharply for both points A and B at both depths (0.0 and 0.75 m); while for point A at 0.0 m depth, when temperatures rose above 40 °C, N_2O production showed a decreasing trend. The generation of N_2O was showed decreasing trend when temperature started to decrease. However, at point C, N_2O concentration was much lower with the highest content of 5 ppm in temperature within the range of 25-30 °C. Oxygen concentration was observed in a range between 0 to 5 % (v/v) except the initial period of high aeration rate; which indicates that release of energy from accelerated decomposition of organic matter was limited by inadequate O_2 content. The data of N_2O concentrations showed that temperature was clearly a potential factor in N_2O formation. N_2O production with temperature in this in-situ aeration study revealed that temperature controlled the nitrification process up to 40 °C, in accordance with results of Robinson, Olufsen, and Last (2015), and N_2O was mainly generated as a by-product of nitrification during aeration in in-situ landfill site.

5.3.5. Evaluation of Greenhouse gases by In-situ Aeration

Results of greenhouse gases (GHGs) had shown the obvious reduction of CH_4 and production of N_2O observed simultaneously. Therefore, total effect of GHGs under the in-situ aeration is evaluated. At minimum aeration rate of 1 l/min, the total average GHGs concentration in CO_2 equivalent were 4.1x 10^6, 4.9 x 10^6 and 5.4 x10^6 ppm in sampling point A, B, and C, respectively, where N_2O was not detected. On the other hand, at high aeration rate of 5 l/min, total average concentrations of GHGs reduced to9.3 x 10^5, 1.9 x 10^6 and 3.3 x 10^6 ppm (CO_2 equivalent) in sampling point A, B, and C, respectively, including N_2O concentration as shown in Fig. 5-5 (2.2 x 104, 1.6 x 10^4, and 3.4 x 10^2 ppm CO_2 equivalent at point A, B, and C, respectively). It has been evaluated that net total CO_2 equivalent concentration reduction at point A, B, and C were 77, 61 and 38 %, respectively, at increased aeration rate from 1 l/min to 5 l/min. The N_2O production impelled by in-situ aeration does not offset the greenhouse effect reduction caused by CH_4 reduction. Therefore, in-situ aeration in MSW landfill is an effective way to minimize greenhouse effects.

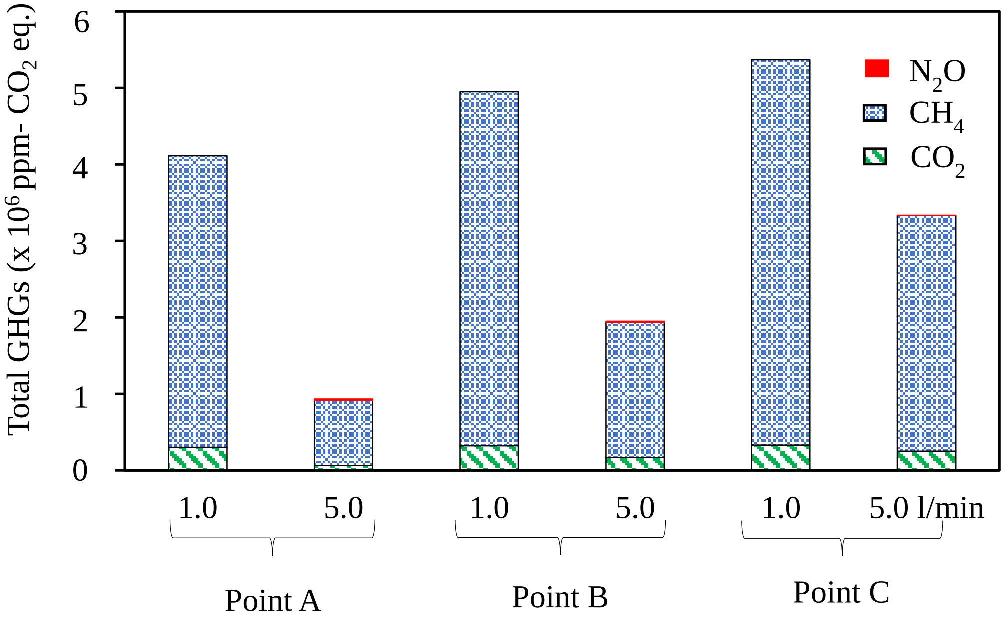


Fig. 5-5 Evaluation of greenhouse gases by in-situ aeration

5.4. Conclusions

In-situ aeration in the MSW landfill accelerates the decomposition of organic matter and therefore released energy that increased temperature and poses a positive impact on N_2O production. The N_2O production varied with temperature range of 30-50 °C and O_2 concentration within range of 5-15 % (v/v). Low O_2 content can lead to N_2O production during nitrification and high O_2 inhibit denitrification which would affect N_2O production. Therefore, optimization of O_2 concentration inside the MSW landfill is a key factor to control N_2O production. It was hypothesized that high air flow rate is a potential parameter of N_2O production because of nitrite accumulation. Nitrification is found the dominant pathway of N_2O production during in-situ aeration. Results of this study should aid in understanding the N and temperature dynamics of N_2O production. N_2O flux measurement in aerated landfill site, with consideration of cover soil materials and adjustment of temperature by a controlled addition of water, and with intermittent aeration would be beneficial in future studies. The findings provide insights concerning the production potential of N_2O in an aerated landfill that may help to minimize with appropriate control of the operational parameters and biological reactions of N turnover, although in-situ aeration helps to minimize the total greenhouse gas emissions.

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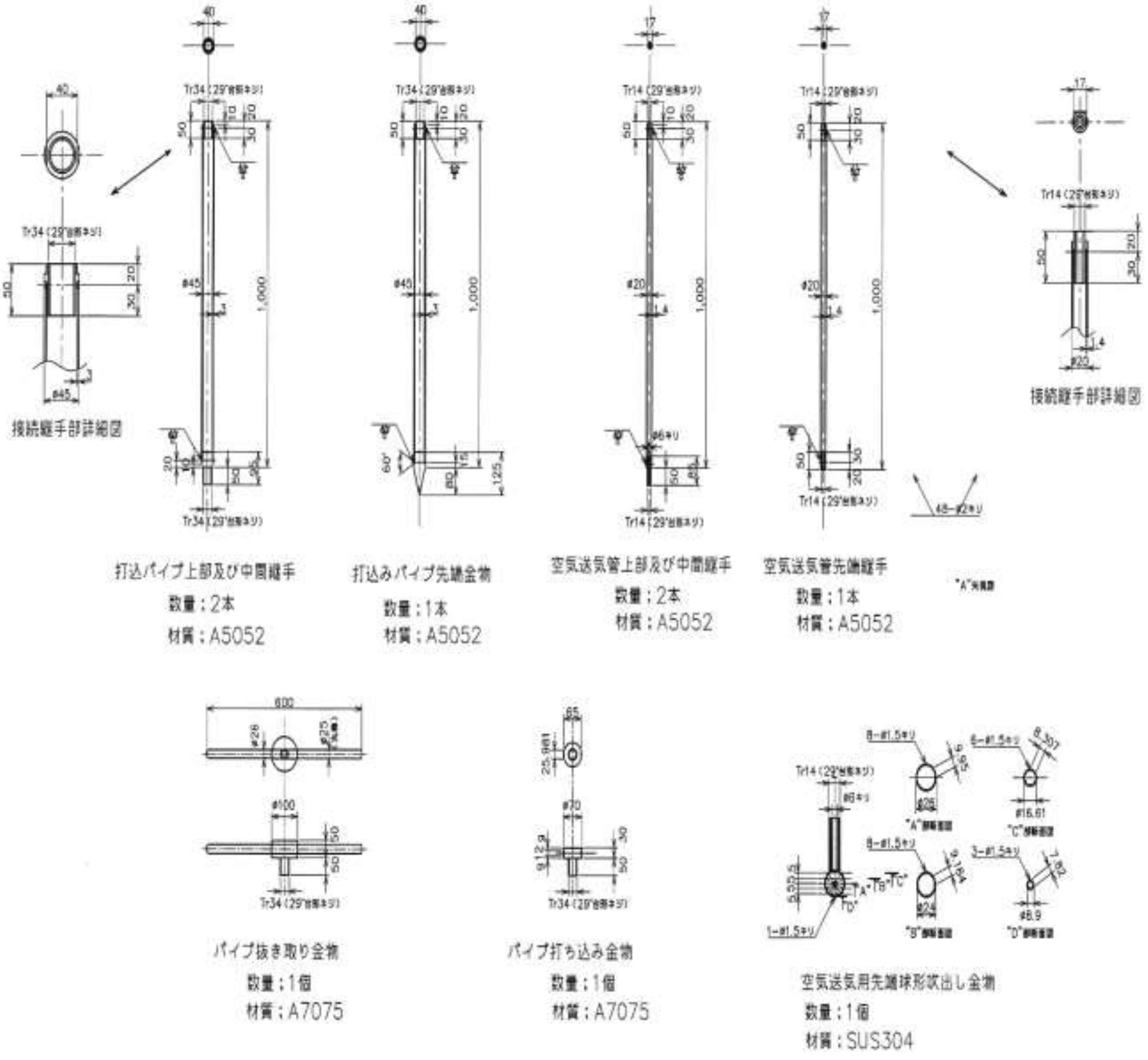
Appendix B



Gas collection pipe design

Final experimental set –up of the gas collection pipes

CHAPTER 6

Conclusions and Recommendations

6.1. Conclusions

According to the estimation of future projections the world's waste production could reach up to 27 billion tons by 2050, a third of which may be generated in Asia, with a significant percentage of that being produced in large economies such as China and India. The types of MSW produced change according to the standard of living in the city. Wastes generated in low- and middle-income cities have a large proportion of organic waste, whereas the wastes in high-income cities are more diversified with relatively larger shares of plastics and paper. The changing composition of waste in turn influences the choice of technology and waste management infrastructure.

Among the MSW disposal strategies, dumping and landfill continue to be extensively used in developing countries due to both economic and technological factors. However, pollutions from traditional sanitary anaerobic landfill may last for longer periods since anaerobic degradation is very slow process. There are still several problems need to be solved in traditional sanitary anaerobic landfill such as, the pollution of leachate on underground water and pollution of the landfill gas. In recent years, various novel landfill technologies have been created with the aim of accelerating biostabilization of landfill. Landfill aeration is considered to be an indispensable method for the controlled and sustainable conversion of conventional anaerobic landfills into a biological stabilized state associated with a significantly lowered or the near elimination of the landfill gas emission potential. Although landfill aeration is not a widely applied concept so far, it has already been successfully applied to several landfills in Europe, North America and Asia. However, adding air can be expensive, thus adding less air may be more economically attractive.

The Aerobic-Anaerobic Landfill Method (AALM), is considered a novel landfill method. In this study, AALM has been adopted by supplying intermittent aeration in column bioreactors to make it environmentally and economically viable. The impact of the operational parameters on the AALM and the transition period phenomena have been studied through column bioreactor experiment. In order to determine the operational parameters influence by intermittent aeration with different aeration rates on AALM, it is necessary to investigate the physic-chemical characteristic and the phenomenon of reducing the pollution loads. In this study, characteristics of leachate pollution and greenhouse gases emission were evaluated through the bioreactor column experiment by intermittent aerations. Additionally, the transition periods phenomenon between aerobic and anaerobic conditions has been studied for AALM.

The experiment was carried out four column bioreactors with 2 m height and 0.1 m diameter where two AALM by intermittent aeration compared with continuous and non-aeration reactors. The results reveal that intermittent aeration has significant impact on leachate and GHGs abatement compared with non-aeration reactor followed by simultaneous nitrification and denitrification processes.

Some specific findings of this column bioreactor experiments have been pointed out below:

- Keeping the other operational parameters such as ambient temperature, moisture content, and precipitation at controlled levels, intermittent aeration had a very significant impact on carbon and nitrogen contaminants removal from organic solid waste.
- Aerobic reactor A (7 days/week) had the highest removal rates of TOC, T-N and NH_4^+-N and lowest CH_4 emissions compared to aerobic-anaerobic reactor B, C and non-aerated reactor D.
- Aerobic-anaerobic reactor C 6hrs/day) exhibited better improvement of leachate quality and reduction of GHGs compared with aerobic-anaerobic reactor B (3 days/week) and

non-aerated reactor D (no aeration), which can save three-fourths of the energy cost of an aerobic landfill, making it cost-effective as well.

- Decomposition of organic carbon and transformation of nitrogen contaminants are rapid processes in aeration periods, which accelerates the degradation of organic matter as well as ammonium removal.
- Transition period phenomena demonstrated that in-situ nitrification and denitrification are feasible in decomposed aerated solid waste environments at various gas-phase oxygen concentrations. Additionally, the results indicate the potential for simultaneous nitrification and denitrification (even under low biodegradable C: N conditions) in field-scale landfills will be significant due to the presence of both aerobic and anoxic areas.
- This concept of AALM method by intermittent aeration is a promising method for reducing leachate pollutants and can abate the potential of greenhouse gases by accelerating the decomposition of waste under aerobic condition. Intermittent aeration can reduce the later phases of landfill aftercare as well and simultaneously reduce the energy cost.

Nitrous oxide is a trace gas for contributing the global warming and stratospheric ozone layer destruction, which is also emitting from landfill site by in situ aeration during nitrogen turnover. Therefore, the nitrous oxide production potential through batch experiment has been studied as well from organic solid waste under nitrification condition. The experiment was investigated at three different temperatures with three different oxygen concentrations. It was investigated that during nitrification optimum temperature and optimum oxygen concentration are important factors for minimizing nitrous oxide production. The results demonstrated that temperature about 20°C and 40°C generated high nitrous oxide with high oxygen concentration (20%). It can be conclude that for proper nitrification condition, temperature range might be 20 to 30°C could reduce the nitrous oxide production with moderate oxygen concentration.

To verify the potential of nitrous oxide production factors during aeration, a field study of in-situ aeration have been conducted in a closed landfill site. The results of in-situ aeration on landfill waste showed the aeration measures as the greenhouse gases reduction, increased temperature of the waste body and so on. The correlations between nitrous oxide production with landfill gases and temperatures have been identified through that experiment. Nitrous oxide production is significantly correlates with temperature range between 35-50°C and with high oxygen concentrations. It was found that during nitrification, low oxygen content led nitrous oxide increase and high oxygen inhibit denitrification might enhanced nitrous oxide production. Therefore, optimization of O_2 concentration inside the MSW landfill is a key factor to control N_2O production. It was hypothesized that high air flow rate is a potential parameter of N_2O production because of nitrite accumulation. Nitrification is found the dominant pathway of N_2O production during in-situ aeration. Results of this study should aid in understanding the N and temperature dynamics of N_2O production. N_2O flux measurement in aerated landfill site, with consideration of cover soil materials and adjustment of temperature by a controlled addition of water, and with intermittent aeration would be beneficial in future studies. The findings provide insights concerning the production potential of N_2O in an aerated landfill that may help to minimize with appropriate control of the operational parameters and biological reactions of N turnover, although in-situ aeration helps to minimize the total greenhouse gas emissions.

6.2. Recommendations

The research carried out in this study performed to govern an optimum aeration strategy for the AALM. Although the outline of the intermittent aeration strategy was proposed on the basis of this research, more in-depth and few studies are required before the field implementation.

Therefore, the following recommendations are suggested for the future studies.

1) Temperature rise impelled by air injection in landfill is a very common phenomenon. However, during the column experiment, temperature insides mainly affect by the controlled atmosphere temperature. The maximum temperature found during the column experiment was about 38^{o}C. Therefore, the optimum aeration proposed in Chapter 2 is suitable for the landfill under moderate temperature conditions. Thus, further studies on the AALM should be conducted in larger scale landfill site which allows the temperature to arise.

2) Continuous monitoring of landfill gas composition, especially oxygen concentration; is very important for evaluate the hybrid condition (aerobic-anaerobic).

3) More parallel studies are necessary to investigate the N_2O production in different aeration rates.

4) During in-situ aeration, N_2O production in source and emission in the atmosphere both are important to estimate the greenhouse gas effect.