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Doctoral Thesis

**Co-additive (Flue gas
desulphurization gypsum and
biochar) supplementation for the
reduction of odor (ammonia
volatilization) from the livestock
facilities**

**병합재(Flue gas desulphurization gypsum과
biochar) 바이오필터가 축산시설 악취(암모니아
휘산) 저감에 미치는 영향**

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**Graduate School of Agricultural Biotechnology
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Co-additive (Flue gas desulphurization gypsum and biochar) supplementation for the reduction of odor (ammonia volatilization) from the livestock facilities

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Abstract

Co-additive (Flue gas desulphurization gypsum and biochar) supplementation for the reduction of odor (ammonia volatilization) from the livestock facilities

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Increased livestock population has led to the escalation of livestock waste generation. The waste from livestock industrial activities (manure and slaughter waste) are the major source of negative environmental impacts (i.e. the release of greenhouse gases or excess nutrients, salt accumulation, growth of pathogenic microorganisms, odor, and so on), which are capable of influencing both humans and animals. Indeed, it has been suggested that odor and gas emissions from livestock farming operations have become an important social problem due to their negative impacts on health and quality life of human being. Livestock manure generated from swine houses is a particularly serious problem compared to other animal species. Moreover, the emission of odors and volatile organic compounds (VOCs) from swine livestock facilities are

representative air pollution problems and are become a crucial public health concern. Accordingly, studies regarding the effort to reduce the pollution affected by the livestock activities were carried out.

In the first phase of the studies, the effectiveness of co-additives for improving livestock waste composting (reduction of air pollution and conservation of nutrients) was investigated. Biochar and Flue gas desulphurization gypsum (FGD gypsum) were used to supplement the composting of a mixture of slaughter waste, swine slurry, and sawdust. Different composition of additives (0% or 5% each, 10% biochar or FGD gypsum) was tested in triplicate on the laboratory scale. In addition, the effects of two different aeration schemes (continuous and intermittent) were also investigated. Ammonia volatilization, physicochemical characteristics, and compost maturity indices were investigated. The results indicated that the use of the co-additive (Biochar and FGD gypsum) during composting of livestock waste led to a reduction of ammonia volatilization by 26–59% and to a 6.7–7.9-fold increase of nitrate accumulation. The total ammonia volatilization of intermittent aeration treatment was lower than that of continuous aeration using co-additives treatment. It was concluded that co-additives (biochar and FGD gypsum) might be utilized in the livestock waste composting to reduce ammonia volatilization and improve nutrient conservation.

Subsequently, the performance of two different types of the integrated ventilation-biofilter system in an experimental pig house was investigated. The wall ventilation system integrated into vertical biofilter (W-VBF), and the pit ventilation system integrated into horizontal biofilter (P-HBF) were tested. The

experiment was conducted for 64 days. Water recycling was used in the biofilter system. Microclimatic variables, aerial pollutant (particulate matter and odorant) and wastewater (slurry and biofilter recycled water) characteristic were observed during the experiment. Results showed that ammonia concentration inside the W-VBF was about 41% higher than that of the P-HBF. Noise from the W-VBF was about 12.1% more than that of P-HBF. Particulate matter and total suspended particle inside the W-VBF house was about 62.2% and 69.9% respectively higher than that of P-HBF. It was concluded that the P-HBF emitted lower air pollution rather than W-VBF. In addition, it could maintain the optimum temperature condition for swine house during cold condition. These results suggested that an integrated ventilation-biofilter system for pig house could be used as an option to minimize the air pollution produced by the swine farming activity as well as to maintain optimum microclimate condition for the pig.

The third phase research investigated the odor emission and its treatment performed by the dual biofilter system supplemented by the co-additive (FGD gypsum and biochar) in the closed livestock confinement. The performance of dual biofilter system was investigated under experimental farm model. The eight weeks (56 days) of tested was performed on the experimental scale pig house. The dual (vertical and horizontal) biofilter system attached to the mechanical wall ventilation system was tested. Water recycling system was equipped in the biofilter system. Microclimatic variables, odor pollutant (ammonia and volatile organic acid), physicochemical of the biofilter pack and wastewater (biofilter recycled water) characteristics were observed during the

experiment. The ammonia removal efficiency of the dual biofilter system supplemented by the co-additive (5% FGD gypsum and 5% biochar) was about 65.28% to 98%. The co-additive supplementation in the biofilter media might increase the ammonia removal efficiency about 57.72% and potent to inhibit the emission of volatile organic odor from the pig house with the average removal efficiency of about 21.36%. The nitrification process, as well as microbial activities in the biofilter media, might be improved by co-additive supplementation. In addition, since it contains higher nitrite nitrate concentration and the macro micro-nutrient (total P, K, Ca, Mg, S, and Fe) availability, the biofilter media might also further be considered as a solid bio-fertilizer product source for the land application. Results indicated that dual biofilter system supplemented by the FGD gypsum and biochar emitted lower air pollution rather than the biofilter without co-additive supplementation. It was concluded that integration of dual biofilter system in the closed livestock house might be used as the option to minimize the air pollution produced by the livestock farming activity.

According to the overall resulted in these studies, it was concluded the co-additive supplementation could be an alternative option to reduce the odor emission from the livestock facilities. An amount of 10% co-additive (FGD gypsum 5% and biochar 5%) could be used as co-additives in livestock waste composting as well as in the biofilter packing material for mitigating the odor emission from the livestock facilities. Co-additive (FGD gypsum 5% and biochar 5%) in the livestock composting reduced ammonia volatilization during the composting process, increased the nitrate accumulation in compost product,

didn't give an adverse effect to composting process, and environmentally save. Co-additive (FGD gypsum 5% and biochar 5%) in the biofilter material increased the ammonia removal efficiency, inhibit the volatile organic compounds emission, increased the macro and micronutrient of used biofilter material for the land application.

Keywords: livestock waste, composting additives combination, ammonia, volatilization, air pollution, odor, biofilter, swine house, volatile organic acid

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List of Abbreviations

AA	: Acetic Acid
BA	: Butyric Acid
BD	: Bulk Density
C/N	: Carbon and Nitrogen Ratio
DO	: Dissolved oxygen
EC	: Electrical Conductivity
FGD	: Flue Gas Desulphurization
IBA	: Isobutyric Acid
IVA	: Isovaleric Acid
ORP	: Oxidation Reduction Potential
PA	: Propionic Acid
PD	: Particle Density
P-HBF	: Pit Ventilation Horizontal Biofilter
PM	: Particulate Matter
RE	: Removal Efficiency
TAN	: Total Ammonia Nitrogen
TDS	: Total Dissolved Solid
TKN	: Total Kjeldahl Nitrogen
TOC	: Total Organic Carbon
TS	: Total Solid
TSP	: Total Suspended Particle
VA	: Valeric Acid
VFA	: Volatile Fatty Acid
VOC	: Volatile Organic Compounds
VS	: Volatile Solid
V/m	: Volume per mass

W-VBF: Wall Ventilation Vertical Biofilter

w/v : weight per volume

w/w : weight per weight

Units and Marks

°C	: Celsius degree
μS/cm	: Micro Siemens/centimeter
dB	: Decibels
g	: Gram
g/cm ³	: Gram per cubic centimeter
g/L	: Gram per Liter
kg	: Kilogram
L	: Liter
L/min	: Liter per minute
CFU/mL	: Colony Forming Unit per milliliter
m	: Meter
m ³ /day	: Cubic meter per day
m ³ /h	: Cubic meter per hour
m ³ /pig head	: Cubic meter per pig head
m ³ /pig·h	: Cubic meter per pig per hour
mg CO ₂ /g TS	: Milligram carbon dioxide per gram dry solid
mg	: Milligram
mg/kg	: Milligram per kilogram
mg/L	: Milligram per Liter
Min	: Minute
mL	: Milliliter
mm	: Millimeter
mol/L	: Moles per liter
mS/cm	: Milli Siemens per centimeter
g N/m ³ ·h	: Gram nitrogen per cubic meter per hour
ppb	: parts per billion
ppm	: parts per million

Rpm	: Revolutions per minute
$\mu\text{g}/\text{m}^3$: Microgram per cubic meter
μm	: Micrometer

CHAPTER 1 . GENERAL

INTRODUCTION

1.1. Background of research

The livestock industry in many Asian countries such as South Korea and China has rapidly grown during the last two decades (Guo et al., 2016a). Korean livestock industry reaches 17 trillion and 471.4 billion won, accounting for 40.2% of the total agro-forestry production (Kim et al., 2012). The high growth population of the livestock has led to the high livestock waste production (Figure 1). The waste from livestock industrial activities (manure and slaughter waste) are the major source of negative environmental impacts (the release of greenhouse gases or excess nutrients, salt accumulation, growth of pathogenic microorganisms, odor, and so on), which are capable of influencing both humans and animals (Franke-whittle and Insam, 2013; ten Hoeve et al., 2014).

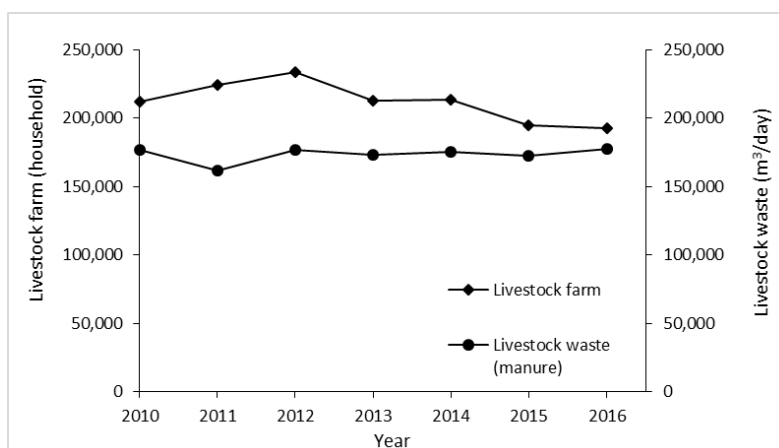


Figure 1.1. Number of the livestock farm and the livestock waste in South Korea 2010 – 2016 (Ministry of Environment - Korea, 2018)

In South Korea, for about 70% of livestock manure is recycled by the composting process (Figure 2). Livestock manure generated from swine houses is a particularly serious problem compared to other animal species (Lee, 2017). Moreover, the emission of odors and volatile organic compounds (VOCs) from swine livestock facilities are representative air pollution problems and are become a crucial public health concern (Yang et al., 2010).

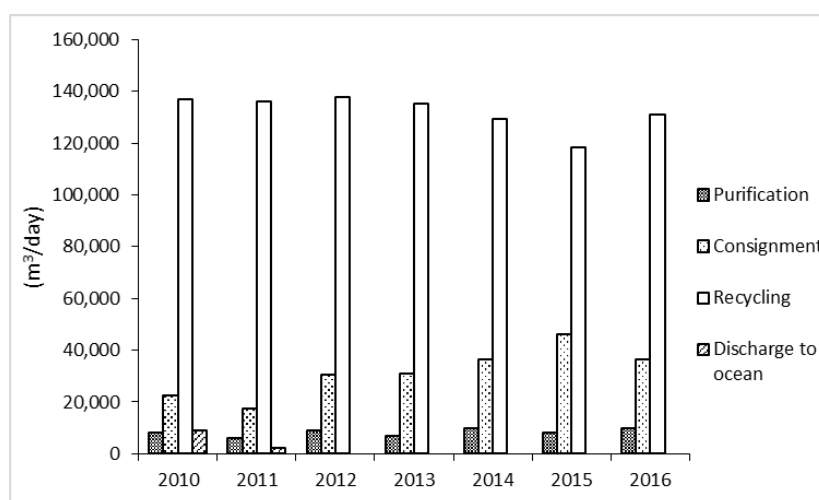


Figure 1.2. Livestock manure treatment status in South Korea 2011 – 2016 (Ministry of Environment - Korea, 2018)

The complaints due to livestock industries related odors increased along with the growth in the number of large livestock operations especially swine operations as well as increasing insensitivity and demand for a clean and pleasant environment of the general public (Rappert and Müller, 2005). About 30% of odor from livestock building and about 20% from manure storage facilities contributed to the emission from livestock production (Mielcarek and Rzeźnik, 2015). Hence the strategies to mitigate the odor problem arose can be focused on those two sectors.

1.1.1. Odor (Ammonia and Volatile Organic Acid) in Swine livestock facilities

The emission of offensive odorants in the livestock housing facilities has increased along with the development of livestock scale and density of operations (Casey and Gay, 2006; Trabue et al., 2011). Livestock facilities such as pig barns, cattle feedlots, and poultry buildings are known as a common source of unpleasant odors (Jo et al., 2015). The decomposition of excessive swine manure and feed commonly caused the emission of volatile odorants on the swine livestock farm. The combined effects of several variables such as feed type, animal population number, and confinement structure regulated the odorant species composition. Waste slurries treatment facilities can also be a major emission source on a pig farm (Jeong et al., 2009; Song et al., 2013; Jo et al., 2015). In general, feed and body odors are not offensive, the considered offensive odor generated from manure and its decomposition during handling, collection, storage, and spreading (Lau et al., 2003; Rappert and Müller, 2005).

Malodorous compounds from livestock buildings and manure storage are mostly byproducts of incomplete anaerobic decomposition of organic matters, especially proteins and, carbohydrates (Rappert and Müller, 2005). The offensive odorous compounds can be divided into four different chemical classes: (1) volatile fatty acids (VFAs, i.e., branched and straight chain VFAs), (2) aromatic compounds (i.e., indoles and phenols), (3) nitrogen-containing compounds (i.e., ammonia and volatile amines), and (4) sulfur-containing compounds (i.e., hydrogen sulfide and mercaptans) (Zhu, 2000; Whitehead and Cotta, 2004; Rappert and Müller, 2005). Malodor generation from stored

manure is considered as caused by the activities of many bacterial species which involves a complex biological process. The possible ammonia volatilization in livestock operation is illustrated in Figure 3.

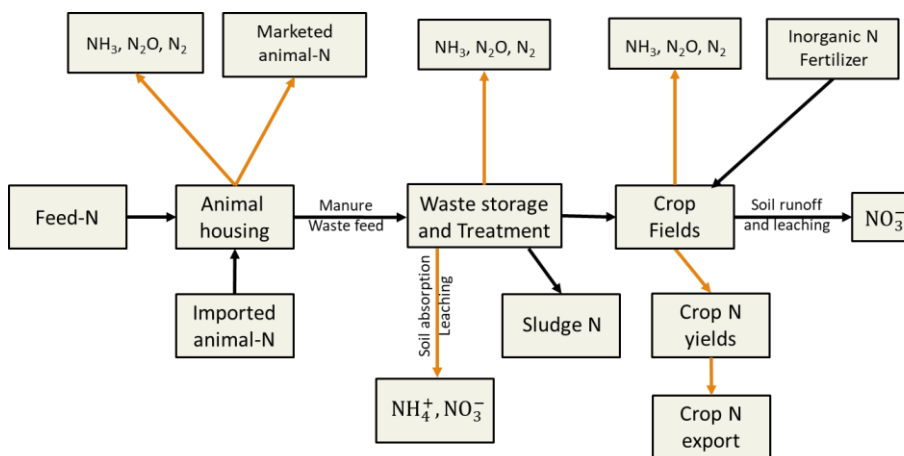
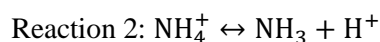
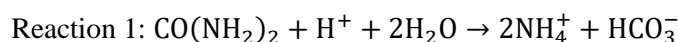


Figure 1.3. Possible ammonia odor volatilization during nitrogen flow in animal feeding operation (Arogo et al., 2003)

The volatile solids to total solids ratio (VS/TS) and crude protein (CP) value of pig manure indicated the pig manure had a high potential for gas and odor emissions. The NH₃, H₂S, CO₂, and VOC concentrations were measured in the ranges of 1.0-13.3 ppm, 0.1-5.7 ppm, 1600-3000 ppm, and 0.0-1.83 ppm, respectively (Kafle and Chen, 2014). Reduced sulfur compounds, ammonia, volatile fatty acids, phenols, indoles, and amines have been generally identified as the dominant odorants on the pig farm (Cho et al., 2013; Parker et al., 2013). However, ammonia and hydrogen sulfide are often identified as the predominant components of all odorous compounds regarding their enormity of emission concentrations (Yang, 2013; Jo et al., 2015).

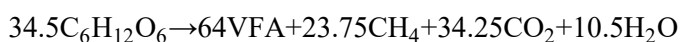
The major source of ammonia in the livestock and poultry production is the biological conversion of urea and uric acid to NH₄⁺ or NH₃ (Arogo et al.,

2001; Yang, 2013). The volatilization of ammonia gas occurs when ammonium N contained in livestock manure is converted by the following reaction (Arogo et al., 2001; Lee, 2017). In the presence of water and urease, $\text{CO}(\text{NH}_2)_2$ (urea) is converted to ammonium N via hydrolysis.

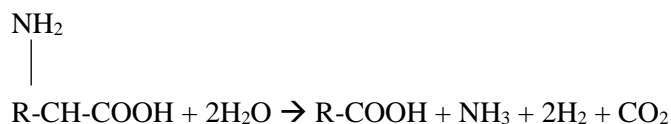


The volatile organic acids with carbon numbers from 2 to 9 showed the highest potential for odor manure. VFAs with long carbon chain (C4-C9, such as butyric, valeric, caproic, and caprylic acids) and branching (such as iso-butyric, iso-valeric, iso-caproic, and iso-caprylic acids) have more offensive odor than those with short carbon chains (such as formic, acetic, and propionic acids) (Rappert and Müller, 2005). VFAs are normally produced as metabolic intermediates or end products from a range of different bacteria. The major contributors to the odorous volatile fatty acids are mostly *Eubacterium* and *Clostridium* (Zhu, 2000).

Sugars are converted into pyruvate via glycolysis pathway in carbohydrate catabolism. Under anaerobic conditions, pyruvate will then go through different fermentation processes and serve as the substrate in the subsequent reactions in which various kinds of VFAs are generated. The sugar metabolism under anaerobic conditions is represented by the following equation (Rappert and Müller, 2005) :



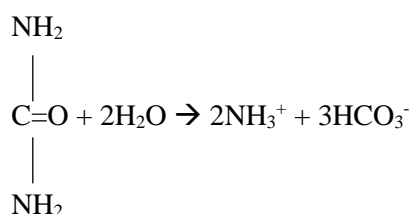
Sugar fermentation usually yields short and straight chain fatty acids (acetic, propionic, lactic, succinic, formic, and butyric acids). However, in the presence of oxygen, pyruvate is oxidized to acetyl CoA and then entered the TCA cycle and the electron transport chain to produce energy. Because no fermentation process occurs under aerobic conditions, no organic acids, especially VFAs, are produced. This fact partially explains why aeration can effectively reduce malodor production (Zhu et al., 1999). Protein and amino acids catabolism are the major sources of microbial VFAs production. The degradation of proteins not only produce straight chain carboxylic acids but also branched chain fatty acids, sulfur compounds, amines, ammonia, phenols, and indoles (Mackie et al., 1998; Alkoaik, 2009). The fate of deaminated amino acids can be diverse. In general, they will be converted to pyruvate, acetyl- CoA, or TCA cycle intermediates and are eventually oxidized in the TCA cycle to release energy (Mackie et al., 1998). The first step in amino acid usage is deamination, the removal of the amino group from an amino acid. The general mechanism for oxidative deamination is as follows (Mackie et al., 1998; Rappert and Müller, 2005):



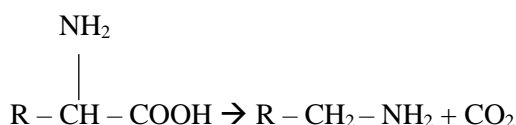
Phenolic compounds such as phenol, p-cresol, 4-ethylphenol, and phenylpropionate are produced from microbial degradation of tyrosine (Rappert and Müller, 2005). The tryptophan metabolism resulted in the production of indole and indole acetate (Ebenau-Jehle et al., 2012). These two

compounds are subsequently converted into 3-methylindole (skatole) (Sasaki-Imamura et al., 2010). Significant further metabolism of these aromatic compounds only occurs under aerobic conditions or in the presence of an inorganic electron acceptor (Smith and Macfarlane, 1997).

Ammonia can be produced from urea and nitrates during deamination of amino acids in manure by anaerobic bacteria. Urea is hydrolyzed to ammonia by enzyme urease from ureolytic bacteria as follows:



More than 50% of the nitrogen from animals could be excreted as urea by cattle, sheep, swine, and poultry. There were relatively large losses of volatilized ammonia converted from urea in urine (Van Horn et al., 1996). The urinary urea is the major source of ammonia in animal manure and wastes. Volatile amines are produced via decarboxylation of amino acids in the gastrointestinal tract and mainly during storage of fresh manure. These processes are induced at pH 5 to 6. The general mechanism for decarboxylation of amino acids is as follows (Chowdhury et al., 2014).



The NH₃-N flux from livestock bio-solids (e.g. cattle feedlot, chicken, cow, horse, and swine manure) was 98.1–99.9% of total N flux, whereas the

trimethylamine (TMA) represented 0.1–1.9% of the total N flux (Schade and Crutzen, 1995; Rosenfeld et al., 2001; Barth et al., 2010). Bacterial genera involved in the activity to produce NH₃ and amine from biosolids, including *Bacteroides*, *Bifidobacterium*, *Peptostreptococcus*, *Selenomonas*, *Streptococcus*, and the *Enterobacteria* (Zhu, 2000; Rappert and Müller, 2005). The average NH₃ emissions for livestock categories are presented in Table 1.

Table 1.1. Average NH₃ emissions for livestock categories (kg NH₃/animal-year) (percentages in brackets are portions of the total) (Schade and Crutzen, 1995)

Livestock type (source of manure)	NH ₃ emission from animal waste			Total
	Livestock house	Spread manure	Grazing animals	
Cattle	7.40 (32%)	12.24 (53%)	3.40 (15%)	23.04
Pigs	2.52 (47%)	2.84 (53%)	0.00 (0%)	5.36
Poultry	0.10 (38%)	0.15 (62%)	0.00 (0%)	0.25
Horses	3.90 (32%)	3.60 (30%)	4.70 (38%)	12.20
Sheep	0.38 (22%)	0.70 (41%)	0.62 (37%)	1.70

Sulfur-containing compounds are produced via sulfate reduction and metabolism of sulfur-containing amino acids (cysteine and methionine) by anaerobic bacteria (Arakawa et al., 2000). In the first process, bacteria produce reduced sulfur for cell biosynthesis of cysteine and methionine by transporting sulfate into the cell and activation to adenosine-50-phosphosulfate. In the dissimilatory process, sulfate is utilized as a terminal electron acceptor, and large quantities of sulfide are produced as follows (Higgins et al., 2003):



Sulfate can be supplied by dietary means or depolymerization and desulfation of endogenously produced sulfated glycoprotein such as mucins. Bacterial genera involved in sulfate reduction are *Desulfovibrio desulfuricans*, *Veillonella*, *Megasphaera*, and the Enterobacteria (Arakawa et al., 2000; Zhu, 2000). Deamination reactions of S-containing amino acids such as cysteine and methionine also give rise to sulfides and mercaptans, respectively (Rappert and Müller, 2005).

Dimethyl disulfide (DMDS) is produced by many bacteria and fungi found in wastewater (Sunesson et al., 1995; Schäfer et al., 2010). DMDS is an important odorant because it has the lowest odor threshold value (OTV, 0.1 $\mu\text{g}/\text{m}^3$) of all odorous compound emissions from biosolids (Rosenfeld et al., 2001) while dimethyl sulfide (DMS) have odor threshold values (OTV) of 2.5 $\mu\text{g}/\text{m}^3$ (Ruth, 1986). DMS and DMDS are formed when the amino acids cysteine and methionine decomposed. Methyl mercaptan and ethyl mercaptan are highly reactive and are easily converted to form disulfides (Hwang et al., 1994; Wang, 2014).

In a typical containment livestock building, ammonia concentration ranges from 0.1 ppm to 30 ppm in a typical livestock building depending on the type of building system and the ventilation rate (Arogo et al., 2003; Kim et al., 2008; Harper et al., 2010). In some extreme cases, ammonia can exceed 200 ppm (Demmers et al., 2003; Manuzon et al., 2007). The ammonia concentration in livestock buildings is usually higher in the cold weather than in the warm weather during to lower ventilation rate (Wheeler et al., 2006; Yang, 2013). Odor from swine operations has become a limiting factor for state and local

governments to permit expanding and new livestock facilities (Radon et al., 2004; Pan et al., 2007; Yang, 2013). Table 1.2 presents the volatile organic odor and ammonia odor emitted from the piggeries.

Table 1.2. Volatile organic odor and ammonia odor emitted from piggeries

Odor	Concentration
Acetic acid	2572 ppm*;
Propionic acid	760 ppm*;
	301.13 – 468.88 ppb**
Butyric acid	1106 ppm*;
	144.17 – 256.08 ppb**
i-Butyric acid	339 ppm*
Valeric acid	6.16 – 14.31 ppb**
i-Valeric acid	337 ppm*;
	8.19 – 20.23 ppb**
SCFA (Acetic acid+Propionic acid+Butyric acid)	3918 ppm*
BCFA (i-Butyric acid+i-Valeric acid)	672 ppm*
Ammonia	6.79 – 17.66 ppm*
Hydrogen sulfide	37.37 – 628.38 ppb**
Methyl mercaptan	6 – 17.52 ppb**
Dimethyl sulfide	1.86 – 6.22 ppb**
Dimethyl disulfide	0.04 – 0.26 ppb**

*(Cho et al., 2013)

** (Kim and Choi, 2013)

1.1.2. Mitigation strategies to reduce odor pollution from swine livestock facilities (from on-farm and livestock waste treatment)

About 30% of odor from livestock building and about 20% from manure storage facilities contributed to the emission from livestock production (Mielcarek and Rzeźnik, 2015). Hence the strategies to mitigate the odor problem from livestock facilities can be focused on those two sectors. As livestock production have been intensified and becoming concentrated in fewer but larger farms over the last decades, interests about the higher risk of negative impacts and the unwillingness of surrounding communities to accept livestock odors have also increased, especially odor from swine facilities (Brisson, 2014; Kafle et al., 2015). Controlling airborne pollutants from livestock buildings is a technical challenge since it is usually composed of low concentration but mixed pollutants, including ammonia, hydrogen sulfide, odorous and potentially hazardous gases, greenhouse gases, particular matters, and microbes, with a large volume (Yang, 2013). These pollutants came from multiple sources and emitted as a mixture (Chen and Hoff, 2009).

Considerable efforts have already been made over the years to minimize malodorous and toxic gases emissions, such as by modifying proteins in pig diets (Kerr et al., 2006), introducing manure additives (Zhu, 2000; McCrory and Hobbs, 2001), and employing various swine manure handling and management strategies (Ndegwa et al., 2002). However, end-of-pipe odor reduction techniques are still necessary. Despite many technologies exist for odor control, for high-volume waste gases containing relatively low concentrations readily biodegradable contaminants such as those emitted from

livestock facilities, biofiltration, biotrickling filters, and bioscrubbers have been found to be the most cost-effective treatment (Sheridan et al., 2002; Raboni and Torretta, 2016; Liu et al., 2017). Water addition to controlling moisture content is commonly incorporated in the biofilters. Microbes are usually fixed to inorganic packing materials and suspended in the water phase of the biotrickling filters. In the bioscrubbers type, air contaminants are removed in a spray tower by water absorption. Biofiltration can be used to mitigate livestock air emissions due to its simplicity and low-cost investment (Nielsen et al., 2008; Chen and Hoff, 2012; Kafle et al., 2015).

By the airflow direction, biofilters can be categorized as vertical or horizontal, and by configuration, the biofilters can be categorized as open or closed type. There are several benefits to using biofilters, (1) low construction and operation costs, as the media are locally available and not expensive (2) capable of dealing with mixed pollutants, for example, both organic and inorganic pollutants can be absorbed or adsorbed and then degraded in biofilters; and (3) reliable for a long-term operation (Sheridan et al., 2003; Jones et al., 2005; Chen et al., 2012). Biofilter media can be operated for years without replacing. Although many technologies exist, biofiltration is still the most attractive method due to its low maintenance and operating costs (Rappert and Müller, 2005).

Livestock waste is a high-strength organic waste and also comprises non-biodegradable substances and highly-concentrated ammonia, and phosphate, and thereby the troublesome pollutant. Livestock waste is a potential reservoir of bacterial, viral, prion and parasitic pathogens, capable of infecting both

animals and humans. A quick, cost-effective and safe disposal method is thus essential to reduce such environmental effect.

As an effective technology for the utilization and minimization of solid organic wastes (Onwosi et al., 2017), composting can convert livestock manure, crop straw, and industrial by-products into a sanitary and marketable organic fertilizer. About 80% of livestock waste produced in South Korea are treated through the composting system (KOSIS, 2016). However, one drawback of composting is that 24–64% of the nitrogen in organic wastes is lost via ammonia volatilization, which reduces the agronomic value of compost as a fertilizer, and leads to air pollution and potential health problems (Gillhespy et al., 2009). For instance, as these impacts both the environment and human health, the European Union has limit their disposal through landfilling and incineration requires the reduction of gaseous emissions from composting.

Methane and nitrous oxide are greenhouse gases contributing to the earth warming trends. Ammonia emissions are responsible for acid rain and eutrophication (Colón et al., 2010). During incomplete nitrification, under the effect of ammonia mono-oxygenase, NH_3 is oxidized sequentially to hydroxylamine (NH_2OH) and nitroxyl (NOH) by hydroxylamine oxidoreductase, which can be converted to N_2O by polymerization and dehydration (Canfield et al., 2011). Thus, development of the alternative method to overcome the nuisance aroused from livestock waste composting is necessary.

The utilization of chemical and biological additives to mitigate ammonia volatilization and nitrogen loss during the composting process has been

extensively studied. These additives have included wood fly ash, lime, phosphor-gypsum, polyethylene glycol, jaggery (Gabhane et al., 2012), zeolite (Nissen et al., 2000), bentonite (Li et al., 2012), superphosphate (Huang et al., 2006), and microbial inoculants (Zhang et al., 2016). Among these additives, biochar was popular to be amended in the composting mixture. Biochar is a carbon-rich material produced by the thermal decomposition of biomass, was effective for reducing volatilization of ammonia during the composting of a mixture of sewage sludge and wood chips (Malińska et al., 2014). The faster decomposition process in the bio-oxidative phase led to lower greenhouse gas emissions being observed when biochar was added at the beginning of the composting process (Vandecasteele et al., 2016; Huang and Xue, 2014). Reduction in readily available P was observed when biochar was applied during compost storage (Vandecasteele et al., 2016). Biochar addition at 3% could reduce the composting time by 20% (Sánchez-García et al., 2015). As a composting additive, biochar favored microbial activity (Jindo et al., 2012; Yoshizawa et al., 2006). Jindo et al., (2012) reported a higher diversity of fungi in biochar-amended compost. Biochar materials strongly sorb polycyclic aromatic hydrocarbons (PAHs) and are thus characterized by very high sorption coefficients (K_D) for PAHs (Mayer et al., 2016).

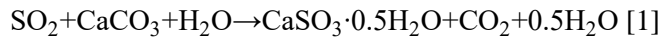
It was also reported that the N loss was mitigated by adding FGD gypsum to composting materials (Guo et al., 2016b; Tubail et al., 2008). Flue gas desulphurization gypsum (FGD gypsum) is a by-product of wet gas desulphurization in coal-fired power stations and generally has a high purity, low heavy metal content, and is rich in essential mineral nutrients for plants

such as S, Si and Ca (Guo et al., 2016a; Chen et al., 2015). The volatilization of ammonia and N loss were reduced by combining organic N-rich organic waste with FGD gypsum during the composting process (Tubail et al., 2008). The nitrogen compounds of humic substance in compost with FGD gypsum was higher than that in compost without FGD gypsum (Guo et al., 2016a). Gypsum has been known as an effective product used in the remediation of sodic soils, by greatly reducing the exchangeable Na, K, and Mg cation concentrations in the sodic soils (Presley, 2016; Ahn et al., 2007). Approximately 1–2 wt % of FGD gypsum was used to reclaim the sodic soil which resulted in no obvious heavy metal contamination (Chen et al., 2015). Guo et al. (Guo et al., 2016a) suggested adding 10% FGD gypsum to compost material to reduce nitrogen loss during composting. Information is still limited to the co-additive means of more than one additive supplemented together when applied with different aeration methods during the composting of livestock waste.

1.1.3. Flue Gas Desulfurization Gypsum

Coal-fired electric utilities use scrubbing technologies to reduce sulfur dioxide (SO₂) emissions during or after coal combustion. The term Flue Gas Desulphurization (FGD) system has traditionally referred to wet scrubbers that remove SO₂ emissions from large electric utility boilers. The FGD systems emerged in the industrial field of the coal-fired power plants and on some industrial processes in the early 1970s in United States (US) and Japan and expanded rapidly in the 1980s in Europe (Córdoba, 2014). In the FGD process, the flue gas interacts with an absorbent medium in either an absorber or a

scrubber vessel to produce high solid S slurry. The flue gasses in such system are usually passed through a suspension of alkaline absorbent in a contact chamber (Figure 3). The reaction of SO₂ with alkaline absorbent yields a solid product such as Ca-sulfite as per reaction [1] (Laperche and Bigham, 2002):



The Ca-sulfite so produced may be partially or fully oxidized to some form of Ca-sulfate as per Reaction [2]:

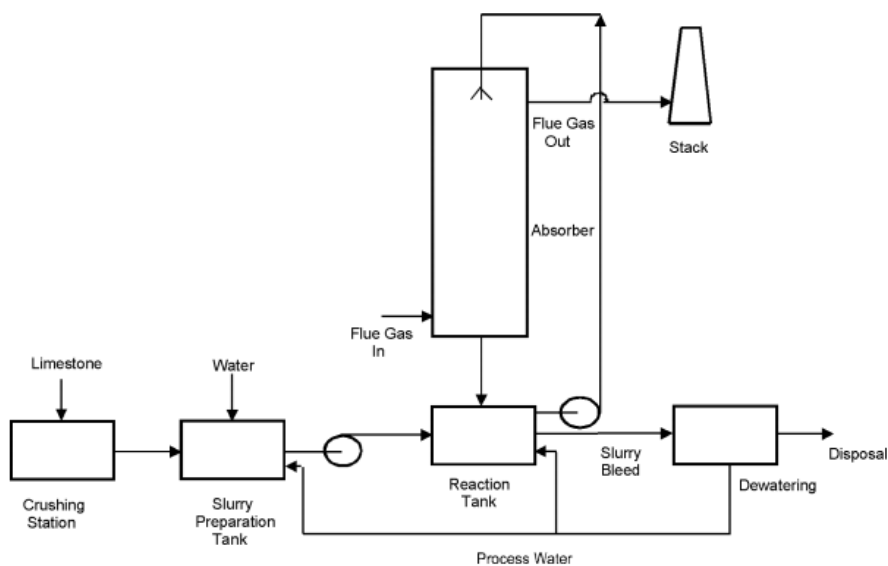
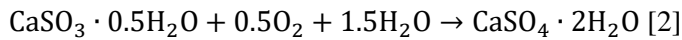


Figure 1.4. Flue gas desulphurization process (Srivastava and Jozewicz, 2011)

Wet flue gas desulfurization gas (FGD) processes often generate large amounts of calcium sulfite (CaSO₃·0.5H₂O) that must be stockpiled and impounded as a waste product (Dick et al., 2000). Calcium sulfite

($\text{CaSO}_3 \cdot 0.5\text{H}_2\text{O}$), a common by-product of coal combustion and flue gas desulphurization (FGD), spontaneously converts to gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) with exposure to air and water (Lee et al., 2008). FGD- CaSO_3 is sometimes an intermediate product in forced oxidation units that generate agricultural-quality or wallboard-quality gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) as a value-added material. More commonly, it is treated as an end waste product and is stockpiled in lagoons or mixed with fly ash for storage in landfills or service as mine by-product (Laperche and Bigham, 2002; Lee et al., 2007)

Flue gas desulphurization gypsum generally has high purity, low heavy metal content, and is rich in essential of beneficial mineral nutrients for plants such as S, Si, and Ca (Guo et al., 2016a). FGD gypsum is a unique synthetic product derived from flue gas desulfurization (FGD) systems at electric power plants. The sulfur dioxide emission control systems used by coal-fired power plants remove sulfur from combustion gases using "scrubbers." One particular type of scrubber that uses lime or limestone reagent (i.e., a substance used in a chemical reaction to produce another substance) and a forced oxidation (i.e., a chemical combination with oxygen) system produces "FGD gypsum," which is chemically nearly identical to mined natural gypsum and provides a wide range of environmentally friendly applications (Lee et al., 2008).

The term "synthetic gypsum" has been used widely to encompass materials produced by a variety of industrial processes. In addition to FGD gypsum, synthetic gypsum includes materials such as phosphogypsum, (a byproduct of processing phosphate ore to make phosphoric acid), Titanogypsum (a byproduct from the production of titanium dioxide),

fluorogypsum (a byproduct from the production of hydrofluoric acid from fluorspar) and citrogypsum (a byproduct of citric acid production) (Laperche and Bigham, 2002).

There are many application uses of FGD gypsum, including in agriculture, gypsum panel products, highway construction, mining applications, cement production, water treatment, and glass making. Gypsum has been shown to reduce runoff on soils prone to crust formation. Increased infiltration from gypsum applications could help reduce runoff P and other nutrient losses from the application of broiler litter (BL), a nutrient-rich fertilizer (Endale et al., 2014; Torbert and Watts, 2014). The addition of FGD gypsum increases the strength of cement and concrete (Jiang et al., 2011). FGD gypsum has the potential to be used as a Ca or S fertilizer, as an acid soil ameliorant, and for reclaiming or mitigating sodium-affected soils (Sakai et al., 2004; DeSutter and Cihacek, 2009).

1.1.4. Biochar

Biochar is the carbon-rich product obtain when biomass, such as wood, manure or leaves, is heated in a closed container with little or no available air. Biochar is produced by the so-called thermal decomposition of organic material under a limited supply of oxygen (O_2), and relatively low temperatures ($<700^\circ C$) (Lehmann and Joseph, 2009)

In contrast to the organic C-rich biochar, burning biomass in a fire creates ash, which mainly contains minerals such as calcium (Ca) or magnesium (Mg) and inorganic carbonates. Also, in most fires, a small portion of the vegetation

is only partially burned in areas of limited O₂ supply, with a portion remaining as char (Kuhlbusch and Crutzen, 1995) in (Lehmann and Joseph, 2009).

‘Char’ is a term that is often used interchangeably with charcoal, but is sometimes applied to refer to a material that is charred to a lesser extent than charcoal, typically as a product of fire (Schmidt and Noack, 2000) in (Lehmann and Joseph, 2009). ‘Activated carbon’ is a term used for biochar-type substances, as well as for coal, that has been ‘activated’ in various ways using, for example, steam or chemicals, often at high temperature (>700°C). This process is intended to increase the surface area for use in industrial processes such as filtration (Boehm, 1994) in (Lehmann and Joseph, 2009). Depending on biomass type and process parameters of thermal conversion of biomass, biochar can consist of 50-90% of organic carbon, 1-15% of moisture content, 0-40% of volatile substances and 0.5-5% of mineral ash. C/N ratio can range from 7 to 500 or more. Biochar has neutral or alkali pH. The contents of P and K in various biochars are different and the content of P and K in biochars in 2.7-480 g/kg and 1.0-58 g/kg, respectively (Verheijen et al., 2009).

Four complementary and often synergistic objectives may motivate biochar applications for environmental management: soil improvement (for improved productivity as well as reduced pollution); waste management; climate change mitigation; and energy production, which individually or in combination must have either a social or a financial benefit or both (Lehmann and Joseph, 2009). Anderson et al., (2011) examined biochar induced soil microbial community changes from the soil where biochar had been incorporated during pasture renewal, and found that compared to control soils

the abundance of the bacterial families *Bradyrhizobiaceae* and *Hyphomicrobiaceae* increased. Anderson et al. (2011) concluded that adding biochar to the soil potentially increased microbial N cycling, especially the abundance of those organisms that may decrease N₂O fluxes and NH₄⁺ concentrations. Noguera et al., (2010) hypothesized that earthworms and biochar would have a synergistic effect on nutrient availability and plant growth. Biochar can improve nutrient availability (Steiner et al., 2017), cation exchange capacity (Liang et al., 2006), bulk density, and water-holding capacity (Tyron, 1948). The metabolizable source of C may stimulate microbial activity and lead to utilization of soil mineral N (immobilization) or release of additional N (mineralization) (Schomberg et al., 2012). The mineralizable N fraction would increase with the addition of biochar with higher volatile matter (%), aliphatic C contents, or high O/C ratios because this biochar might increase C availability to the microbial (Schomberg et al., 2012).

Incorporating biochar into the soil can significantly decrease NH₃ volatilization from ruminant urine and that the NH₃-N adsorbed onto the biochar is bioavailable (Taghizadeh-toosi et al., 2012). Biochar manufactured from wood at low temperature can adsorb NH₃, reducing volatilization losses from ruminant urine affected the soil. The urine-N adsorbed by the biochar under a urine patch is plant available. Biochar has the potential for use in intensively grazed pastures to decrease NH₃ volatilization and act as a transient store of plant available N (Taghizadeh-toosi et al., 2012). The addition of biochar reduced volatilization of ammonia significantly during the first week of the process. Also, the addition of biochar increased temperature and organic

matter decomposition (Malińska et al., 2014). Biochar accelerated the humification progress of sludge organics. Biochar increased oxygen uptake rates of sewage sludge during aerobic degradation. SEM showed the porosity of the sludge surface increased with 12-18% biochar amended (Zhang et al., 2014). The use of biochar as a bulking agent can result in decreasing bulk density and increasing aeration conditions (Steiner et al., 2010), improving microbial growth and microbial respiration rates (Jindo et al., 2012a), as well as enhancing the absorption of gaseous NH_3 and water-soluble NH_4^+ (Hua et al., 2009; Steiner et al., 2011).

1.2. Research objectives

While much has been done to monitor ammonia and VOA odor in the swine livestock facilities (Harper et al., 2010; Kalantarifard et al., 2013; Cho et al., 2013; Kim and Choi, 2013), relatively less is known about the co-additive application rule to mitigate the ammonia and VOC emission in the swine livestock facilities. Therefore the objectives of this study were:

- 1) To investigate the effect of additive (Flue Gas Desulphurization Gypsum (FGDG) combined with Biochar) to the performance of livestock waste composting process and its product;
- 2) To evaluate the effect of two different ventilation type (wall ventilation and pit ventilation) attached to different biofilter system on odor emissions, aerial environments and microclimate conditions of the pig house.
- 3) To evaluate the performance of dual biofiltering system supplemented by the co-additive FGD-gypsum and biochar in the integrated pilot pig house-biofilter system.

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CHAPTER 2 . THE EFFECT OF CO-ADDITIVES (BIOCHAR AND FGD GYPSUM) ON AMMONIA VOLATILIZATION DURING THE COMPOSTING OF LIVESTOCK WASTE

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2.1. Abstract

The effectiveness of co-additives for improving livestock waste composting (reduction of air pollution and conservation of nutrients) was investigated. Biochar and Flue gas desulphurization gypsum (FGD gypsum) were used to supplement the composting of a mixture of slaughter waste, swine slurry, and sawdust. Different compositions of additives (0% or 5% each, 10% biochar or FGD gypsum) were tested in triplicate on the laboratory scale. In addition, the effects of two different aeration schemes (continuous and intermittent) were also investigated. Ammonia volatilization, physicochemical characteristics, and compost maturity indices were investigated. The results indicated that the use of the co-additive (Biochar and FGD gypsum) during composting of livestock waste led to a reduction of ammonia volatilization by

26–59% and to a 6.7–7.9-fold increase of nitrate accumulation. The total ammonia volatilization of intermittent aeration treatment was lower than that of continuous aeration using co-additives treatment. It was concluded that co-additives (biochar and FGD gypsum) might be utilized in livestock waste composting to reduce ammonia volatilization and improve nutrient conservation.

Keywords: livestock waste; composting additives combination; ammonia; volatilization

2.2. Introduction

The livestock sector globally is highly dynamic and is organized in long market chains that employ at least 1.3 billion people worldwide and directly support the livelihoods of 600 million poor smallholder farmers in the developing world (Thornton, 2010). However, the waste from livestock industrial activities (for example manure, slaughter waste, is the major source of negative environmental impacts (that is, the release of greenhouse gases or excess nutrients, salt accumulation, growth of pathogenic microorganisms, odor, and so on), which are capable of influencing both humans and animals (Franke-whittle and Insam, 2013; ten Hoeve et al., 2014). Composting is an effective technology for treatment and disposal of livestock and agricultural waste because it can be converted to sanitary and marketable organic fertilizer (Gamroth, 2012; Sivakumar et al., 2008). The stabilization process during composting kills most parasites, pathogens, and viruses contained in the livestock waste (Bharathy et al., 2012; Pan et al., 2012).

Nevertheless, the process of aerobic composting can cause problems. Several chemical compounds (for example, hydrogen sulfide (H_2S), trimethylamine (TMA), ammonia (NH_3), 2-pentanone, 1-propanol-2-methyl, dimethyl sulfide, dimethyl disulfide, dimethyl trisulphide, and acetophenone) were identified as the main potential odor contributors from composting, which were correlated with both the aeration rate and the ratio of bulking agent to waste during composting (Blazy et al., 2014; Sun et al., 2014). Among the potential odor compounds, ammonia (NH_3) is a major component that causes odors nuisance and toxicity to humans and plants (Colón et al., 2010). Nitrogen losses that occurred during composting resulted in the emission of ammonia (NH_3) gas (ammonia volatilization) accounted for 24–33% of the initial N in household waste (Beck-Friis et al., 2001), 46.8–77.4% of the initial N of a mixture of straw and manure (Martins and Dewes, 1992), 62% of the initial N was during composting of poultry layer manure (Kithome et al., 1999), and 24–64% of the nitrogen was in organic waste (Zeng et al., 2012). The volatilization of ammonia deteriorating both human health and the environment would reduce the fertilizer value of organic waste. Barrington et al. (Barrington et al., 2002) reported that carbon availability, bulking agent, particle size, moisture content, and aeration regime are the factors that determine whether composting results in N volatilization as odor or N immobilization into organic components. The odor emission rate was correlated with both the aeration rate and the ratio of bulking agent to waste when composting (Blazy et al., 2014).

The utilization of chemical and biological additives to mitigate ammonia volatilization and nitrogen loss during the composting process has been

extensively studied. These additives have included wood fly ash, lime, phosphor-gypsum, polyethylene glycol, jaggery (Gabhane et al., 2012), zeolite (Nissen et al., 2000), bentonite (Li et al., 2012), superphosphate (Huang et al., 2006), and microbial inoculants (Zhang et al., 2016). Among these additives, biochar was popular to be amended in the composting mixture. Biochar is a carbon-rich material produced by the thermal decomposition of biomass, was effective for reducing volatilization of ammonia during the composting of a mixture of sewage sludge and wood chips (Malińska et al., 2014). The faster decomposition process in the bio-oxidative phase led to lower greenhouse gas emissions being observed when biochar was added at the beginning of the composting process (Vandecasteele et al., 2016; Huang and Xue, 2014). Reduction in readily available P was observed when biochar was applied during compost storage (Vandecasteele et al., 2016). Biochar addition at 3% could reduce the composting time by 20% (Sánchez-García et al., 2015). As a composting additive, biochar favored microbial activity (Jindo et al., 2012; Yoshizawa et al., 2006). Jindo et al., (2012) reported a higher diversity of fungi in biochar-amended compost. Biochar materials strongly sorb polycyclic aromatic hydrocarbons (PAHs) and are thus characterized by very high sorption coefficients (K_D) for PAHs (Mayer et al., 2016).

It was also reported that the N loss was mitigated by adding FGD gypsum to composting materials (Guo et al., 2016b; Tubail et al., 2008). Flue gas desulphurization gypsum (FGD gypsum) is a by-product of wet gas desulphurization in coal-fired power stations and generally has a high purity, low heavy metal content, and is rich in essential mineral nutrients for plants

such as S, Si and Ca (Guo et al., 2016a; Chen et al., 2015). The volatilization of ammonia and N loss were reduced by combining organic N-rich organic waste with FGD gypsum during the composting process (Tubail et al., 2008). The nitrogen compounds of humic substance in compost with FGD gypsum was higher than that in compost without FGD gypsum (Guo et al., 2016a). Gypsum has been known as an effective product used in the remediation of sodic soils, by greatly reducing the exchangeable Na, K, and Mg cation concentrations in the sodic soils (Presley, 2016; Ahn et al., 2007). Approximately 1–2 wt % of FGD gypsum was used to reclaim the sodic soil which resulted in no obvious heavy metal contamination (Chen et al., 2015). Guo et al. (Guo et al., 2016a) suggested adding 10% FGD gypsum to compost material to reduce nitrogen loss during composting. Information is still limited to the co-additive means of more than one additive supplemented together when applied with different aeration methods during the composting of livestock waste. Accordingly, the objectives of this research were to investigate the effect of co-additives (FGD gypsum and biochar) and the different aeration schemes (Continuous and Intermittent) on the performance of livestock waste composting and on its product.

2.3. Methods

2.3.1. Feedstock Preparation and Experimental Setup

The mixture of swine slurry and slaughter waste were utilized as livestock waste in this study. Mixtures of swine slurry and slaughter waste for composting trials were not commonly applied yet in South Korea. The swine slurry was obtained from the swine farm station of Seoul National University.

The slaughter waste was obtained from a local slaughter house in Yeongcheon City (North Gyeongsang Province, South Korea). These livestock wastes functioned as the main substrate for the composting process. Sawdust was utilized as a bulking agent and was obtained from the local municipality. The Biochar was obtained from the local producer in Gyeonggi province, South Korea. It was produced from a combination of hardwood (80%) and softwood (20%) by the pyrolysis process at 550 °C. FGD gypsum was obtained from the KEPCO coal-fired power plant of South Korea. The physicochemical properties of the raw materials prior to composting were given in Table 2.1.

The experiment was prepared with eight different sets of composting mixtures with different co-additive combinations, and aeration schemes labeled CC, BC, FG, FB, CC-I, BC-I, FG-I, and FB-I respectively (Table 2.2). Mixed raw materials of an amount of 5 kg were composted in 15 L cylindrical laboratory scale composter reactors (height 30 cm × inner diameter 25 cm) covered with polyurethane insulation material to prevent heat loss (Figure 2.1).

The experiment was conducted for 28 days. The compost pile was stirred on Day 7, 14, and 21. Samples of approximately 150 g each were collected just after the materials were stirred; then they were kept at 4 °C for later physicochemical analysis. The aeration rate of 0.75 L/min·kg VS was set to keep the oxygen concentration above 15%, as suggested by References (Ahn et al., 2007) and (Lu et al., 2001). For the intermittent aeration, the air pump was left off for 135 minutes then a timer switched on the high flow air pump for 15 min. to provide air at about 0.75 L/min·kg VS. The composition ratio of

raw material was considered to reach the initial moisture content of about 55–60% as the most suitable condition for the composting process.

Table 2.1. Properties of raw materials used for composting experiment

	Swine Slurry	Slaughter Waste	Sawdust	Biochar	FGD Gypsum
pH	8.09 ±0.40	6.23 ±0.42	7.59 ±0.38	10.99 ±0.55	8.54 ±0.43
EC	22.25 ±1.67	6.85 ±0.28	0.15 ±0.01	0.73 ±0.05	2.69 ±0.20
BD (g/L)	1059.75 ±63.59	1133.92 ±110.82	243.57 ±14.61	515.29 ±30.92	703.33 ±42.20
TS (%)	6.27 ±0.56	26.11 ±1.48	81.49 ±7.33	90.35 ±8.13	77.75 ±7.00
VS	72.15 ±3.61	94.91 ±4.58	98.99 ±4.95	65.90 ±3.30	3.04 ±0.15
Ash (%)	27.85 ±2.09	5.09 ±0.64	0.94 ±0.07	34.10 ±2.56	96.06 ±7.20
TOC	41.85 ±2.51	55.05 ±3.19	57.42 ±3.45	38.23 ±2.29	2.41 ±0.14
TKN	6.74 ±0.61	6.26 ±0.82	0.37 ±0.03	0.36 ±0.03	n.d
C/N	6.21 ±0.31	8.79 ±0.29	155.18 ±7.76	105.02 ±5.25	n.d

EC = Electrical conductivity; BD = Bulk density; TS = Total solid; VS = Volatile solid; TOC = Total organic carbon; TKN = Total Kjeldahl Nitrogen; C/N = C/N ratio.
Values are expressed as mean ± standard deviation

Table 2.2. Feedstock composition ratio of treatment compost

Treatment	Feedstock Ratio (% Wet Weight)			Additive (% Raw Feedstock on Wet Weight)		Aeration
	Slaughter Waste (%)	Swine Slurry (%)	Sawdust (%)	FGD Gypsum (%)	Biochar (%)	
CC	41.7	16.7	41.6	-	-	continuous
BC	41.7	16.7	41.6	-	10	continuous
FG	41.7	16.7	41.6	10	-	continuous
FB	41.7	16.7	41.6	5	5	continuous
CC-I	41.7	16.7	41.6	-	-	intermittent
BC-I	41.7	16.7	41.6	-	10	intermittent
FG-I	41.7	16.7	41.6	10	-	intermittent
FB-I	41.7	16.7	41.6	5	5	intermittent

2.3.2. Physicochemical Parameter Analysis

Samples of the raw materials and treatment mixtures were air dried, ground up into small particles using porcelain mortar, and passed through a 2-mm sieve. The sieve samples were analyzed for physicochemical properties. The physicochemical characteristics of samples and raw materials were analyzed using methods on dry weight basis (APHA, 2005). The moisture content was determined by drying fresh samples at 105 °C until the mass loss in 24 h was less than 0.5% relative to the previous day. Volatile solid was measured by dry combustion of dried samples (550 °C for four h). The pH and electrical conductivity (EC) were determined using a pH meter (Inolab, WTW, GmbH, Weilheim, Germany) and an EC214 conductivity meter (Hanna Instruments, Ltd., Sarmeola di aarubano, Italy) respectively.

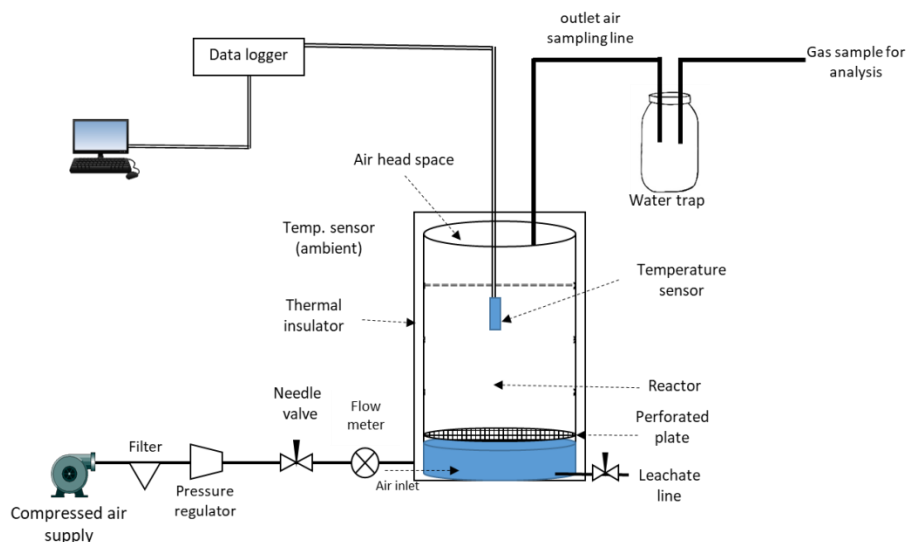


Figure 2.1. Reactor for composting experiment

The sample was suspended in distilled water in a ratio of 1:10 (w/v). This suspension was shaken in a mechanical shaker at 230 rpm for 30 min and allowed to stand for an hour before pH and EC measurement. The bulk density of compost material was defined as its weight per unit volume according to Thompson (Thompson et al., 2002). The Kjeldahl Nitrogen (TKN) content was determined according to the Kjeldahl digestion method (Bremmer et al., 1996). The TOC was determined by the Walkey Black method (Nelson and Sommers, 1996). C/N ratio was estimated by divided the TOC to the TKN. Inorganic nitrogen NH_4^+ , NO_3^- and NO_2^- were extracted using 2 mol/L KCL (10:1 V/m), and then analyzed using DR 5000 UV-Vis spectrophotometry (Hach Co., Loveland, CO, USA). The ambient temperature and compost temperature in the center of the composting mixture were recorded hourly by a temperature sensor and data logger (T&D recorder TR52i, Shinagawa, Japan). NH_3 was measured by washing bottle according to the method presented by Ren et al. (Ren et al., 2010). Ammonia gas released during composting was captured in boric acid

and was determined by the spectrophotometric method (Hach Co., Loveland, CO, USA).

The phytotoxicity was evaluated according to the method proposed by Sellami et al. (Sellami et al., 2008) with modification. The Germination Index (GI) was determined by extracting 20 g (dry weight) of compost with 200 mL of distilled water, stirring for two hours, and then centrifuging at 9000 r/min. Ten *Raphanus sativus* (white radish) seeds were evenly distributed on filter paper in Petri dishes (10 cm diameter) and moistened with 5 mL of compost extract. Three replicate dishes for each sample were incubated at 25 °C for three days. The number of seeds that germinated was recorded. As a control, 5 mL of distilled water was used to replace the compost extract. The GI was calculated according to the Equation 2.1:

$$GI (\%) = \frac{\text{seed germination} \times \text{root length of treatment}}{\text{seed germination} \times \text{root length of control}} \times 100\% \quad (2.1)$$

Elemental analysis (that is, P, K, Na, Ca, Mg, Fe, Mn, Cu, Zn, and S) of the samples were performed according to standard nitric acid-hydrochloric acid digestion (3030 F, APHA) (APHA, 2005) and using inductively coupled plasma (ICP) atomic emission spectroscopy (AES) (ICPS-7510; Shimadzu Corp., Kyoto, Japan).

2.3.3. Statistical Analysis

The physicochemical parameter data reported in this study were analyzed by ANOVA using the statistical package SPSS 21. The average values of the quantitative factors under evaluation were compared by means of Tukey's test.

2.4. Results and Discussion

2.4.1. The Effect on Temperature, pH, and EC

The temperature change during composting is associated with many biological reactions that take place (Bustamante et al., 2008). The increase of temperature has been widely reported to correlate with microbial activities (Vandecasteele et al., 2016; Huang and Xue, 2014). The composting temperature during this experiment is shown in Figure 2.2A,B. Initially, the temperature in the piles was about 23 °C. On the first day, the temperature increased rapidly in all treatments. The temperature remained above 55 °C for 1–2 days. According to the temperature profiles, the compost with the continuous aeration scheme reached a higher temperature than that of the compost with intermittent aeration. Composting with continuous aeration reached a temperature of 55–60 °C for two consecutive days, while the temperature in the intermittently aerated compost reached 50–55 °C for almost three consecutive days. The composting temperature with intermittent aeration was lower than that with continuous aeration, likely because the oxygen supplied was not constantly adequate for complete bio oxidation (Gao et al., 2010; Tiquia et al., 2002). The longer thermophilic phase that occurred with intermittent aeration composting was likely a result of the slower degradation of partially decomposed materials transferred from the anaerobic areas to aerobic areas, as explained by Jiang et al. (2011). The fact that the composting temperature remained above 55 °C for three days was adequate to provide standard sterilization of the compost (Canadian Council of Ministers of the Environment (CCME), 2005). This phase was followed by a mesophilic phase.

This condition corresponded to a typical composting temperature profile at the laboratory scale. According to the temperature profiles, the FGD gypsum additive supplementation did not disturb the composting process. This result was in accordance with the results of Li et al., (2012) who reported that composting supplemented with bentonite did not show any stimulatory or inhibitory effects on the composting temperature profile. In this study, the highest temperature was reached in the biochar additive treatment. López-Cano et al., 2016) reported that the addition of biochar favored activation of the composting process.

The pH change throughout the composting process is shown in Figure 2.2C. The pH of all treatments showed a gradually decrease. The decline of pH is linked to natural acidification as a result of nitrification during the composting process (Liu et al., 2017). The co-additives of FGD gypsum and biochar had a significant effect on the compost pH ($p < 0.05$). FGD gypsum supplementation had a greater influence on lowering pH, which was shown by the pH decline from 8.10–8.60 to 5.45–6.93 within 28 days of composting. The gradual decline of the pH was likely due to the formation of low-molecular-weight organic acids and CO₂ during the decomposition of organic matter, volatilization of ammonia, and nitrification during the composting process (Jindo et al., 2012; Guo et al., 2016b).

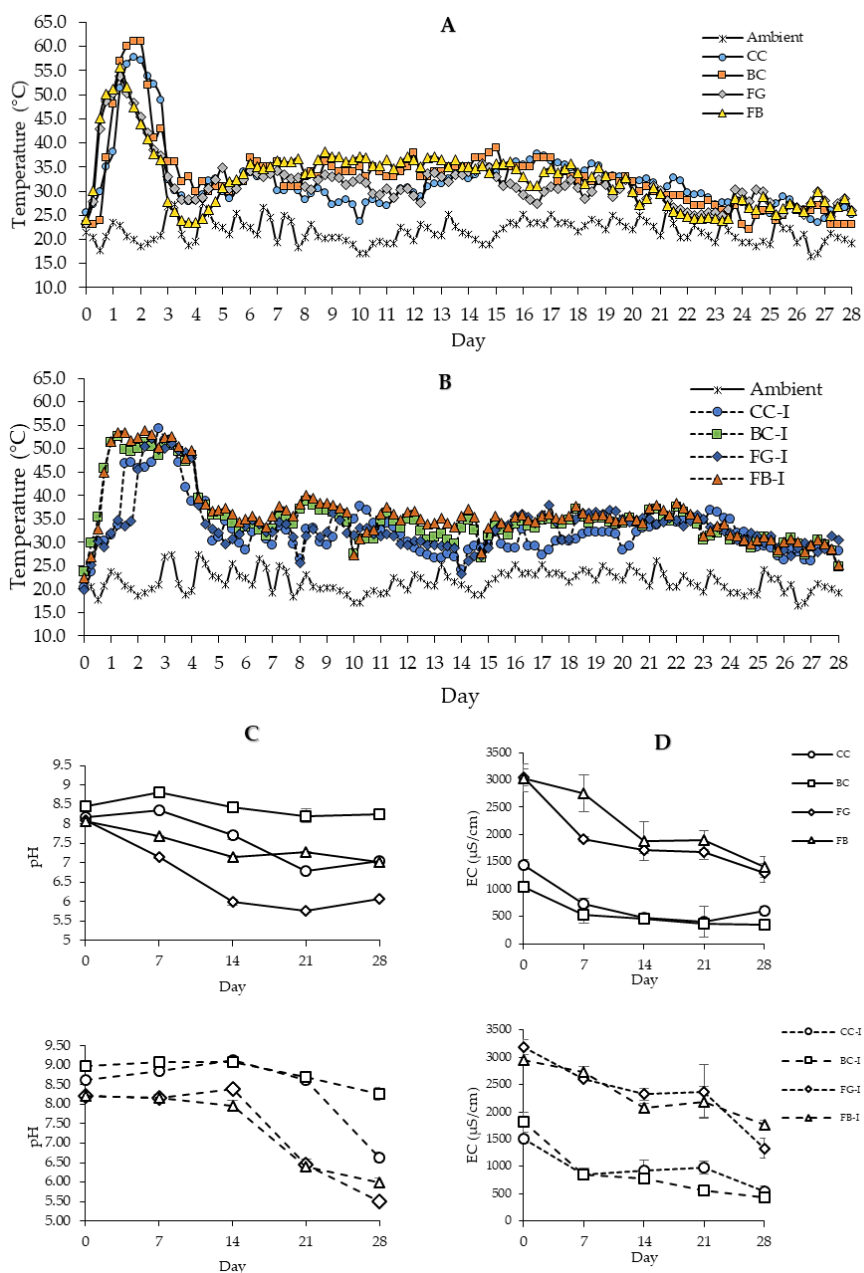


Figure 2.2. Temperature (continuous (A), intermittent (B)), pH (C) and EC (D) profiles throughout composting process

The pH decrease was also attributed to the replacement of Na^+ by Ca^{2+} from the FGD gypsum, by which the reaction $\text{CO}_3^{2-} + \text{HCO}_3^{2-}$ generated deposits of CaCO_3 (Du et al., 2017). The pH of the compost supplemented with

biochar only remained higher. This was probably attributable to the initially high pH. In addition, the rate of ammonia released due to ammonification was higher in this treatment. The pH increase was a result of ammonification and mineralization during composting (Gao et al., 2010). The different aeration schemes resulted in no significant difference on the compost pH in this study.

Soluble salt contents were estimated through electrical conductivity (EC). The EC values throughout the composting process are presented in Figure 2.2D. The initial EC of the treatment with only supplemented by the FGD gypsum showed higher than that of other treatments. Supplementation with FGD gypsum significantly increased the initial EC of the compost substrate and was likely due to the contribution of the high mineral concentration in FGD gypsum. Possible phytotoxic effects could result in compost with high EC values, which indicates high salinity in the composting product. In this study, we observed that EC values decreased with the composting time in all treatments. The decrease of EC could be caused by the volatilization of ammonia and the precipitation of mineral salts during the composting process (Gao et al., 2010). The results observed in this study suggested that the FGD gypsum supplementation did not have a negative effect on the composting process.

2.4.2. The Effect on Ammonia Volatilization

The results of ammonia emission throughout composting in all treatments are shown in Figure 2.3. Compared with the control, the cumulative ammonia emitted from composting with FGD gypsum was significantly less with both the continuous or intermittent aeration scheme. Biochar addition resulted in higher ammonia emission than did the addition of FGD gypsum.

Intermittent aeration resulted in lower cumulative ammonia emission. In composting with continuous aeration, 90% of the ammonia was emitted during the first ten days after commencement of composting whereas, with intermittent aeration, the ammonia emission was distributed over 20 days after the composting start. The higher ammonia emission with the biochar additive could be attributed to higher pH and temperature. Tiquia and Tam (2000) reported that the loss of N by NH_3 volatilization was significant at pH levels above 7.0 and higher temperatures ($>40^\circ\text{C}$). Furthermore, at a higher pH, non-volatile ammonia ions were converted to the volatile ammonia form (Pagans et al., 2006). The highest NH_3 volatilization rates in the present study occurred at 50°C . It was likely because the high temperature affects ammonia volatilization by inhibiting the nitrification process (Koenig et al., 2005).

The reduction of ammonia volatilization in this experiment could be attributed to a lower pH and to ammonia absorption affected by the addition of FGD gypsum. Jeong and Kim, (2001) found that using amendments such as bauxite residues or biodegradable plastics could control compost pH. Guo et al., (2016a) reported that FGD gypsum has the ability to absorb ammonia. In addition, gypsum addition converts the ammonium carbonate to ammonium sulfate along with the formation of calcium carbonate (Guo et al., 2016a). The N in ammonium carbonate is much more apt to be lost as ammonia than when it is in the form of ammonium sulfate (Tubail et al., 2008).

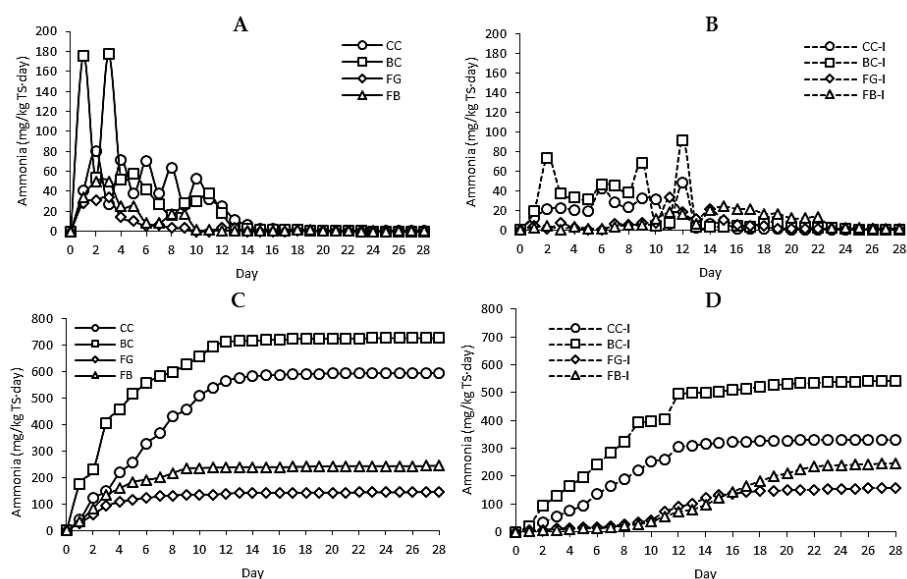
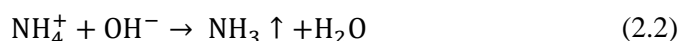


Figure 2.3. Daily and cumulative ammonia production throughout composting process (A) Daily, continuous aeration; (B) Daily, intermittent aeration; (C) Cumulative, continuous aeration; (D) Cumulative, intermittent aeration

We observed that by FGD gypsum supplementation, NH_3 volatilization could be reduced by 75.46% with continuous aeration, in comparison to the control. Co-additive supplementation with FGD gypsum and biochar reduced the ammonia volatilization by 58.93% with continuous aeration, in comparison to the control. Supplementation by biochar increased the ammonia volatilization by about 22.17% in comparison to that of the control. By intermittent aeration, FGD gypsum alone and co-additive supplementation reduced the ammonia volatilization by 52.64% and 25.87%, respectively. These results were in accordance with the results of (Koenig et al., 2005) who demonstrated that aluminum sulfate; chloride salts of aluminum, calcium and magnesium; gypsum; and surface-applied sodium bisulfate reduced the NH_3 volatilization by 0–87%, depending on the rate and intervals of incubation. We observed in the present study that the lower NH_3 volatilization rates mostly

related to the reduction in the pH of the substrate. The results of the present study show that ammonia volatilization after biochar treatment was significantly higher than that of other treatments. Biochar supplementation increased ammonia volatilization by 63.24% in comparison to that of the control. These results contrast with earlier findings that suggested that co-composting with biochar might reduce ammonia emission and increase nitrification (Khan et al., 2014). The possible explanation of this result is that the high initial pH and temperature of the composting substrate induced the higher volatilization of ammonia. Schomberg et al., (2012) found that biochar and biochar mixed with ash have the potential to raise the substrate pH. Mandal et al. (Mandal et al., 2016) explained the significant linear relationship between the soil pH values and the cumulative NH_3 volatilization from soils. Ammonia volatilization is favored at alkaline pH and when a high concentration of NH_4^+ is present. When NH_4^+ and OH^- are present in the substrate, the OH^- ions readily react with NH_4^+ ions, thereby resulting in the release of NH_3 gas (Bolan et al., 2004), as described in Equation (2.2).



Schomberg et al., (2012) demonstrated that NH_3 volatilization increased when biochar with a high pH was applied. This was because at high pH, the ammonification process was accelerated, thereby increasing the NH_3 volatilization. A corresponding increase in NH_3 volatilization was observed as a result of the higher pH (increased OH^- concentration) during the first seven days of composting unless treatment with FGD gypsum occurred along with continuous aeration. The high ammonification process seems to have stopped

after the first seven days of composting, and the nitrification process started with a corresponding decline in NH_3 volatilization and compost pH. The decline of the compost pH could be attributed to the nitrification reaction. NH_4^+ ions oxidized directly to NO_3^- ions and released H^+ ions, which subsequently reduced NH_3 volatilization. The slow mineralization of N was reported to be one possible reason for the low NH_3 volatilization (Mandal et al., 2016). The explanation of low NH_3 and high NH_4^+ with FGD gypsum treatment could be that acidic surfaces (low pH) can protonate NH_3 gas to form NH_4^+ ions, as in Equation (2.3).



The treatment with intermittent aeration shows many peaks of ammonia emission even after 14 days composting (Figure 2.3B). According to Tiquia and Tam, (2000) and Liang et al., (2006), the occurrence of several NH_3 emissions peaks could be explained in three ways. (1) Part of the organic N was first degraded to some simple N-containing compounds and stored in the compost. These compounds were mineralized after biodegradation slowed down. (2) A slowly degradable fraction with a high content of biodegradable N was mineralized. (3) NH_4^+ released from ammonification at the beginning of the treatment was first immobilized in the biomass; then, the remineralization of the biomass was responsible for the next peak of NH_3 emissions. Furthermore, it was explained that during incomplete nitrification, under the effect of ammonia monooxygenase, NH_3 is oxidized sequentially to hydroxylamine (NH_2OH) and nitroxyl (NOH) by hydroxylamine oxidoreductase, which can be

converted to N_2O (Canfield et al., 2010). Koenig et al., (2005) also reported that the increase of NH_3 volatilization could be due to the physical effects from changes in materials and pore-space causing enhanced ventilation, as well as of improved aerobic decomposition during composting. In later work, de Guardia et al. (de Guardia et al., 2010) demonstrated that NH_3 emissions were much higher at higher aeration rates and suggested the importance of the mineralization of organic N.

2.4.3. The Effect on Organic Matter and nitrogen fraction

The effects of treatments on TOC throughout the composting are shown in Figure 2.4A. TOC content slightly decreased in all treatments. There was no significant effect from the aeration schemes on the decrease of TOC during the composting in this experiment, however by co-additive treatment, the decrease in TOC was 4.98% during the 28 days composting. This could be due to the ability of FGD gypsum and biochar to increase the porosity of the compost substrate. Li et al., (2012) reported that the decomposition of organic matter was enhanced by the addition of bentonite, clinoptilolite, and zeolite. Gao et al., (2010) reported a decrease of 15.4% in 60 days of composting chicken manure. Tiquia and Tam, (2000) reported 9% organic matter loss during 120 days of chicken manure composting.

The variation of TKN during the composting process is presented in Figure 4B. The TKN content gradually increased under all treatments. The result in the present study was in accordance with Li et al., (2012) who found that the TKN content improved by the addition of bentonite during swine manure composting. The TKN increase can be attributed to a concentration

effect as a consequence of the degradation of organic compounds because composting biomass is reduced faster than nitrogen (Liu et al., 2017). The loss of TKN during the first week of composting was likely because of the loss of ammonia due to the volatilization at a relatively high temperature. TKN increased the most in co-additive treatment (5% FGD gypsum and 5% biochar). This result was likely associated with the additive combination speeding up the degradation rate during the composting process. The large specific surface area of the biochar was more favorable for microbial attachment. In addition, the porosity of the biochar made air and moisture available to the compost material, which is favorable for the decomposition process in composting (Liu et al., 2017). On the other hand, FGD gypsum supplied more mineral nutrient sources for microbial growth, which accelerated the decomposition of organic matter.

The mineralization of organic matter during composting led to the gradual decrease of the C/N ratio (presented in Figure 2.4C). The C/N ratio of the control and biochar treatment slightly increased in the first week of composting; then gradually decreased as the composting progressed. The slight increase of the C/N ratio in the first week of the composting process could be due to vigorous ammonia volatilization (Tiquia and Tam, 2000). Organic matter degradation and nitrogen compound mineralization then responded to the reduction of the C/N ratio during composting (Li et al., 2012). The C/N ratio in all treatments during composting in this experiment decreased from around 34.64 to 25.36. These results were in accordance with those in another reported by Tiquia et al., (2002), in which the decline of the C/N ratio from 28 to 17 was reported during the composting of chicken manure.

The ammonification and nitrification processes were also monitored by determining the concentrations of NH_4^+ and NO_3^- . The ammonification reaction is indicated by the release of NH_4^+ , while nitrification is indicated by the production of NO_3^- (Mandal et al., 2016). The NH_4^+ variation throughout the composting process is shown in Figure 2.4D. The NH_4^+ content in all treatments decreased gradually during composting. We observed the $\text{NH}_4\text{-N}$ decrease rapidly only in the treatments of composting with biochar (BC and BC-I) either by continuous and intermittent aeration (Figure 2.4D). Reduction of the ammonification rate could be attributed to the alkalinity and high absorptive capacity of the biochar (Chen et al., 2015; Sellami et al., 2008). Chen et al., (2013) reported that the application of alkaline biochar with a high absorptive capacity played a major role in maintaining the soil pH, enhanced the adsorption of $\text{NH}_4^+/\text{NH}_3$, and thereby reduced the NH_3 volatilization.

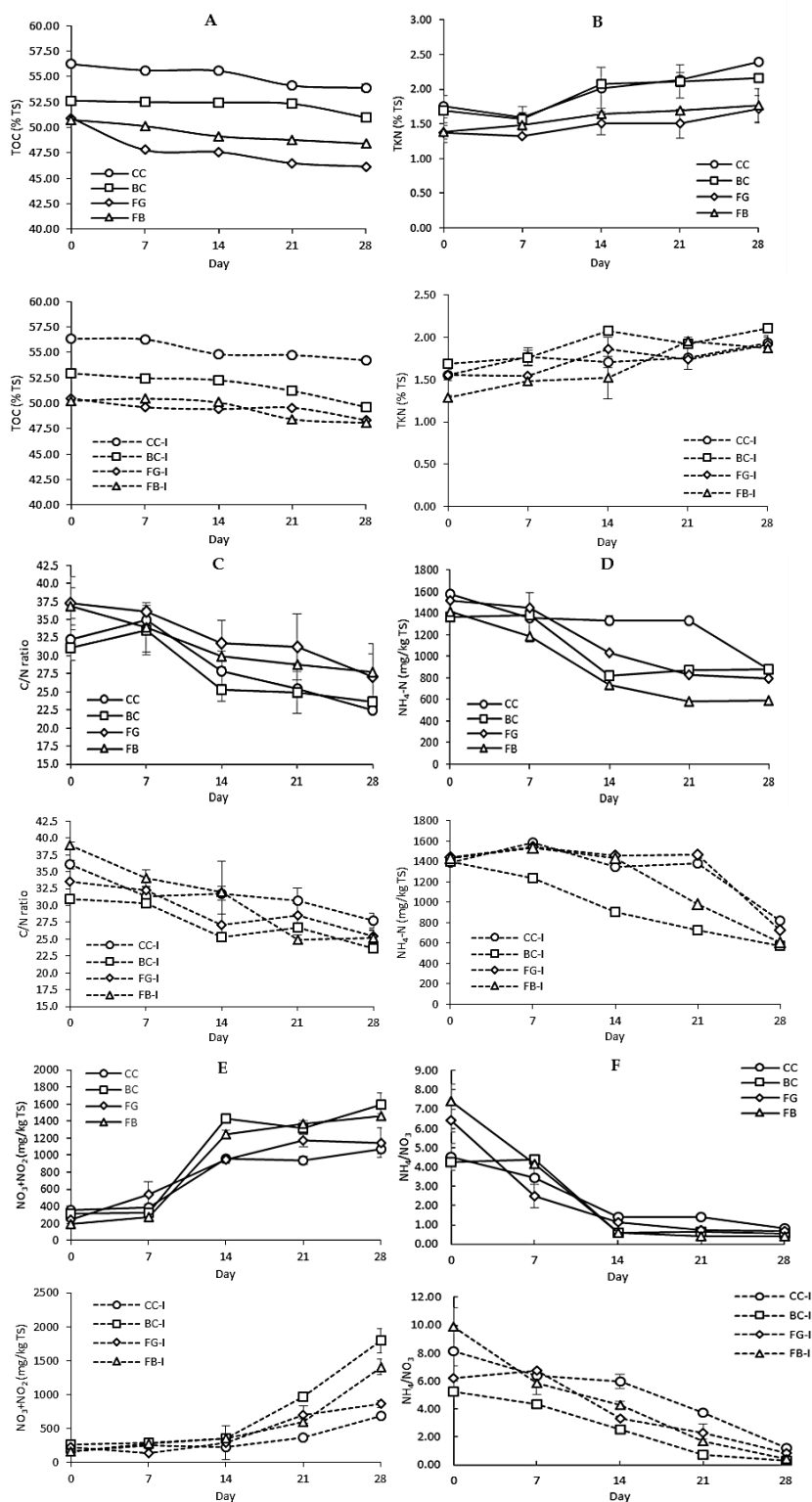


Figure 2.4. TOC (A), TKN (B), C/N ratio (C), $\text{NH}_4\text{-N}$ (D), NO_3+NO_2 (E), NH_4/NO_3 (F) throughout the composting process.

The treatment with Co-additives and continuous aeration showed a rapid decrease of NH_4^+ . In biochar additive treatment, NH_4^+ decreased rapidly after the second week of composting while other treatments showed rapid decreases after the third week of composting (Figure 2.4D). The increase of NH_4^+ during the first week of composting could be due to the decomposition of nitrogen-containing organic matter to form ammonia. The decrease of NH_4^+ during composting was attributed to assimilation by microorganisms, to volatilization, and to nitrification (Gao et al., 2010).

The $\text{NO}_3^- + \text{NO}_2^-$ transformation during composting are shown in Figure 4E. Different aeration schemes showed a different pattern of $\text{NO}_3^- + \text{NO}_2^-$ transformation. In continuous aeration, $\text{NO}_3^- + \text{NO}_2^-$ accumulation started rapidly from the second week of composting, while rapid $\text{NO}_3^- + \text{NO}_2^-$ accumulation under intermittent aeration started after three weeks of composting. Late accumulation of $\text{NO}_3^- + \text{NO}_2^-$ in intermittent aeration treatments could be due to the late activation of nitrifying bacteria. The growth and activity of nitrifying bacteria are inhibited during the temperature conditions of more than 40 °C (Gao et al., 2010). The temperature goes down slowly under intermittent aeration rather than under continuous aeration, thus the condition in which nitrifying bacteria could actively proliferate was delayed. Co-additives treatment of FGD gypsum and biochar (FB and FB-I) showed the highest $\text{NO}_3^- + \text{NO}_2^-$ accumulation rate (6.72 and 7.90-fold) during the 28 days composting. As the composting progressed to the cooling phase, the autotrophic nitrifying bacteria increased and oxidized NH_4^+ to NO_2^- , which was further

oxidized to NO_3^- by nitrite-oxidizing microorganisms. During the intermittent aeration treatment, the presence of biochar increased the porosity of the compost pile, which may induce oxygen penetration and accelerate the nitrification or conversion of NH_4^+ to $\text{NO}_3^- + \text{NO}_2^-$. Furthermore, absorption via the transfer of NO_3^- along with soil pore solution into biochar pores could also be possible. The adsorption of NO_3^- (an anion) to biochar may also be related to its anion exchange capacity (Khan et al., 2014).

Figure 2.4F shows the $\text{NH}_4^+/\text{NO}_3^-$ ratio throughout the composting process. The $\text{NH}_4^+/\text{NO}_3^-$ ratio is a desirable candidate indicator for compost maturity (Guo et al., 2016a). The $\text{NH}_4^+/\text{NO}_3^-$ ratio of all treatments in the present study decreased gradually. The $\text{NH}_4^+/\text{NO}_3^-$ ratio of FG-I increased in the first week of composting, which could be due to the inhibition of nitrification and accumulation of NH_4^+ . The addition of FGD gypsum increased the soluble salt, as indicated by the higher EC. The high EC or pH of the additive might disrupt nitrification by inhibiting the growth of nitrifying bacteria (Martins and Dewes, 1992; Barrington et al., 2002). Because the presence of biochar provides benefits that induce nitrifying bacteria growth (Liu et al., 2017), we observed that biochar might serve to overcome the limitation of adding FGD gypsum alone in composting. We also observed a similar pattern of $\text{NH}_4^+/\text{NO}_3^-$ decrease during composting by both continuous and intermittent aeration.

2.4.4. The Effects on Macro-, Micronutrients and Maturity Indices

The effects of the co-additive treatment and aeration scheme on the concentrations of macro- and micro- mineral nutrients are summarized in Tables 2.3 and 2.4, respectively. FGD gypsum treatment tends to increase the concentrations of P, Ca, Mg, and S in the compost ($p < 0.05$), whereas compost with biochar had elevated amounts of Fe and Mn ($p < 0.05$). These high concentrations of minerals can be explained by the nature of the additives and the slaughter waste substrate. In addition, the mineralization process during composting increased the concentration of mineral nutrients. Hu et al. (Hu et al., 2009) suggested that mature compost should contain a variety of nutrients such as phosphorus (2.15 g/kg), potassium (0.79 g/kg), calcium (12.9 g/kg), magnesium (8.0 g/kg), sulphur (3 g/kg), sodium (1.15 g/kg), iron (1.2 g/kg), along with trace amounts of zinc (24.9 mg/kg), manganese (80.4 mg/kg), copper (5.7 mg/kg), and boron (5.7 mg/kg). The balance of macro and micronutrients of compost observed in the present study was achieved by the co-additive treatment (combined FGD gypsum and biochar). The compost treated with the co-additive contained no extremely high macro- and micro-mineral concentrations. The observed results show that compost aerated by continuous aeration mostly contains comparatively higher macro- and micro-mineral nutrient concentrations. This indicated that a higher rate of mineralization occurred under continuous aeration than under intermittent aeration. The high mineral concentration of the compost could also be the effect of the mineral content in the raw materials (slaughterhouse and pig slurry), which initially contained a high concentration of minerals.

The maturity indices from the composting test are presented in Table 2.5. The pH of the compost derived from the present study varied in a range of about 5.50–8.69. The lowest pH was achieved in FG-I (5.50) while the highest pH was achieved in BC-I (8.69). These were significantly different ($p < 0.05$) from the controls CC (7.05) and CC-I (6.64). Hogg et al. (Hogg et al., 2002) reported that in most European national standards, compost should have a pH value within the range of 6.0–8.5 to ensure the safety of most plants. Hu et al. (Hu et al., 2009) suggested that the mature compost was slightly acidic. Most of the compost in this study was within the safe range limit of pH except the treatments with only FGD gypsum additive (FG-I) and only the biochar additive (BC-I). The pH of FG-I (5.5) was slightly more acidic, whereas the pH of BC-I (8.69) was slightly more alkaline. This condition could be attributable to the nature and effect of the additive treatments used in this study.

Table 2.3. The concentration of macronutrients after 28 days composting in this study

Treatment	P	K	Na	Ca	Mg
	(mg/kg)				
CC	494.99 ± 1.27 ^{cd}	3125.03 ± 38.09 ^e	429.66 ± 5.40 ^f	3853.11 ± 7.22 ^b	269.12 ± 3.55 ^a
BC	434.28 ± 1.11 ^b	3203.257 ± 7.78 ^f	321.92 ± 1.51 ^c	11,075.89 ± 19.03 ^d	443.36 ± 2.94 ^d
FG	470.12 ± 0.85 ^c	2939.59 ± 13.09 ^d	302.94 ± 2.60 ^b	26,624.35 ± 71.91 ^g	694.01 ± 7.79 ^f
FB	513.54 ± 0.68 ^d	3634.01 ± 22.32 ^g	399.04 ± 1.73 ^e	26,113.85 ± 86.57 ^f	602.85 ± 3.47 ^e
CC-I	651.69 ± 0.64 ^e	1674.51 ± 19.13 ^a	251.23 ± 2.71 ^a	2261.39 ± 3.62 ^a	301.69 ± 1.79 ^b
BC-I	461.12 ± 0.61 ^{bc}	1991.08 ± 12.28 ^c	244.98 ± 1.06 ^a	6685.14 ± 22.16 ^c	306.98 ± 1.76 ^b
FG-I	386.95 ± 35.66 ^a	1787.36 ± 9.07 ^b	381.61 ± 8.14 ^d	14,448.42 ± 209.08 ^e	434.25 ± 2.50 ^d
FB-I	650.15 ± 0.79 ^e	1679.82 ± 10.42 ^a	313.78 ± 1.36 ^{bc}	11,123.09 ± 36.87 ^d	328.59 ± 0.82 ^c

Values are expressed as mean ± standard deviation

Different superscripts within a column indicate a statistical difference ($p < 0.05$)

FGD gypsum might induce low pH while biochar might induce high pH in the compost product. The acidic compost has potential use for horticultural growing media because of its pH of 5–6.5, as suggested by Careces et al. (Careces et al., 2006). Several elements such as phosphorus, calcium, and magnesium are likely 25–75% available under acidic conditions, as reported by Hu et al. (Hu et al., 2009).

Table 2.4. The concentration of micronutrients after 28 days composting in this study

Treatment	Fe	Mn	Cu	Zn	S
	(mg/kg)				
CC	225.37 ± 0.55 ^e	53.07 ± 1.54 ^{bc}	21.16 ± 0.27 ^e	51.98 ± 0.17 ^b	973.46 ± 2.91 ^a
BC	628.39 ± 0.57 ^g	147.24 ± 3.34 ^f	19.37 ± 0.09 ^d	36.67 ± 0.07 ^a	1580.28 ± 3.41 ^b
FG	460.48 ± 1.79 ^f	46.57 ± 1.90 ^b	15.01 ± 0.15 ^c	38.09 ± 0.06 ^a	14,420.03 ± 36.23 ^f
FB	629.88 ± 1.01 ^g	119.09 ± 4.32 ^e	19.45 ± 0.11 ^d	52.73 ± 0.10 ^b	11,325.19 ± 36.23 ^e
CC-I	135.77 ± 0.27 ^a	27.23 ± 0.77 ^a	14.65 ± 0.13 ^c	105.88 ± 0.17 ^d	1500.77 ± 1.46 ^b
BC-I	401.44 ± 0.65 ^e	86.61 ± 3.14 ^d	13.70 ± 0.08 ^b	71.41 ± 0.13 ^c	1638.19 ± 5.24 ^b
FG-I	192.48 ± 2.09 ^b	21.56 ± 1.04 ^a	8.68 ± 0.12 ^a	69.96 ± 2.64 ^{bc}	10,752.07 ± 88.65 ^d
FB-I	330.02 ± 0.53 ^d	54.72 ± 1.98 ^c	13.45 ± 0.08 ^b	120.91 ± 0.22 ^e	6828.30 ± 21.69 ^c

Values are expressed as mean ± standard deviation

Different superscripts within a column indicate a statistical difference ($p < 0.05$)

The EC varied across the eight observed composting treatments (Table 2.5). FGD gypsum and biochar (as co-additives) significantly altered the EC concentration of livestock waste during composting ($p < 0.05$). Compost derived from slaughter waste/slurry/sawdust mix with FGD gypsum and biochar supplementation had a higher EC ($p < 0.05$) (Table 2.5). The high EC value of treatment FG, FB, FG-I, and FB-I might be attributable to the nature of FGD gypsum, which initially has high EC (Liu and Price, 2011), and the

high EC from amendments applied to soils might induce phytotoxic effects in plants. However, the EC of all the compost produced still could be considered as acceptable for nursery production, the limit of which is about 3000 $\mu\text{S}/\text{cm}$, as suggested by Careces et al. (Careces et al., 2006). The C/N ratios in the product from all the compost treatments are shown in Table 2.5. Different additive and aeration schemes did not have a significant effect on the C/N ratio of the compost produced in this study. The compost produced in the present study had a higher C/N ratio than that established by Hu et al. (Hu et al., 2009) of about 14.6, and by Hogg et al., (2002) (<25).

Table 2.5. Characteristics of maturity indices from the composting test

Treatment	pH	Electrical Conductivity ($\mu\text{S}/\text{cm}$)	C/N Ratio	Germination Index (%)
CC	7.05 ± 0.05^c	610 ± 31.11^a	22.47 ± 0.22^a	104.56 ± 6.88^a
BC	8.25 ± 0.03^d	354 ± 47.38^a	23.64 ± 0.13^a	109.02 ± 6.30^a
FG	6.06 ± 0.04^b	1295 ± 176.78^b	27.08 ± 3.14^a	139.43 ± 9.14^a
FB	7.03 ± 0.11^c	1405 ± 190.92^b	27.69 ± 3.95^a	125.89 ± 2.51^a
CC-I	6.64 ± 0.13^c	553 ± 90.19^a	27.76 ± 1.04^a	117.42 ± 6.91^a
BC-I	8.69 ± 0.15^d	570 ± 28.28^a	26.68 ± 0.49^a	104.07 ± 9.28^a
FG-I	5.5 ± 0.05^a	1330 ± 183.85^b	25.40 ± 0.95^a	129.92 ± 0.99^a
FB-I	5.98 ± 0.12^b	1769 ± 82.73^c	25.23 ± 0.98^a	131.64 ± 39.22^a

Values are expressed as mean \pm standard deviation

Different superscripts within a column indicate a statistical difference ($p < 0.05$)

2.4.5. The Effect on Germination and Respiration Rate

The germination index of all treatments gradually increased during composting (Figure 2.5A). The germination index may reflect the toxicity level of the final compost; a low germination index equals to high toxicity contained by the compost, and a high germination index means low toxicity. Because this compost substrate originated from slaughterhouse waste and pig slurry, there is

some potential for the presence of toxic substances. The increase of the germination index during composting indicated that the toxicity was decreased. The decline of the toxic level was associated with the decomposition of organic matter and conversion of toxic matter to non-toxic matter. This involved the release of toxic substances (for example, ammonia and low-molecular-weight short-chain volatile fatty acids, primarily acetic acid) (Gopinathan and Thirumurthy, 2012). We observed from the present study that the co-additive and aeration scheme treatments did not have any significant effect on the germination index and respiration rate of the composting process.

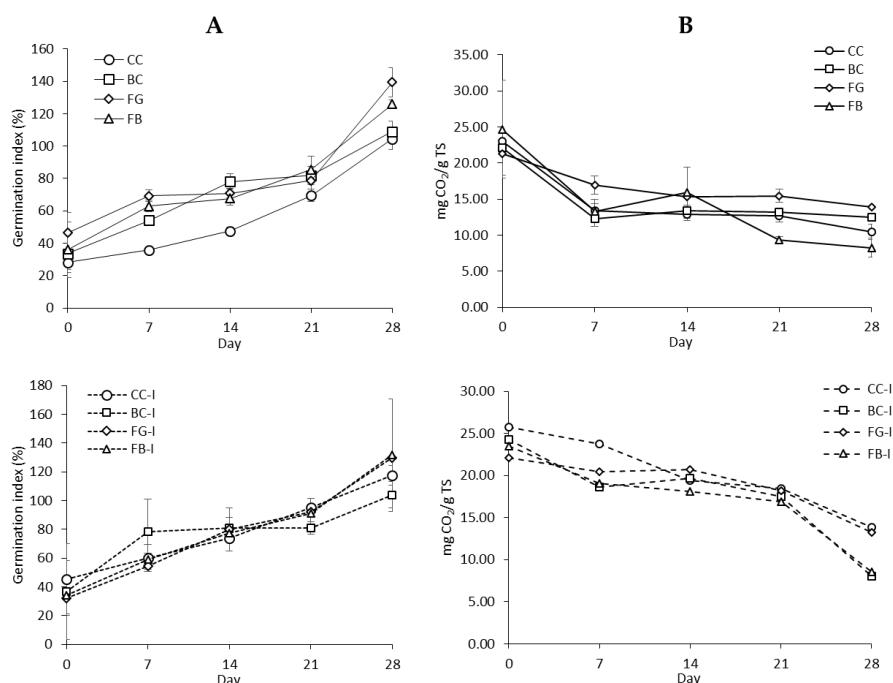


Figure 2.5. Change in (A) Germination index; (B) CO₂ respiration rate during composting. Values are expressed as mean \pm standard deviation

Figure 2.5B shows the respiration rate in all the composting treatments. According to the results, the rate of respiration of composting with continuous

aeration decreased more rapidly than with intermittent aeration treatments. Low rates of respiration are indicative of highly stabilized compost. It was reported that the final compost has a CO₂ respiration rate in a range 0.3–0.6 mg CO₂/g substrate, which was reached after 80–90 days of composting (Wang et al., 2004). We observed that for the compost products from the present study, the CO₂ respiration rate values were in the range of 8.03–13.83 mgCO₂/g·TS after 28 days of composting, which indicated the need for more time to be more stabilized. However, the use of co-additives did not have any negative effects on the compost respiration rate.

2.5. Conclusion

According to the results of this study, it can be concluded that co-additives treatment using FGD gypsum and biochar did not give adverse effect during the 28 days livestock waste composting process. The selection of appropriate additives needs to be adjusted for the purpose of composting. FGD gypsum supplementation may reduce ammonia volatilization more than biochar supplementation, however, biochar supplementation may give a faster biodegradation process during composting. Thus co-additive treatment during composting might be an alternative method to reduce ammonia volatilization as well as induce the biodegradation process.

Supplementation of livestock waste composting with the co-additives FGD gypsum and biochar reduced ammonia loss and enhanced nutrient recovery. The utilization of these co-additives led to the reduction of ammonia volatilization by 26–59% and to an increase in the nitrate (NO₃⁻) accumulation by 6.7–7.9 fold compared to the initial value. The total ammonia volatilization

of the intermittent aeration treatment was lower than that of continuous aeration using co-additive treatments. The macro- and micro-nutrient content were enhanced by the supplementation with these co-additives. The use of co-additives did not have any negative effect on the compost respiration rate and reduced the phytotoxic effect. Some parameter showed that livestock waste composting using co-additives during the 28 days results in waste that is safe to be discharged to the environment, however, the waste will not have matured yet to be utilized as compost fertilizer. Studies on the co-additive application on the pilot and large-scale composting are necessary to investigate the effectiveness and economic visibility of this method.

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CHAPTER 3 . EVALUATION OF DIFFERENT INTEGRATED VENTILATION-BIOFILTER SYSTEM FOR MITIGATING THE AIR POLLUTION FROM PIG HOUSE

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3.1. Abstract

The present study aimed to observe the air pollution emission performed by the integrated pig house-biofilter system. The performance of two different types of the integrated ventilation-biofilter system was investigated by experimental pig house. The wall ventilation system integrated into vertical biofilter (W-VBF), and the pit ventilation system integrated into horizontal biofilter (P-HBF) were tested. The experiment was conducted for nine weeks (64 days). Water recycling was used in the biofilter system. Microclimatic variables, aerial pollutant (particulate matter and odorant) and wastewater (slurry and biofilter recycled water) characteristic were observed during the experiment. Results showed that ammonia concentration inside the W-VBF was about 41% higher than that of the P-HBF. Noise from the W-VBF was about 12.1% more than that of P-HBF. Particulate matter and Total suspended particle inside the W-

VBF house was about 62.2% and 69.9% respectively higher than that of P-HBF. It was concluded that the P-HBF emitted lower air pollution rather than W-VBF. In addition, it could maintain the optimum temperature condition for swine house during cold condition. The design of an integrated ventilation-biofilter system for pig house might be used as the option to minimize the air pollution produced by the swine farming activity as well as maintain optimum microclimate condition for the pig.

Keywords: air pollution, ammonia, odor, ventilation, biofilter, pig house

3.2. Introduction

High density and intensive confinement livestock operation have increased drastically to supply the higher demand for meat products in the last few decades (Hoff and Harmon, 2006; Nimmermark, 2011). Air pollution problems arose as the number and size of confinement facilities increased. In the swine farm, the ventilation air is exhausted from the barn may contain concentrated odorous gases and particulate matter. The emission of odors and volatile organic compounds (VOCs) from swine livestock facilities are representative air pollution problems and are become a crucial public health concern (Yang et al., 2010). VOCs contribute the formation of photochemical smog, ozone and respirable suspended particulates in the atmosphere (Le Leuch et al., 2003). Particulate matter from pig houses contains micro-organisms and bio-aerosols which increase the prevalence of respiratory infections to people living in a neighboring area (Cai et al., 2006).

About 50% of odor emission from livestock production is contributed from livestock buildings (30%) and manure storage facilities (20%) (Mielcarek

and Rzeźnik, 2015). From piggeries, gases such as ammonia (NH_3), carbon dioxide (CO_2), methane (CH_4), and sulfur compounds (hydrogen sulfide and mercaptans) are the most abundant (Dumont et al., 2014). The importance of managing odorous compounds, VOC's and particulate matter from livestock facilities have recently been increasing. Effective methods of handling a wide range of airborne compounds are needed to reduce the negative environmental problems caused by the swine industry.

Various air pollution control technologies have been developed such as activated carbon adsorption, wet scrubbing, and masking agents (Chung et al., 2007). Biofiltration has been recognized as a reliable technology compared with other physicochemical technologies to control low and moderate-concentrations of waste gases containing odors and VOCs (Ryu et al., 2011; Estrada et al., 2012). Biofilters are used to reduce odor emissions from livestock facilities located to residential areas and the odor loads can be reduced about 40 – 83% (Martens et al., 2001). Biofilters need adequate air contact time to effectively reduce air pollutants (Sheridan, et al., 2002). In other words, the ventilation rate of livestock facilities can play an important role in the effective operation of biofilters. Zong et al., (2015) investigated the application of different ventilation system in the pig house to find out the cost-effective methods for exhaust air purification from pig buildings.

However, there is a little study on pig housing that incorporates a ventilation system and biofilters to reduce the emission of air pollutants in pig houses. Therefore, the purpose of this study was to assess the effects of two types of integrated ventilation and biofilter system (i.e., W-VBF, wall

ventilation system integrated into vertical biofilter; P-HBF, pit ventilation system integrated into horizontal biofilter) on odor emissions, air environments, and microclimate conditions.

3.3. Materials and Methods

3.3.1. Experimental pig house

Two trials were carried out in experimental pig houses that were designed to facilitate tests of various biofilters incorporated with ventilation systems. The experiment was conducted for nine weeks (64 days) in the autumn – early winter season (October – December 2017). The pig house research facilities consist of 9 fattening pigs placed in two barns (3 m wide x 4 m long, i.e., 1.3 m²/pig), facilitated with the different ventilation system and attached to the different biofilter system (Figure 3.1). The biofilter pads were made of cellulose. Recycling water was applied to moisturize the biofilter pad during the operation. Pigs were fed on an ad libitum basis (unrestricted ration). The pigs also have unrestricted access to water. Ratios of crude proteins contained in the food intake are 16% and 14.5% during growing and finishing periods, respectively. The floor of the piggery is fully-slatted with manure storage on underneath.

The air ventilation inlet of W-VBF and P-HBF was installed in the ceiling. In the W-VBF system, air flows into the building above the slatted floor and going into the casing on which the fan was placed on the wall of the building. In the P-HBF pig house, air flows from ceiling to the slatted floor before going into the outlet duct placed on the stored manure. The exhausted fan is placed outside the building through the outlet duct. The flow rate is

adapted to keep a constant temperature in the building (23 °C) independently of the external weather conditions, with the highest flow at 60 m³/pig·h (Dumont et al., 2014). Ventilation rates through the exhaust channels were automatically controlled according to the set room temperature (Veng System A/S, Roslev, Denmark). Description of each experimental pig house-biofilter type is presented in Table 3.1.

Table 3.1. The experimental ventilation-biofilter system type used in the present study

Experimental house type	Description
W-VBF	Wall ventilation – vertical biofilter
P-HBF	Pit ventilation – underground horizontal biofilter

3.3.2. Microclimatic variables measurement and odorant analysis

Indoor (room and pit) and outdoor air temperature and humidity were observed. A hygrothermograph was used to measure air temperature and relative humidity (SK-110TRH, SATO, Tokyo, Japan). Particulate matter (PM) was analyzed using an aerosol mass monitor (GT-331, SIBATA, Soka-city, Japan) and the mass concentrations of PM₁₀ (PM average aerodynamic diameter: 10 µm), PM₇ (PM average aerodynamic diameter: 7 µm), PM_{2.5} (PM mean aerodynamic diameter: 2.5 µm), PM₁ (PM mean aerodynamic diameter: 1 µm), and total suspended particles were obtained simultaneously, with a flow rate of 2.83 L/min.

A total of 11 different types of VOCs were analyzed in this study according to (Kumari et al., 2016), which included two sulfuric compounds (dimethyl sulfide [DMS] and dimethyl disulfide [DMDS]); six volatile fatty acids (VFA) odor (acetic acid, propionic acid, butyric acid, iso-butyric acid,

valeric acid and isovaleric acid); two indolics (indole and skatole); and one phenolic (p-cresol) (Indolic-Phenolic odor). The air was sampled for 5 min using a 1 L Tedlar bag (No.22053, Restek, Bellefonte, PA, USA). The collected air samples were analyzed using gas chromatography/mass spectrometry (GC/MS) (Agilent GC6890N/5975C MS, Youngin, Korea) in the laboratory, and all samples were analyzed within 24 hours of sample collection. Gastec tube (Gastec Co., Ltd., Kanagawa, Japan) were used to measure the concentrations of ammonia (NH_3).

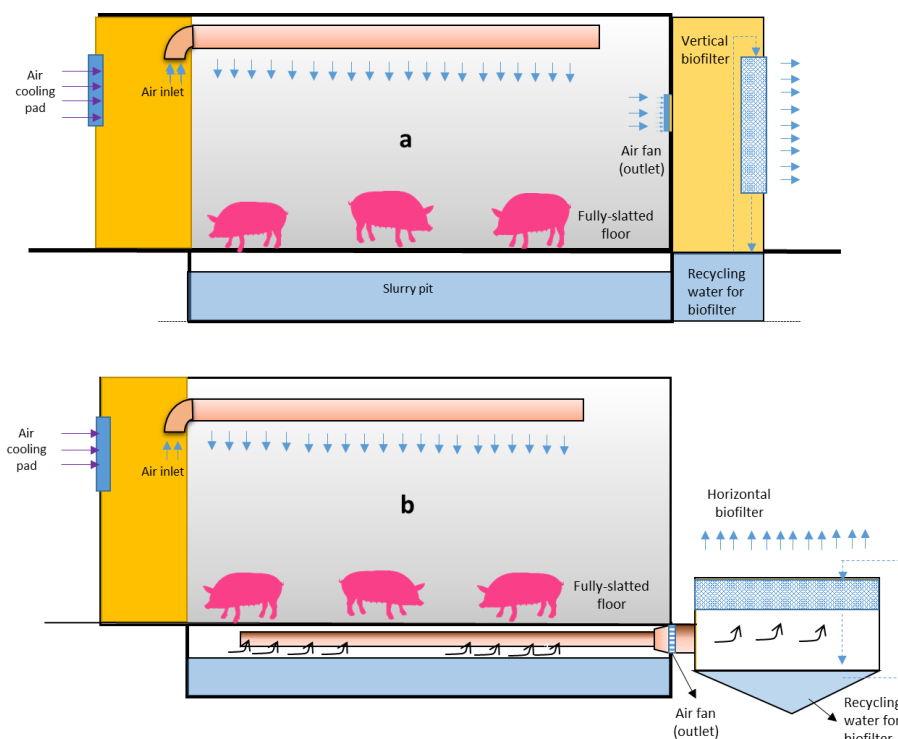


Figure 3.1. The experimental pig house (a) The W-VBF (wall ventilation-vertical biofilter); (b) P-HBF (pit ventilation-horizontal biofilter)

The differences of NH_3 concentrations across the biofilter were used to calculate the biofilter NH_3 removal efficiency as described by Ashtari et al.,

(2016). Equation (3.1) was used for calculating NH₃ removal efficiencies (RE) based on inlet NH₃ concentrations (ppmv) (C₁) and exhaust NH₃ concentrations (C₂).

$$RE = \left(\frac{C_1 - C_2}{C_1} \right) \times 100 \quad \text{Eq. (3.1)}$$

3.3.3. Wastewater analysis

The slurry samples characteristics of each pig house were monitored weekly for pH, Electrical Conductivity (EC), Dissolved Oxygen (DO) and Oxidation-reduction potential (ORP). The pH and electrical conductivity (EC) were determined using a pH meter (Inolab, WTW, GmbH, Weilheim, Germany) and an EC214 conductivity meter (Hanna Instruments, Ltd., Sarmeola di aarubano, Italy) respectively. The DO and ORP were measured using DO meter (Hanna instruments) and ORP meter (Hanna instruments). Similar parameters were also used to monitor the recycling water for biofilter. Additionally, NH₄⁺, NO₃⁻ and NO₂⁻ were analyzed using DR 5000 UV-Vis spectrophotometry (Hach Co., Loveland, CO, USA).

3.3.4. Statistical analysis

Statistical evaluations were carried out using an SPSS software package (SPSS Inc., Chicago, IL, USA). Comparisons of each variable were first performed with ANOVA at a 5% significance level. The average values of the quantitative factors under evaluation were compared by means of Tukey's test.

3.4. Results and Discussion

3.4.1. House microclimate condition and ammonia concentration

The average temperature and relative humidity of the W-VBF house during the experiment are shown in Figure 3.2. The average environmental temperature during the experiment was 20.1°C in the first two weeks and gradually decreased to 16.5 °C in the fourth week of the experiment. The environmental temperature continued to decrease to the average of 6 °C in the last two weeks of the experiment (Figure 3.2a).

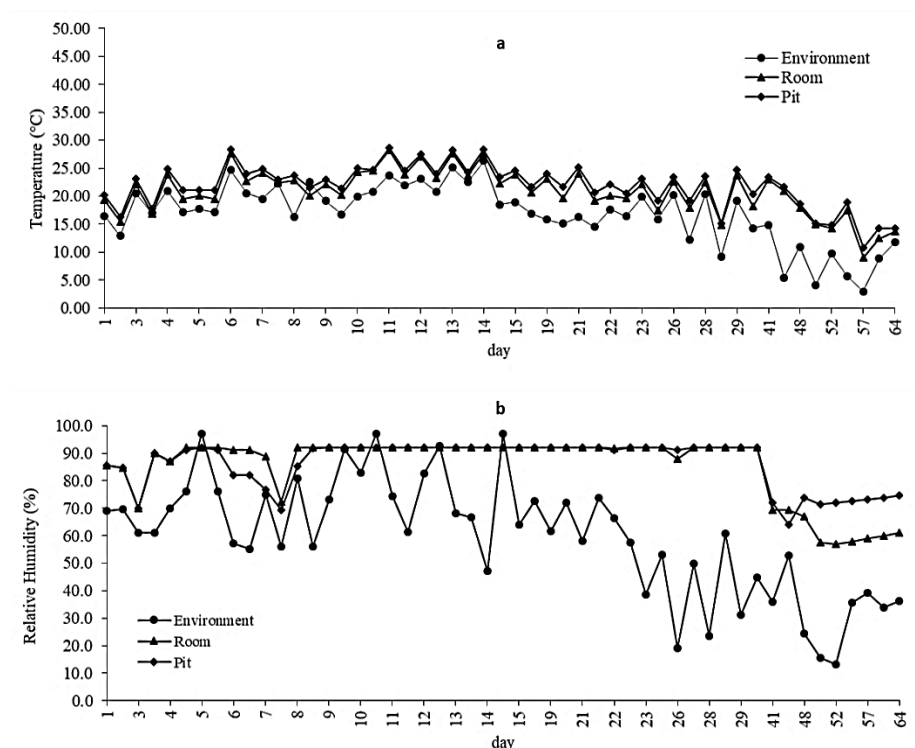


Figure 3.2. The average temperature, relative humidity in the wall ventilation-vertical biofilter (W-VBF) pilot pig house during the experiment (a) temperature; (b) humidity

The temperature in the room and pit inside the W-VBF house during the experiment is presented in Figure 3.2a. The average temperature in the room area of the W-VBF during the experiment was 22.7 °C in the first two weeks and slightly decreased to 20.6 °C in the fourth week of the experiment. The W-VBF's room area temperature decrease to the average of 18.4°C in the latest two weeks of the experiment (Figure 3.2a). The average temperature in the pit area of the W-VBF during the experiment was 23.6 °C in the first two weeks and slightly decreased to 21.8°C in the fourth week of the experiment. The W-VBF's pit area temperature decrease to the average of 19.2 °C in the latest two weeks of the experiment (Figure 3.2a).

During the experiment, there was a significant difference ($p < 0.05$) temperature between environment and inside the W-VBF house. The average temperature of the room area of the W-VBF was about 3.9 °C higher than the outside environment temperature. The temperature difference between room and pit area of the W-VBF was about 1 °C, higher in the pit area than that of room area. The higher temperature in the pit area was likely because of the air flow pattern affected by the ventilation fan installed in the wall. The turbulence air flow probably mostly occurred in the room area. Thus the air exchange mostly only occurs in the room rather than that in the pit.

The average environmental relative humidity during the experiment was 73 % in the first two weeks of the experiment and gradually decreased to 36.2 % in the last two weeks of the experiment (Figure 3.2a). The average relative humidity in the room area of W-VBF was 89.4 % during the first two weeks of

the experiment then slightly increased to the average of 91.5 % in the fourth weeks of the experiment. The average W-VBF's room area relative humidity gradually decreased to 72.6 % during the latest week of the experiment. The average relative humidity in the pit area of W-VBF was 87.8 % during the first two weeks of the experiment then slightly increased to the average of 91.8 % in the fourth weeks of the experiment. The average relative humidity in the pit area of W-VBF gradually decreased to 78 % during the latest week of the experiment (Figure 3.2b). The lower relative humidity inside the W-VBF room area during the latest period of experiment could be in related to lower relative humidity of the inlet air. In addition, the water vapor production inside the pilot pig house could possibly lower since the effect of lower environmental temperature might decrease the evaporation rate from the pig slurry in the pit.

During the experiment, there was a difference relative humidity between the outside environment and the inside of W-VBF pilot pig house. The relative humidity difference in the room area of the W-VBF pilot pig house was significantly higher ($p<0.05$) about the average of 25.9 % than the relative humidity in the outside environment. The average relative humidity in the pit area of W-VBF was significantly higher ($p<0.05$) than that in the room area during the latest week of the experimental period. This condition could be related to the lower ventilation rate provided. The lower ventilation rate occurred to maintain the designated room temperature (18 °C – 23 °C). By the lower ventilation rate, the carrying capacity of the air to carry out the water vapor was decreased.

The average temperature and relative humidity of the P-HBF pilot pig house during the experiment are shown in Figure 3.3. The temperature in the room and pit inside the W-VBF house during the experiment are presented in Figure 3.3a. The average temperature in the room area of the P-HBF during the experiment was 23.3 °C in the first two weeks and slightly decreased to 21.4 °C in the fourth week of the experiment. The P-HBF's room area temperature decreased to the average of 20.6°C in the latest two weeks of the experiment (Figure 3.3a). The average temperature in the pit area of the P-HBF during the experiment was 24.09 °C in the first two weeks and slightly decreased to 22.7°C in the fourth week of the experiment. The P-HBF's pit area temperature decreased to the average of 21.5 °C in the latest two weeks of the experiment (Figure 3.3a).

The temperature of the room area of the P-HBF was about 4.9°C higher than the temperature of the outside environment. The temperature difference between room and pit area (0.9 °C) of the W-VBF was not significantly different ($p>0.05$). The similar temperature condition between room and pit area could be due to the ventilation system applied in the P-HBF house. In the P-HBF house, the air from the room was coming out through the outlet duct installed in the pit surface by mechanically ventilated using negative air pressure ventilation system. By flowing the air through the pit before exhausted, the air is much distributed evenly in the room and pit area.

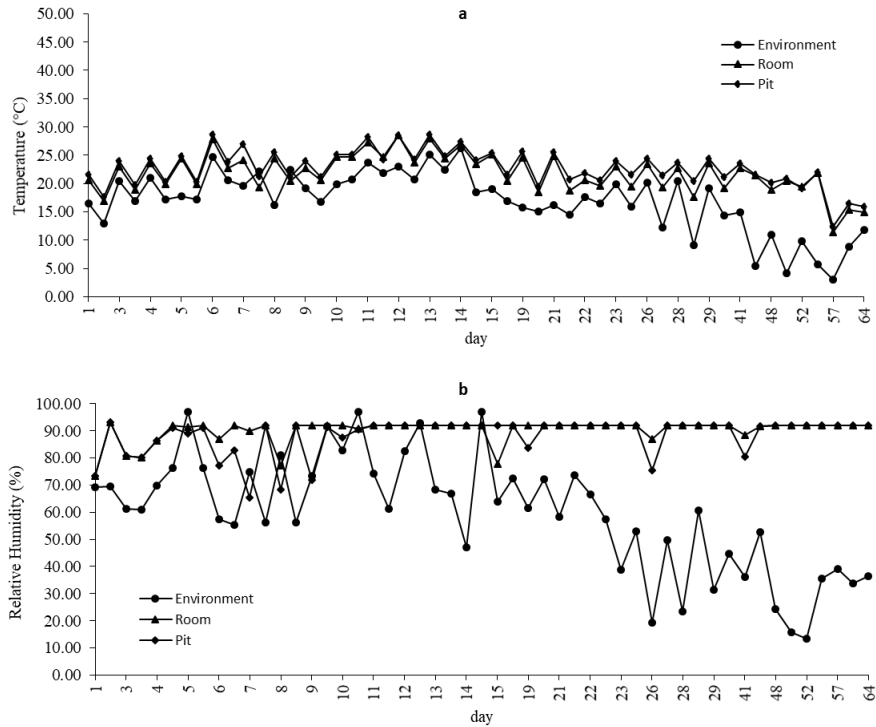


Figure 3.3. The average temperature, relative humidity in the P-HBF pilot pig house during the experiment (a) temperature; (b) humidity

The average relative humidity in the room area of P-HBF was 94.3 % during the first two weeks of the experiment then slightly increased to the average of 96.4 % in the fourth weeks of the experiment. The average P-HBF's room area relative humidity stays constant in the average of 95.6 % during the latest week of the experiment. The average relative humidity in the pit area of P-HBF was 91.1 % during the first two weeks of the experiment then slightly increased to the average of 95.2 % in the fourth weeks of the experiment. The average relative humidity in the pit area of P-HBF stays constant in the average of 95.7 % during the latest week of the experiment (Figure 3.3b). The relative humidity in the room and pit area of P-HBF were not significantly different ($p>0.05$) during the experiment. However, the difference of relative humidity

between inside the room of P-HBF house and outside environment was getting higher in line with the experimental period. The relative humidity inside the room area was constant while the outside relative humidity decreased. This condition was likely due to the insufficient ventilation rate to carry out the water vapor inside the pig house.

The W-VBF and P-HBF house have similar pattern regarding the house temperature condition during fall to winter. However, the higher temperature during cold season can be maintained better in P-HBF pilot pig house rather than W-VBF pilot pig house. Temperature condition in W-VBF house dropped much more than that of P-HBF house despite the inside room temperature was set at 23 °C to regulate ventilation rate. It means in the W-VBF house, the set temperature cannot be maintained longer despite the ventilation rate has been reached to the minimum. Thus the supplemental heat was required earlier in the W-VBF for keeping the designated temperature. According to the resulted, it demonstrated that pit ventilation system was better to maintain house temperature condition during the cold season under the minimum ventilation rate. This condition could be due to slower heat loss released from inside the P-HBF house to the outside environment. The temperature in the pit was observed comparatively similar than that of room temperature either in W-VBF and P-HBF housing type.

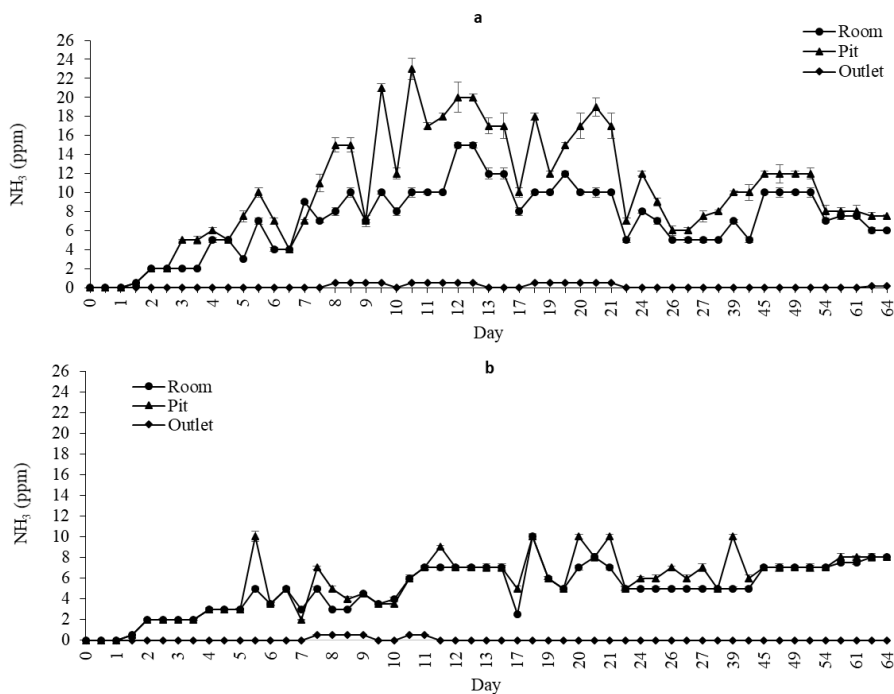


Figure 3.4. The average ammonia concentration in the pilot pig house during the experiment (a) wall ventilation-vertical biofilter (W-VBF) house; (b) pit ventilation-horizontal biofilter (P-HBF). Values are expressed as mean \pm standard deviation

The W-VBF and P-HBF house have different pattern regarding the house relative humidity during late fall to early winter. The relative humidity in the P-HBF house comparatively higher than that of the W-VBF house. Wall ventilation system was better to maintain ideal house humidity for pig growth, however in extremely cold weather condition, there was no difference between wall and pit ventilation on the humidity. Relative humidity observed in this experiment reached 90% which was higher than optimal range of relative humidity for pig growth comfort area of about 50 – 70% (Schauberger et al., 2000) and the disparities between environment and room relative humidity was also high. The possible explanation of this condition is likely because of the cooling evaporation unit installed before the air duct inlet. The air passing the

evaporative cooling unit may bring some water vapor, thus increased the air relative humidity before coming into the pig house. Water vapor production from pig activities also contributed the high relative humidity inside the pig house (Lu et al., 2017).

The measured ammonia concentration results during the experiment are shown in Figure 3.4. In the W-VBF pig house, ammonia concentration in the pit area was higher than that of the room during the experiment. The average ammonia concentration in the room, pit, and outlet of the W-VBF house were 7.09 ppm, 10.27 ppm, and 0.15 ppm respectively (Figure 3.4a). In the P-HBF pig house, ammonia concentration in pit and room are comparatively similar. The average ammonia concentration in the room, pit and outside of the P-HBF pig house were 4.93 ppm, 5.56 ppm, and 0.06 ppm respectively (Figure 3.4b). Generally, ammonia concentration inside the W-VBF pig house was higher than that of the P-HBF pig house. Thus it was demonstrated that different ventilation method resulted in the different ammonia emission. The higher ammonia concentration in the pig house with wall ventilation could be due to the airflow pattern inside the pig house affected by the ventilation type. Zong et al., (2015b) found that a big turbulence flow was found in the system with wall ventilation. Furthermore, it was also explained that a partial pit ventilation system (PPV) had been proved capable to significantly improve indoor air quality and reduce ammonia emission if combined with an air purification systems. The ammonia concentration from either W-VBF or P-HBF pig house decreased after passing the biofilter system to the level under 0.5 ppm. This ammonia concentration value was lower than the permitted ammonia emission

from industrial area regulated by the South Korea government of about 1 ppm (Park, 2005).

3.4.2. Effect on volatile organic odor, particulate matter, and noise level

All volatile organic compounds were detected with the exception of valeric acid, P-cresol, and skatole (Table 3.2). According to the results, among the volatile fatty acid compounds, Propionic acid and Butyric acid showed different between W-VBF and P-HBF pig house during the experiment. Propionic acid and Butyric acid concentration in the house with pit ventilation (8.6 ppbv and 3.9 ppbv respectively) were significantly higher ($p<0.05$) than that of the house with wall ventilation (4.6 ppbv and 0.8 ppbv respectively). After biofilter system, the concentration of Propionic acid between W-VBF and P-HBF were not significantly different ($p>0.05$) whereas Butyric acid showed higher ($p<0.05$) in the outlet of W-VBF (5.2 ppbv) with vertical biofilter than that of P-HBF pig house (1.6 ppbv). Results show that different house and biofilter type led to different VFA odor compounds.

The average Dimethyl disulfide and Dimethyl trisulfide concentration observed during the experiment were not significantly different ($p>0.05$) between W-VBF (6.7 ppbv) and P-HBF (6.4 ppbv). However, after the biofilter system, dimethyl disulfide of P-HBF (3.3 ppbv) showed significantly higher ($p<0.05$) than that of W-VBF (0.3 ppbv). The higher concentration of Dimethyl disulfide and Dimethyl trisulfide concentration after the horizontal biofilter showed could be due to the pit ventilation system application and the contribution from the recycling water used for the biofilter. The higher biomass particulate matter containing sulfuric odor compound from the inside of the pig

house possibly came out throughout the pit ventilation duct. Also, a limited oxygen concentration may cause biological reactions that produce sulfuric odor compounds during the storage of wastewater.

Table 3.2. The odor and particulate matter concentration in the experimental pig house-biofilter system

Parameter	W-VBF		P-HBF	
	inside	outlet	inside	outlet
Odorant (ppbv)	----- ppbv -----			
Acetic acid (AA)	31.8±11.2	29.2±9.3	31.7±11.7	30.9±5.3
Propanoic acid (PA)	4.6±1.3 ^a	5.2±1.9 ^{ab}	8.6±3.3 ^b	8.0±3.2 ^{ab}
Isobutyric acid (I-BA)	1.4±0.3	1.6±0.3	1.5±0.2	1.3±0.2
Butyric acid (BA)	0.8±0.3 ^a	5.2±0.6 ^c	3.9±1.4 ^b	1.6±0.3 ^a
Isovaleric acid (I-VA)	3.6±0.3	3.6±0.6	3.6±0.4	3.6±0.4
Valeric acid (VA)	ND	ND	ND	ND
P-Cresol	ND	ND	ND	ND
Indole	1.6±0.1	2.2±0.9	1.7±0.3	1.6±0.4
Skatole	ND	ND	ND	ND
Dimethyl sulfide (DMS)	6.7±2.5 ^c	0.3±0.1 ^a	6.4±1.2 ^c	3.3±1.2 ^b
Dimethyl disulfide (DMDS)	0.4±0.1 ^{ab}	0.3±0.0 ^a	0.6±0.3 ^b	0.5±0.2 ^b
Particulate matter (PM)	----- µg/m ³ -----			
PM10	203.1±30.4 ^c	33.0±25.0 ^a	122.8.0±36.8 ^b	32.5±7.6 ^a
PM7	114.3±41.0 ^c	25.0±12.9 ^a	68.3±15.3 ^b	26.1±11.4 ^a
PM2.5	13.7±2.7	9.3±5.4	11.7±1.8	10.4±7.2
PM1	6.5±2.1	5.6±3.2	5.3±1.2	6.1±4.5
Total Suspended Particle (TSP)	443.3±79.7 ^c	59.2±45.1 ^a	260.9±98.3 ^b	54.6±21.8 ^a

Values are expressed as mean ± standard deviation

Data with different letters in the same row are significantly different (p -value < 0.05).

ND represents not detected.

The pig house with pit ventilation system showed significantly lower ($p < 0.05$) particulate matter PM10 (122.8 µg/m³) and PM7 (68.3 µg/m³) in comparison with that of wall ventilation system house (Table 3.2), however after passing the biofilter system, they were not significantly different ($p > 0.05$). The TSP of P-HBF house (260.9 µg/m³) was significantly lower ($p < 0.05$) than

that of W-VBF house (443.3 $\mu\text{g}/\text{m}^3$). The higher PM and TSP in the pig house with wall ventilation system occurred could be due to higher turbulence intensity as explained by Zong et al., (2015b). These results show that the utilization of a pit ventilation system in the pig house might increase the air quality inside the house. There was no significant effect on particulate matter emission after biofilter between both types of biofilter systems.

Table 3.3. Pearson correlation of measured odorant and microenvironment variables in the present study

Pearson correlation	Temperature	Humidity	PM10	PM7	PM2.5	PM1	TSP
Ammonia	0.4	0.55**	0.54**	0.50*	0.18	0.21	0.56**
AA	-0.06	0.15	-0.16	-0.21	-0.06	0.10	-0.16
PA	0.23	-0.22	-0.35	-0.34	0.07	0.15	-0.34
IBA	0.59**	-0.20	0.03	-0.08	0.00	0.04	0.10
BA	0.05	-0.11	-0.41*	-0.39	-0.09	-0.01	-0.39
IVA	0.13	-0.02	0.14	0.16	0.07	0.09	0.15
Indole	0.15	0.09	-0.14	-0.07	0.26	0.39	-0.17
DMS	0.32	0.59**	0.66**	0.55**	0.28	0.09	0.63**
DMDS	0.51*	0.10	0.31	0.28	0.24	0.28	0.30

*correlation is significant at the 0.01 level

**correlation is significant at the 0.05 level

The Pearson correlation of measured odorant and microenvironment variables in the present study is presented in Table 3.3. According to the results in this study, the ammonia had significant correlation with the humidity ($r = 0.55$), PM10 ($r = 0.54$), PM7 ($r = 0.50$) and TSP ($r = 0.56$). The Iso-butyric acid odor (IBA) had significant correlation with temperature ($r = 0.59$). The butyric acid odor had significant negative correlation with PM10 ($r = 0.41$). The DMS odor had significant correlation with the humidity ($r = 0.59$), PM10 ($r = 0.66$),

PM7 ($r = 0.55$) and TSP ($r = 0.63$). The DMDS had significant correlation with temperature ($r = 0.51$).

Table 3.4. Pearson correlation of odorant variables in the present study

Pearson correlation	Ammonia	AA	PA	IBA	BA	IVA	Indole	DMS	DMDS
Ammonia	1	0.39	-0.03	0.19	-0.03	0.02	-0.07	0.54**	0.57**
AA	0.39	1	0.41*	-0.12	0.13	-0.37	-0.18	0.13	0.22
PA	-0.03	0.41*	1	-0.14	0.16	-0.30	-0.08	-0.03	0.37
IBA	0.19	-0.12	-0.14	1	0.36	0.35	0.28	-0.22	0.08
BA	-0.03	0.13	0.16	0.36	1	0.02	0.48*	-0.41*	-0.32
IVA	0.02	-0.37	-0.30	0.35	0.03	1	0.36	0.02	-0.08
Indole	-0.07	-0.18	-0.08	0.28	0.49*	0.36	1	-0.26	-0.22
DMS	0.54**	0.13	-0.03	-0.22	-0.41*	0.02	-0.26	1	0.47*
DMDS	0.57**	0.22	0.37	0.08	-0.32	-0.08	-0.22	0.47*	1

*correlation is significant at the 0.01 level

**correlation is significant at the 0.05 level

The Pearson correlation of odorant variables in the present study is presented in Table 3.4. The ammonia odor had a significant correlation with DMS ($r = 0.54$) odor and DMDS odor ($r = 0.57$). The acetic acid odor had a significant correlation with propionic acid odor ($r = 0.41$). The butyric acid odor a significant correlation with indole ($r = 0.48$) and had a significantly negative correlation with DMS odor ($r = -0.41$). Ammonia odor had a significant correlation with DMS ($r = 0.54$) and DMDS ($r = 0.57$).

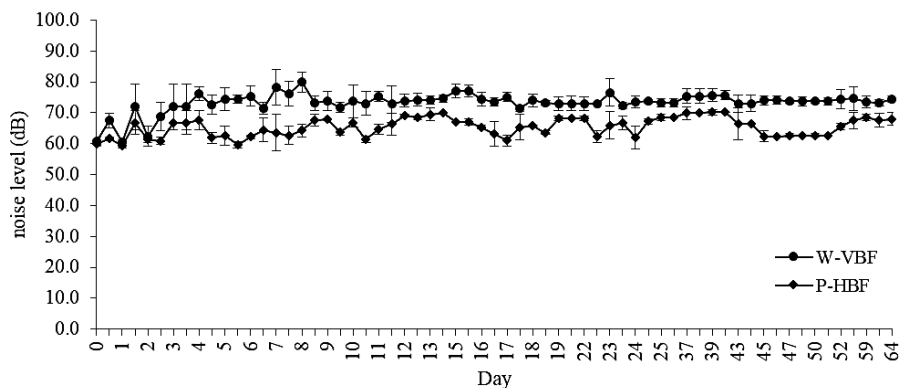


Figure 3.5. The average of the noise level in the wall ventilation-vertical biofilter (W-VBF) and pit ventilation-horizontal biofilter (P-HBF) pilot pig house during the experiment. Values are expressed as mean \pm standard deviation.

The average of the noise level in the W-VBF and P-HBF of experimental pig house during the experiment are presented in Figure 3.5. The Average noise level inside the W-VBF pig house (73.28 dB) was significantly higher ($p < 0.05$) than that of the P-HBF pig house (65.37 dB), it shows that the wall ventilation house was noisier of about 12.10 % than that of pit ventilation house. Average noise level difference between wall ventilation and pit ventilation was around 7.92 dB. The higher noise in the pig house with wall ventilation system is likely because the contribution from the sound coming from the exhaust fan operation.

3.4.3. Biofilter condition and performance

Biofilter pad temperature of the W-VBF and B pig house during the experiment is presented in Figure 3.4. Biofilter pad temperature in the W-VBF and the P-HBF was gradually decreased in line with the environment temperature. In the warm environmental conditions above 14°C, the temperature of the biofilter pad in the W-VBF was higher than that of P-HBF. Under cool temperatures below 14°C, the temperature of the biofilter pad in the

W-VBF was found to be lower than that of the P-HBF. This fact indicates that horizontal biofilters are capable of responding to changes in ambient temperature and provide much suitable microbial growth environment.

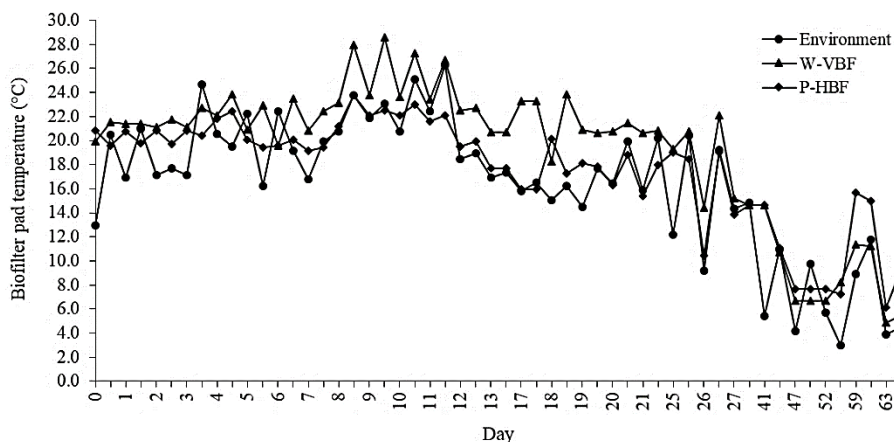


Figure 3.6. Biofilter pad temperature of the wall ventilation-vertical biofilter (W-VBF) and pit ventilation-horizontal biofilter (P-HBF) pilot pig house during the experiment

The average of ammonia reduction efficiency of the biofilter in W-VBF and P-HBF pig house during the experiment are shown in Figure 3.5. Ammonia reduction efficiency of W-VBF and P-HBF were not significantly different. Average ammonia reduction efficiency of W-VBF (98.45%) and P-HBF (98.68%). According to our observation results, the different type of biofilter showed no significant effect ($p>0.05$) to the ammonia reduction efficiency. Until the end of the experiment (pig harvesting), the performance of both biofilter systems was still on the average above 95%. It can be inferred that regeneration of the biofilter system every production period was not required.

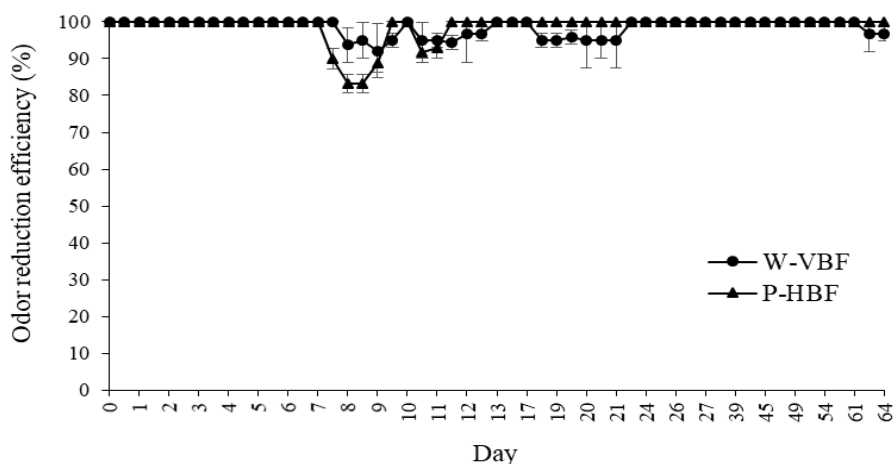


Figure 3.7. Average ammonia reduction efficiency of the biofilter in the wall ventilation-vertical biofilter (W-VBF) and pit ventilation-horizontal biofilter (P-HBF) pilot pig house during the experiment. Values are expressed as mean \pm standard deviation

3.4.4. Effect on Biofilter recycled water characteristic

About 200 L of water is circulated to the biofilter pad during the experiment. The chemical characteristics of recycling water are presented in Figure 3.6. The pH of the recycling water of W-VBF biofilter showed higher than that of P-HBF throughout the experiment period (Figure 3.6a). The final pH level of W-VBF biofilter (6.65) was significantly higher than that of P-HBF (5.70). The different pH of recycling water was likely because of the difference of recycling water storage condition. In the P-HBF, the recycling water was placed underneath the biofilter pad, which might provide lower oxygen penetration in comparison to the water recycling storage in W-VBF pig house. During the lower oxygen penetration or anaerobic condition, acetic, propionic and butyric acids are produced in the liquid. For instance, in slurries with near neutral pH, the production of these acids results in the release of H^+ into the medium causing the slurry pH to decrease (Paul and Beauchamp, 1989).

Another possible explanation of lower pH of recycling water in the P-HBF pig house was due to the lower NH_4^+ concentration in comparison with the recycling water of biofilter in W-VBF pig house. In addition, higher nitrate concentration in the recycled water of biofilter in P-HBF pig house (Figure 3.6e) might correlate with lower pH. Dadgar and Payandeh (2017) reported that the increasing levels of nitrate in water, the pH levels decline.

The EC and nitrate of the P-HBF recycling water increased drastically rather than that of W-VBF recycling water during the experiment (Figure 3.6b and 3.6e). The EC of W-VBF recycling water reached 3.0 mS/cm, whereas the EC value of P-HBF recycling water reached 6.0. The higher EC value was likely because of the higher content of NH_4^+ and nitrate in the water. Suresh et al., (2009) reported the positive relationships were noticed between EC and ammonia nitrogen (NH_4^+). Gent (2003) and Miyamoto et al. (2015) reported the positive correlation between EC and nitrate in the solution as showed as well in this experiment.

The alteration of ORP, pH and DO value were comparatively similar between the recycling water of biofilter in W-VBF and P-HBF pig house during the experiment. The nitrite concentration of W-VBF biofilter recycling water increased rapidly after three weeks whereas nitrite concentration of P-HBF recycling water increased gradually since the first week. However, the final nitrite concentrations were comparatively similar between the recycling water of W-VBF biofilter and P-HBF.

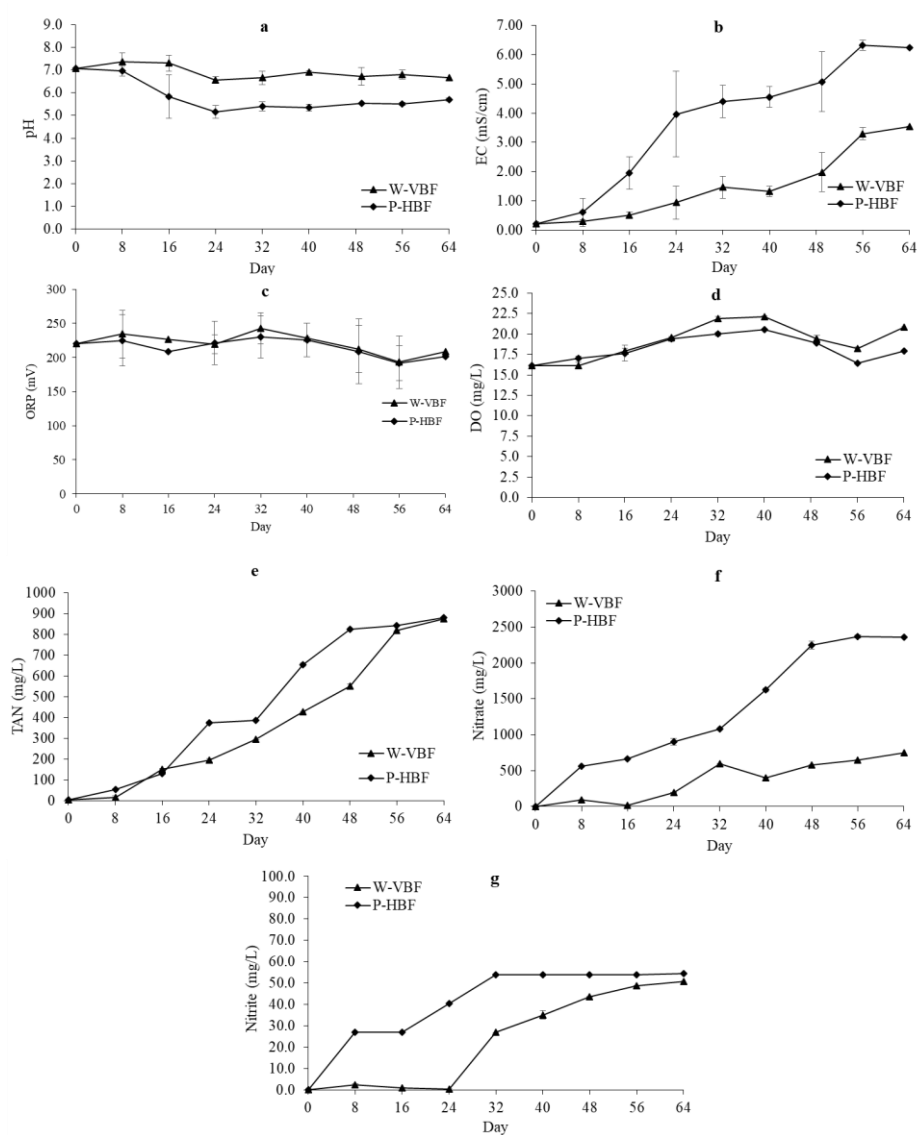


Figure 3.8. Physicochemical characteristics of recycled water for biofilter (a) pH; (b) Electrical conductivity (EC); (c) Oxidation Reduction Potential (ORP); (d) Dissolved Oxygen (DO); (e) Total Ammonia Nitrogen (TAN); (f) Nitrate; (g) Nitrite

The trend of Total Ammonia Nitrogen (TAN) concentrations in recycling water was comparatively similar between that in W-VBF and P-HBF, TAN concentration in recycling water increased gradually during the experiment (Figure 3.6e). The final TAN concentrations in recycling water of W-VBF and

P-HBF pig house biofilter were not significantly different ($p>0.05$). According to the TAN concentration results in this study, it was indicated that the different biofilter type didn't give a significant effect on the ammonia captured in the recycling water. At pH of aqueous solution less than 7.0, NH_4^+ is formed and dominant, for instance, the NH_4^+ and NH_3 concentrations in aqueous NH_3 solution at pH 6.0 and $t = 21^\circ\text{C}$ are about 99.96% and 0.04% respectively (Thurston et al., 1979) which is agreed with the results in this study

The higher NH_3 removal efficiencies would result in higher NH_4^+ in the recycled water solution. The fate of the NH_3 captured from the air depends on the pH of the recycled water solution. If the pH is low, then the most of the captured NH_3 is converted to ammonium ion NH_4^+ . When the pH is high, more of the captured NH_3 remains as NH_3 , so a portion of NH_3 may be released from the solution to the atmosphere (Ashtari et al., 2016).

3.4.5. Effect on the slurry characteristic

Figure 3.9 shows the physicochemical characteristics of the slurry produced during the experiment from the W-VBF and P-HBF pig house. The pH of the slurry from W-VBF and P-HBF were constant along with the period on the range of 6.95 to 7.25. EC of the slurry from W-VBF and P-HBF house increased in line with the growing time from initial 6.65 to 10.20 mS/cm on the final, was likely because of the accumulation of TAN concentration in the slurry. The final TAN concentration of slurry from P-HBF pig house was significantly higher ($p<0.05$) than that of slurry from W-VBF pig house. The higher TAN concentration of the slurry from P-HBF pig house could be due to the

ventilation type in P-HBF might induce the particulate matter and ammonia in the air penetrated to the slurry stronger than that of W-VBF.

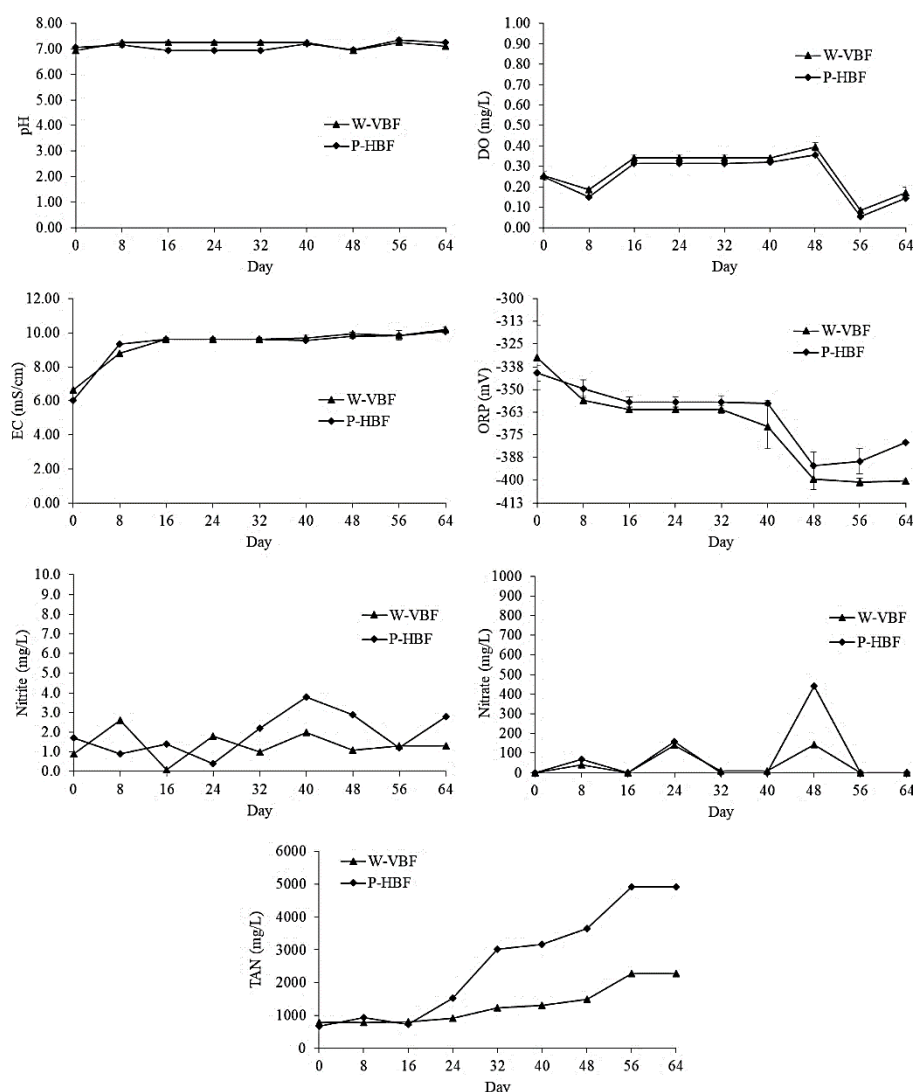


Figure 3.9. Physicochemical characteristics of the slurry produced in the wall ventilation-vertical biofilter (W-VBF) and pit ventilation-horizontal biofilter (P-HBF) pilot pig house during the experiment. Values are expressed as mean \pm standard deviation

It was indicated by the lower particulate matter in the P-HBF pig house room (Table 3.1) rather than W-VBF pig house and explanation for this condition

was the particulate matter might be dropped to the slurry in the pit of P-HBF pig house. The particulate matter brings some organic substance and induce a biological reaction in the slurry. The results of ORP in this study showed around -375 to -400 which indicated anaerobic zone occurred in the slurry, as shown by the low DO. Zhu et al., (2002) reported that when ORP reached about -125 mV, the aerobic counts almost approached zero and a large of anaerobes would coexist. Furthermore, Colmenarejo et al., (2004) characterized that hydrolysis occurred under the ORP value around -300 mV.

3.5. Conclusion

This present study evaluated the performance of two different models of integrated pig house-biofilter system on reducing the air pollutant emission. Results show that pit ventilation system integrated with the horizontal biofilter emitted lower air pollution rather than wall ventilation integrated to vertical biofilter system. The ammonia concentration inside the W-VBF was about 41% higher than that of the P-HBF. The particulate matter inside the W-VBF was about 62% higher than that of P-HBF. The total suspended particle in the room area correlated positively with the concentration of ammonia odor and volatile organic acid odorant. Noise from the W-VBF was about 12.1% more than that of P-HBF. Particulate matter and Total suspended particle inside the W-VBF house was about 62.2% and 69.9% respectively higher than that of P-HBF. The P-HBF application could maintain the optimum temperature condition better than that of W-VBF for swine house during the cold condition. According to the results, it was concluded that integration design of ventilation system and biofilter system of swine house might be used as the option to minimize the air

pollution produced by the swine farming activity as well as maintain optimum microclimate condition for the pig.

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CHAPTER 4 . THE EVALUATION OF CO-ADDITIVE (FGD GYPSUM AND BIOCHAR) SUPPLEMENTATION IN THE LIVESTOCK HOUSE AIR BIOFILTERING SYSTEM

This chapter will be published in SCIE Journal, as a partial fulfillment of Seoul National University Graduate School, Department of Agricultural Biotechnology, Animal Science and Biotechnology's Ph.D. program for Andi Febrisiantosa

4.1. Abstract

The present study aimed to investigate the odor emission and its treatment performed by the dual biofilter system supplemented by the co-additive (FGD gypsum and biochar) in the closed livestock confinement. The performance of dual biofilter system was investigated under experimental farm model. The eight weeks (56 days) of tested was performed on the experimental scale pig house. The dual (vertical and horizontal) biofilter system attached to the mechanical wall ventilation system was tested. Water recycling system was equipped in the biofilter system. Microclimatic variables, odor pollutant (ammonia and volatile organic acid), physicochemical characteristics of the biofilter pack and wastewater (biofilter recycled water) were observed during the experiment. The ammonia removal efficiency of the dual biofilter system supplemented by the co-additive (5% FGD gypsum and 5% biochar) was about 65.28% to 98%. The co-additive supplementation in the biofilter media might

increase the ammonia removal efficiency about 57.72% and potent to inhibit the emission of volatile organic odor from the pig house with the average removal efficiency of about 21.36%. The nitrification process, as well as microbial activities in the biofilter media, might be improved by co-additive supplementation. Also, since it contains higher nitrite nitrate concentration and the macro micro-nutrient (total P, K, Ca, Mg, S, and Fe) availability, the biofilter media might also further be considered as a solid bio-fertilizer product source for the land application. Results indicated that dual biofilter system supplemented by the FGD gypsum and biochar emitted lower air pollution rather than the biofilter without co-additive supplementation. It was concluded that integration of dual biofilter system in the closed livestock house might be used as the option to minimize the air pollution produced by the livestock farming activity.

Keywords: air pollution, ammonia, odor, biofilter, swine house

4.2. Introduction

Odor and gas emissions from livestock farming operations have become an important social problem due to their negative impacts on the local human health and quality of life (Radon et al., 2004). As livestock production has become more concentrated, neighbor complaints about odors from livestock operations have increased in number and became a drawback at the local level to grow livestock operations, associated with this, the odor-related complaints in areas where pig production facilities are more frequent and intensified (Kafle and Chen, 2014).

Concentrated animal feeding operations (CAFOs), including pig facilities, are sources of air pollutants, such as ammonia (NH_3), hydrogen sulfide (H_2S), carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), volatile organic compounds (VOCs), particulate matter (PM), and odor that affect the air quality (Mielcarek and Rzeźnik, 2015). Odors from livestock wastes have resulted in public annoying that prevent the livestock producers from developing new facilities for expanding their business and have decreased the effectiveness of farmland utilization (Wiles et al., 2001). The volatile fatty acids (VFAs), including acetic, propionic, isobutyric, butyric, isovaleric, and valeric acids, as well as aromatic compounds, including phenol, p-cresol, indole, and skatole have been identified as the major malodorous compounds in the livestock manure (Chen et al., 1994; Jo et al., 2015). Ammonia is primarily an onsite problem due to increased volatility during aerobic treatments though ammonia can be formed anaerobically and aerobically (Wiles et al., 2001).

The barns, feedlot surfaces, composting structures, manure storage, and treatment facilities, and land application areas are sources of odor emissions from the livestock sector industries (Hood et al., 2011; Kafle and Chen, 2014). Gases emissions such as ammonia (NH_3), carbon dioxide (CO_2), methane (CH_4), and sulfur compounds (hydrogen sulfide (H_2S) and mercaptans are the most abundant from piggeries (Dumont et al., 2014). In swine facilities, these emissions mainly originate from the decomposition of manure (Rappert and Müller, 2005; Martel et al., 2017). About 50% of odor emission from livestock production is contributed from livestock buildings (30%) and manure storage facilities (20%) (Mielcarek and Rzeźnik, 2015).

Quantification of ammonia (NH_3) emissions was always considered as an indicator of overall odor concentrations in the livestock environment (Phillips et al., 2001; Dennis et al., 2010). The ammonia is commonly present in the livestock environment. It has a relative higher odor threshold value of 0.3 – 53 ppm than other odorous compounds. However the contribution of ammonia to odor intensity is insignificant (Lacey et al., 2004). Despite the insignificant odor intensity, the emission of ammonia from animal houses can contribute significantly to eutrophication and acidification of the environmental and much effort is directed to quantify and reduce the emissions (Sommer et al., 2006; Nielsen et al., 2008).

Considerable efforts have already been made over the years to minimize production of malodorous and toxic gases from the livestock, for example in the swine farm facilities, by modifying proteins in pig diets (Kerr et al., 2006), introducing manure additives (Heber et al., 2000; McCrory and Hobbs, 2001; Banhazi et al., 2009), and employing various swine manure handling and management strategies (Zhang and Westerman, 1997; Ndegwa et al., 2002; Burton, 2007), however exhausted air abatement techniques are still necessary. Various air pollution control technologies have been developed such as activated carbon adsorption, wet scrubbing, and masking agents (Chung et al., 2007; Chen and Hoff, 2012). Among many technologies exist for odor control, bioreactors (e.g., biofilters, biotrickling filters, and bioscrubbers) have been found to be the most cost-effective treatment for high volume waste gases containing readily biodegradable contaminants in relatively low concentrations,

such as those emitted from farm facilities (Raboni and Torretta, 2016; Liu et al., 2017; Kafle et al., 2015; Martel et al., 2017).

Biofiltration can be an efficient method to reduce ammonia and odor emission from the animal house (Nielsen et al., 2008). Biofiltration has also been recognized as a reliable technology compared with other physicochemical technologies to control low and moderate-concentrations of waste gases containing odors and VOCs (Ryu et al., 2011; Estrada et al., 2012). Biofilters are used to reduce odor emissions from livestock facilities located to residential areas, and the odor loads can be reduced about 40 – 83% (Martens et al., 2001). Zong et al., (2015) investigated the application of different ventilation system in the pig house to find out the cost-effective methods for exhaust air purification from pig buildings. The previous chapter in this dissertation investigated the performance of the integrated pig house-biofilter system to reduce ammonia and VOA odor emitted from swine production. It was suggested that a combination of horizontal and vertical biofilter might result in the optimal odor removal efficiency as well as maintain the optimal inside pig barn comfort zone during the extreme external season.

The effectiveness of co-additives for the reduction of air pollution and conservation of the nutrients in the livestock waste composting has been investigated (Febrisiantosa et al., 2018). Biochar and Flue gas desulphurization gypsum (FGD gypsum) were used to supplement the composting of a mixture of slaughter waste, swine slurry, and sawdust. The results indicated that the use of the co-additive (5% Biochar and 5% FGD gypsum) during composting of livestock waste led to a reduction of ammonia volatilization by 26–59% and to

a 6.7–7.9-fold increase of nitrate accumulation, hence the co-additives (biochar and FGD gypsum) can be utilized in livestock waste composting to reduce ammonia volatilization and improve nutrient conservation (Febrisiantosa et al., 2018). The utilization of the co-additive for improving the performance of the biofilter system integrated with the animal barn could be explored. Accordingly, the present study aimed to observe the performance of dual biofiltering system supplemented by the co-additive FGD-gypsum and biochar in the integrated house-biofilter system under the experimental pilot scale.

4.3. Material and Methods

4.3.1. Experimental house-biofilter facilities

The experiment was performed on the experimental scale pig house. The wall ventilation system integrated with the vertical biofilter system and the horizontal biofilter system was utilized. The pilot pig houses were mechanically ventilated under a negative pressure system and were equipped with an air inlet duct, an exhaust fan, and a temperature-humidity probe. Fresh air from outside the building passed through a heating/cooling system before entering the pilot pig house through the air inlet. The pilot pig house (3.0 m wide \times 4.0 m long \times 2.0 m high) housed nine growers/finisher pigs (Figure 4.1). The pigs were raised from 25 to 110 kg and assigned in such a manner that the chambers contained similar average weights. The air temperatures in the chambers were initially maintained at a set point of 28°C and then gradually decreased to 22°C following the pigs growth. The animals had free access to feed and water 24 h a day (Martel et al., 2017). The ventilation air flow rate is adapted to keep a constant temperature in the building (23 °C) independently of the external

weather conditions, with the highest flow at $60 \text{ m}^3/\text{pig}\cdot\text{h}$, i.e., $600 \text{ m}^3/\text{h}$ (Dumont et al., 2014). Ventilation rates through the exhaust channels were automatically controlled according to the set room temperature (Veng System A/S, Roslev, Denmark).

The biofilter chamber was made of galvanized steel with internal dimensions of $1.1 \text{ m} \times 1.5 \text{ m} \times 2.0 \text{ m}$. The dual biofilter system was constructed inside the biofilter chamber. It consists of two-step biofiltration (combination of vertical and horizontal type biofiltration). The unit was equipped with a water recirculation system with a sump holding a total water volume of 130 L. In the first step of biofiltration, the polluted air was filtered by the vertical biofilter pad equipped with the recycled water (Figure 4.2). After passing the first biofiltration, the air coming out and pass the second biofiltration system. The second biofiltration system consists of a three-layer horizontal biofiltration module. Each layer composted by moisturized organic material (sawdust) add with the co-additive material (FGD gypsum 5% w/w and Biochar 5% w/w) to increase odor absorption ability. The air was flowed by zigzag direction between each layer which aimed to increase empty bed contact time as well as to reduce the pressure drop commonly occur in the conventional biofilter pack structure.

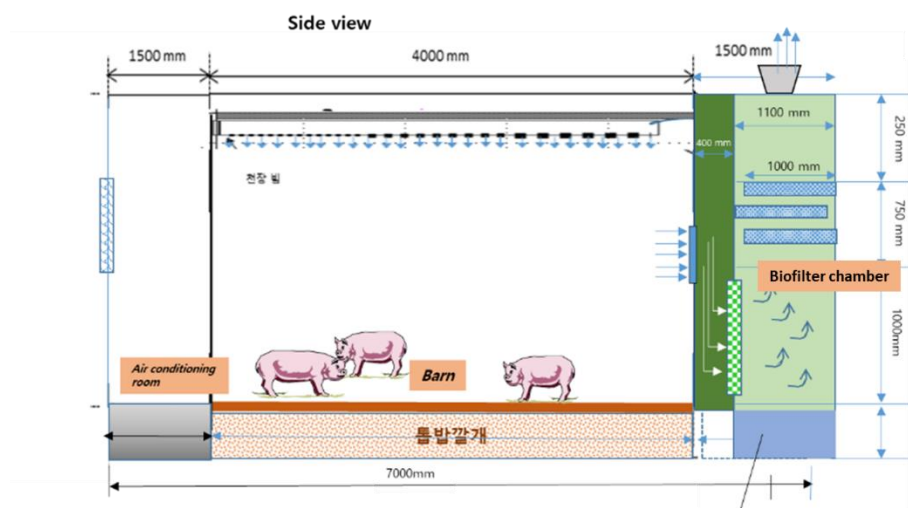


Figure 4.1. Experimental integrated pig house - biofilter system

The experiment was carried out in the bedding system pilot pig house (Figure 4.1). Two treatment of co-additive supplementation in biofilter pack were tested. The co-additive was the mixture of biochar and FGD gypsum 1:1 w/w. The treatments were T1: without co-additive and T2: with 10% (w/w) co-additive supplementation (5% (w/w) FGD gypsum, 5% (w/w) biochar). The Biochar was obtained from the local producer in Gyeonggi province, South Korea. It was produced from a combination of hardwood (80%) and softwood (20%) by the pyrolysis process at 550 °C. FGD gypsum was obtained from the KEPCO coal-fired power plant of South Korea. The experiment was carried out for about 56 days. Description of each experimental pig house-biofilter type is presented in Table 4.1. The physicochemical properties of the raw materials for biofilter media were given in Table 4.2, whilst the physicochemical properties of the mixtures of the biofilter media were given in Table 4.3.

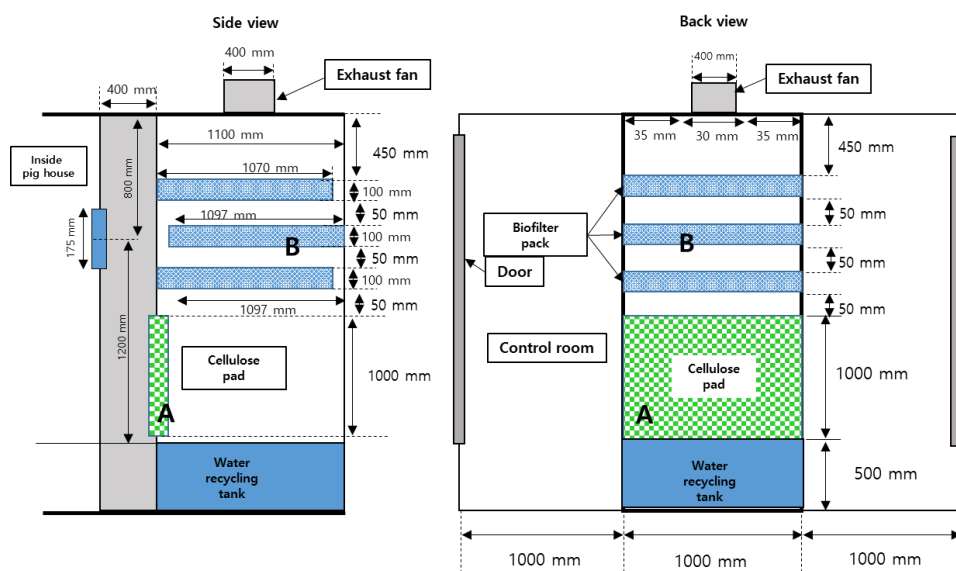
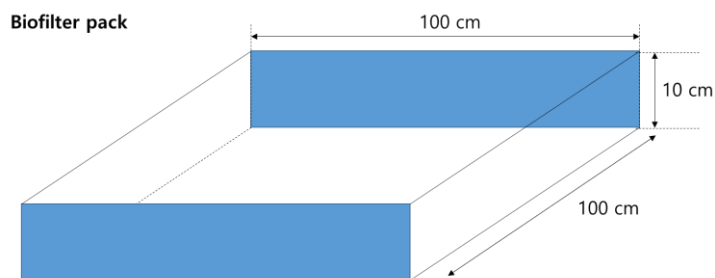


Figure 4.2. The experimental of dual biofilter system (A) vertical biotrickling (B) horizontal biofilter



Dimension	$h = 10 \text{ cm}; l = 100 \text{ cm}; w = 100 \text{ cm} = 100,000 \text{ cm}^3 = 0.1 \text{ m}^3$
Biofilter pack bulk density	400 kg/m^3
Biofilter weight	$400 \text{ kg/m}^3 \times 0.1 \text{ m}^3 = 40 \text{ kg}$
Sawdust	$90\% \times 40 \text{ kg} = 36 \text{ kg/pack}$; Total = 3 pack \times 36 kg = 108 kg
FGD gypsum	$5\% \times 40 \text{ kg} = 2 \text{ kg}$; Total = 3 pack \times 2 kg = 6 kg
Biochar	$5\% \times 40 \text{ kg} = 2 \text{ kg}$; Total = 3 pack \times 2 kg = 6 kg

Figure 4.3. Biofilter packing material

Table 4.1. Description of the treatment in this study

Treatment	Description
T1	Biofilter system without co-additive treatment
T2	Biofilter system supplemented by the FGD gypsum (5% w/w) and biochar (5% w/w)

Table 4.2. Characteristic of the raw material for the biofilter media

	Sawdust	Biochar	FGD Gypsum
pH	7.59 ± 0.65	10.99 ± 0.82	8.54 ± 0.64
EC (µS/cm)	0.15 ± 0.03	0.73 ± 0.05	2.69 ± 0.20
BD (g/L)	243.57 ± 20.70	515.29 ± 38.65	703.33 ± 52.75
TS (%)	81.49 ± 6.93	90.35 ± 6.78	77.75 ± 5.83
VS (%TS)	98.99 ± 8.41	65.90 ± 4.94	3.04 ± 0.23
Ash (%)	0.94 ± 0.08	34.10 ± 2.56	96.06 ± 7.20
TOC (%TS)	57.42 ± 4.88	38.23 ± 2.87	2.41 ± 0.18
TKN (%TS)	0.37 ± 0.05	0.36 ± 0.03	n.d
C/N	155.18 ± 13.19	105.02 ± 7.88	n.d

Values are expressed as mean ± standard deviation

Table 4.3. The physicochemical characteristics of the biofilter media mixture in this study

	T1	T2
TS (%)	47.59 ± 0.30	45.34 ± 2.70
VS (%TS)	98.04 ± 0.45	88.11 ± 3.72
pH	7.2 ± 0.04	7.69 ± 0.61
EC (mS/cm)	0.03 ± 0.01	1.40 ± 0.33
BD (g/cm ³)	0.46 ± 0.01	0.41 ± 0.00
PD (g/cm ³)	1.22 ± 0.00	0.82 ± 0.01
TOC (%TS)	30.94 ± 1.19	23.87 ± 0.59
TKN (%TS)	0.38 ± 0.01	0.22 ± 0.01
C/N ratio	80.38 ± 3.09	110.19 ± 2.75
P (mg/kg)	54.57 ± 1.36	163.57 ± 4.09
K (mg/kg)	253.2 ± 6.33	1990.15 ± 49.75
Na (mg/kg)	21.72 ± 0.54	71.11 ± 1.78
Ca (mg/kg)	2769.73 ± 69.24	17484.15 ± 473.10
Mg (mg/kg)	149.97 ± 3.75	533.28 ± 13.33
Fe (mg/kg)	24.43 ± 0.61	588.05 ± 14.70
S (mg/kg)	132.26 ± 3.3	7733.03 ± 193.33

Values are expressed as mean ± standard deviation

4.3.2. Ammonia and volatile organic acid odorant analysis

A total of 11 different types of VOCs were analyzed in this study according to Kumari et al., (2016), which included two sulfuric compounds (dimethyl sulfide [DMS] and dimethyl disulfide [DMDS]); six volatile fatty acids (acetic acid, propionic acid, butyric acid, iso-butyric acid, valeric acid and isovaleric acid); two indolics (indole and skatole); and one phenolic (p-cresol). The air was sampled for 5 min using a 1 L Tedlar bag (No.22053, Restek, Bellefonte, PA, USA) which was connected to a Teflon septum fitting to a low-volume air sampler (No. 800510; Gillian, USA) with a flow rate of 2.0 L/min. The collected air samples were analyzed using gas chromatography/mass spectrometry (GC/MS) (Agilent GC6890N/5975C MS, Youngin, Korea) in the laboratory, and all samples were immediately transported to the laboratory and analyzed within 5 h by using SPME fibers (Solid Phase Microextraction Fiber, Supelco, Bellefonte, PA, USA) – 75 μ m Carboxen/PDMS. Samples were extracted by SPME fibers for 10 min with a manual fiber holder from Supelco (Bellefonte, PA, USA). After extraction, the SPME fiber was removed from the Tedlar bag and immediately inserted into the injection port of the GC/MS for analysis. The desorption time of an SPME fiber was 10 min at 250 °C with a 0.75 mm injection port liner. Conventional GC/MS (Agilent GC6890N/5975C MS, Palo Alto, CA, USA) was the base platform. The system was equipped with 60 m (DB-5MS (J&W Scientific, Folsom, CA)) 0.250 mm id; 1.00 μ m film thickness; initial temperature, 35 °C for 5 min; 6 °C/min to 140°C, hold for 0 min; 15°C/min to 220 °C, hold for 3 min (Agilent Technology, 2007-2008).

Table 4.4. Detection thresholds of the target odorous compounds (Yao, 2010)

Compounds	Detection threshold
Ammonia	0.44 ppm
Dimethyl sulfide	0.11 ppb
Dimethyl disulfide	0.26 ppb
Indole	1.36 ppb
Skatole	0.13 ppb
p-cresol	2.90 ppb
Acetic acid	9.33 ppb
Propionic acid	0.009 ppb
n-butyric acid	0.10 ppb
i-butyric acid	1.27 ppb
n-valeric acid	0.48 ppb
i-valeric acid	3.17 ppb

The standard gases of sulfuric compounds – DMS and DMDS were purchased from RIGAS (Daejung, South Korea) with the approximate concentration of 10 ppmv. Headspace method was used to calibrate the indolic compounds, phenolic compound and volatile fatty acids. Based upon the Henry's Law constants of all the chemicals, the concentrations in the head space area were calculated as the concentration of ppbv and milligram per cubic meter. The detection threshold of the target odorous compounds is shown in Table 4.9.

Gastec tube (Gastec Co., Ltd., Kanagawa, Japan) were used to measure the concentrations of ammonia (NH₃). The differences in NH₃ concentrations across the biofilter were used to calculate the biofilter NH₃ removal efficiency as described by Ashtari et al., (2016). Equation (4.1) was used for calculating NH₃ removal efficiencies (RE) based on inlet NH₃ concentrations (ppmv) (C₁) and exhaust NH₃ concentrations (C₂).

$$RE = \left(\frac{C_1 - C_2}{C_1} \right) \times 100 \quad \text{Eq. (4.1)}$$

4.3.3. Biofilter media and recycled water characteristic analysis

Samples of the biofilter media were air dried, ground up into small particles using porcelain mortar, and passed through a 2-mm sieve. Physicochemical properties of the sieved samples were analyzed. The physicochemical characteristics of the samples were analyzed using methods on the dry weight basis (APHA, 2005). The moisture content was determined by drying fresh samples at 105 °C until the mass loss in 24 h was less than 0.5% relative to the previous day. Volatile solid was measured by dry combustion of dried samples (550 °C for four h). The pH and electrical conductivity (EC) were determined using a pH meter (Inolab, WTW, GmbH, Weilheim, Germany) and an EC214 conductivity meter (Hanna Instruments, Ltd., Sarmeola di aarubano, Italy) respectively.

The biofilter media sample was suspended in distilled water in a ratio of 1:10 (w/v). This suspension was shaken in a mechanical shaker at 230 rpm for 30 min and allowed to stand for an hour prior to pH and EC measurement. The bulk density of biofilter media material was defined as its weight per unit volume according to Thompson (Thompson et al., 2002). The Kjeldahl Nitrogen (TKN) content was determined according to the Kjeldahl digestion method (Bremmer et al., 1996) using Kjeltec Auto 1035 apparatus. The TOC was determined by the TOC analyzer Siever 5310 C. C/N ratio was estimated by divided the TOC to the TKN. Inorganic nitrogen NH_4^+ , NO_3^- and NO_2^- were extracted using 2 mol/L KCL (10:1 V/m), and then analyzed using the DR 5000 UV-Vis spectrophotometry method (Hach Co., Loveland, CO, USA).

The biofilter media's microbial (total bacterial, yeast and molds population) characteristic were analyzed refers to the method described in Suresh and Choi (2009). An amount of 1 g of samples was transferred to 9 mL of sterile distilled water and was subsequently serially diluted up to 10^{-8} . Estimation of the total heterotrophic bacterial count was quantified by spread plating (0.1 mL on Tryptic soy agar plates (in triplicate) and incubated at 35°C for 48H. The Potatoes dextrose agar plates incubated at 27°C for 72H was performed for total yeast and molds quantification.

The recycled water samples characteristics of each biofilter were monitored weekly for pH, Electrical Conductivity (EC), Dissolved Oxygen (DO) and Total Dissolved solid (TDS). The pH and electrical conductivity (EC) were determined using a pH meter (Inolab, WTW, GmbH, Weilheim, Germany) and an EC214 conductivity meter (Hanna Instruments, Ltd., Sarmeola di aarubano, Italy) respectively. The DO and TDS were measured using DO meter (Hanna instruments) and TDS meter (Hanna Instruments). Additionally, NH_4^+ , NO_3^- and NO_2^- were analyzed using DR 5000 UV-Vis spectrophotometry (Hach Co., Loveland, CO, USA).

4.3.4. Statistical analysis

Statistical evaluations were carried out using an SPSS software package (SPSS Inc., Chicago, IL, USA). Comparisons of each variable were first performed with ANOVA at a 5% significance level. The average values of the quantitative factors under evaluation were compared by means of an Independent sample T-test.

4.4. Results and Discussion

4.4.1. Ammonia and Volatile Organic Acid odor

Daily ammonia odor concentration before and after dual biofilter without co-additive treatment during the experiment is presented in Figure 4.4. According to the data showed in Figure 4.4, the ammonia concentration reduced after passing the biofilter system. The daily ammonia increased in line with time during the experiment. The highest ammonia concentration inside the pilot pig house (inlet) was about 15.45 ppm, and the lowest ammonia concentration was 2.52 ppm. It is known that NH_3 and sulfur compounds are released during the decomposition process of organic nitrogen compounds such as proteins (Higgins et al., 2008). However, after passing the biofilter system, the average ammonia concentration was significantly reduced ($p < 0.05$). The highest ammonia concentration after passing the biofilter system was 7.67 ppm, and the lowest ammonia concentration after passing the biofilter system was 1.33 ppm. The ammonia concentration emitted after the biofilter system increased following the increase of the ammonia concentration from inlet source (Figure 4.4). Results of this study were comparable to those measured in other studies which found that the NH_3 and H_2S concentration in the pig chambers ranged from 2 to 25 ppmv and from 3 to 1300 ppbv, respectively (Sun et al., 2010; Chen and Hoff, 2012; Martel et al., 2017).

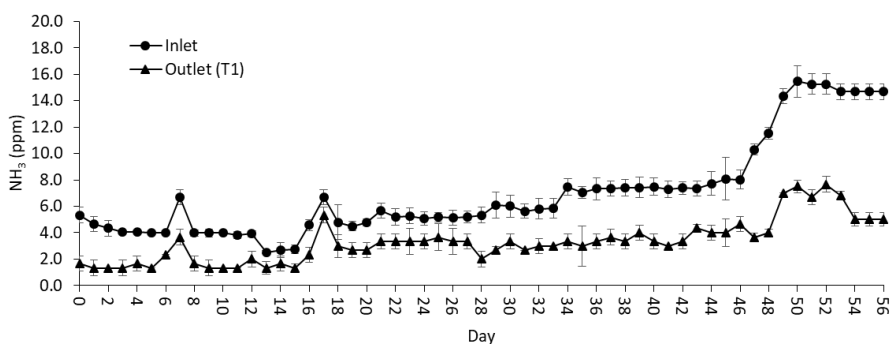


Figure 4.4. Daily ammonia odor concentration before and after dual biofilter without co-additive treatment. Values are expressed as mean \pm standard deviation

The increasing of the inlet ammonia concentration could be related to the pig grow which produced more feces and urine that dropped to the bedding material. The dropped waste might increase the nitrogenous organic matter composition in the bedding material which might induce the aerobic decomposition process rate and release more amount of ammonia. Higher accumulation of nitrogenous organic matter might decrease the C/N ratio in the bedding litter. Lower C/N ratio caused higher NH_3 emission (Jiang et al., 2011). The season also might affect the variation NH_3 emission as observed by Petersen et al., (2013) and Chowdhury et al., (2014) who found that higher NH_3 emissions during the summer season, which was the similar season when this experiment was carried out.

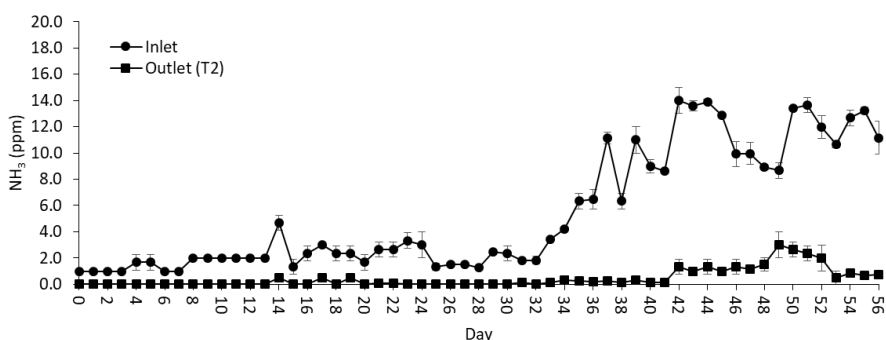


Figure 4.5. Daily ammonia odor concentration before and after dual biofilter with co-additive treatment. Values are expressed as mean \pm standard deviation

Figure 4.5 shows the daily ammonia odor concentration before and after dual biofilter with co-additive treatment during the experiment. According to the data showed in Figure 4.5, the daily ammonia increased in line with time during the experiment. The highest ammonia concentration inside the pilot pig house (inlet) was about 14 ppm, and the lowest ammonia concentration was 1 ppm. The average ammonia concentration was significantly reduced ($p < 0.05$) after passing the biofilter system. The highest level of ammonia concentration after passing the biofilter system was 3 ppm and the lowest level of ammonia concentration after passing the biofilter system was 0 ppm. The trend of ammonia concentration in the T2 was similar to those of T1. The ammonia concentration emitted after the biofilter system increased following the increase of the ammonia concentration from inlet source. However, in the T2 treatment with co-additive, the ammonia concentration can be maintained less than those of T1 treatment without co-additive.

The ammonia removal efficiency of the biofilter system investigated in this study was estimated. Figure 4.6 shows the ammonia odor removal

efficiency of dual biofilter system during the experiment. The removal efficiency of T1 was significantly lower ($p<0.05$) than that of the T2. The average removal efficiency of T1 and T2 were 51.23% and 94.85% respectively. The ammonia removal efficiency of T1 fluctuated, the maximum and minimum ammonia removal efficiency of T1 was 70% and 19.84% respectively. The ammonia removal efficiency of T1 decreased until the second week of experiment and increased gradually after day 20. The low ammonia removal efficiency of T1 could be due to the adaptation period of the ammonia-oxidizing bacteria in the biofilter media. The ammonia removal efficiency of T2 was constant until the second weeks of the experiment and slightly decreased. However, the ammonia removal efficiency was going back to increase until the sixth weeks of the experiment. According to the data showed in Figure 4.6, the co-additive treatment might significantly increase ($p<0.05$) the ammonia removal efficiency of the biofilter media in this study. The ammonia removal efficiency increased by about 57.72% after the co-additive treatment. These study results were in accordance with those reported by (Hartung et al., 2001) which explained that the average efficiency of odor reduction of biofilter media (coconut fiber and peat fiber mixture) was about 80% with a minimum of 25% and a maximum 95%. The efficiency of ammonia reduction was mainly influenced by the air flow rate, which determined the air retention time in the filter media. Increasing the moisture content of the filter material from 20% to 50% resulted in a 30% relative improvement of the cleaning efficiency of the biofilter (Hartung et al., 2001). However, Hanajima et al., (2010) have reported that the biotricking filter was better than chemical scrubber and biofilters for

eliminating the concentrations of odors, including those caused by NH_3 and sulfur compounds.

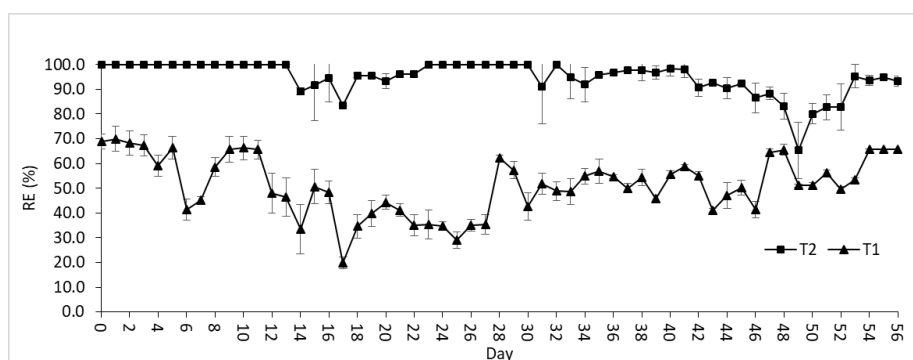


Figure 4.6. Ammonia odor Removal efficiency of dual biofilter during the experiment. Values are expressed as mean \pm standard deviation

The exhausted air treatment of animal house investigated by Tsang et al., (2015) resulted in removal efficiencies for NH_3 were on average 88% and 67% during fall/winter and spring/summer, respectively by a coal slag-packed biotrickling filter. Melse et al., (2012) reported that the NH_3 removal efficiencies in pig facilities achieved to 82% using the biotrickling filter packing that was made from vertically bundled polyethylene plastic net tube, which was in accordance with the results in the present study (achieved 51.23% and 94.85% removal efficiency). Nielsen et al., (2008) reported that analysis of nitrogen chemistry and biological potentials showed that most ammonia was removed in a restricted zone, the biological processes occur in combination with evaporation and counter flow of air and water generated distinct chemical gradients in the filters. The properties of NH_3 such as high water solubility and weak alkalinity, are suitable for water or acid scrubbing treatments. In addition, NH_3 is easily biologically transformed into non-smelling substances through

the process of nitrification and denitrification (Yasuda et al., 2009). According to the results in this study, the performance of the biofilter was fluctuate (Figure 4.6). It is likely because the growth and loss of ammonia oxidizer biomass and the occasional establishment of nitrite-oxidizing bacteria could also change filter performance. Sectioning of the filter together with biomass control and proper water management was essential for optimal ammonia and odor removal (Nielsen et al., 2008).

In addition, (Liang et al., 2006) reported that available carbon amendment to daily manure had been reported to be effective in preventing NH_3 volatilization, this explains the function of the biochar in this study for reducing ammonia emission from the pilot pig house. The FGD gypsum characteristic could also explain the higher ammonia removal efficiency in the T2. The FGD gypsum can convert the ammonium carbonate to ammonium sulfate along with the formation of calcium carbonate (Guo et al., 2016). The N form in ammonium carbonate is much more appropriate to be lost as ammonia than when it is in the form of ammonium sulfate (Tubail et al., 2008).

The volatile fatty acids odorants were also frequently found inside the pilot pig house airspace. Total VFA odor fluctuation during the experiment of dual biofilter system without co-additive treatment is presented in Figure 4.7. The VFA odor concentration increased after biofilter without co-additive supplementation. The VFA odor concentration inside the pig house before passing the biofilter system varied in the range of 1.34 ppbv to 58.51 ppbv. After passing the biofilter system, the VFA concentration was in the range of 48.34 ppbv to 103.64 ppbv. VFA Odor significantly increased ($p < 0.05$) after

the biofilter system in T1 treatment was possibly caused by the additional odor production from the biofilter media and from the recirculated water.

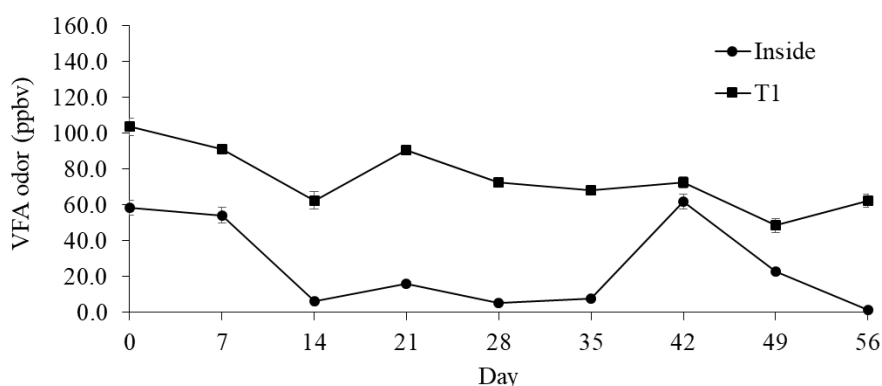


Figure 4.7. Total VFA odor fluctuation of dual biofilter system without co-additive treatment during the experiment. Values are expressed as mean \pm standard deviation

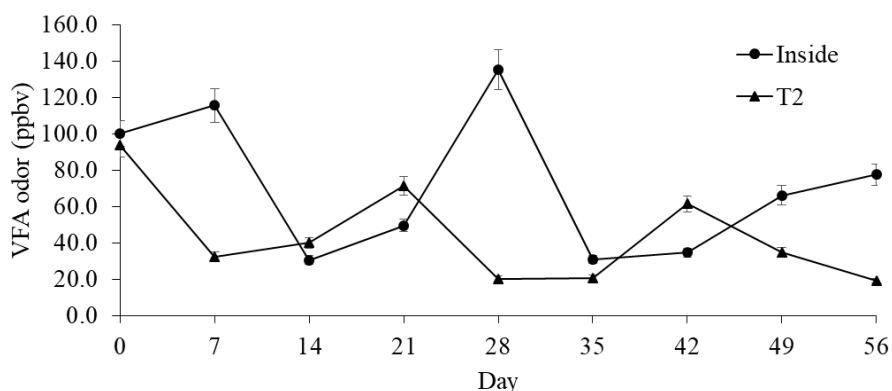


Figure 4.8. Total VFA odor fluctuation before and after dual biofilter with co-additive treatment during the experiment. Values are expressed as mean \pm standard deviation

Total VFA odor fluctuation during the experiment of dual biofilter system with co-additive treatment is presented in Figure 4.8. The VFA odor concentration from the inside of the pilot pig house before passing the biofilter system varied in the range of 30.59 ppbv to 135.22 ppbv. After passing the biofilter system, the VFA concentration was in the range of 19.44 ppbv to 93.81

ppbv. Ahn and Choi (2005) reported a total VFA concentration of 0.23 g/L corresponds to the beginning of offensive odors. VFA is strongly pH dependent, and the VFA's volatility is higher at lower pH. However, it can be retained in the compost material due to an alkaline condition caused by high NH_3 production or decomposed in the compost mass (Maeda et al., 2009). Rather than from composting, the contribution of VFAs could be more important for odors from barns, manure pits or manure spreading, (Hanajima et al., 2010).

VFA Odor on the inlet of dual biofilter system in T2 was not significantly different ($p>0.05$) with that on its outlet. However, in comparison to that of T1, the odor did not increase after passing the T2 biofilter system, while increased in the T1 biofilter system. It can be inferred that the co-additive supplementation might inhibit the emission of VFA odor from pilot pig house and did not give a negative effect to the biofilter media performance. The VFA was almost completely removed in the biofilters, as they seldom appeared in the samples collected at the exhaust of the biofilter, which could be attributed to the relatively high solubility of these compounds in water (Martel et al., 2017).

The phenolic-indolic odor fluctuation during the experiment of dual biofilter system without co-additive supplementation (T1) is presented in Figure 4.9. The phenolic-indolic odor concentration from inside the pig house before passing the biofilter system (biofilter inlet) varied in the range of 0 - 1.55 ppbv. After passing the biofilter system (biofilter outlet), the VFA concentration was in the range of 0 - 0.98 ppbv. The average phenolic-indolic

odor concentration on the inlet of T1 was not significantly different ($p>0.05$) with that of outlet the biofilter system.

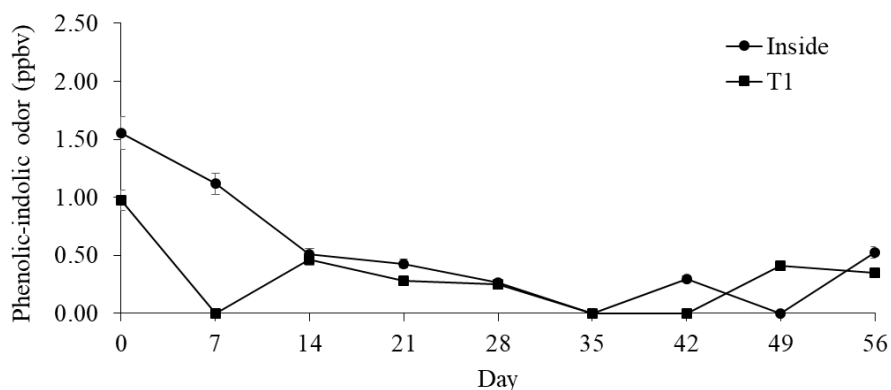


Figure 4.9. Total phenolic-indolic odor fluctuation before and after dual biofilter without co-additive treatment during the experiment. Values are expressed as mean \pm standard deviation

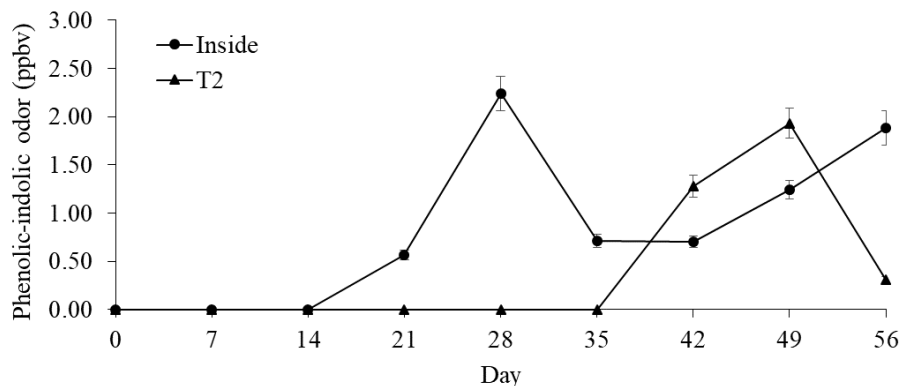


Figure 4.10. Total phenolic-indolic odor fluctuation before and after dual biofilter with co-additive treatment during the experiment

Figure 4.10 shows the total phenolic-indolic odor fluctuation before and after dual biofilter with co-additive supplementation treatment (T2) during the experiment. The total phenolic-indolic odor concentration in the biofilter inlet of T2 was in the range of 0 - 2.24 ppbv. The phenolic-indolic odor on the inlet of the biofilter fluctuated. However, it tends to increase in line with time. The phenolic-indolic odor concentration on the biofilter outlet of T2 was in the

range of 0 - 1.93 ppbv. The average phenolic-indolic odor concentration between the biofilter inlet and the biofilter outlet of T2 was not significantly different ($p>0.05$). However, the phenolic odor concentration in the biofilter outlet of T2 was significantly lower ($p<0.05$) than that of biofilter outlet at day 21 – 28 and at day 56. The co-additive supplementation in the biofilter media might delay the phenolic-indolic odor dispersion from the pilot pig house. However, the emission still cannot be reduced.

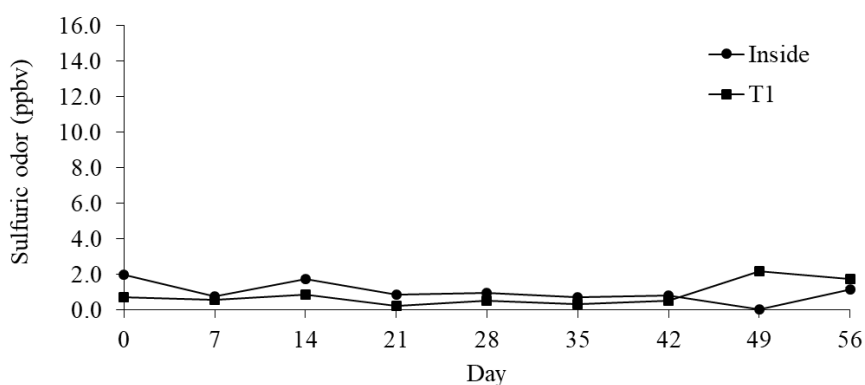


Figure 4.11. Total Sulfuric odor fluctuation before and after dual biofilter without co-additive treatment during the experiment. Values are expressed as mean \pm standard deviation

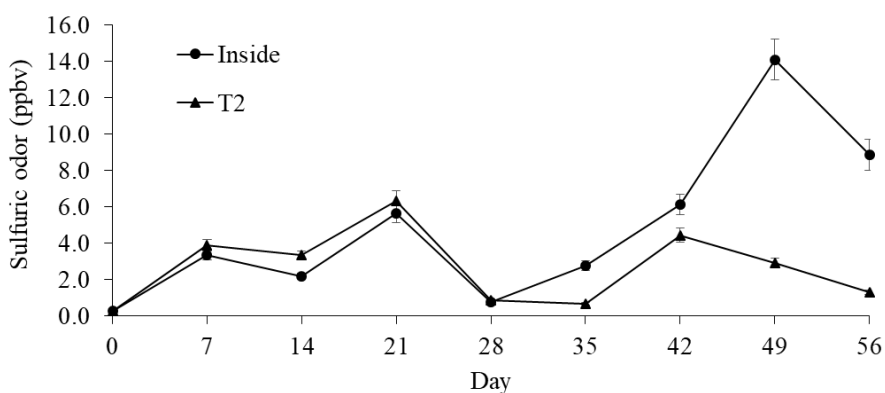


Figure 4.12. Total Sulfuric odor fluctuation before and after dual biofilter with co-additive treatment during the experiment. Values are expressed as mean \pm standard deviation

Total sulfuric odor fluctuation during the experiment of dual biofilter system without co-additive supplementation (T1) is presented in Figure 4.11. The sulfuric odor concentration in the biofilter inlet of T1 was constant in the range 0.04 - 1.99 ppbv during the experiment. The sulfuric odor concentration in the biofilter outlet of T1 was also constant in the range of 0.21 – 2.17 ppbv. The average of sulfuric odor concentration in the biofilter inlet and outlet of T1 was not significantly different ($p>0.05$). The sulfuric odor concentration in the biofilter outlet of T1 was significantly higher ($p<0.05$) than that of biofilter inlet on day 49 of the experiment. The capability of the biofilter to filter the sulfuric odor was likely reduced after day 49.

Figure 4.12 shows the sulfuric odor fluctuation during the experiment of dual biofilter with co-additive supplementation (T2). According to the data showed in Figure 4.12, during the experiment, the sulfuric odor concentration in the biofilter inlet of T2 tend to increase from 0.29 ppbv initially to 14.09 ppbv on the day 49. However, the sulfuric odor concentration in the biofilter outlet of T2 can be maintained stay constant in the level below 4 ppbv (Figure 4.12). Blanes-Vidal et al. (2009) found the sulfur-containing compounds, which include H_2S , to have high correlations with odor emitted from swine slurry. Sulfur compounds are found during the decomposition process of amino acids containing sulfur (Higgins et al., 2008). Dimethyl disulfide (DMDS) is one of the major odor compounds that emitted simultaneously with that of NH_3 and Methyl mercaptan (MM) and is indicated to have a high correlation. MM can be readily oxidized to form DMDS (Higgins et al., 2006). The production of MM was found to mainly occur from the degradation of methionine and the

methylation of hydrogen sulfide. Dimethyl sulfide (DMS) was formed through the methylation of MM. The average of sulfuric odor concentration in biofilter inlet of T2 (4.89 ppbv) was not significantly different ($p>0.05$) than that of biofilter outlet of T2 (2.66 ppbv). It can be inferred from this study that co-additive supplementation did not give effect to the biofilter performance to reduce sulfuric odor emitted from the pig house. The explanation of this fact was the water solubility characteristic of MM (2.33 g/100 ml at 20 °C) and DMS (insoluble in water) are much lower than that of NH_3 (52.6 g/100 ml at 20 °C). The use of chemicals such as hypochlorite, ozone or hydrogen peroxide as the scrubbing liquids can remove the volatile organic sulfur compounds more effective (Smet and Van Langenhove, 1998) rather than using co-additive in the solid biofilter media.

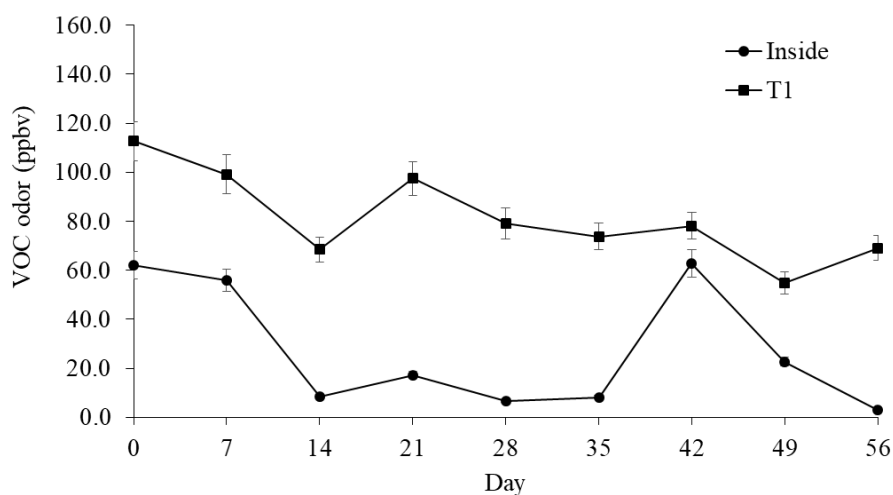


Figure 4.13. Total VOC odor fluctuation before and after dual biofilter without co-additive treatment during the experiment. Values are expressed as mean \pm standard deviation

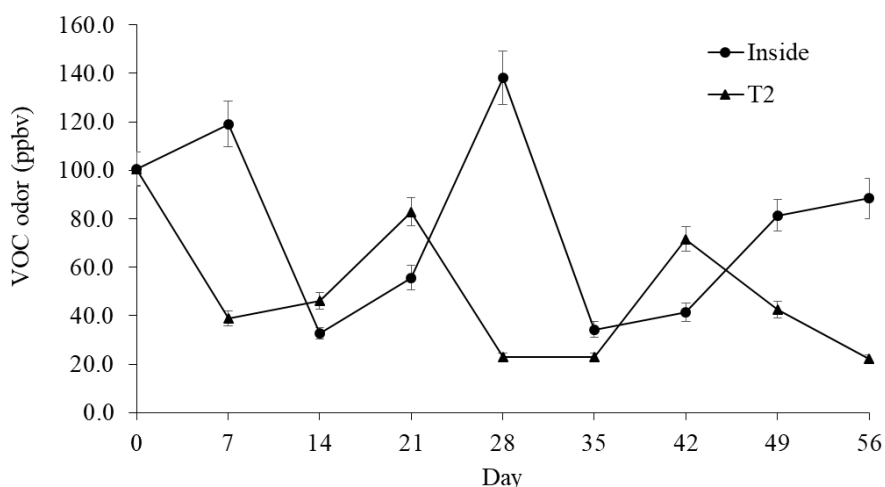


Figure 4.14. Total VOC odor fluctuation before and after dual biofilter without co-additive treatment during the experiment. Values are expressed as mean \pm standard deviation

Total VOC odor fluctuation before and after dual biofilter without co-additive treatment (T1) during the experiment is presented in Figure 4.13. According to the data shown in Figure 4.13, the average volatile organic acid odor concentration in the biofilter inlet was significantly lower ($p < 0.05$) than that of biofilter outlet. The volatile organic acid odor concentration in the biofilter inlet was in the range of 3.02 – 62.83 ppbv. The volatile organic acid odor concentration in the biofilter outlet was in the range of 50.93 – 105.32 ppbv.

Total VOC odor fluctuation before and after dual biofilter with co-additive treatment (T2) during the experiment is presented in Figure 4.14. According to the data shown in Figure 4.14, the average volatile organic acid odor concentration in the biofilter inlet was not significantly different ($p > 0.05$) than that of biofilter outlet. The volatile organic acid odor concentration in the

biofilter inlet was in the range of 32.77 – 138.21 ppbv. The volatile organic acid odor concentration in the biofilter outlet was in the range of 21.21 – 94.06 ppbv. The volatile organic acid concentration in the biofilter outlet of T2 showed lower than that of biofilter inlet on several days of the experiment (Figure 4.14). The performance for VOC odor filtration of T2 still fluctuated, however, it seems to be improved rather than that of T1. On average, the T2 had removal efficiency (RE) for VOA odor about 21.36 %, and it was significantly higher ($p < 0.05$) than that of T1 (-572% RE). The co-additive supplementation might potent to inhibit the emission of volatile organic odor from the pig house.

Based on the results of the present study, among the odorant compounds, the ammonia was the most significant odor that can be removed by the biofilter and the performance was significantly improved by co-additive supplementation. Akdeniz et al., (2012) found that NH_3 to be one of the compounds that had the most significant impact on odor from livestock buildings. Furthermore, Akdeniz et al., (2012) reported that higher molecular weight compounds such as phenol, 4-methyl phenol, 4-ethyl phenol, and 1H-indole were also observed to be significant predictors of sensory odor. Significant correlations between NH_3 and H_2S odor concentrations were also observed. Kumari et al., (2016) found that microclimate variables did not correlate significantly with most of the airborne particles and odorous compounds, suggesting that microclimate variables not influence their emission from compost facilities.

Results from this study indicated that most of the VOCs had the highest correlations with the volatile fatty acids. This result agrees with the findings of

other studies (Blanes-Vidal et al., 2009; Hanajima et al., 2010; Martel et al., 2017) in which volatile fatty acids were found to be good indicators of swine odor. Furthermore, Martel et al., (2017) described that butanoic acid, 3-methyl butanoic acid, 2-methyl butanoic acid, p-cresol, 2-methyl propionic acid, pentanoic acid 2,3-butanedione, and dimethyl sulfide were found to have the largest odor impacts. In another case, methyl mercaptan also had a relatively high correlation with odor concentration as observed by Hanajima et al., (2010) and Martel et al., (2017).

Table 4.5. Pearson correlation of measured odorant during the experiment

Pearson	Total VOC	Total VFA	Total Sulfuric	Total Phenolic	Ammonia
Total VOC	1	0.99**	0.13	0.27	-0.21
Total VFA	0.99**	1	0.04	0.23	0.2
Total Sulfuric	0.13	0.04	1	0.37*	0.34*
Total Phenolic	0.27	0.23	0.37*	1	0.27
Ammonia	-0.21	0.2	0.34*	0.27	1

** Correlation is significant at the 0.01 level

*Correlation is significant at the level 0.05

The Pearson correlation coefficient of measured odorant during the experiment is presented in Table 4.5. Based on the results of the current study, it illustrated that total VOC had a significantly positive correlation with the total VFA since the most VOC odorant consists of VFA odor. The total sulfuric had a significantly positive correlation with total phenolic ($r = 0.37$) as well as ammonia ($r = 0.34$).

4.4.2. Biofilter media and recirculated water

The physicochemical characteristic of biofilter media during the experiment are present in Figure 4.15. The moisture content of the biofilter pack

was maintained in the range of 50 – 55% (Figure 4.15a). There was no significantly different ($p>0.05$) of moisture content in the biofilter media of T1 and T2. Yang et al., (2014) reported that ammonia removal efficiency was greatly improved when media moisture content was increased from 35 – 55% due to the abundance of the ammonia oxidizers microbial community.

The volatile solid of the biofilter media during the experiment is presented in Figure 4.15b. The volatile solid or organic matter of the T2 was significantly lower than that of T1. The volatile solid of T2 was in the range of 85.78% – 89.72%, while the volatile solid of T1 was in the range of 98.05% - 98.66%. The lower volatile solid of T2 was likely because of the co-additive composted in the biofilter media contain much more amount of mineral (inorganic mineral) rather than organic matter. The biofilter T1 pH showed slightly increased during the experiment whilst the pH of the biofilter T2 showed constantly. In the final day of the experiment, the pH of T1 (8.48) was significantly higher ($p<0.05$) than that of T2 (7.81). The lower pH of T2 was likely because of the co-additive supplementation; the pH decrease was attributed to the replacement of Na^+ by Ca^{2+} from the FGD gypsum, by which the reaction $\text{CO}_3^{2-} + \text{HCO}_3^{2-}$ generated deposits of CaCO_3 (Du et al., 2017). The pH drop was also due to unexpected colonization of the filter by nitrite oxidizing bacteria. By converting nitrite into nitrate, these bacteria allow pH to drop more before the “nitrous acid brake” works as reported by Nielsen et al., (2008).

Higher mineral content in the biofilter media will affect the higher EC as shown in Figure 4.15d. The EC of T1 was in the range of 0 mS/cm – 0.5 mS/cm,

and the EC of T2 was in the range of 1.40 mS/cm – 1.87 mS/cm. Co-additive supplementation of the biofilter media increased the EC of the biofilter media. The average of the bulk density of T1 and T2 biofilter media was 0.382 g/cm³ and 0.418 g/cm³ respectively, the bulk density of biofilter media T2 was significantly higher ($p < 0.05$) than that of T1. According to the data showed in Figure 4.15d, the co-additive supplementation increased the bulk density, this could be related to the nature of the co-additive mixture of FGD-gypsum and biochar which has higher bulk density. The final particle density of T1 and T2 were not significantly different ($p > 0.05$) (Figure 4.15f). The final particle density of T1 and T2 were 1.03 g/cm³. The porosity was calculated by using bulk density and particle density. The porosity of the biofilter media in this experiment is presented in Figure 4.15g. According to the data showed in Figure 4.15g, the average porosity of the T1 biofilter media (63.36%) was significantly higher ($p < 0.05$) than that of T2 biofilter media (52.93%). It showed that co-additive supplementation might decrease the porosity of the biofilter media. The higher the porosity might allow the contaminant air to pass the biofilter easily and the odor might not be filtered properly. In addition the lower porosity, the lower empty bed retention time, thus the reaction time of the odor compound with the biofilter media was shorter. Contrary, the proper porosity might optimize the odor filtration by physically giving more reaction or contact time for air containing odor in the biofilter media. It can be inferred that co-additive supplementation might improve the porosity of the biofilter media, hence improve the odor reduction capability.

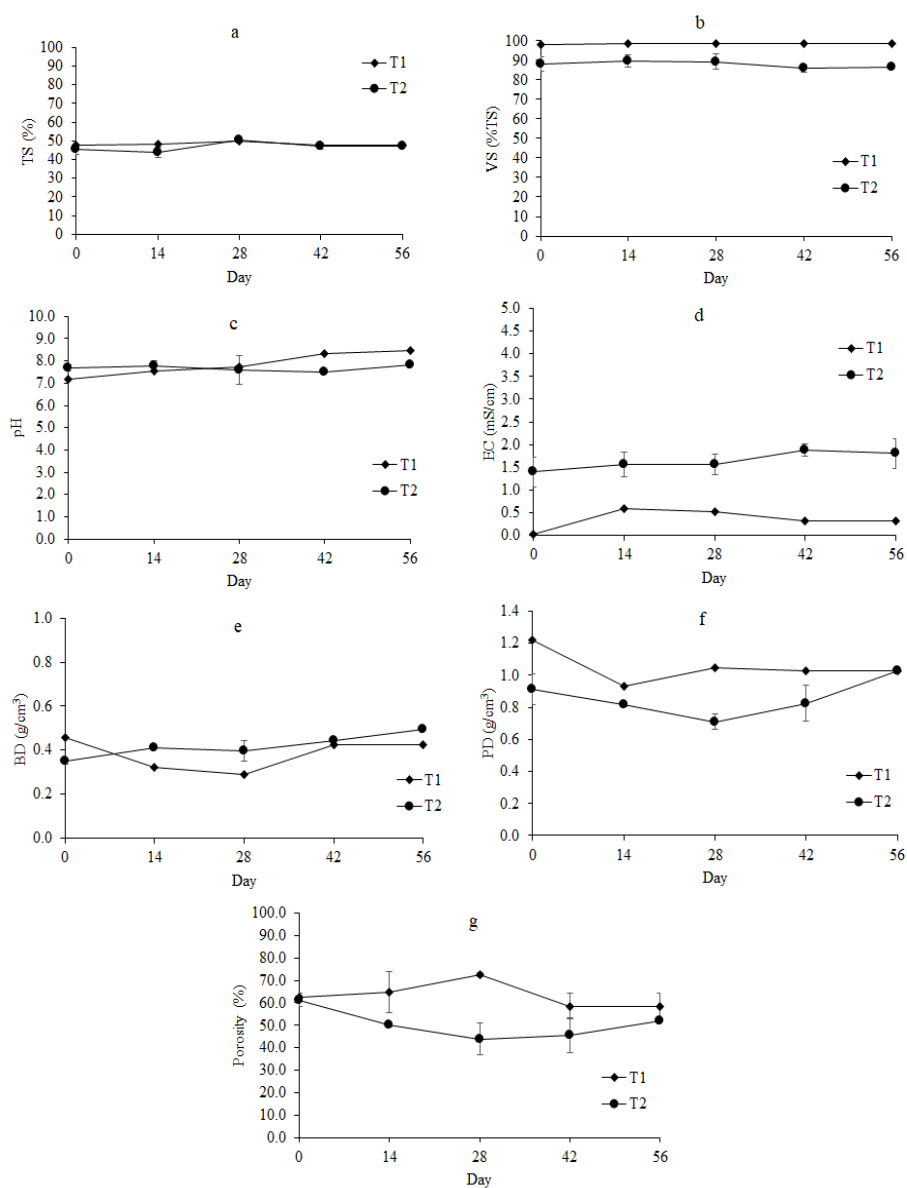


Figure 4.15. Physicochemical characteristic of biofilter media during the experiment (a) TS; (b) VS; (c) pH; (d) EC, (e) Bulk Density, (f) Particle Density, (g) porosity. Values are expressed as mean \pm standard deviation

Figure 4.16 shows the Nitrogen fraction of the biofilter media during the experiment. The final total ammonia nitrogen of the T1 biofilter media (1591.87 mg/kg TS) showed significantly higher ($p < 0.05$) than that of T2 biofilter media (612.48 mg/kg TS). The total ammonia nitrogen in the T1

biofilter media gradually increased along with the experiment time. Higher total ammonia nitrogen in the T1 biofilter media might come from the contaminant air from the pilot pig house room and be absorbed in the biofilter media. However, the higher total ammonia concentration in the biofilter media also might increase the possibility of the ammonia to be released easily as gas phase if an environmental condition such as higher temperature support the equilibrium for ammonium nitrogen to transform became ammonia gas. In addition, after considering the nitrite-nitrate concentration, the higher amount of ammonia in the biofilter media could be the indicator for less ammonification process occurred. Hence the nitrite-nitrate concentration was also observed in this study. The nitrification rate can be identified by estimate the ratio of $\text{NH}_4^+/\text{NO}_3^-$ in the biofilter media. From the results shown in Figure 4.16c, it can be seen that the final $\text{NH}_4^+/\text{NO}_3^-$ ratio of T1 biofilter media (4.30) was higher than that of $\text{NH}_4^+/\text{NO}_3^-$ ratio of T2 biofilter media (1.00). Higher $\text{NH}_4^+/\text{NO}_3^-$ ratio means that higher TAN present in comparison to nitrate, this condition indicated that nitrification rate was lower in the T1 biofilter media. Thus it can be inferred that co-additive supplementation might improve nitrification process in the biofilter media.

The $\text{NO}_3^- + \text{NO}_2^-$ concentration of the T1 and T2 biofilter media during the experiment is presented in Figure 4.16b. The final $\text{NO}_3^- + \text{NO}_2^-$ concentration in T1 (607.51 mg/kg TS) showed significantly higher ($p < 0.05$) than that of T2 (376.96 mg/kg TS). Higher amount of $\text{NO}_3^- + \text{NO}_2^-$ fraction in the T1 was likely because of lower nitrification rate occurred in the T1 biofilter media. The co-additive supplementation might affect to the nitrification process improvement

in the biofilter media which induce the microorganism activities transforming dissolved ammonium into other nitrogen forms, such as nitrite, nitrate nitrogen, and nitrous oxide (Yang et al., 2014). In addition, since it contains higher nitrite nitrate concentration, the biofilter media might also further be considered as a bio-fertilizer product source for land application.

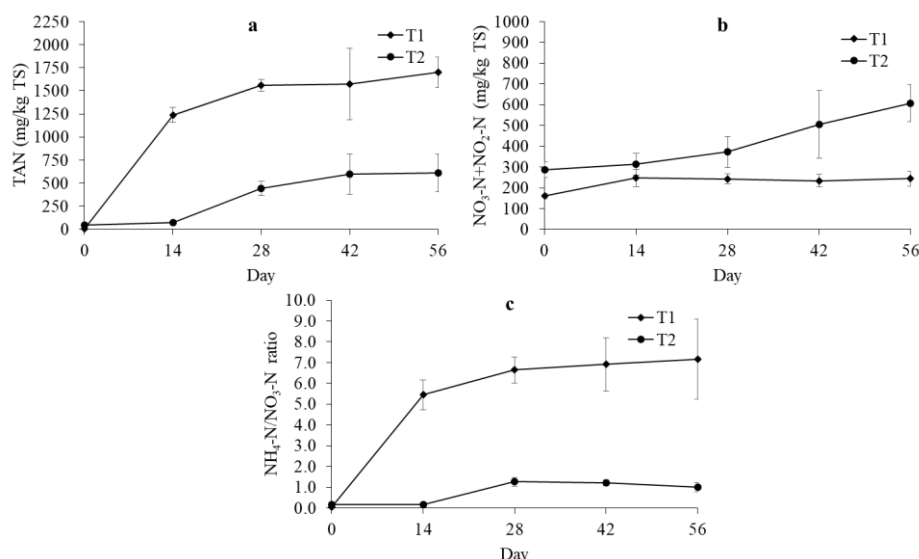


Figure 4.16. Nitrogen fraction of the biofilter media during the experiment (a) Total Ammonia Nitrogen (TAN); (b) NO₃⁻+NO₂⁻; (c) NH₄⁺/NO₃⁻. Values are expressed as mean \pm standard deviation

Moussavi et al., (2011) found that the developed biotrickling filter could completely remove 100 ppm ammonia from a waste stream, with an empty bed retention time of 60 s and 98.4% nitrogen removal. It was found that both autotrophic and heterotrophic bacteria were involved in the nitrogen removal through the simultaneous nitrification/denitrification process in the biotrickling filter. Additionally, the elimination capacity of total nitrogen by the biotrickling filter increased from 3.5 to 18.4 g N/m³·h with an inlet load of 20.6 g N/m³·h (73.6%) which was in accordance with the results of this study.

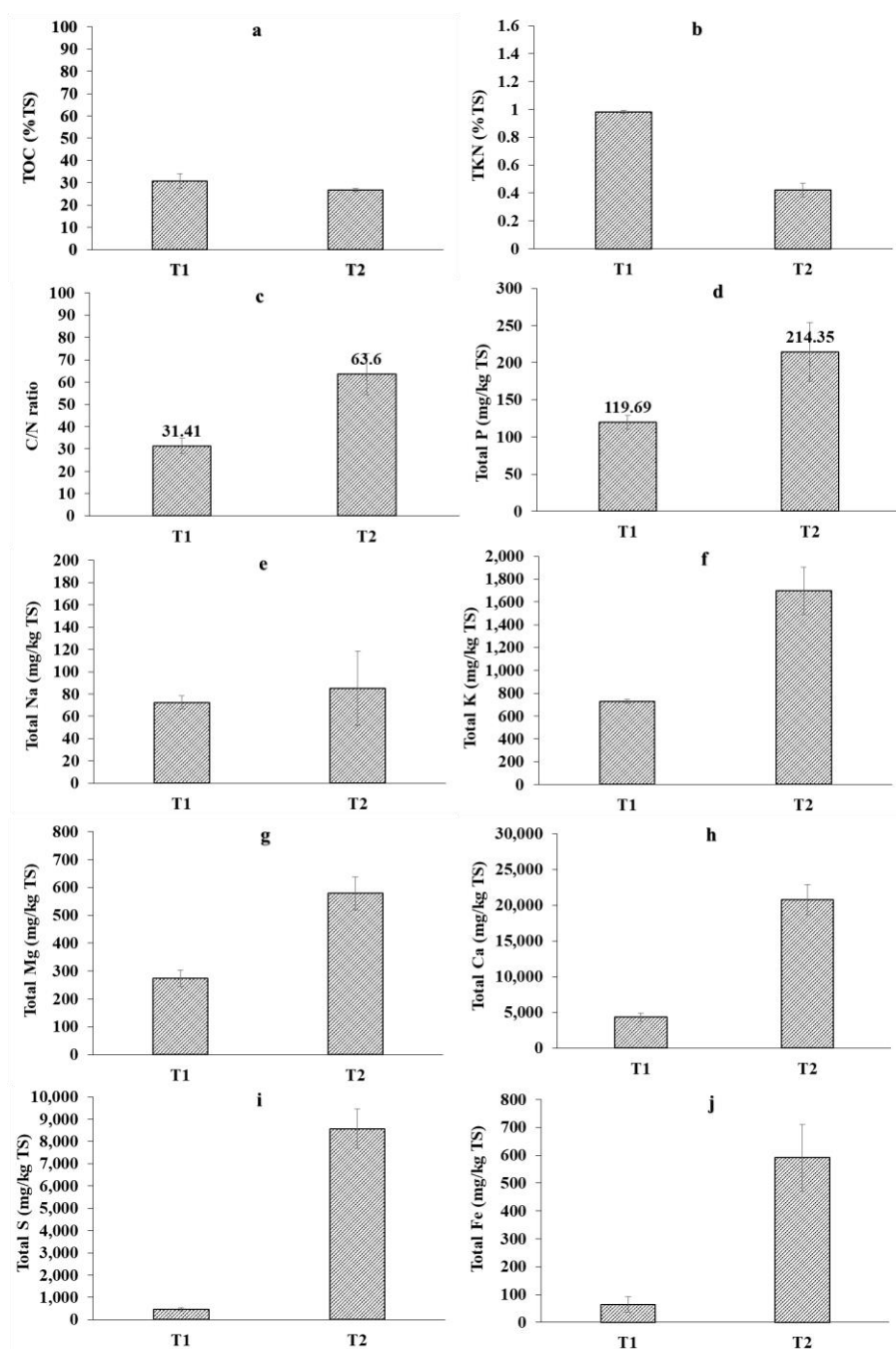


Figure 4.17. The C/N ratio and minerals concentration of the biofilter media (a) TOC; (b) TKN; (c) C/N ratio; (d) Total P; (e) Total Na; (f) Total K; (g) Total Mg; (h) Total Ca; (i) Total S; (j) Total Fe. Values are expressed as mean \pm standard deviation

The C/N ratio and mineral concentration of the biofilter media samples taken at the end of the experiment are presented in Figure 4.17. The C/N ratio of T2 (63.60) was significantly higher ($p < 0.05$) than that of T1 (31.41). Higher C/N ratio means that the biofilter media contain lower ammonia nitrogen fraction concentration in comparison to the carbon material in T2 as shown in Figure 4.17c. The total ammonia nitrogen that contributes to the TKN concentration was lower in T2, could be due to the higher rate of ammonia oxidation to be transformed to nitrate and nitrite, while the TOC concentration of T1 and T2 was not significantly different ($p > 0.05$).

Total phosphorus (P) concentration was found to be significantly higher ($p < 0.05$) in the T2 (214.35 mg/kg) rather than that of T1 (119.69 mg/kg) (Figure 4.17d). The co-additive supplementation of biofilter media by FGD gypsum might contribute the higher phosphorus. The higher phosphorus in the biofilter media might be potent the further utilization of discharged biofilter media as the bio-fertilizer since the phosphorus was essential nutrition for crop plant. The total sodium (Na) of T1 (72.29 mg/kg) and T2 (85.10 mg/kg) was not significantly different ($p > 0.05$) as shown in Figure 4.17e. It means that co-additive supplementation did not change the sodium concentration in the biofilter media. Total potassium (K) concentration of the biofilter media was found to be significantly higher ($p < 0.05$) in T2 (1,696.05 mg/kg) than that of T1 (729.83 mg/kg) (Figure 4.17f). Potassium is one of the macro-nutrient for plant growth. Thus the higher amount of potassium contained in T2 might give the beneficial potent of the T2 biofilter media for further utilization as the biofertilizer product. The higher potassium contained in the T2 was likely

contributed to the co-additive supplementation. The average concentration of Mg, Ca, S and Fe in T2 biofilter media were 578.75 mg/kg, 20,752.66 mg/kg, 8,570.93 mg/kg, and 590.83 mg/kg respectively, and it was significantly higher ($p<0.05$) than that of T1 which contain about 274.23 mg/kg, 4,342.01 mg/kg, 464.60 mg/kg and 64.98 mg/kg of Mg, Ca, S and Fe respectively. According to the results of the present study, it can be illustrated that the co-additive supplementation might increase the plant macro and micronutrient concentration of the biofilter media, especially the total P, K, Ca, Mg, S, and Fe.

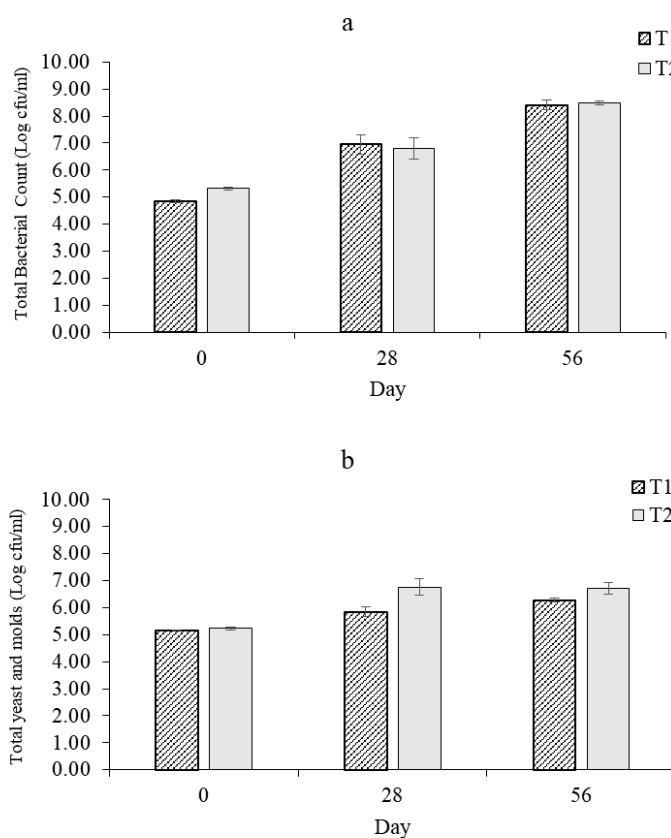


Figure 4.18. The microbial population in the biofilter media during the experiment (a) Total Bacterial Count (TBC); (b) Total yeast and molds count. Values are expressed as mean \pm standard deviation

The total bacterial, yeast and molds population in the biofilter media during the experiment is presented in Figure 4.18. The population of bacteria, yeast, and molds increased in line with time. The total bacterial population of T1 significantly increased ($p < 0.05$) from initially 4.8 Log CFU/mL to 8.4 Log CFU/mL in the final (day 56). The total bacterial population of T2 significantly increased ($p < 0.05$) from initially 5.33 Log CFU/mL to 8.48 Log CFU/mL (Figure 4.18a). There was not significantly different ($p > 0.05$) of total bacterial count between T1 and T2 at the end of the experiment. The co-additive supplementation did not result in a significant effect on the total bacterial population in the biofilter media. The higher ammonia odor removal efficiency in the T2 could be due to the higher population of ammonia-oxidizing bacteria in T2 rather than that of in T1. Further analysis of the bacterial type living in the biofilter media such as ammonia oxidizing bacteria could be necessary to be investigated since the ammonia removal efficiency in the T2 was significantly higher than that of T1.

Total yeast and molds population in the biofilter media during the experiment is presented in Figure 4.18b. The total yeasts and molds in the T2 (6.71 Log CFU/mL) was significantly higher than that of T1 (6.28 Log CFU/mL). The significant difference of the yeast and molds population between T1 and T2 was showed since the day 28 of the experiment. The higher total yeast and molds population in T2 could be due to the higher macro and micro mineral composition in the T2 by the co-additive supplementation. The

higher macro and micro mineral composition source might induce higher microbes metabolism. The co-additive supplementation to the biofilter media might induce the yeast and molds growth.

Pearson correlation coefficients of measured physicochemical characteristics of the biofilter media during the experiment are presented in the Table 4.5. We found that the TS content of the biofilter media had a significantly positive correlation with TAN ($r = 0.58$) and $\text{NH}_4^+/\text{NO}_3^-$ ($r = 0.47$) whilst had a significantly negative correlation with BD ($r = -0.34$). The biofilter media's VS had significantly positive correlation with PD ($r = 0.51$), TAN ($r = 0.51$), and $\text{NH}_4^+/\text{NO}_3^-$ ($r = 0.62$). Pearson correlation matrix showed the highest positive correlation between TAN and $\text{NH}_4^+/\text{NO}_3^-$ ($r = 0.96$), whilst the higher negative correlation were between VS and EC ($r = -0.88$).

Table 4.6. Pearson correlations of measured physicochemical characteristics of the biofilter media during the experiment

Pearson Correlation	TS	VS	pH	EC	BD	PD	TAN	$\text{NH}_4^+ + \text{NO}_3^-$	$\text{NH}_4^+/\text{NO}_3^-$	Porosity
TS	1	0.15	-0.04	-0.05	-0.34**	-0.15	0.58**	0.11	0.47**	0.13
VS	0.15	1	0.18	-0.88**	-0.31*	0.52**	0.51**	-0.62**	0.62**	0.62**
pH	-0.04	0.18	1	-0.17	-0.01	0.01	0.45**	-0.15	0.49**	0.06
EC	-0.05	-0.88**	-0.17	1	0.17	-0.61**	-0.38	-0.68**	-0.50**	-0.59**
BD	-0.34**	-0.31*	-0.01	0.17	1	0.13	-0.39**	0.33**	-0.48**	-0.63**
PD	-0.15	0.52**	0.01	-0.61**	0.13	1	0.09	-0.45**	0.21	0.67**
TAN	0.58**	0.51**	0.45**	-0.38	-0.39**	0.09	1	-0.08	0.96**	0.36**
$\text{NO}_3^-/\text{NO}_2^-$	0.11	-0.62**	-0.15	-0.68**	0.33**	-0.45**	-0.08	1	-0.31*	-0.59**
$\text{NH}_4\text{-N}/\text{NO}_3^-$	0.47**	0.62**	0.49**	-0.50**	-0.48**	0.21	0.96**	-0.31*	1	0.51**
Porosity	0.13	0.62**	0.06	-0.59**	-0.63**	0.67**	0.36**	-0.59**	0.51**	1

**Correlation is significant at the 0.01 level

*Correlation is significant at the level 0.05 level

There was a two-step biofiltration process occurred in the dual biofilter system. The first phase was the vertical biofiltration which used the vertically

cellulose pad equipped with the circulating water system flowed to the cellulose pad. The circulated water was expected to increase the ammonia odor absorption as well as reduced particulate matter emitted from the pig house. The physicochemical characterization of the recirculated water of the dual biofilter system is presented in Figure 4.19. The pH of the T1 and T2 recirculated water were not significantly different in the level of 7 and stay constant during the experiment period (Figure 4.19a). The lower pH stimulates ammonia uptake in the filter, but adverse effects on the odor-removing bacteria (Nielsen et al., 2008).

The electrical conductivity increased in line with time during the experiment on both T1 and T2. The electrical conductivity of T1 increased from 3.36 mS/cm at the beginning to 14.40 mS/cm at the end of the experiment. The electrical conductivity of T2 increased from 1.22 mS/cm at the beginning to 13.72 mS/cm at the end of the experiment (Figure 4.19c). The increasing of the electrical conductivity within time was likely because of the accumulation of ammonia nitrogen in the recirculated water. Total dissolved solids of the recirculated water also increased during the experiment on both T1 and T2 treatment.

The total dissolved solids of T1 recirculated water increased from 208.33 ppm at the beginning to 493.83 ppm at final. The total dissolved solids of T2 recirculated water increased from 204 ppm at the beginning to 398.33 ppm at the end of the experiment. The increasing of the TDS in the circulated water was likely because of the particle matter emitted from the pig house filtered in the biofilter pad and was carried by the water that was recirculated. There was

no standard yet how much the acceptable TDS concentration might be allowed in the recirculated water, however higher TDS concentration might increase the possibility for the biodegradation process in the recirculated water that further might result in the other problem such as odor production. Another possible explanation of higher TDS could be related to the increasing of the microbial biomass that was carried from the biofilter pad. The dissolved oxygen concentration of the dual biofilter recirculated water is presented in Figure 4.17d. The dissolved oxygen level in both of T1 and T2 recirculated water was constant during the experiment. The average dissolved oxygen level in both of T1 and T2 recirculated water were not significantly different ($p>0.05$). The average oxygen level in T1 and T2 are 14.22 mg/L and 14.84 mg/L respectively. According to the results, it can be inferred that co-additive supplementation did not affect the biological recirculated water performance.

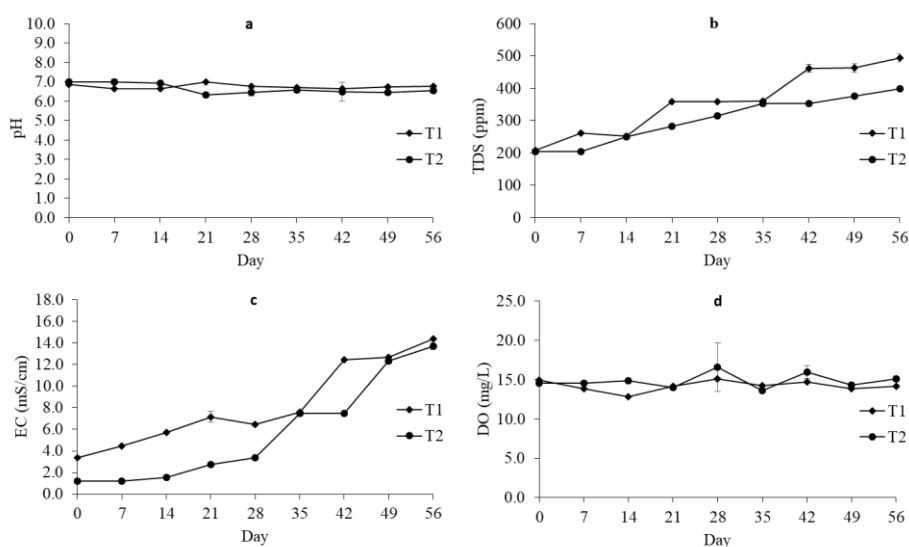


Figure 4.19. Biofilter recirculated water characteristic during the experiment (a) pH, (b) EC, (c) TDS, (d) DO. Values are expressed as mean \pm standard deviation

The nitrogen fraction characteristic of the recirculated water during the experiment was observed and is presented in Figure 4.20. Total ammonia nitrogen (TAN) of the recirculated water in T1 significantly increased ($p<0.05$) from 516.67 mg/L at the beginning to 3800.65 mg/L at the end day of the experiment (Figure 4.20a). TAN concentration of the recirculated water in T2 increased from 188.33 mg/L to 1450 mg/L. The final TAN concentration of T1 recirculated water was significantly higher ($p<0.05$) than that of T2. The higher TAN could be related to the higher ammonia loading rate emitted from the pig house during the experiment. Nielsen et al., (2008) studied to drain off the wastewater frequently to keep the total concentration of nitrogen compounds below 6,800 mg/L in the recycled water and resulted in the ammonia reduction more than 70% after passing the biofilter.

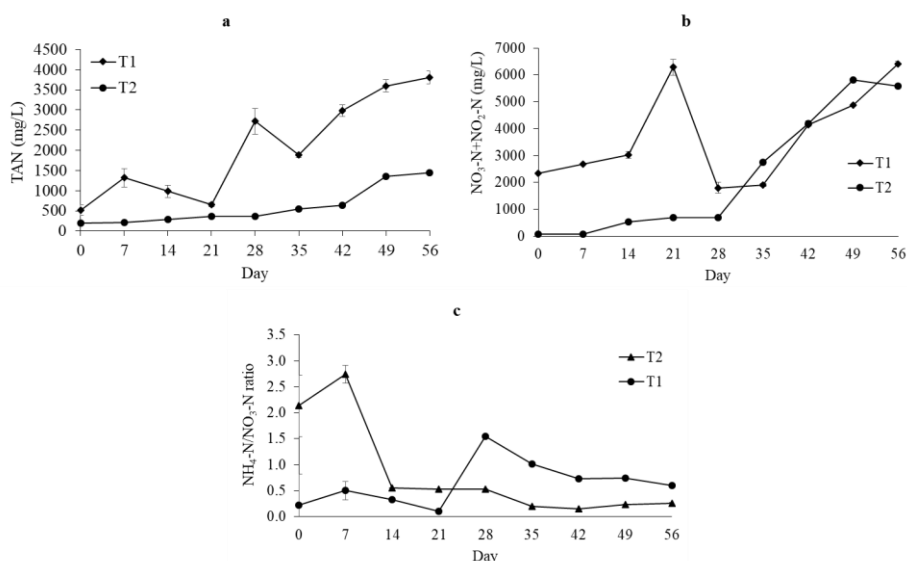


Figure 4.20. Nitrogen fraction characteristics of biofilter recirculated water during the experiment (a) TAN, (b) NO₃+NO₂; (c) NH₄⁺/NO₃

In this study, the recycled water amount reduced after certain time, due to the evaporation, thus to maintain the low nitrogen concentration, the recycled water

volume was maintained by automatic leveling system in the level of 130 L. However, the concentration of nitrogen compounds still increased along with operation time as seen in Figure 4.20.

The $\text{NO}_3^- + \text{NO}_2^-$ concentration of the recirculated water during the biofilter performance experiment is presented in Figure 4.20b. During the experiment, the $\text{NO}_3^- + \text{NO}_2^-$ concentration in the T1 and T2 recirculated water increased gradually. The $\text{NO}_3^- + \text{NO}_2^-$ concentration of T1 recirculated water was 2,336.34 mg/L initially and increased to 6,420.05 mg/L at the end of the experiment. The $\text{NO}_3^- + \text{NO}_2^-$ concentration of T2 recirculated water was 88.77 mg/L initially and increased to 5,590.57 mg/L at the end of the experiment. The increased $\text{NO}_3^- + \text{NO}_2^-$ concentration in both treatments was likely because of nitrification process going on the recirculated water resulting in the accumulation of in NO_3^- and NO_2^- .

Table 4.7. Pearson correlations of measured recycled water physicochemical variables during the experiment

Pearson correlation	pH	EC	TDS	DO	TAN	$\text{NO}_3^- + \text{NO}_2^-$	$\text{NH}_4^+ / \text{NO}_3^-$
pH	1	-0.30	-0.30	-0.11	-0.03	-0.17	0.54*
EC	-0.30	1	0.92**	-0.12	0.77**	0.87**	-0.40
TDS	-0.30	0.92**	1	0.003	0.82**	0.75**	-0.35
DO	-0.11	-0.12	0.003	1	-0.17	-0.17	0.04
TAN	-0.03	0.77**	0.82**	-0.17	1	0.55*	-0.04
$\text{NO}_3^- + \text{NO}_2^-$	-0.17	0.87**	0.75**	-0.17	0.55*	1	-0.57*
$\text{NH}_4^+ / \text{NO}_3^-$	0.54*	-0.40	-0.35	0.04	-0.04	-0.57*	1

**Correlation is significant at the 0.01 level

*Correlation is significant at the level 0.05 level

The $\text{NH}_4^+ / \text{NO}_3^-$ ratio of the recirculated water in the dual biofilter system during the co-additive supplementation is presented in Figure 4.20c. The $\text{NH}_4^+ / \text{NO}_3^-$ ratio of the T1 was relatively constant during the experiment, it

increased in the middle of the experiment time (day 21-28) to the level of 1.55 and decreased to the level of 0.60 at the end of the experiment. The $\text{NH}_4^+/\text{NO}_3^-$ ratio of the T2 decreased rapidly in day 14 of the experiment from 2.74 to the level of 0.26 at the end of the experiment. The decreased of the $\text{NH}_4^+/\text{NO}_3^-$ ratio is likely because of higher nitrification rate occurred and higher nitrate accumulation in the recirculated water. According to the results showed in Figure 4.20, the co-additive supplementation did not give significant effect to the recirculated water characteristic fluctuation.

Pearson coefficient correlation of measured recycled water's physicochemical variables during the experiment is illustrated in Table 4.6. According to the results, the highest correlation between the physicochemical variables of the recirculated water showed between EC and TDS ($r = 0.92$), followed by EC and $\text{NO}_3^- + \text{NO}_2^-$ ($r = 0.87$). The pH had a significantly positive correlation with $\text{NH}_4^+/\text{NO}_3^-$ ($r = 0.54$). According to the results from the present study, it can be inferred that the pH and EC might be used in the field as variables for monitoring the recycled water quality condition for the biofilter system since these variables had a significant correlation with the other important parameters which could not be simply analyzed.

4.5. Conclusion

The present study investigated the performance of dual biofilter system with co-additive (FGD gypsum and biochar) to reduce the ammonia and volatile organic acid odorant emission. The ammonia removal efficiency of the dual biofilter system supplemented by the co-additive (5% FGD gypsum and 5% biochar) resulted in the ammonia removal efficiency about 65.28% to 98%.

According to the results of this study, the co-additive supplementation in the biofilter media might increase the ammonia removal efficiency of about 57.72%. The role of FGD gypsum supplementation in the biofilter media was controlling the pH and supply the micronutrient for the microbes, while biochar might have a role to enhance the absorbance capacity in the biofilter media. The VFA odor emitted from the pilot pig house was not increased after passing the dual biofilter supplemented by co-additive supplementation. The co-additive supplementation in the biofilter media might delay the phenolic-indolic odor dispersion from the pilot pig house. However, the emission still cannot be reduced. The co-additive supplementation did not give effect to the biofilter performance to reduce sulfuric odor emitted from the pig house. The co-additive supplementation might potent to inhibit the emission of volatile organic odor from the pig house with the average removal efficiency of about 21.36%. Based on the results of the present study, among the odorant compounds, the ammonia was the most significant odor that can be removed by the biofilter and the performance was significantly improved by co-additive supplementation.

Results from this study indicated that most of the VOCs had the highest correlations with the volatile fatty acids. Based on the results of the current study, it illustrated that total VOA had a significantly positive correlation with the total VFA since the most VOA odorant consists of VFA odor compound. The total sulfuric had significantly positive correlation with total phenolic ($r = 0.37$) and with ammonia ($r = 0.34$). The co-additive supplementation might improve the porosity of the biofilter media, hence improve the odor reduction

capability. It can be inferred that co-additive supplementation might improve nitrification process in the biofilter media. In addition, since it contains higher nitrite nitrate concentration, the biofilter media might also further be considered as a such bio-fertilizer product source for land application. The co-additive supplementation might increase the plant macro and micronutrient concentration of the biofilter media, especially the total P, K, Ca, Mg, S, and Fe. The co-additive supplementation did not result in a significant effect to the total bacterial population in the biofilter media. The co-additive supplementation to the biofilter media might induce the yeast and molds growth.

TS content of the biofilter media had significantly positive correlation with TAN ($r = 0.58$) and $\text{NH}_4^+/\text{NO}_3^-$ ($r = 0.47$) whilst had significantly negative correlation with BD ($r = -0.34$). The biofilter media's VS had significantly positive correlation with PD ($r = 0.51$), TAN ($r = 0.51$), and $\text{NH}_4^+/\text{NO}_3^-$ ($r = 0.62$). Pearson correlation matrix showed the highest positive correlation between TAN and $\text{NH}_4^+/\text{NO}_3^-$ ($r = 0.96$), whilst the higher negative correlation were between VS and EC ($r = -0.88$). According to the results, it can be inferred that co-additive supplementation did not affect the biological recirculated water performance. The co-additive supplementation did not give significant effect to the recirculated water characteristic fluctuation. The highest correlation between the physicochemical variables of the recirculated water showed between EC and TDS ($r = 0.92$), followed by EC and $\text{NO}_3^- + \text{NO}_2^-$ ($r = 0.87$). The pH had a significantly positive correlation with $\text{NH}_4^+/\text{NO}_3^-$ ($r = 0.54$). According to the results from the present study, it can be inferred that the pH and EC might be used in the field as variables for monitoring the recycled water quality

condition for the biofilter system since these variables had a significant correlation with the other important parameters which could not be simply analyzed. The microbial diversity which plays important role in the biofilter media by the co-additive supplementation is necessary to be further investigated. Based on the results of this study, it was concluded that the co-additive (FGD gypsum and biochar) supplementation to the biofilter media might become the option to reduce the odor emission from the livestock facilities. The performance of the dual biofiltration system supplemented by the co-additive in the larger scale is necessary to investigate its effectiveness and evaluate its economic visibility. We also suggest that optimizing strategy for the removal of low water-soluble components such as sulfuric compound might improve the performance of dual biofilter in reducing the odor emitted from swine livestock facilities.

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CHAPTER 5 . CONCLUSION

5.1. General summary

The expansion of animal feeding scale and density of operations led to the increasing of offensive odorants emission in animal housing facilities (Jo et al., 2015). The livestock facilities (such as pig barns, cattle feedlots, and poultry buildings) are commonly known as unpleasant odors sources (Kim and Choi, 2013). Effort to reduce and mitigate the odor emission from the livestock facilities is necessary. This dissertation studied the role of co-additive consisted of the FGD gypsum and biochar mixture to reduce the odor pollutant from livestock facilities especially for ammonia and VOA odor (Figure 5.1). As a result, the following conclusions were obtained.

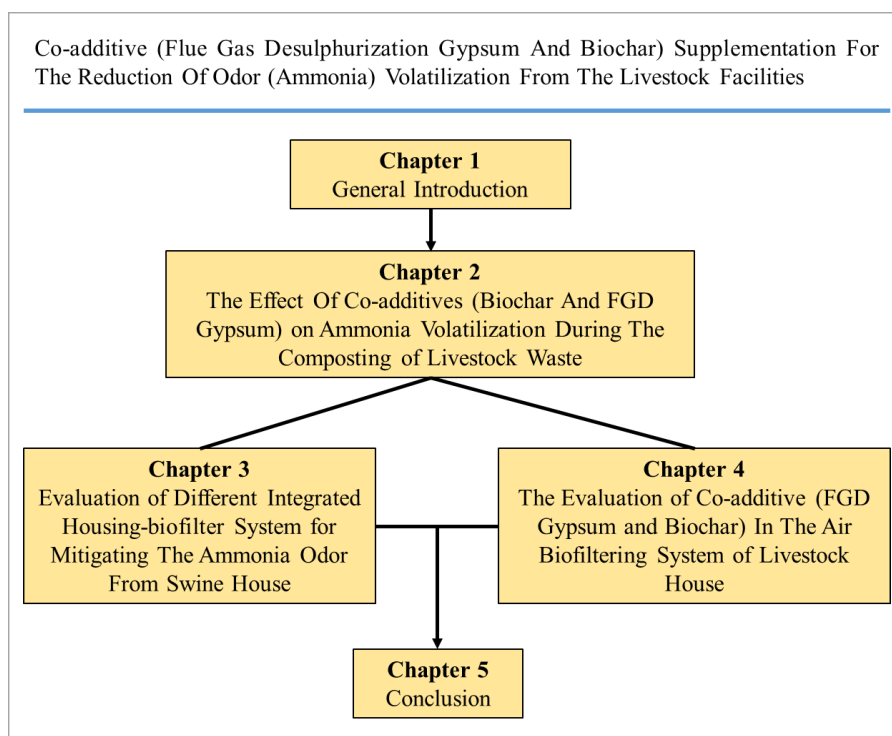


Figure 5.1. Scheme of each chapter in this thesis

The co-additives treatment using FGD gypsum and biochar did not give adverse effect during the 28 days livestock waste composting process. The selection of appropriate additives needs to be adjusted for the purpose of composting process. FGD gypsum supplementation may reduce ammonia volatilization more than biochar supplementation, however, biochar supplementation may give a faster biodegradation process during composting. Thus co-additive treatment during composting might be an alternative method to reduce ammonia volatilization as well as induce the biodegradation process.

Supplementation of livestock waste composting with the co-additives FGD gypsum and biochar reduced ammonia loss and enhanced nutrient recovery. The utilization of these co-additives led to the reduction of ammonia volatilization by 26–59% and an increase in the nitrate (NO_3^-) accumulation by 6.7–7.9 fold compared to the initial value. The total ammonia volatilization of the intermittent aeration treatment was lower than that of continuous aeration using co-additive treatments. The supplementation of composting with these co-additives enhanced the macro- and micro-nutrient content in the compost product. The use of co-additives did not have any negative effect on the compost respiration rate and reduced the phytotoxic effect. Some parameter showed that livestock waste composting using co-additives during the 28 days results in waste that is safe to be discharged to the environment, however, the waste will not have matured yet to be utilized as compost or organic fertilizer. Studies on the co-additive application on the pilot and large-scale composting are necessary to investigate the effectiveness and economic visibility of this method.

The performance of two different model of the integrated ventilation-biofilter system on reducing the air pollutant emission from swine house was evaluated. Results show that pit ventilation system integrated with the horizontal biofilter emitted lower air pollution rather than wall ventilation integrated to vertical biofilter system. The ammonia concentration inside the W-VBF was about 41% higher than that of the P-HBF. The particulate matter inside the W-VBF was about 62% higher than that of P-HBF. The total suspended particle in the room area correlated positively with the concentration of ammonia odor and volatile organic acid odorant. Noise from the W-VBF was about 12.1% more than that of P-HBF. Particulate matter and Total suspended particle inside the W-VBF house was about 62.2% and 69.9% respectively higher than that of P-HBF. The P-HBF application could maintain the optimum temperature condition better than that of W-VBF for swine house during the cold condition. According to the results, it was concluded that integration design of ventilation system and biofilter system of swine house might be used as the option to minimize the air pollution produced by the swine farming activity as well as maintain optimum microclimate condition for the pig.

The ammonia removal efficiency of the dual biofilter system supplemented by the co-additive (5% FGD gypsum and 5% biochar) resulted the ammonia removal efficiency about 65.28% to 98%. According to the results of this study, the co-additive supplementation in the biofilter media might increase the ammonia removal efficiency of about 57.72%. The role of FGD gypsum supplementation in the biofilter media was to control the pH and supply the micronutrient for the microbes. While biochar might have a role to enhance

the absorbance capacity in the biofilter media. The VFA odor emitted from the pilot pig house was not increased after passing the dual biofilter supplemented by the co-additive supplementation. The co-additive supplementation in the biofilter media might delay the phenolic-indolic odor dispersion from the pilot pig house, however, the total emission still cannot be reduced. The co-additive supplementation did not give adverse effect to the biofilter performance reducing sulfuric odor emitted from the pig house. The co-additive supplementation might potent to inhibit the emission of volatile organic odor from the pig house with the average removal efficiency of about 21.36%. Based on the results of the present study, among the odorant compounds, the ammonia was the most significant odor that can be removed by the biofilter and the performance was significantly improved by co-additive supplementation.

Results from this study indicated that most of the VOCs had the highest correlations with the volatile fatty acids. Total VOC had a significantly positive correlation with the total VFA since the most VOC odorant consists of VFA odor compound. The total sulfuric had a significantly positive correlation with total phenolic and with ammonia odor. The co-additive supplementation might improve the porosity of the biofilter media, hence improve the odor reduction capability. It can be inferred that co-additive supplementation might improve nitrification process in the biofilter media. In addition, since it contains higher nitrite nitrate concentration, the biofilter media might also further be considered as such bio-fertilizer product sources for land application. The co-additive supplementation might increase the plant macro and micronutrient concentration of the biofilter media, especially the total P, K, Ca, Mg, S, and

Fe. The co-additive supplementation did not result in a significant effect to the total bacterial population in the biofilter media. The co-additive supplementation to the biofilter media might induce the yeast and molds growth.

TS content of the biofilter media had significantly positive correlation with TAN and $\text{NH}_4^+/\text{NO}_3^-$. Whilst had a significantly negative correlation with bulk density. The highest positive correlation was shown between TAN and $\text{NH}_4^+/\text{NO}_3^-$, while the highest negative correlation was between VS and EC. According to the results, it can be inferred that co-additive supplementation did not affect the biological recirculated water performance. The co-additive supplementation did not give significant effect to the recirculated water characteristic fluctuation. The highest correlation between the physicochemical variables of the recirculated water showed between EC and TDS, followed by EC and $\text{NO}_3^- + \text{NO}_2^-$. The pH had a significantly positive correlation with $\text{NH}_4^+/\text{NO}_3^-$. According to the results from the present study, it can be inferred that the pH and EC might be used in the field as variables for monitoring the recycled water quality condition for the biofilter system since these variables had a significant correlation with the other important parameters which could not be simply analyzed.

The microbial diversity which plays an important role in the biofilter media by the co-additive supplementation is necessary to be further investigated. Based on the results of this study, it was concluded that the co-additive (FGD gypsum and biochar) supplementation to the biofilter media might become the option to reduce the odor emission from the livestock facilities. The performance test of the dual biofilter system supplemented by

the co-additive in the larger scale is necessary to investigate its effectiveness and evaluate its economic visibility. We also suggest that optimizing strategy for the removal of low water-soluble components such as sulfuric compound might improve the performance of dual biofilter in reducing the odor emitted from swine livestock facilities.

5.2. Overall conclusion and implication

According to the overall resulted in these studies, it was concluded the co-additive supplementation could be an alternative option to reduce the odor emission from the livestock facilities. An amount of 10% co-additive (FGD gypsum 5% and biochar 5%) could be used as co-additives in livestock waste composting as well as in the biofilter packing material for mitigating the odor emission from the livestock facilities. Co-additive (FGD gypsum 5% and biochar 5%) in the livestock composting reduced ammonia volatilization during the composting process, increased the nitrate accumulation in compost product, didn't give an adverse effect to composting process, and environmentally save. Co-additive (FGD gypsum 5% and biochar 5%) in the biofilter material increased the ammonia removal efficiency, inhibit the volatile organic compounds emission, increased the macro and micronutrient of used biofilter material for the land application.

Further research related to the ammonia oxidizing bacteria and the microbial community in the livestock waste composting and the biofilter media with the co-additive supplementation is necessary for finding the optimum strategies to reduce odor emission from the livestock facilities. Development of odor absorber biomaterial based on the mixture of FGD gypsum and biochar

can be further investigated. The application simplicity and economical visibility should also be considered in implementation of co-additives technology to reduce odor from livestock facilities.

Abstract in Korean

병합재(Flue gas desulphurization gypsum과 biochar) 바이오필터가 축산시설 악취(암모니아 취산) 저감에 미치는 영향

요약(국문초록)

가축 수의 증가는 축산 폐기물 양을 단계적으로 증가시킨다. 축산 활동(거름이나 도축 폐기물)의 폐기물은 인간들과 동물들 모두에게 영향을 줄 수 있는 부정적인 환경 영향들(예를 들어, 온실 가스 방출이나 과영양화, 염 축적, 병원성 미생물의 증가, 악취 등)의 주요 원인이다. 실제로, 목축업 운영으로 발생하는 악취와 가스 방출이 인류의 건강과 삶의 질에 미치는 악영향 때문에 중요한 사회적 문제가 되고 있다고 제기 되어왔다. 돈사로부터 발생하는 가축 분뇨는 다른 동물 종에 비해 특히 심각한 문제이다. 더 나아가, 돼지 축산 시설로부터의 악취와 휘발성 유기 화합물(VOCs)의 방출은 대표적인 대기 오염 문제들이며, 중대한 공공 보건 문제가 되었다. 이에 따라, 축산 활동들에 영향 받은 공해를 감소시키는 노력에 대한 연구들이 수행 되었다.

연구의 첫 단계로 축산 폐기물 퇴비화(대기 오염 감소와 양분 보존)를 향상시키는 보조 첨가제들의 효과가 조사되었다. 바이오차와

배연탈황석고(FGD gypsum)들은 도축 폐기물, 돼지 슬러리 그리고 톱밥 배합물 퇴비화에 보조제로 사용되었다. 첨가제의 상이한 조성(각각 0%, 5%, 10%의 바이오차나 배연탈황석고)은 실험실 규모로 3 번씩 실험되었다. 덧붙여, 두개의 다른 공기 주입법(연속적 그리고 비연속적)의 효과 또한 조사되었다. 이 결과들은 축산 폐기물 부식 시 보조 첨가제(바이오 차나 배연탈황석고)의 사용이 암모니아 발생을 26~59% 감소시키고, 질소 축적을 6.7~7.9 배 증가시킨다고 나타났다. 보조 첨가제 사용 시 비연속적 공기 주입법에 의한 총 암모니아 휘발량은 연속적 공기 주입법을 사용했을 때보다 낮았다. 이는 보조 첨가제(바이오차 그리고 배연탈황석고)가 암모니아 발생을 감소시키고 양분 보존을 향상시키기 위해 축산 폐기물 퇴비화에 실용화될 수 있다고 결론내려진다.

이어서 실험적 돈사 내의 상이한 두 가지 종류의 통합 환기 바이오 필터 장치의 성능을 조사했다. 수직 바이오필터(W-VBF)로 통합된 벽형 환기 장치와 수평 바이오필터(P-HBF)로 통합된 바닥형 환기장치가 시험되었다. 실험은 64 일간 수행되었다. 바이오 필터 장치내에 순환 여과가 사용되었다. 미기후 변수들, 공기 오염 물질(미세먼지와 악취) 그리고 폐수(슬러리와 바이오 필터 재사용수) 특징이 실험 중 관찰되었다. 결과들은 W-VBF 내 암모니아 농도가 P-HBF 내의 농도보다 약 41% 높다고 나타났다. W-VBF 에서 P-HBF 보다 소음이 약 12.1% 더 발생되었다. W-VBF 내 미세 먼지와 총

부유 입자량은 P-HBF 보다 각각 62.2%, 69.9% 가량 높았다. 이는 P-HBF 가 W-VBF 에 비해 공기 오염을 적게 배출한다고 결론지어졌다. 덧붙여 이는 추울 때 돈사를 최적 온도 조건으로 유지시킬 수도 있다. 이러한 결과는 돈사의 통합 환기 바이오 필터 장치가 돼지를 위한 최적 미기후 조건을 유지할 뿐만 아니라 양돈에서 발생된 대기 오염을 최소화하는 선택이 될 수 있다고 시사한다.

세번째 단계에서는 폐쇄식 축사내 악취 방출과 보조 첨가제(배연탈황석고와 바이오차)가 첨가된 이중 바이오 필터 장치를 사용한 악취의 처리를 연구했다. 이중 바이오 필터 장치의 성능은 실험 축사 모델 하에 연구되었다. 8 주(56 일)의 실험일이 실험실 규모의 돈사에서 수행되었다. 기계적 벽형 환기 장치에 부착된 이중(수직과 수평) 바이오 필터 장치가 실험되었다. 순환 여과 장치가 바이오 필터 장치 내에 설치되었다. 미기후 변수, 악취 물질(암모니아와 휘발성 유기산), 바이오 필터 팩의 생화학물질, 그리고 폐수(바이오 필터 재사용수)의 특징들이 실험 중 관찰되었다. 보조 첨가제(5% 배연탈황석고와 5% 바이오차)로 보완된 이중 바이오 필터 장치의 암모니아 제거 효율은 약 65.25%에서 98%였다. 바이오 필터 담체에 보조 첨가제를 첨가하여 암모니아 제거 효율을 약 57.72% 증가시킬 수 있을 것이고, 약 21.36%의 평균 제거율로 돈사의 휘발성 유기 악취의 방출을 강력하게 방지할 수 있다. 바이오 필터 담체 내 미생물 활동뿐 아니라 질화 과정 역시 보조 첨가제 첨가로 향상될 수 있다. 덧붙여, 이는

더 높은 아질산염, 질산염 농도와 다량, 미량 영양소(P, K, Ca, Mg, S, Fe 모두) 유효도를 가지기에 바이오 필터 담체는 더 나아가 토양환원을 위한 견고한 바이오 비료 물질로도 고려될 수 있다. 결과들은 배연탈황석고와 바이오차가 첨가된 이중 바이오 필터 장치가 보조 첨가제로 보완되지 않은 바이오 필터에 비해 대기 오염 물질을 덜 배출시킨다고 나타낸다. 이는 폐쇄식 축사 내 이중 바이오 필터 장치의 통합이 축산 활동에서 생산되는 대기 오염을 최소화하는 선택지가 될 수 있다고 결론 지어진다.

이러한 연구들의 전반적인 결과에 따라, 보조 첨가제 첨가는 축산 시설의 악취 배출을 줄이는 대안이 될 수 있다고 판단된다. 10%의 보조 첨가제(5% 배연탈황석고와 5% 바이오차)는 축산 시설의 악취 배출을 완화시키는 바이오 필터 구성물이자 축산 폐기물 비료화를 위한 보조 첨가제로 사용될 수 있다. 축산 비료화 시 보조 첨가제(5% 배연탈황석고와 5% 바이오차)는 암모니아 휘발을 감소시키고, 퇴비의 질산염 축적을 증가시키며, 비료화 과정에 부정적인 영향을 주지 않으며 환경을 보호한다. 바이오 필터 물질 내 보조 첨가제(5%배연탈황석고와 5% 바이오차)는 암모니아 제거 효율을 신장시키며, 휘발성 유기 화합물 배출을 방지하며, 토양환원을 위한 사용된 바이오 필터 물질의 다량/미량 영양소를 증가시킨다.

주요 단어: 축산 폐기물, 비료화 첨가제 조합물, 암모니아, 휘발, 대기
오염, 악취, 바이오 필터, 돈사, 휘발성 유기산

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