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Greenhouse Gas Emissions from Composting and Anaerobic Digestion Plants

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Summary

Almost all biowaste collected in Germany is treated either by composting or by anaerobic digestion (AD). Both treatments result in greenhouse gas (GHG) emissions. The main focus of the thesis was to quantify the contribution of composting and anaerobic digestion plants to global warming. Data collection and measurements were performed at 9 composting plants and 9 AD plants treating biowaste. In the thesis, GHG emission factors were calculated in g per Mg fresh waste.

GHG emissions from composting plants were measured by using a tunnel method, which used a plastic tunnel covering a complete part of a windrow. The tunnel was ventiled by 2 vetilators. The concentration differences of GHG between incoming and outgoing air were used to calculate the emission from the waste under the tunnel. In average, composting resulted in 4060g (Mg biowaste)$^{-1}$ CH_4 emissions, 55g (Mg biowaste)$^{-1}$ N_2O emissions and 157g (Mg biowaste)$^{-1}$ NH_3 emissions, giving a total emission factor of 118kg CO_2 equivalents (Mg biowaste)$^{-1}$.

The emission sources at the AD consisted of biofilters, combined heat and power units (CHP), liquid digestate treatment systems (LTS) and open composting windrows of the solid digestate. In average, the biofilters removed 30% of total organic carbon (TOC), 50% of non methane volatile organic carbon (NMVOC) and 51% NH_3, whereas N_2O concentrations increased by 26%. For CH_4 the biofilters had only a small removal effect (6%). Overall, the emission factors were reported as 1.2-16kg (Mg biowaste)$^{-1}$ for CH_4, 9-172g (Mg biowaste)$^{-1}$ for N_2O and 41-6,031g (Mg biowaste)$^{-1}$ for NH_3, giving total emission factors from 50 to 457 kg CO_2 equivalents (Mg biowaste)$^{-1}$. The post-treatment of solid digestate resulted in highest GHG emissions.

In order to reduce GHG emissions from composting windrows, small windrows with high turning frequencies and a short composting duration would result in the lowest emissions. For anaerobic digestion plants, potentially there are great GHG emissions benefits if biowaste is treated by wet anaerobic digestion. Wet digestion results in liquid digestate only, which should be directly applied without being composted.

Zusammenfassung (summary in German)

Fast alle gesammelten Bioabfälle werden in Deutschland entweder durch Kompostierung oder durch Vergärung behandelt. Bei der Behandlung entstehen Treibhausgase (THG) wie Methan und Lachgas. Ziel der Arbeit war es, diese Emissionen zu quantifizieren.

Messungen erfolgten an 9 Kompostierungsanlagen (KoA) und 9 Vergärungsanlagen (VA). THG-Emissionen von KoA wurden mit Hilfe einer Tunnel-Methode, bei der ein kompletter Teil einer Miete mit einem Kunststoff-Tunnel abgedeckt wurde, gemessen. Der Tunnel wurde mit Hilfe von zwei Ventilatoren belüftet. Die Konzentrationsdifferenzen der Treibhausgase zwischen Zu- und Abluft wurden genutzt, um die Emission der Abfälle unter dem Tunnel zu bestimmen. Im Mittel führte die Kompostierung zu 4.060g (Mg Bioabfälle)$^{-1}$ CH_4 Emissionen, 55g (Mg Bioabfälle)$^{-1}$ N_2O Emissionen und 157g (Mg Bioabfälle) $^{-1}$ NH_3 Emissionen. Daraus ergibt sich ein Emissionsfaktor von 118kg CO_2 -Äquivalenten (Mg Bioabfälle)$^{-1}$.

Die Emissionsquellen an VA waren der Biofilter, das Blockheizkraftwerk (BHKW), die flüssigen Gärrest-Behandlungssysteme und die offenen Kompostmieten der festen Gärreste. Im Durchschnitt reduzierten die Biofilter den gesamten organischen Kohlenstoffs (TOC) um 30%, bzw um 50% des Non-Methane Voltile Organic Carbon (NMVOC). CH_4 wurde am Biofilter nur sehr wenig abgebaut (6%), dagegen reduzierte sich die NH_3-Fracht um 51%. Die N_2O Emissionen wurden um 26% erhöht. Insgesamt wurden 1,2-16kg CH_4 (Mg Bioabfälle)$^{-1}$, 9-172g N_2O (Mg Bioabfälle)$^{-1}$ und 41-6.031g NH_3 (Mg Bioabfälle)$^{-1}$ emittiert. Daraus ergibt sich ein Emissionsfaktor von 50 bis 457 kg CO_2 -Äquivalenten (Mg Bioabfälle)$^{-1}$. Die höchsten Emissionen wurden in der Nachbehandlung der festen Gärreste freigesetzt.

In der Kompostierung können die Emissionen durch kleine Mieten, öfteres Umsetzen und eine kürzere Behandlungsdauer reduziert werden. Die geringsten Emissionen an THG sind von der Nassvergärung ohne Separierung zu erwarten. Die Nachbehandlung von Gärresten führt zu hohen Treibhausgas-Emissionen. Deshalb sollte Gärrest ohne vorherige Kompostierung direkt angewendet werden.

Erklärung (Declaration)

Ich versichere, dass ich diese Arbeit selbständig verfaßt habe, keine anderen Quellen und Hilfsmateralien als die angegebenen benutzt und die Stellen der Arbeit, die anderen Werken dem Wortlaut oder dem Sinn nach entnommen sind, kenntlich gemacht habe. Die Arbeit hat in gleicher oder ähnlicher Form keiner anderen Prüfungsbehörde vorgelegen.

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List of Abbreviations

AD	Anaerobic digestion
CHP	Combined heat and power
ECD	Electron capture detector
FID	Flame ionisation detector
GC	Gas chromatography
GHG	Greenhouse gas
IPCC	the Intergovernmental Panel on Climate Change
LCA	Life cycle analysis
LTS	Liquid treatment system
NMVOC	Non methane volatile organic carbon
TOC	Total organic carbon
TS	Total solid
VFA	Volatile fatty acid
VDI	Verein Deutscher Ingenieure (German engineers association)

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1. CH_4, N_2O AND NH_3 EMISSIONS FROM OPEN-WINDROW COMPOSTING IN GERMANY

Abstract

The study investigated CH_4, N_2O and NH_3 emissions from open composting plants treating biowaste, green waste and anaerobic digestion (AD) material. To assess the gaseous formation in the composting windrows, CH_4, CO_2, N_2O and O_2 volume concentrations were measured at different depths. The tunnel method was used to perform emission measurements. CH_4 concentrations in the tabular and trapezoidal windrows were much higher than in the triangular windrows. The emission factors of CH_4 were 4,060g Mg^{-1} for biowaste, 4,415g Mg^{-1} for green waste and 10,254g Mg^{-1} for AD material. The N_2O emissions were 55g Mg^{-1} for biowaste, 24g Mg^{-1} for green waste and 105g Mg^{-1} for AD material. The NH_3 emissions were 157g Mg^{-1} for biowaste, 2g Mg^{-1} for green waste and 699g Mg^{-1} for AD material.

1.1 Introduction

Biowaste and green waste are treated in Germany either by composting or by a combination of anaerobic digestion and composting (Jäckel *et al.*, 2005; Font *et al.*, 2011). In recent years, composting has become a promising technique because operating costs and technology requirements are low (Barth, 2006). However, composting plants which treat biowaste may create environmental problems such as gas emissions and unpleasant odours. In a composting plant, composting windrows generate undesirable gases such as CH_4, N_2O and NH_3 (Illmer, 1996; Beck-Friis *et al.*, 2000; Hobson *et al.*, 2005; Font *et al.,* 2011). CH_4 and N_2O are considered to be strong greenhouse gases (GHGs), whereas NH_3 is identified as an odour component and an indirect GHG. According to the Intergovernmental Panel on Climate Change (IPCC, 2007), the global warming potential of CH_4 and N_2O in 100 years are respectively 25 and 298 times higher than CO_2.

GHG emissions during composting processes can be reduced by designing and managing windrows appropriately (Hellebrand, 1998). However, data concerning GHG emissions from composting windrows is limited and results are sometimes conflicting. The aim of this research is to quantify the emissions of CH_4, N_2O and NH_3 from different full-scale composting plants and to compare the emissions of different windrows. In addition, mitigation options are proposed for minimising emissions. Investigations were undertaken at nine composting plants in Germany treating biowaste and green waste, AD and green waste or just green waste.

1.2 Hypotheses/ Objectives

1.2.1 Hypotheses

It is hypothesised that composting windrows generate significant quantities of CH_4, N_2O and NH_3.

The size of the windrow, turning frequency and composting duration are key factors in reducing CH_4, N_2O and NH_3 emissions.

1.2.2 Objectives

The objectives were:

- to quantify the emissions of CH_4, N_2O and NH_3 from composting plants treating biowaste, green waste and AD material.
- to analyse GHG concentrations in windrows and emissions of them.
- to define mitigation options for reducing GHG emissions from composting plants.

1.3 General background

1.3.1 Generation of biowaste, green waste and AD material in Germany

1.3.1.1 Biowaste and green waste generation in Germany

According to VDI 3475 (2005), “biowaste is recyclable waste of animal or plant origin that is amenable to biodegradation by microorganisms”. Biowaste consists of garden waste, organic household waste, and waste from commerce and the food manufacturing industry (Boldrin *et al.*, 2009).

Green waste consists of woody materials, grasses, branches and leaves *etc.* from private gardens and public parks (Boldrin *et al.*, 2009).

In Germany, approximately 4 million Mg of green waste and 6-7 million Mg of biowaste are produced per annum. There are currently around 1,000 composting plants treating biowaste and green waste (Kranert *et al.*, 2010), of which 600 treat just biowaste (Weißenfel, 2001 as cited by Jäckel, 2005).

1.3.1.2 AD material generation in Germany

Anaerobic digestion treats organic waste and produces biogas. Usually, the organic waste is digested first in anaerobic thermophilic (at 50-55^{o}C) fermenters with a retention time of around 3 weeks. Biogas produced in the fermenter is stored and subsequently routed to gas engines for power generation. After the anaerobic digestion process, the residue is dewatered and the solid residues, called AD material, are post-treated by composting (Smet *et al.*, 1999). This post-treatment is essential for obtaining a stable end-product (Mata-Alvarez *et al.*, 2000). In fact, a considerable amount of CH_4 may be contained in this AD material (Lissens *et al.*, 2004).

1.3.2 Composting process

Composting is an aerobic biological process in which organic material is decomposed by microorganisms present in the organic material (Paul, 2009, Figure 1.1). The aims of composting are to achieve a rapid breakdown of organic substances and a hygienisation effect. The composting process consists of three phases: thermophilic, maturing and cooling. The thermophilic phase is characterised by high temperatures when fresh raw material is just mixed. This phase is important for reducing pathogens and bacteria. The maturing phase is necessary in order to achieve a stable product. In this phase, the material is moderately stable. The final phase of the process is the cooling phase, when the material gets very stable and mature (Paul, 2009).

Oxygen availability, C/N ratio and moisture are the most important parameters for composting processes (de Guardia *et al.*, 2008; Maeda *et al.*, 2010). In fact oxygen is required for the composting process because the microorganisms and bacteria consume oxygen to degrade the organic material. The products are CO_2, H_2O and heat (Xu *et al.*, 2007; Maeda *et al.,* 2010). High temperatures in the windrow kill worm eggs, pathogens and bacteria that may be health hazards to people or animals. If oxygen is limited during the active phase of composting, aerobic microbial activities may be inhibited and the temperature gradually decreases (Fernandes *et al.*, 1994). A C/N ratio of 15 to 30 is recommended for rapid composting (de Guardia *et al.,* 2008). A high C/N ratio limits microorganism activities, whereas a low C/N ratio leads to greater NH_3 emissions (He *et al.*, 2000; Pagans *et al.*, 2006; de Guardia *et al.,* 2008; Möller and Stinner, 2009). The optimum moisture content for composting is in the range of 50 to 60% (Poincelot, 1974), as cited by Fernandes *et al.,* (2007). A higher moisture content leads to higher GHG emissions because it creates anaerobic conditions (Tamura and Osada, 2006).

1.3.3 GHG formations and emissions

The production of CO_2, CH_4, N_2O and NH_3 is the result of microbial activities during the composting process. Composting is an aerobic treatment, but an anaerobic process in a composting windrow is unavoidable. CH_4 is formed due to insufficient diffusion of O_2 into the core of the windrow (Font *et al.,* 2011). It was also reported that CO_2 and CH_4 were highest in the centre of the windrows, where no O_2 was detected (Hao, 2001).

Nitrous oxide is a by-product of nitrification and denitrification found in both aerobic and anaerobic conditions (Paul, 2009). Anoxic conditions are defined as the absence of oxygen, but oxygen is still available in the form of NO_3^- and SO_4^{2-}. Under anaerobic conditions oxygen is absent. The emissions of N_2O are affected by temperature, nitrate content and the aeration rate (Hellebrand, 1998). The emissions of N_2O can be kept at a low rate if aerobic conditions prevail in the windrows (Hellebrand, 1998). N_2O produced during denitrification can be further reduced to N_2 (Hao, 2001). On the other hand, the aerobic nitrification of NH_3 to NO_2^- and NO_3^- may also generate N_2O as a by-product (Clemens and Ahlgrimm, 2001; Sánchez-Monedero *et al.*, 2010).

The pH value, NH_4^+/NH_3 equilibrium, quantity and mineralisation intensity of N-compounds, C/N ratio, temperature, dry matter content and wind speed are the most important factors influencing NH_3 emissions (Martins and Dewes, 1992; Hellebrand and Kalk, 2001a). During the composting process of animal excrement, 47-77% of the N content in the compost material is emitted in the form of NH_3 (Martins and Dewes, 1992). The elevation in temperature and pH increases NH_3 emissions. The emission of NH_3 is high immediately after the windrow is established and usually declines after a few weeks (Hansen *et al.*, 2006).

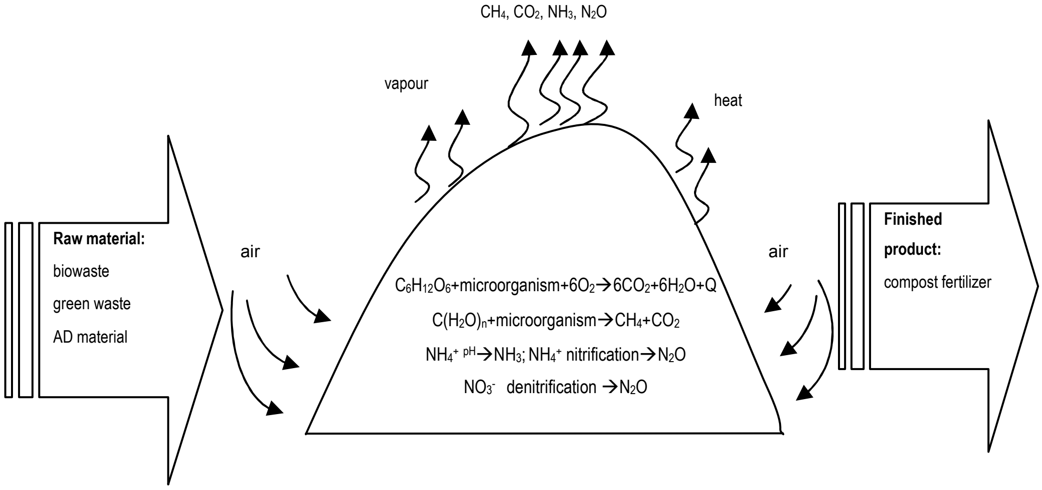

Figure 1.1: The composting process in a windrow

1.3.4 Methane oxidation in a composting windrow

Jäckel *et al.,* (2005) reported that 46-98% of the CH_4 produced in windrows is oxidised by methanotrophic bacteria before it escapes the windrow. The optimum temperature for methane oxidation in the windrow ranges from 45 to 55°C. The authors assumed that CH_4 is oxidised by microorganisms in the upper layer of the compost material. This has been confirmed by other studies (Hao, 2001; Szanto *et al.*, 2007; Andersen, 2010). CH_4 is oxidised as it moves through the material towards the surface. CH_4 oxidation is correlated with the availability of O_2.

Humer *et al.,* (2011) found that bulk density, organic matter, total organic carbon and nutrient content, water holding capacity and electricity conductivity influence methane oxidation efficiency as well. Furthermore, high NH_4 concentrations may inhibit CH_4 oxidation.

1.3.5 Open composting windrows

Composting plants generally use typical windrow geometries:

Triangular windrows: according to VDI 3457 (2005), triangular windrows are long and narrow at 1.5 to 4m wide and 1 to 2m high (Figure 1.2). Their triangular shape means that they have a larger ratio of surface to volume compared to trapezoidal and tabular windrows. The larger surface results in fairly high aeration due to natural convection. Heat is produced by respiration in the windrow. Ambient air is sucked into the windrow at the sides. The warm air with its lower density leaves the windrow at the highest point. This effect is called the “chimney effect”. Triangular windrows are turned frequently either by front-end loaders or specialised windrow turners. Turning frequencies vary depending on the substrate and composting process. Frequent turning during the first phase of the composting process supplies O_2 and increases homogeneity.

Trapezoidal windrows: these windrows are 4m to 10m wide and 2 to 4m high (Figure 1.2). The trapezoidal windrow can also be turned using similar technologies to those used with triangle windrows. They have a lower surface to volume ratio than triangular windrows. To improve the natural aeration rate, a high proportion of bulking agent is needed for ambient air to penetrate to the windrows (VDI 3475, 2005).

Tabular windrows: these windrows are 2 to 3m high and 10 to 50m wide (VDI 3475, 2005) (Figure 1.2). Tabular windrows are common in composting plants where space is limited. Maturation in a tabular windrow takes longer because of inadequate aeration.

Active aerated windrows: these windrows either press or suck air into the windrow to increase O_2 availability. In some composting plants, windrows are covered to protect them from rewetting or to reduce odour emissions (VDI 3475, 2005).

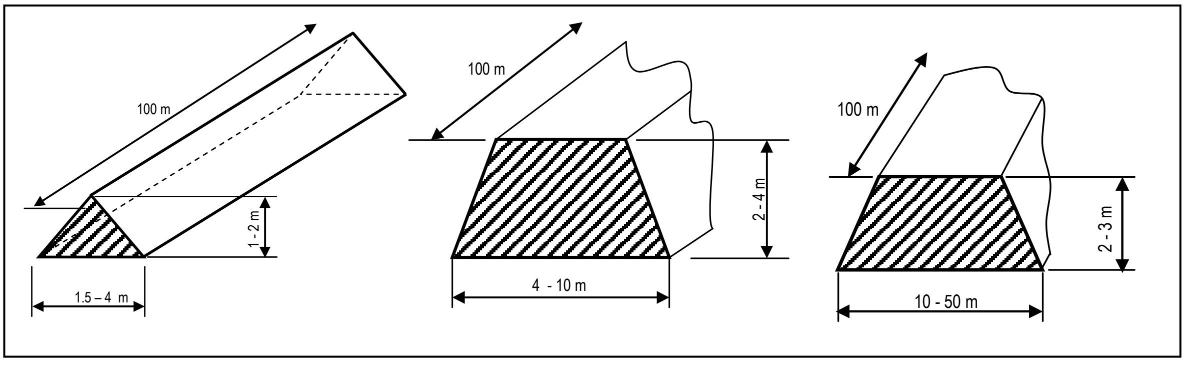

Figure 1.2: Typical compost windrows (VDI 3475, 2005): left to right: triangular windrow, trapezoidal windrow and tabular windrow

1.3.6 Greenhouse gases

According to Insam and Wett (2008), CH_4 is the major contributor to GHG emissions from waste treatment. Landfills and waste water are the largest sources of CH_4 emissions, accounting for 90% of CH_4 emissions linked to the waste sector. In developed countries, CH_4 produced in landfills is collected and used as a renewable energy source (Bogner *et al.*, 2008). In addition, N_2O and NH_3 also contribute to global warming (Table 1.1). The emission of N_2O from the composting of organic and green waste in Germany has been calculated to be in the range of 0.15 – 1.2% (Schenk, 1997).

Table 1.1: Atmospheric lifetime and GWP relative to CO_2 for several greenhouse gases (IPCC, 2007)

Gas name	**Chemical formula**	**Lifetime (years)**	**Global warming potential (GWP) for given time horizon**		
			20 years	100 years	500 years
Carbon dioxide	CO_2	30-95	1	1	1
Methane	CH_4	12	72	25	7.6
Nitrous oxide	N_2O	114	289	298	153
Ammonia (indirect GHG)	NH_3	-	-	2.98	-

1.3.7 Quantification of GHG emissions from composting windrows

1.3.7.1 Static chambers

Static chambers are widely used to measure CH_4 and N_2O emissions (Czepiel *et al.*, 1996; Clemens *et al.*, 1997; Hellmann, 1997; Schenk, 1997; B. Beck Friis, 2000; Hao, 2000; Sommer, 2000; Hellebrand and Kalk, 2001; Hobson *et al.,* 2005; Chiumenti *et al.*, 2007; Chang *et al.*, 2009; Andersen, 2010; Sánchez-Monedero *et al.,* 2010). A chamber is placed on top of a windrow and inserted approximately 5-10cm deep into the windrow to seal the chamber against atmospheric influences. The chamber covers only a small part of the windrow's surface (usually up to $1m^2$). Emitted gases are captured inside the chamber. The emission rate is determined from the increase of gas concentration under the cover.

1.3.7.2 Flux chambers

This method is an improvement of the static chamber method. Air is continuously flushed through the chamber by a small ventilator. Gas concentrations in the incoming and outgoing air and the flow rate are used to calculate emission rates. Flux chambers are usually the same size as static chambers (Schenk, 1997; Beck-Friis *et al.,* 2000; Hao, 2001; Hao, 2007; Xu *et al.,* 2007; Andersen, 2010).

1.3.7.3 Funnel method

This method was described by Andersen (2010). An upside-down funnel covers $1m^2$ of a windrow and a vent pipe is attached to the top of the funnel. Gases emitted from the windrow escape from the funnel through the vent pipe. The flux is calculated by gas concentration and air flow velocity in the vent pipe.

1.3.7.4 Large open dynamic chamber

The large open dynamic chamber can be regarded as a large flux chamber. The cover is usually tunnel shaped. The tunnels were used in studies of gas emissions from municipal solid waste (MSW) and farmyard manure composting (Wolter *et al.*, 2004; El Kader *et al.*, 2007; Amlinger *et al.,* 2008; Park *et al.*, 2011). Table 1.2 shows the emission factors of CH_4, N_2O and NH_3 for green waste, organic household waste, biowaste, municipal solid waste and animal manure derived from previous studies.

Table 1.2: Review of previous studies of GHG emission quantification conducted both in the laboratory and at different composting plants: green waste (GW), organic household waste (OHW), biowaste (BW); municipal solid waste (MSW), animal manure (AM)

Authors	**Method performed for the quantification of GHG emissions**	**Waste**	**CH_4 (g Mg^{-1} ww)**	**N_2O (g Mg^{-1} ww)**	**NH_3 (g Mg^{-1} ww)**	**CO_2 equiv. (kg Mg^{-1} ww)**
(Andersen, 2010)	Flux chamber method	GW	2,400±500	60±30	nd	111±30
(Amlinger *et al.*, 2008)	Large open dynamic chamber	GW	49-604	25-178	25-354	9-68
(Hellebrand, 1998)	Concentration and transport model	GW	4939	53.38	nd	139.38
(Fisher, 2006)	nd	BW	30.3	16.5	nd	5.67
(Smet *et al.*, 1999)	Pilot scale composting bins	BW	nd	nd	152	nd
(Amlinger *et al.*, 2008)	Large open dynamic chamber	BW	243-293	27-116	52-576	14-41
(Clemens and Cuhls, 2003)	Large open dynamic chamber	MSW	$6\text{-}12.10^3$	1.44-378	18-115	0.63-413
(Andersen *et al.*, 2010)	Flux chamber method	OHW	400-4,200	300-550	nd	100-239
(Amon *et al.*, 2006)	Large open dynamic chamber	AM	nd	24-56	201-552	14-64

ww: wet waste

nd: no data

1.4 Materials and methods

1.4.1 Site description and compost system

The primary objective was to estimate the emissions of CH_4, N_2O and NH_3 from different composting plants using different composting management (Figure 1.3). In all, four biowaste composting plants, two green waste composting plants and three AD composting plants were investigated.

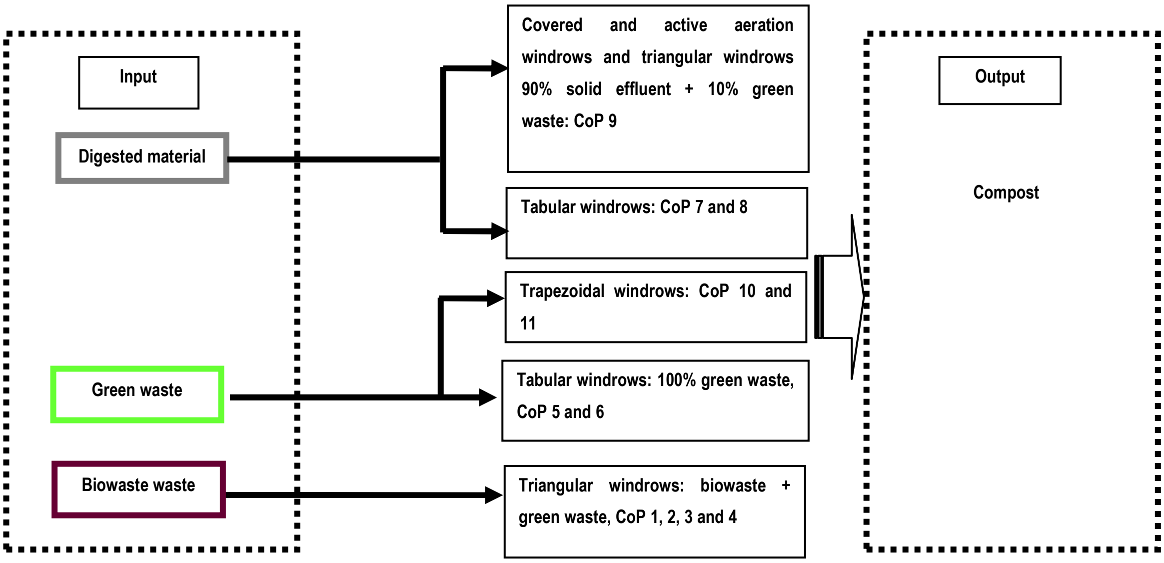

Figure 1.3: Schematic of composting material and windrows. CoP: composting plant

1.4.2 Biowaste composting

Measurements were taken at four plants (Table 1.3). All the plants receive biowaste and compost it in a continuous process. CoP 1, 2 and 3 were analysed in September 2009. At CoP 4, measurements were taken on four occasions in September and October 2010 and January and March 2011. Twenty-two triangular windrows were measured in CoP 4, hence CoP 4 is described in more detail. At CoP 4, the material was arranged in triangular windrows, each 40m long, 4m wide (on the ground) and 1.5m high. The composting process was limited to 10.5 weeks. All the windrows were turned twice a week using a special windrow turner (Figure 1.4).

Figure 1.4: Triangular composting window is turned by a special windrow turner

At the beginning of the composting process, the biowaste and green waste are placed on a concrete floor. After mixing the biowaste and green waste, triangular windrows are formed. It takes one to two days to form one windrow.

Table 1.3: Processing parameters for the different composting plants treating biowaste

Composting plants	**CoP 1**	**CoP 2**	**CoP 3**	**CoP 4a**	**CoP 4b**	**CoP 4c**	**CoP 4d**
Time of measurement	Autumn	Autumn	Autumn	Autumn	Winter	Summer	Spring
Waste	Bio+green	Bio+green	Bio+green	Bio+green	Bio+green	Bio+green	Bio+green
Windrow	Triangular	Triangular	Triangular	Triangular	Triangular	Triangular	Triangular
(length x width x height) (m)	(62x5x2.6)	(35x5-6.8x2-2.3)	(53x4.5x1.6)	(40x4x1.5)	(40x4x1.5)	(40x4x1.5)	(40x4x1.5)
Number of windrows	3	3	4	5	6	6	6
Turning (times)	1	1	6	7	21	11	20
Treatment duration (weeks)	ca. 5	ca. 47	ca. 14	3.5	10	4.5	10
Capacity (Mg waste /a)	12,000	6,500	6,500	n.d	n.d	n.d	n.d
Time of measurement	Sept 2009	Sept 2009	Sept 2009	Oct 2010	Jan 2011	Sept 2010	Mar 2011

n.d: no data

1.4.3 Green waste composting

The emissions of CH_4, N_2O and NH_3 from green waste composting plants with tabular windrows were measured in September 2009 and in September 2010 (CoP 5 and CoP 6, Table 1.4). The plants had three (CoP 5) and four (CoP 6) open tabular composting windrows (each 50m long, 20m wide and 3-3.5m high) on a concrete base. At CoP 5, the windrows were turned every 5-7 weeks, whereas the windrows at CoP 6 were turned only once during the whole composting process. Composting at CoP 5 took over a year; at CoP 6, it took four months. Due to limited space, the tabular windrows at CoP 6 were arranged very close together, therefore the four tabular windrows were considered as one large windrow 80m wide.

Table 1.4: Processing parameters for the different composting plants treating green waste

Composting plants	**CoP 5**	**CoP 6**
Waste	Green waste	Green waste
Windrow	Tabular	Tabular
(length x width x height) (m)	(50x20x3)	(50x20x3.5)
Number of windrows	3	4
Turning (times)	2	1
Treatment duration (weeks)	ca. 21	ca. 16
Capacity (Mg/a)	10,000	-
Time of measurement	Sept 2009	Oct 2010

With the trapezoidal windrows, measurements were taken at two composting plants (CoP 10 and CoP 11) in March and September 2011. At CoP 10, the three windrows investigated were initially 10m long, 6m wide and 2.5m high. After 9.5 weeks of composting, measurements were taken of the GHG emissions and CH_4 concentration in the windrows. At CoP 11, only one windrow (42m long x 12m wide x 3m high) was investigated. The measurements were taken after two weeks of composting. Two measurements were performed at each plant.

1.4.4 AD material composting

The measurements were performed at two anaerobic digestion plants with post-treatment in the form of composting (CoP 7 and 8). Two series of AD composting measurements were taken in the summer and winter in each composting plant. In CoP 7 and CoP 8 the compost material consisted of AD material mixed with 90% and 10% respectively of tree branches and bushes as bulking agents. The tabular windrows are each 80-90m long, 7-10m wide and 5m high with a compost age varying from two weeks to one year (Table 1.5). All windrows were turned once during the composting period. Four measurements were taken in August 2010 (twice), January 2011 and April 2011.

Active aerated windrows: measurements were taken at one anaerobic digestion plant, CoP 9, with post-treatment in the form of composting. The windrows consisted solely of AD material and were covered by semi-permeable sheets. The windrows were placed on a floor with an aeration system (four ventilators) connected to sensors continuously measuring O_2 in the windrows. The aeration process lasted three to four weeks. After the aeration process, the AD material is mixed with green waste as a bulking agent and piled up into smaller triangular windrows for 14 weeks of composting. During the aeration phase, the windrows are not turned, whereas during the non-aeration process the smaller windrows are turned once a week. The windrows initially have a large triangular shape (2.5m high), but after the aeration process the windrows are transformed into windrows with a smaller triangular shape 1.4m high. Two measurements were performed in August 2010 and in April 2011.

Table 1.5: Processing parameters for the different composting plants treating AD material

Composting plants	**CoP 7**	**CoP 8**	**CoP 9**	
			Phase I	Phase II
Waste	AD +green	AD +green	AD +green	AD +green
Windrow	Tabular	Tabular	Triangular	Triangular
(length x width x height) (m)	(80x10x5)	(90x7x5)	(37x7x2.5)	(40x4x1.4)
Number of windrows	4	2	4	3
Active aeration	No	No	Yes	No
Turning (times)	1 time	1 time	No	1/week
Treatment duration (weeks)	24-35	16-18	ca. 4	ca.13-14
Capacity (Mg/a)	17,000	-	10,600	10,600
Time of measurement	Aug 2010 April 2011	Aug 2010 April 2011	Aug 2010 April 2011	Aug 2010 April 2011

1.5 Gas concentration and emission measurements

1.5.1 Tunnel method

Emissions from open windrows are difficult to determine because there is no suitable measuring technique. GHG emissions have widely been determined by what is known as the static chamber (Czepiel *et al.,* 1996; Clemens *et al.,* 1997; Hellmann, 1997; B. Beck Friis, 2000; Hao, 2000; Sommer, 2000; Hobson *et al.,* 2005; Andersen, 2010). However, the chambers used covered only a small surface of the windrow (usually up to 1m^2). This method has another disadvantage: gas emissions via convection cannot be measured using the static chamber method (Sommer *et al.*, 2004; Andersen, 2010). Convection is an important factor, because air flows through the windrow and transports gases away from the windrow into the atmosphere.

To overcome these limitations and improve the accuracy of gas emission estimates, a tunnel method has been developed to measure GHG emissions from open composting windrows. The tunnel covers an area of around 50m^2 with a length of 10m and a width of 5m. The height of the tunnel may vary from 1.5 to 2m. Two ventilators are used to ventilate the tunnel from one side (Figure 1.5). The ventilation rate is fixed at 1,000m^3 h^{-1}. Fresh air enters the tunnel from the front. In the tunnel, gas is emitted into the fresh air and leaves the tunnel at the rear. At the outlet, a Teflon tube (4mm in diameter) is installed 0.5m above the windrows and used for gas sampling. The gas is pumped via a cooler to an infrared gas N_2O and CH_4 analyser (Uras, ABB). The infrared detector has a sensitivity of 0.1mg/m^3 for N_2O and 1mg/m^3 for CH_4. When the tunnel was installed, it took ten to twenty minutes for GHG concentrations to be constant. GHG concentrations were then recorded every minute for one hour. To control the accuracy of the online analyser, gas samples were collected manually using an evacuated headspace vial. The samples were analysed by gas chromatography (ECD/FID, SRI 8610C, USA) in the laboratory. The differences in GHG concentration between incoming and outgoing air were used to calculate emissions from the waste in the tunnel. Air fluxes were determined using an anemometer (testo 435) or a micromanometer (Müller Instruments EPM-300-BA, Germany).

In parallel, 60L of outgoing air were flushed through two flasks containing 40mL of a 0.05M H_2SO_4 solution. NH_3 was trapped in the solution as NH_4^+ and subsequently analysed colorimetrically in the laboratory.

1.5.2 Pore gas measurements

Gas concentrations in the windrow were analysed. Below the tunnel, three gradients were sampled in the windrow. At each sampling point, a gas probe was drilled into the windrow to a depth of 0.2, 0.4, 0.6, 0.8 and 1m (Figure 1.5). Gas from the pore space was sucked into an infrared analyser by a pump to detect CH_4, CO_2 and O_2 . As a control, gas was sampled using 20ml evacuated headspace vials. These were subsequently analysed for N_2O and CH_4 in the laboratory using GC (ECD/FID). The temperature was also measured at each depth.

1.5.3 General overview of emission factors and calculation of CH_4, N_2O and NH_3 emissions

CH_4, N_2O and NH_3 emissions were calculated using the aeration rate and different concentrations of incoming and outgoing gases. The emission rate and emission factor for each gas were calculated using the following formula:

$$E_r = \frac{(E_{out} - E_{in})}{A_{tunnel}} \times Q_{tunnel} \qquad (\mathrm{mg \times h^{-1} \times m^{-2}})$$

$$E_{fd} = \left(\frac{E_r \times 24[h/d]}{1000[mg/g]} \right) \times \frac{A_w}{M_w} \qquad \mathrm{g\ (Mg\ d)^{-1}}$$

$$E_f = \sum E_{fd} \times T \qquad \mathrm{g\ (Mg\ waste)^{-1}}$$

With

E_r: emission rate (mg x h^{-1} x m^{-2})

$E_{\text{in and out}}$: concentration of inlet and outlet (mg x m^{-3})

Q_{tunnel}: input air (m^3 x h^{-1})

A_{tunnel}: surface under tunnel (m^{-2})

E_{fd}: emission factor per day g (Mg d)$^{-1}$

A_w: total surface of the windrow (m^2)

M_w: total mass of the windrow (Mg)

E_f: emission factor g (Mg waste)$^{-1}$

T: composting duration (d)

To access the contribution of emitted gases from the composting plants, emission factors (kg CO_2 equivalent emitted per tonne of substrate) were used.

$$E_{fCO_2 equiv.} = \left(E_{fCH_4} \times 25 + E_{fN_2O} \times 298 + E_{fNH_3} \times 2.98 \right) \qquad \mathrm{g\ (Mg\ waste)^{-1}}$$

All emission factors were calculated as a mass ratio of gas emitted per initial fresh matter mass or wet weight (kg Mg^{-1}) of the input material. Only CH_4, N_2O and NH_3 emissions from the composting process were considered.

Examples of tunnel measurements at full-scale composting plants are shown in Figure 1.5 and Figure 1.6. Triangular windrows are fairly small, therefore the tunnel covered part of the windrow completely. In contrast, trapezoidal windrows are larger in size. The tunnel could not cover a complete part of it, so only one side of the windrow was covered and measured using the tunnel method. It was assumed here that emissions from both sides of the windrow were similar. For tabular windrows, emissions were only measured on the surface.

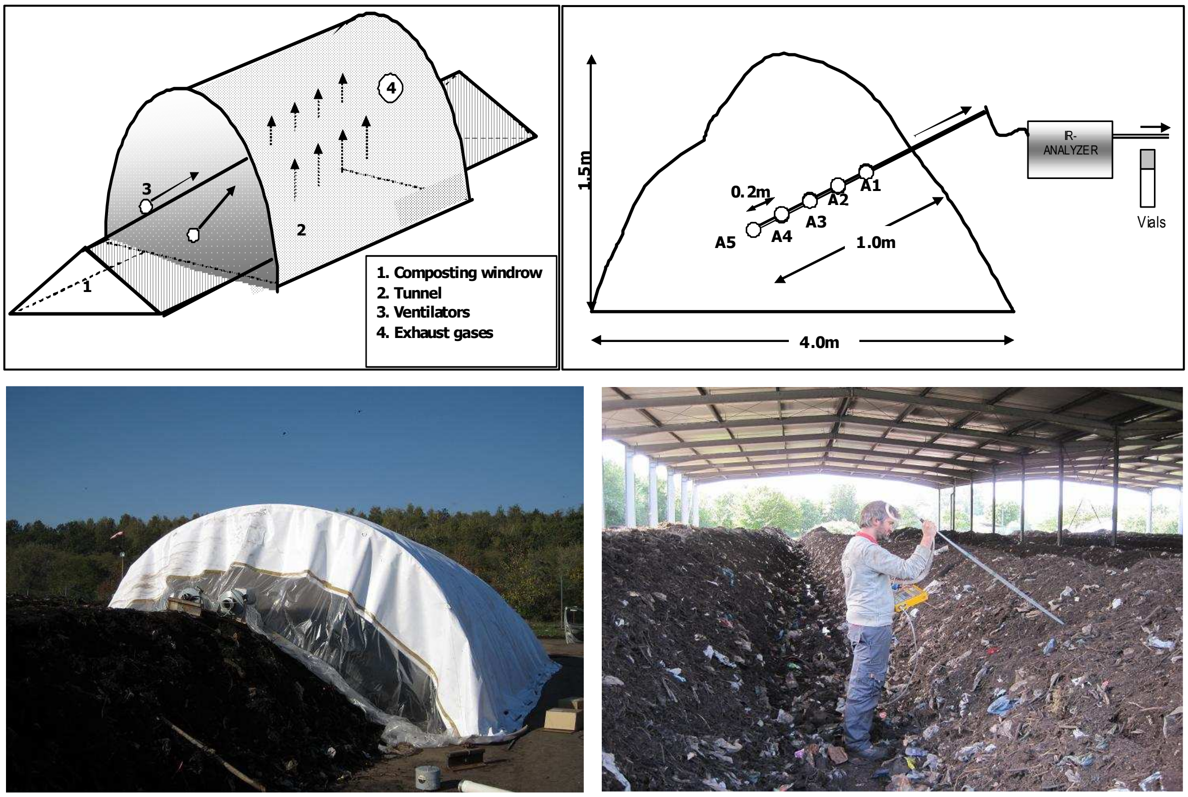

Figure 1.5: Left: Schematic drawing of the tunnel over a compost windrow for analysing N_2O and CH_4 emissions. Right: Schematic drawing of the probe in a compost windrow, gas is sampled at sampling points A1 to A5 and pumped into an IR analyser

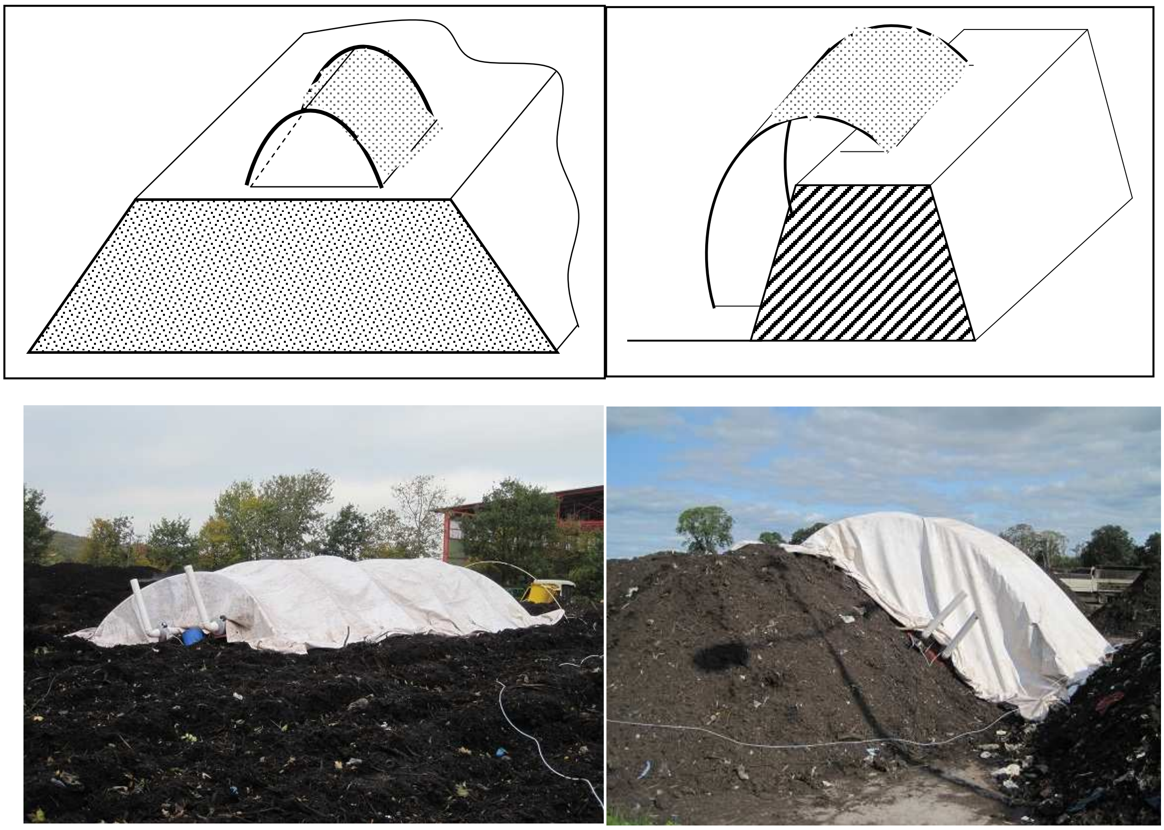

Figure 1.6: Left: Schematic drawing of the tunnel at a tabular windrow for analysing N_2O and CH_4 emissions. Right: Schematic drawing of the tunnel on a trapezoidal windrow for analysing N_2O and CH_4 emissions

1.6 Results and discussion

1.6.1 Profiles of gas concentrations in triangular, trapezoidal and tabular windrows

1.6.1.1 CH_4, CO_2, O_2, N_2 profiles

Gas concentrations of CH_4, O_2, N_2 and CO_2 in triangular, trapezoidal and tabular windrows differed (Figure 1.7). O_2 and N_2 concentration in the pores of all windrows decreased significantly from 0.2 to 1m. Concentrations of CO_2 and CH_4 increased with depth. The decrease of O_2 from the surface to the lowest point was due to biological activity that consumed O_2 (Xu *et al*., 2007). The highest O_2 concentration was at 0.2m and the lowest at 1m. In the triangular windrows, at a depth of 0.2m the mean concentration of O_2 was 16.03 ± 3.6%, while at a 1m depth it was 0.3 ± 0.5%. In the trapezoidal windrows, at a depth of 0.2m the mean concentration of O_2 was 11.8 ± 4.8% and at a 1m depth it was 4.5 ± 5.8%. Similarly, in the tabular windrows, at a depth of 0.2m the mean concentration of O_2 was 16.4 ± 4.7%, while at a 1m depth it was 0.8 ± 1.7%.

The highest concentrations of CO_2 and CH_4 were found at a depth of 1m, where the lowest O_2 concentrations were found. The highest CH_4 concentrations were 16% for triangular, 31.3% for trapezoidal and 31.5% for tabular windrows. The highest CH_4 concentration in triangular windrows was similar to the windrows (1.3 - 2m high) reported by Georgaki *et al.,* (2009). The highest CO_2 concentrations were 58.6% for triangular, 51.6% for trapezoidal and 45.8% for tabular windrows. The high concentration of CH_4 in the windrows indicates that the anaerobic process occurred partly in the windrows (Sommer, 2001). The pattern of CH_4 concentration in trapezoidal and tabular windrows was fairly similar: the highest CH_4 concentration was twice as high as that in triangular windrows. The highest CH_4 concentrations found in the study are of the same magnitude as those measured by Beck-Friis *et al.,* (2000) who found that the concentration of CH_4 in large windrows was much higher than in small windrows.

In both trapezoidal and tabular windrows, mean values of CH_4 and CO_2 increased in parallel from 0.6 to 1m. This pattern did not occur in the triangular windrows. High O_2 and CO_2 concentrations in the composting windrow are evidence of aerobic decomposition (Hao, 2001). For example, the decomposition of carbon hydrate $C_m(H_2O)_n$ represents organic material that decomposes under anaerobic conditions to CH_4 and CO_2, whereas the decomposition of $C_m(H_2O)_n$ with the presence of O_2 produces CO_2 and H_2O. If the mean value of CO_2 in windrows is much higher than CH_4, this is an indicator of an adequate O_2 supply in the windrows for the composting process.

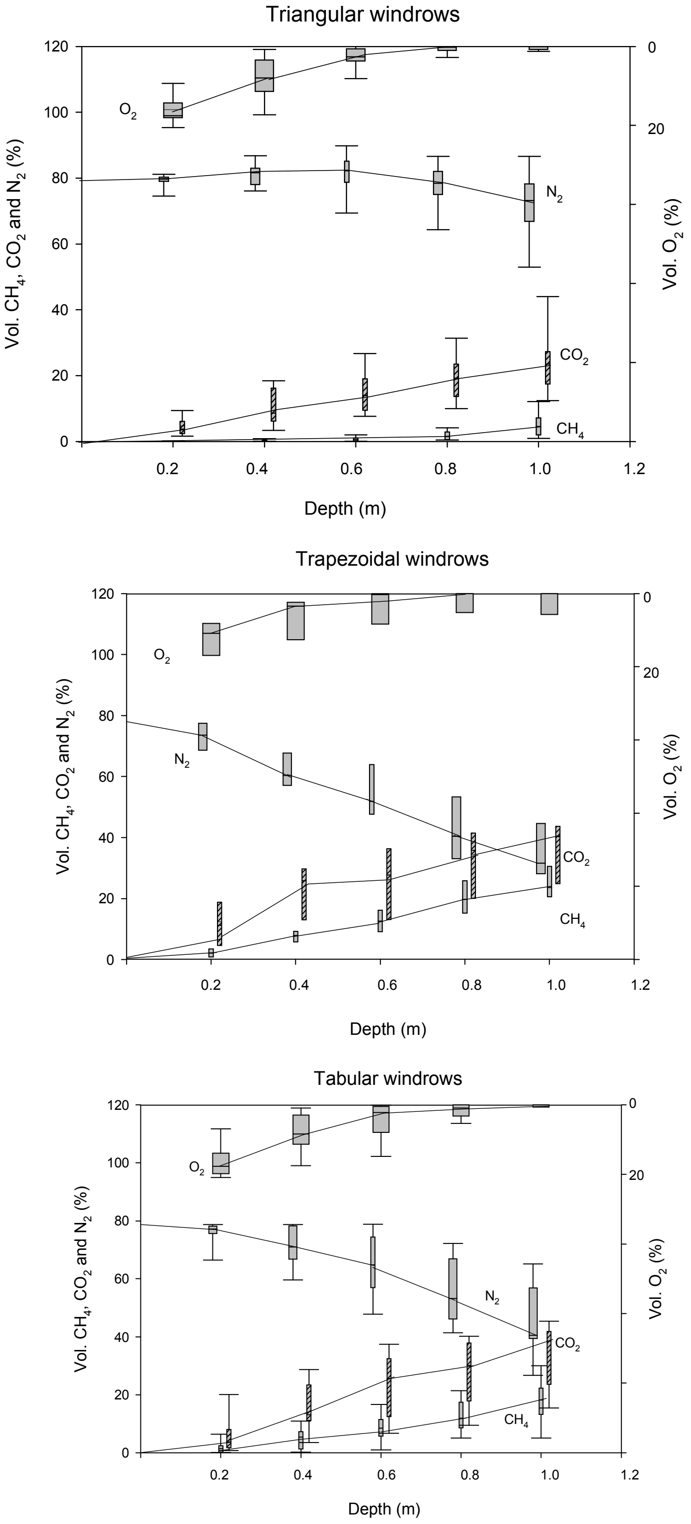

Figure 1.7: Box plot: Profiles of O_2, CO_2, CH_4 and N_2 (calculated) in triangular windrows, trapezoidal windrows and tabular windrows. Box plots show mean values (triangular (n=22), trapezoidal (n=8), tabular (n=16)), the 20 and 75% percentile and minimum and maximum values

1.6.1.2 N_2O profiles

N_2O concentrations differed from those of CH_4, CO_2 and O_2 (Figure 1.8). In the triangular windrows, N_2O concentration was low at 0.2m, gradually increasing to the maximum between 0.4 and 0.6m and slowly decreasing at 1m. The results are in line with those measured by Hellebrand (1998) and Andersen (2010), but Czepiel *et al.,* (1996) and Hao (2001) reported that N_2O production was highest between 20 and 30cm. In both trapezoidal and tabular windrows, the highest concentration of N_2O was between depths of 0.2 and 0.4m. High N_2O concentration near the surface can be explained by optimal O_2 concentration. Mean concentration of O_2 at 0.4m was 9%. Previous studies have shown that N_2O production is affected by moisture and O_2 (Czepiel *et al.,* 1996). For example, N_2O production was highest when O_2 concentration ranged from 6 to 9% (Hao, 2001).

Mean concentrations of N_2O in triangular, trapezoidal and tabular windrows were 12.6, 1.2 and 8.3ppm respectively. The low concentration of N_2O in the trapezoidal windrows may be explained by the low N content (presumably resulting in low NO_2^- and NO_3^- content) in the material because the trapezoidal windrow consisted of green waste alone. This is a qualified inference, since NO_2^- and NO_3^- content was not measured. However, previous reports have shown that the NO_3^- content in biowaste is much higher than that in green waste (Schenk, 1997; Dalal *et al.*, 2009). Higher NO_3^- content in waste increased the production of N_2O (Schenk, 1997; Beck-Friis *et al.,* 2000). Furthermore, Hao (2001) and Paul (2009) stated that N_2O is an intermediate product in the conversion of NO_3^- to N_2 under anoxic conditions during denitrification. In addition, He *et al.,* (2001) found a good correlation between NO_2^- and N_2O production and suggested that N_2O originated from NO_2^-. Osada *et al.,* (2000) studied N_2O emissions from swine waste composting processes. The author confirmed that N_2O production was closely related to both NO_2^- and NO_3^- content.

N_2O concentration in the triangular windrows was higher than N_2O concentration in the tabular windrows. This may be explained by high O_2 concentrations. O_2 in the windrow is replenished by air drawn in at the side of the windrow (Sommer, 2000; Veeken Adrie, 2002). The triangular windrows were small (1.5m high) compared to tabular windrows with a height of 4-5m. Thus O_2 penetrated more effectively in the triangular windrows than in the tabular windrows. Czepiel *et al.* (1996) found that availability of O_2 is a determining factor for N_2O production. An increase of CH_4 production seemed to cause a decrease of N_2O production and vice versa (Park *et al.,* 2011). The production of CH_4 in the tabular windrows was much higher than CH_4 production in the triangular windrows.

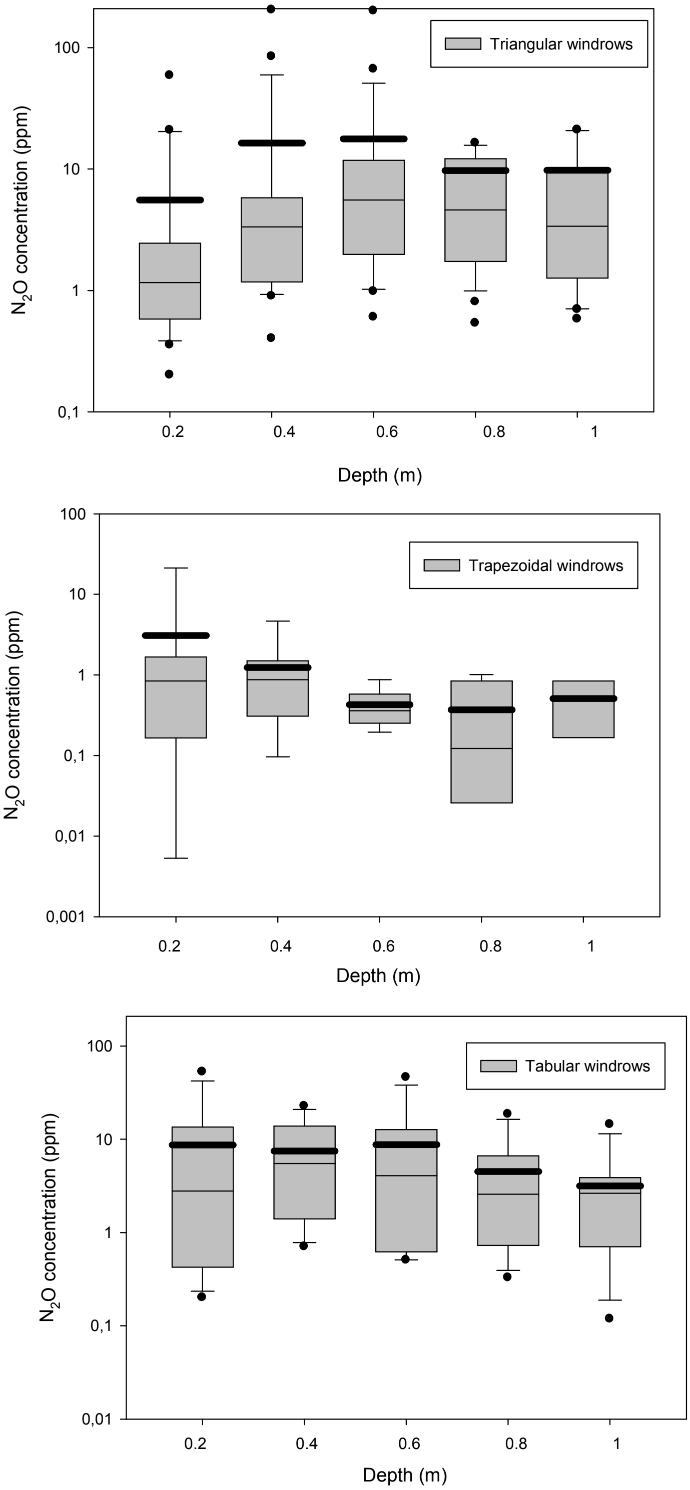

Figure 1.8: Box plot: Profiles of N_2O in triangular windrows, trapezoidal windrows and tabular windrows. Box plots show ▬ mean values (triangular windrows: n=22, trapezoidal windrows: n=8, tabular windrows: n=16), the 25 and 75% percentile and minimum and maximum values

In the windrows, the CH_4 and N_2O concentrations (%) were negatively correlated (Figure 1.9). An increase in CH_4 concentration led to a decrease in N_2O concentration and vice versa. This result is in line with (Sommer *et al.*, 2004; Thompson *et al.*, 2004 and Park *et al.*, 2011). When CH_4 concentrations were above 15%, N_2O concentrations were below 0.4ppm. Chiumenti *et al.,* (2007) reported that CH_4 production was intensive during the thermophilic phase and low O_2 concentrations in the windrows, whereas N_2O occurred mainly in the cooling phase with the availability of O_2 (Sommer, 2001).

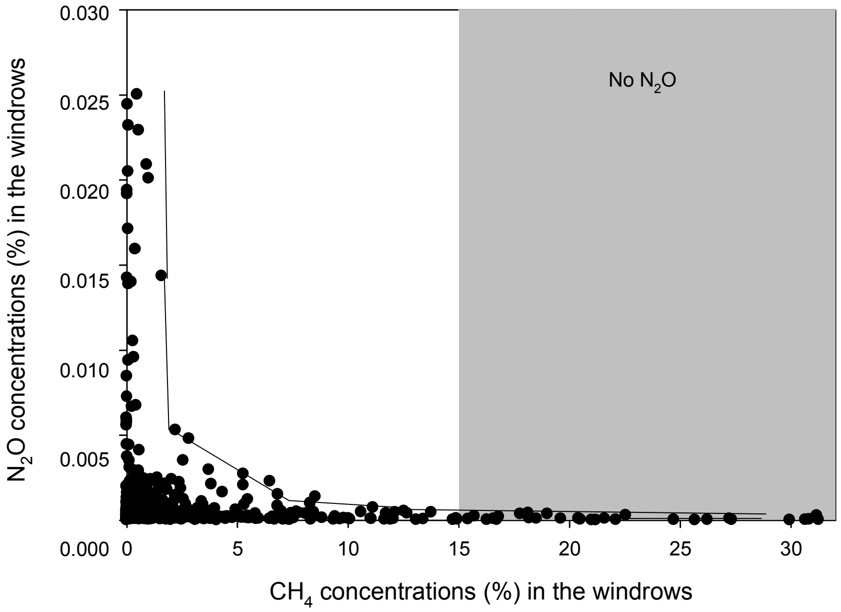

Figure 1.9: Concentration of N_2O and CH_4 in windrows

1.6.2 Ratio of CO_2/CH_4 in compost windrows

In all windrows, the CO_2/CH_4 ratio indicates whether the zones are anaerobic or aerobic. When the ratio was around 1, pore gas contained a typical composition of biogas. In contrast, a higher ratio indicated that aerobic processes prevailed. The CO_2/CH_4 ratios varied from 1.2 to 3,000 in triangular windrows, from 0.97 to 12 in trapezoidal windrows and from 1.1 to 6.4 in tabular windrows (Figure 1.10). The lowest ratio of 0.97 was measured at a depth of 1m in a trapezoidal windrow, while the highest one of 3,000 was detected at a depth of 0.2m in a triangular windrow. The ratios gradually decreased with the depth profile, starting from 0.2m to 1m depths.

Mean CO_2/CH_4 ratios in triangular windrows were above 10, while mean CO_2/CH_4 ratios in trapezoidal and tabular windrows were below 10. The low CO_2/CH_4 ratio in trapezoidal and tabular windrows is an indicator that those windrows established more anaerobic zones than triangular windrows. Therefore it is likely that more CH_4 was formed in the trapezoidal and tabular windrows. This is in agreement with previous studies: CH_4 production was much higher in a large windrow than in a small windrow (Beck-Friis *et al*., 2000; Thompson *et al*., 2004). Fukumoto *et al.,* (2003) evaluated CH_4, N_2O and NH_3 emissions from swine manure windrows. The authors concluded that the scale of the windrows is a key factor in reducing the potential of the windrow producing and emitting CH_4. Andersen (2010) pointed out that a small windrow and more frequent turning could reduce CH_4 production due to more O_2 being supplied to the windrows.

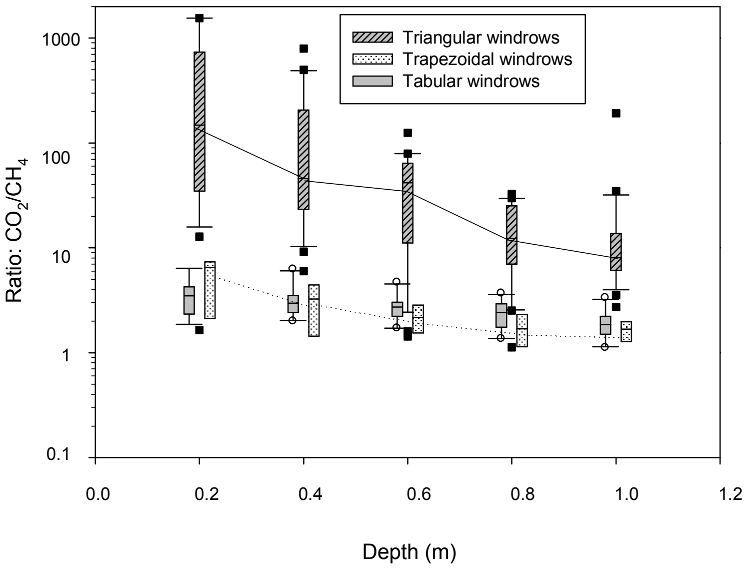

Figure 1.10: Box plots show mean ratio values (logarithmic scale) of CO_2/CH_4 in different depths of triangular windrows▨, trapezoidal windrows ░ and tabular windrows▬ (triangular windrows: n=22; trapezoidal windrows: n=8; tabular windrows: n=16). The box indicates the 25 and 75% percentiles and minimum and maximum values

In all windrows, gas concentrations at a depth of 1m ranged from 0.3-32% CH_4, 5.2-58.6% CO_2, 23-87% N_2 and 0-16% O_2 (Figure 1.11). In triangular windrows, the CH_4 concentration was 0.2 to 16%. In trapezoidal windrows, the CH_4 concentration varied from 3 to 31%. In tabular windrows, the CH_4 concentration ranged from 3 to 32%. CH_4 concentrations were negatively correlated with the CO_2/CH_4 ratio. The CO_2/CH_4 ratio of tabular windrows ranged from 1 to 3 and in trapezoidal windrows ranged from 1 to 4, whereas in triangular windrows it varied from 3 to 200. The differences may be explained by the share of the overall free air space pore volume in the windrows: tabular and trapezoidal windrows are 4-5m high and the materials in lower layers are compacted because of the weight of the material. Consequently, only some air may enter the windrows by convection and diffusion. In contrast, triangular windrows are small with a height of only 1.5-2m. Air may permeate the windrow more easily. More O_2 is available and aerobic processes are more likely.

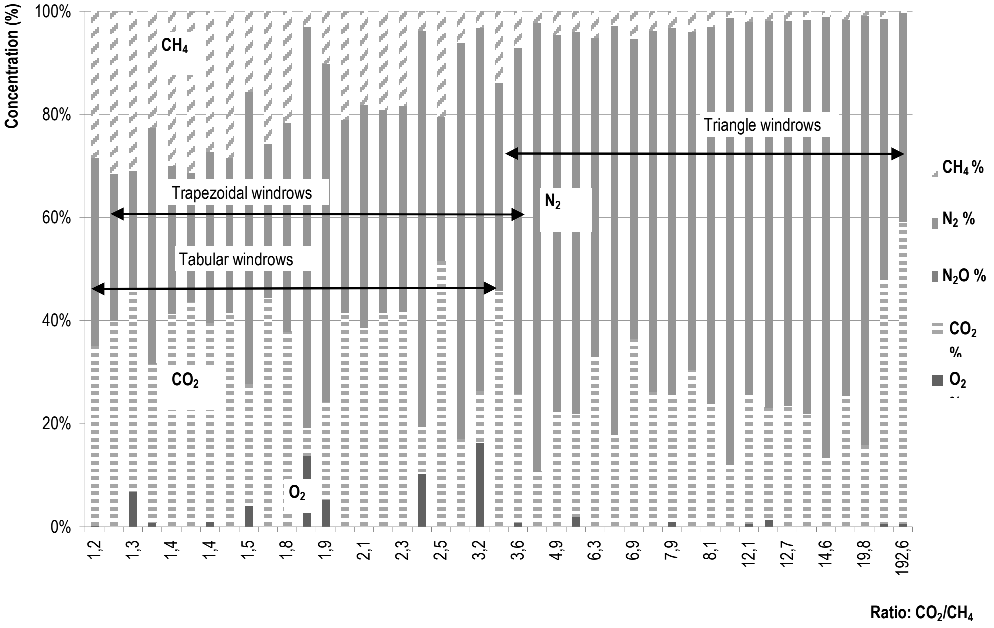

Figure 1.11: Gas concentrations in all windrows sorted by CO_2/CH_4 ratio at a depth of 1m**.** It is assumed that the remaining gas was N_2 (CH_4, CO_2, O_2 and N_2O were measured). Results are mean concentrations of tripled measurements (n=3 for each windrow)

Two different types of windrows may be classified. Although the concentration of O_2 was nil below 0.6m in both windrows A and B (Figure 1.12), the CO_2 concentration increased with depth in windrow A. CH_4 was relative low (<10%). In windrow B concentrations of CH_4 and CO_2 increased with depth. The highest concentrations of CH_4 and CO_2 were 20% and 40% respectively at a depth of 1m. Concentration of O_2 is considered to be an important factor in indicating aerobic or anaerobic processes in a windrow (Amon *et al.*, 2006). However, the gas concentration data in the study indicate that the O_2 factor alone is not enough to determine whether the windrow is experiencing aerobic or anaerobic conditions. Although concentration of O_2 was nil below 0.6m in both windrows, the degradation processes in windrow A still seemed to be aerobic due to high concentrations of CO_2 (37%) and N_2 (58%): the ambient air contains 78% nitrogen and 21% oxygen. O_2 and N_2 enter at the sides of the windrow, but only O_2 is consumed and transformed into CO_2 via respiration. Theoretically, a windrow with a CO_2

concentration of 21% could be still aerobic. N_2 and gases produced by microorganism activities emit through the middle at the top. N_2 is not affected by chemical and biological activities in the compost windrow (with the exception of complete denitrification). Therefore N_2 provided evidence of air exchange in the windrow. Low concentration of O_2 but high concentration of N_2 indicated that a chimney effect had developed in the windrow due to the convective transportation of gases.

In contrast, concentrations of N_2, CO_2 and CH_4 were 45%, 39% and 16% respectively at 0.8m in the windrow B. Stoichiometrically, such a distribution of pore gas hints at a mix of an aerobic and an anaerobic process. Nitrogen is still in the windrow, but CO_2 and CH_4 are formed via anaerobic processes as well. As a consequence, N_2 concentration decreases and CO_2 and CH_4 increase. In a completely anaerobic windrow, the gas pore space would be completely filled with CO_2 and CH_4.

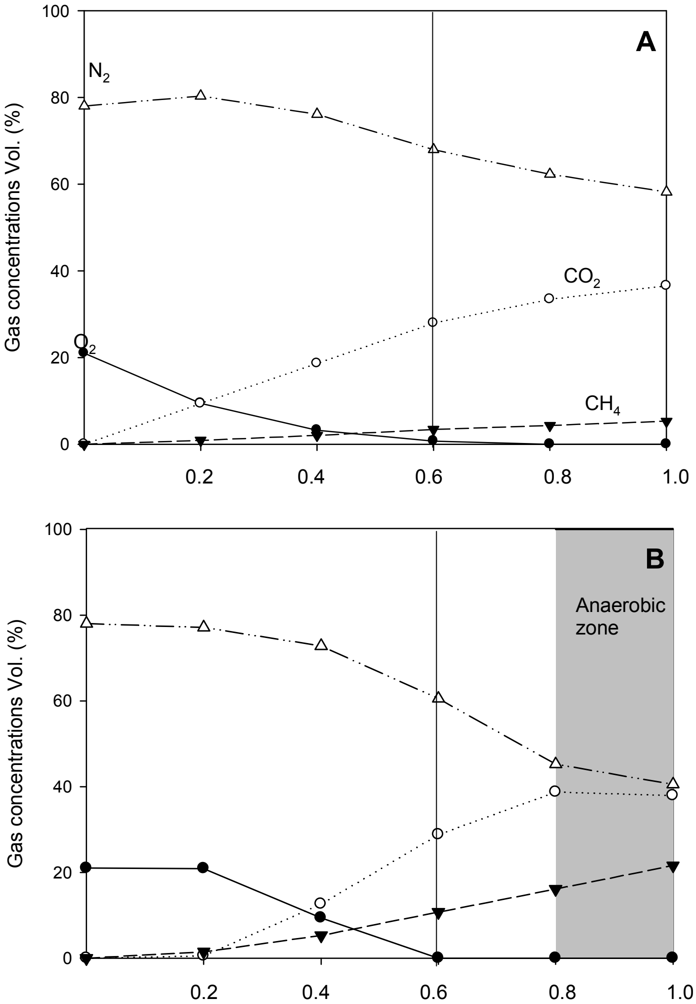

Figure 1.12: Profiles of O_2, CO_2, CH_4 and N_2 in two typical windrows

1.6.3 Correlation between gas production and emission

There was no correlation between the pore gas concentration in all windrows and their emissions. The 46 composting windrows measured were very different in size, form and material. Some composting plants treated biowaste or green waste to produce compost, others used a mixture of anaerobic digestion residues and green waste or just green waste. Different characteristic input materials might cause a variation of CH_4 and N_2O formation, degradations and emissions (Smet *et al.*, 1999; Lissens *et al.*, 2004). In addition, different operating conditions may contribute to different gas emissions (Illmer, 1996; Hao, 2000; Hobson *et al.*, 2005). In this investigation, the frequency of turning varied from 0 to 20 times. The overall composting duration also varied from 5 weeks to 1 year.

However, 22 of the 46 windrows had similar operating conditions and used similar biowaste as input material. In this case, CH_4 in the windrows and its emissions were correlated (Figure 1.13). The result agrees with the finding of Sommer (2000) that the increase of CH_4 in the windrow led to an increase in its emission. Although CH_4 emission was related to CH_4 concentration in the windrow, the emission did not increase significantly until the concentration inside the windrow reached 2% CH_4. Where CH_4 concentrations in the pore gases were below 5%, CH_4 emissions were around 30g (Mg waste)$^{-1}$ per day. However, at maximum concentration of CH_4 (16%), the windrow emitted nearly 80g CH_4 (Mg waste)$^{-1}$ per day.

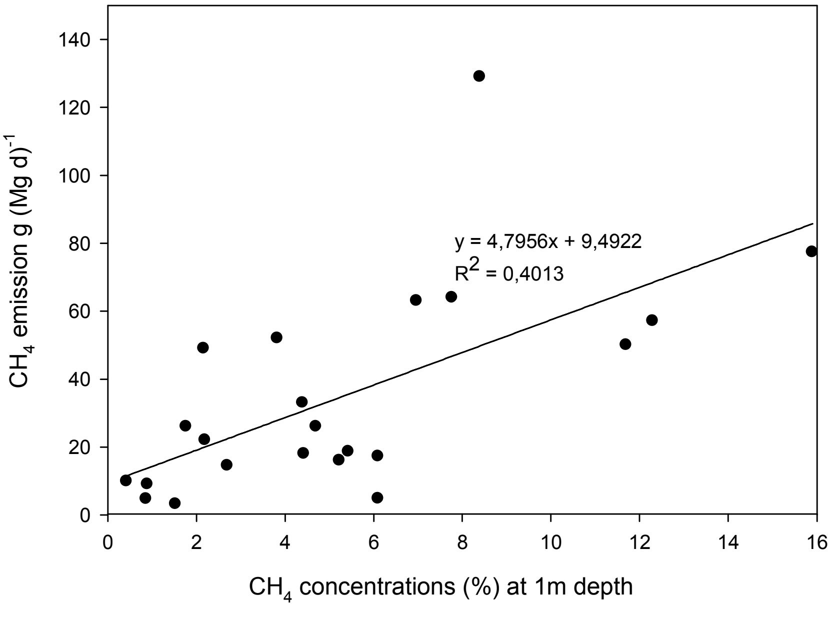

Figure 1.13: Correlated CH_4 concentration at 1m depth in the windrows and its emissions

1.6.4 Emissions of CH_4, N_2O and NH_3 from biowaste composting

The cumulative CH_4, N_2O and NH_3 emission factors g (Mg waste)$^{-1}$ of composting plants varied during the composting process time (Figure 1.14). At CoPs 1, 2, 4a, 4b, 4c and 4d, emissions of CH_4 increased sharply during the first five weeks of the composting process (Figure 1.14A), which confirmed previous results (Hellmann, 1997; Hao, 2000; Hellebrand and Kalk, 2001). At CoP 3, emissions of CH_4 were fairly low at the beginning but started to increase after four weeks. Before biowaste can be treated at the composting plants, it is stored in waste bins in households for a certain period (1-2 weeks). Thus, part of the composting process may have taken place in the waste bins already. Furthermore, different input material may also contribute to different CH_4 emissions.

The N_2O emission patterns from CoP 2 and CoP 4a and 4c were fairly similar (Figure 1.14B). The emission of N_2O increased rapidly in the first four weeks. The emission patterns of N_2O resembled those found by Wolter *et al.,* (2004), Chadwick (2005) and El Kader *et al.,* (2007). However, the emission pattern from CoP 3, 4b and 4d had a lag phase at the beginning and increased sharply after six weeks. Similarly, Hellmann (1997) and Beck-Friis *et al.,* (2000) stated that the N_2O emissions increase with compost age. He *et al.,* (2001) reported that over 95% of the N_2O was produced and emitted during the last stage of composting. Emissions of N_2O at CoP 1 remained low during the whole composting process. Hellmann (1997) studied the release of GHGs during the maturation of open-windrow composting of biowaste. The study showed that N_2O was emitted during the first few days and last few days of the process, when temperatures were low again. Czepiel *et al.,* (1996) found no significant statistical correlation between N_2O emission and compost age, but the availability of O_2 was a determining factor for N_2O production.

Emissions of NH_3 at CoP 4b were highest in the first two weeks, whereas at CoPs 4c and 4d NH_3 emissions were high between weeks 3 and 5. NH_3 emissions at CoPs 1, 2, 3 and 4a were quite low (Figure 1.14 C). In general, the emission of NH_3 was high at the beginning and declined after a few weeks. During the curing phase, there were no NH_3 emissions (Beck-Friis *et al.*, 2001).

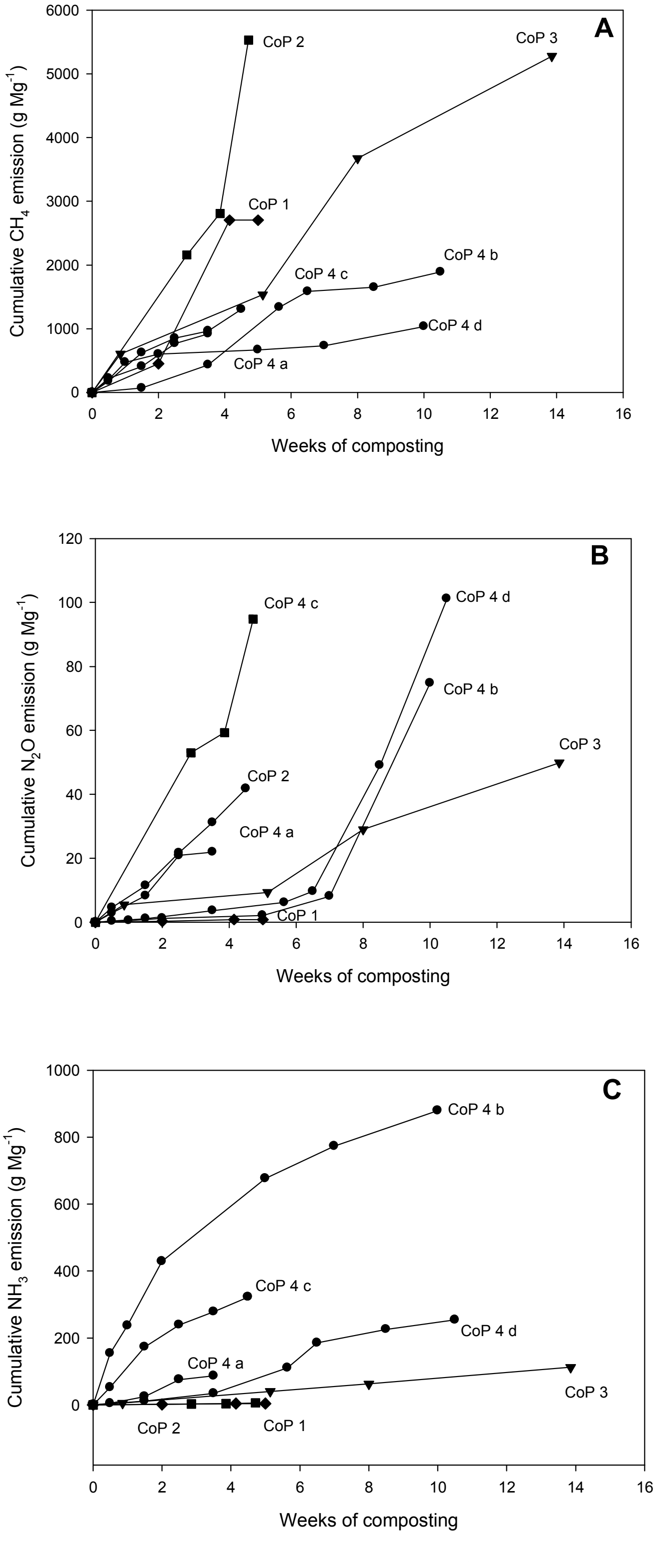

Figure 1.14: Cumulative emissions of CH_4 (Figure A), N_2O (Figure B) and NH_3 (Figure C) from biowaste composting plants: ♦: CoP 1, ■: CoP 2, ▼: CoP 3 and ●: CoP 4. Four measurements were performed at CoP 4 (a, b, c and d)

GHGs emissions varied between the composting plants (Figure 1.15). The emission factor of CH_4 ranged from 920 to 5,500g CH_4 (Mg biowaste)$^{-1}$. Nitrous oxide emissions ranged from 1 to 100g (Mg biowaste)$^{-1}$ and the emission factor of NH_3 varied from 5.8 to 879g (Mg biowaste)$^{-1}$. The results of CH_4, N_2O and NH_3 emissions from the study were comparable with those in other studies (Smet *et al.*, 1999; Amon *et al.*, 2006; Amlinger *et al.*, 2008; Andersen *et al.*, 2010).

Frequent turning is likely to reduce CH_4 emissions but increase N_2O and NH_3 emissions (Figure 1.15A). Turning provides a better O_2 supply for microbial activity due to exposure of particles to O_2 and enhances porosity. It may lead to a higher diffusion of gases from the interior to the surface and vice versa. The windrows at CoP 1 were turned only once during the whole composting process (5 weeks), whereas at CoP 4 the windrows were turned twice a week during the composting process. The emission factor of CH_4 at CoP 1 was 3,600g (Mg biowaste)$^{-1}$, while at CoP 4 it was 1,829g (Mg biowaste)$^{-1}$. Previous studies showed similar results. Szanto *et al.* (2007) and Amon *et al.* (2006) described how frequent mixing reduces CH_4 emissions during manure composting.

A shorter composting treatment results in lower CH_4 emissions (Figure 1.15B). CoP 2, which had a long composting process (47 weeks), emitted five times more than CoP 4 which had a short composting duration (3.5 to 10 weeks). In fact, CH_4 emissions at CoP 2 were 5,500g (Mg biowaste)$^{-1}$, whereas CH_4 emissions at CoP 4a were just 920g (Mg biowaste)$^{-1}$.

However, at CoP 1, the composting duration was 5 weeks, but CH_4 emissions were twice as high as at CoP 4. This might be explained by the fact that the windrows at CoP 1 (height: 2.6m) are larger than the windrows at CoP 4 (height: 1.5m) and the turning frequency at CoP 1 was 0.2 times a week compared to twice a week at CoP 4. Large windrows with a low turning frequency result in high CH_4 emissions (Sommer and Moller, 2000; Fukumoto *et al.,* 2003).

For N_2O, there was a relationship between N_2O emissions and turning. Frequent turning is likely to increase N_2O emissions. There was a positive linear correlation between emission factors of NH_3 and turning frequency (r^2 = 0.9338). While windrows at CoP 4 emitted high amounts of NH_3, the emissions from CoPs 1 and 2 were quite low. It is known that high NH_3 emission is a consequence of a higher concentration of O_2 in a windrow and that this is related to the mineralisation of organic nitrogen (Amon *et al.*, 2006; Szanto *et al.*, 2007). Furthermore, frequent turning leads to the release of NH_3 accumulated within the windrow (Parkinson *et al.*, 2004).

The overall CO_2 emissions were highest at CoP 2 (166 kg Mg^{-1} biowaste) followed by CoP 3 (148kg Mg^{-1}), CoP 1 (90kg Mg^{-1}) and CoP 4 (69kg Mg^{-1}). The CO_2 emissions tended to be lower from windrows with a high turning frequency (Table 1.6). In contrast, Hao (2001) and Andersen *et al.* (2010) found that CO_2-equivalent emissions from turned windrows were 1.67 times higher than from static windrows. Ahn *et al.* (2011) reported 20% higher GHG emissions from turned windrows of dairy manure compared with static windrows. Turned windrows released 3.5 times more N_2O and 16% more CH_4 than static windrows. After turning, the windrows had adequate O_2 (21%) and CH_4 production decreased immediately. However, the intense aerobic activity consumed the O_2 rapidly and promoted anaerobic conditions, therefore increasing CH_4 production again (Chiumenti *et al.*, 2007).

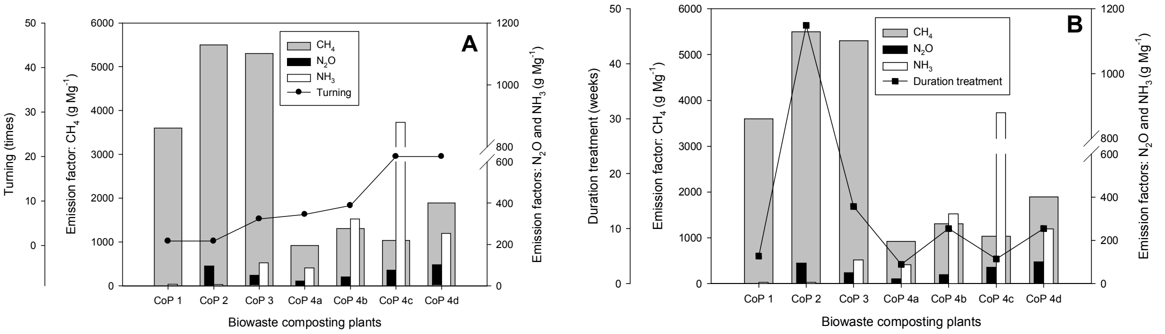

Figure 1.15: CH_4, N_2O and NH_3 emission factors g (Mg biowaste)$^{-1}$ from different composting plants; white bars: emission factor of NH_3; grey bar: emission factor of CH_4; black bar: emission factor of N_2O; ●: turning frequency. ■: duration treatment

Table 1.6: Processing parameters and emission factors for biowaste composting plants

Composting plants	**CoP 1**	**CoP 2**	**CoP 3**	**CoP 4 a**	**CoP 4 b**	**CoP 4 c**	**CoP 4 d**
Turning (times/week)	0.2	0.02	0.4	2	2	2	2
Treatment duration (weeks)	5	47	14	3.5	10	4.5	10
Emission factor CO_2 equivalent (kg Mg^{-1})	90	166	148	26	92	83	75

1.6.5 Emissions of CH_4, N_2O and NH_3 from green waste composting

The emissions of CH_4, N_2O and NH_3 at CoP 5 and CoP 6 are shown in Figure 1.16. Emissions of CH_4 increased gradually after the windrows were set up. The highest CH_4 emission at CoP 6 was from 8-11 weeks. The emission factors of CH_4 at CoP 5 and CoP 6 were 1,700 and 7,129g CH_4 (Mg green waste)$^{-1}$ respectively. The emissions of N_2O at CoP 6 increased with composting duration, while emissions of N_2O at CoP 5 were low at the beginning but started to rise after 12 weeks. The emission factors of N_2O at CoP 5 and CoP 6 were 36 and 13g N_2O (Mg green waste)$^{-1}$ respectively. The emission of NH_3 was measured at CoP 5 but not at CoP 6. The emission factor of NH_3 was only 2g (Mg green waste)$^{-1}$ at CoP 5.

Windrows at CoP 6 were larger than at CoP 5. The results indicate that large windrows significantly increase the emissions of CH_4 but decrease the emissions of N_2O. The NH_3 emission was comparatively low. The results are in agreement with previous reports. Fukumoto *et al.,* (2003) stated that small windrows reduce CH_4 emissions but increase N_2O emissions during swine manure composting. Beck-Friis *et al.,* (2000) and Thompson *et al.* (2004) found that emissions of CH_4 in large windrows were higher than in small ones.

The overall CO_2 emissions varied between 53 and 182 kg CO_2 equivalent (Mg green waste)$^{-1}$ (Table 1.7). The results are in line with those found in previous studies. Amlinger *et al.,* (2008) reported 9-68 kg CO_2 equivalent (Mg green waste)$^{-1}$, whereas Andersen *et al.,* (2010) estimated 91-161 kg CO_2 equivalent (Mg green waste)$^{-1}$. Hellebrand (1998) measured 139 kg CO_2 equivalent (Mg green waste)$^{-1}$.

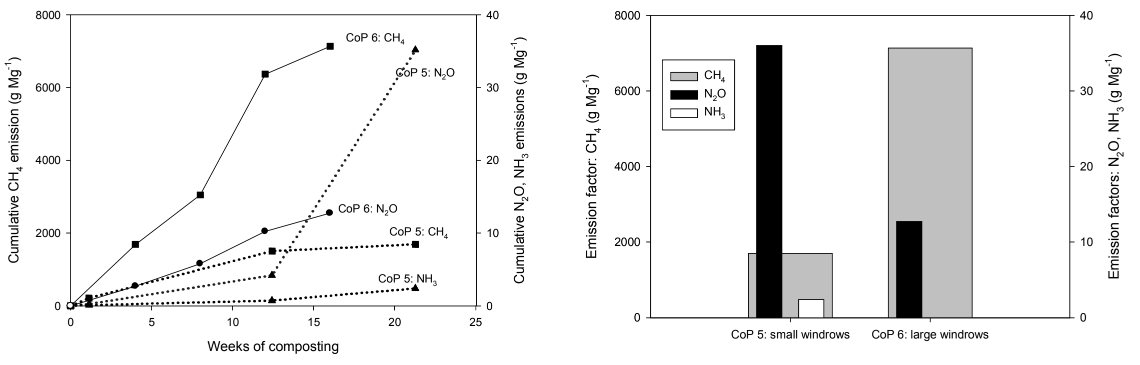

Figure 1.16: Left: Cumulative emissions of CH_4, N_2O, and NH_3 from green waste composting plants, ■: CH_4, ●: N_2O and ▲: NH_3. Right: emission factors of CH_4, N_2O and NH_3 from CoPs 5 and 6: white bars: emission factor of NH_3; grey bar: emission factor of CH_4; black bar: emission factor of N_2O

Table 1.7: Processing parameters for green waste windrows

Composting plants	**CoP 5**	**CoP 6**
Waste	Green waste	Green waste
Windrow	Tabular	Tabular
(length x width x height) (m)	Small (50 x 20 x 3 high)	Large (50 x 80 x 3.5 high)
Turning (times/week)	0.1	0.06
Treatment duration (weeks)	21	16
CO_2 equivalent (kg Mg^{-1})	53	182

1.6.6 Emissions of CH_4, N_2O and NH_3 from AD material composting

Mean CH_4 emission factors were 15,452 ± 2,204 and 5,763 ± 644 g (Mg waste)$^{-1}$ at CoP 7 and CoP 8 respectively (Figure 1.17A). Over 90% of the CH_4 was emitted during the initial 15 and 20 weeks at CoP 8 and CoP 7 respectively. Composting duration at CoP 7 ranged from 24-35 weeks, while composting duration at CoP 8 was 16-18 weeks (Table 1.8). The low emission factors of CH_4 at CoP 8 may be related to the short period of composting compared with CoP 7.

N_2O emissions at CoP 7 and CoP 8 were similar. Emissions of N_2O at CoP 7 and CoP 8 were 82 ± 22 and 71 ± 22g (Mg waste)$^{-1}$ respectively. The emission patterns of NH_3 from CoP 7 and 8 were fairly similar. NH_3 emissions increased rapidly at the beginning. The emission factors of NH_3 at CoP 7 and 8 were 103 ± 6 and 76 ± 16g (Mg waste)$^{-1}$ respectively. Expressed as CO_2 equivalents, the overall emissions from composted AD material varied from 143 to 460kg CO_2 equivalent (Mg waste)$^{-1}$ (Table 1.8).

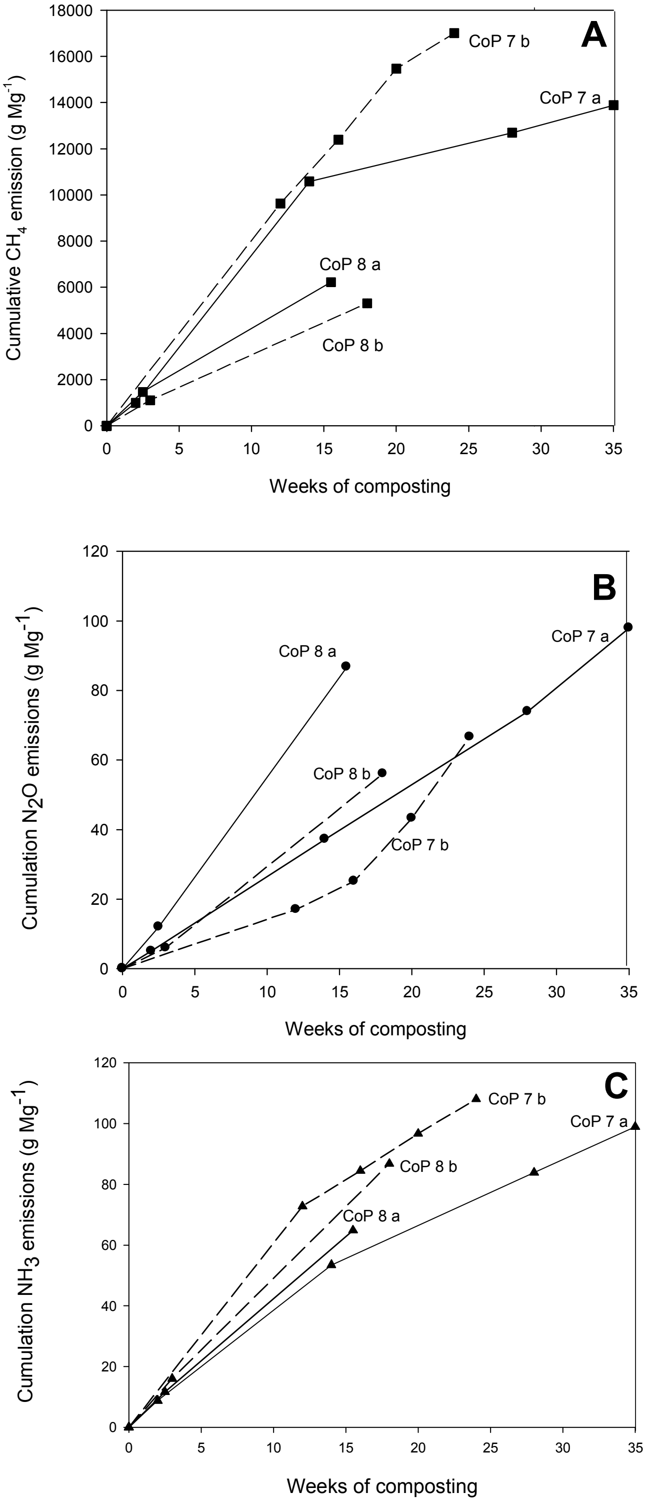

Figure 1.17: Cumulative emissions of CH_4 (Figure A), N_2O (Figure B), and NH_3 (Figure C) from AD material composting plants**.** Two measurements were performed at CoPs 7 and 8 (a and b)

Table 1.8: Processing parameters and emission factors for AD material composting plants

Composting plants	**CoP 7**	**CoP 8**	**CoP 9**	
			Phase I	Phase II
Waste	10% AD + 90% green	90% AD + 10% green	100% AD	70%AD + 30% green
Active aeration	No	No	Yes	No
Turning (times/week)	0.04-0.03	0.06	0	1
Composting duration (weeks)	24-35	6-11	4	13-14
Emission factor CO_2 equivalent (kg Mg^{-1})	411 ± 49	166 ± 23	20 ± 8.8	272 ± 167

1.6.7 Effect of aeration on CH_4, N_2O and NH_3 emissions

The influence of aeration on CH_4 emissions is clearly demonstrated (Figure 1.18). During the aeration stage, only 607g CH_4 (Mg waste)$^{-1}$ was emitted while the total emission factor of CH_4 was 14,826g CH_4 (Mg waste)$^{-1}$. This observation is higher than the results of Clemens and Cuhls (2003) who found that 6-12.10^{3}g CH_4 (Mg waste)$^{-1}$ were emitted from the composting of municipal solid waste with intensive aeration.

During aeration (4 weeks), only 4% of the total CH_4 emissions were emitted. Over 95% of the total CH_4 emissions occurred in subsequent composting without aeration. The extreme increases in CH_4 emissions after three weeks' aeration were probably due to a lack of O_2 in the windrows. This result matched the findings of Osada *et al*., (2000), Sommer (2000), Thompson *et al.,* (2004), de Guardia *et al*., (2008), Jiang *et al.,* (2010) and Shen *et al.,* (2011).

The emission factor of N_2O was 120g (Mg waste)$^{-1}$. The result is comparable with that reported by Clemens and Cuhls (2003) which demonstrated that emission factors of N_2O ranged from 1.44-378g N_2O (Mg MSW)$^{-1}$. As shown in Figure 1.18, N_2O emissions were controlled by active aeration. Only 24% of the total N_2O emissions were emitted during the first three weeks. After stopping aeration, emissions gradually increased during composting. The same results were observed by Hellebrand (1998), Osada *et al.,* (2000), Sommer (2000), de Guardia *et al.,* (2008) and Shen *et al.,* (2011). In contrast, He *et al.* (2001) and Jiang *et al.* (2010) found that a low aeration rate results in low emissions of N_2O.

The total emission factor of NH_3 was 3,327g (Mg waste)$^{-1}$. During the active aeration period, emissions of NH_3 were extremely high in the first four days, but gradually decreased thereafter. An increase in the aeration rate leads to an increase in NH_3 emissions (Osada *et al.,* 2000; Sommer, 2000; de Guardia *et al.*, 2008; Jiang *et al.*, 2010; Shen *et al.*, 2011). However, too high an aeration rate leads to a low moisture content and therefore inhibits microorganism activities (Osada *et al.*, 2000).

On the other hand, Figure 1.18 shows a clear trend of NH_3 increasing after the active aeration stage. During the non-aeration period, 2,423g NH_3 (Mg waste)$^{-1}$ were emitted, contributing to 73% of total NH_3 emissions. The result is much higher than those found in a laboratory-scale and full-scale composting plant in previous studies. The emissions of NH_3 during the intensive aeration composting of digestate were studied on a laboratory scale by Smet *et al.* (1999). The accumulation of NH_3 emissions was 87g (Mg digestate)$^{-1}$. Clemens and Cuhls (2003) reported that NH_3 emission factors were between 19-1,150g (Mg waste)$^{-1}$ at a full-scale composting plant treating MSW.

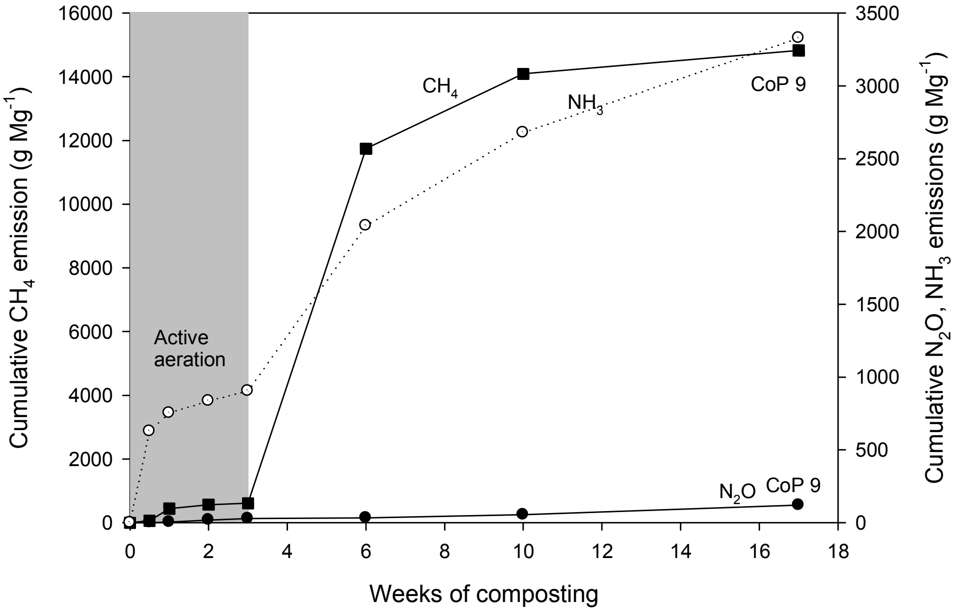

Figure 1.18: Cumulative emissions of CH_4, N_2O and NH_3 in the active aeration stage and non-aeration stage: ■ CH_4; ● N_2O; ○ NH_3. ▨ Windrows were covered by a semi-membrane and aerated

1.6.8 CO_2 equivalent emissions

The overall CO_2 emissions for composting were 118kg (Mg biowaste)$^{-1}$, 118kg (Mg green waste)$^{-1}$ and 290kg (Mg AD material)$^{-1}$. The emissions of CH_4 from composting windrows were much greater than the emissions derived from N_2O and NH_3 (Figure 1.19). CH_4 emissions accounted for 66-99%, while the emission of N_2O and NH_3 contributed from 0.4 to 32% and from 0 to 2.2% respectively to the overall CO_2 emissions. This is in line with the findings of Amon *et al.,* (2001) who established that CH_4 emissions from composting windrows contributed 78% of total GHG emissions. The composting of biowaste, green waste and AD material is a source of greenhouse gases (especially CH_4).

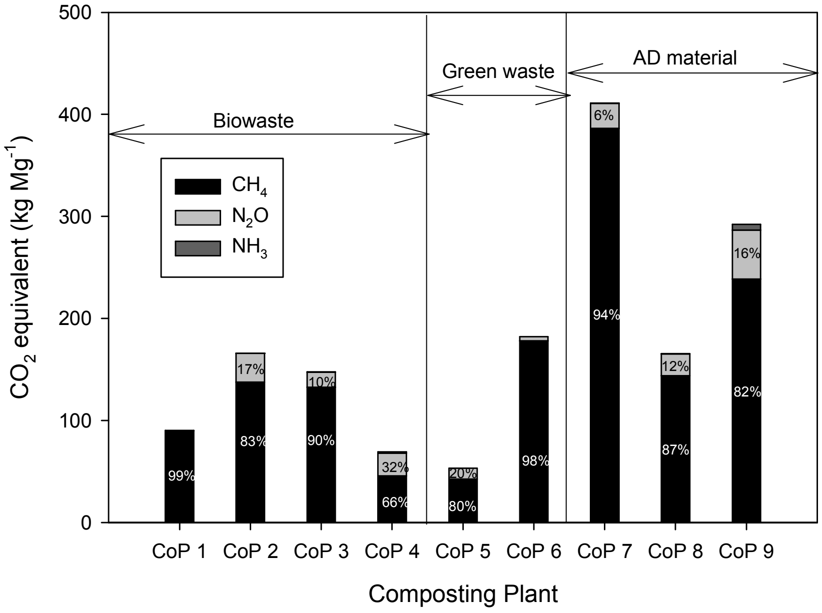

Figure 1.19: Total global warming emission factors calculated in kg CO_2 equivalent (Mg waste)$^{-1}$ from biowaste, green waste and AD material composting plants

The emission factors of CH_4, N_2O and NH_3 showed differences between biowaste, green waste and AD material composting (Figure 1.20). Average emission factors of CH_4 were 4,060 ± 1,713g (Mg biowaste)$^{-1}$, 4,415 ± 3,839g (Mg green waste)$^{-1}$ and 10,254 ± 5,593g (Mg AD material)$^{-1}$. N_2O emission factors were 55 ± 40g (Mg biowaste)$^{-1}$, 24 ± 16g (Mg green waste)$^{-1}$ and 105 ± 52g (Mg AD material)$^{-1}$. NH_3 emission factors were 157 ± 239g (Mg biowaste)$^{-1}$, 2g (Mg green waste)$^{-1}$, 699 ± 1,298g (Mg AD material)$^{-1}$. AD material, which was treated either in tabular windrows or by active aeration with semi-permeable covers, showed the highest CH_4, N_2O and NH_3 emissions. Biowaste-composting windrows emitted more N_2O

and NH_3 than green waste-composting windrows. The emission factors of N_2O and NH_3 in biowaste windrows were respectively 2.3 and 79 times higher than green waste windrows. The average emission factors of CH_4 from biowaste and green waste were fairly similar.

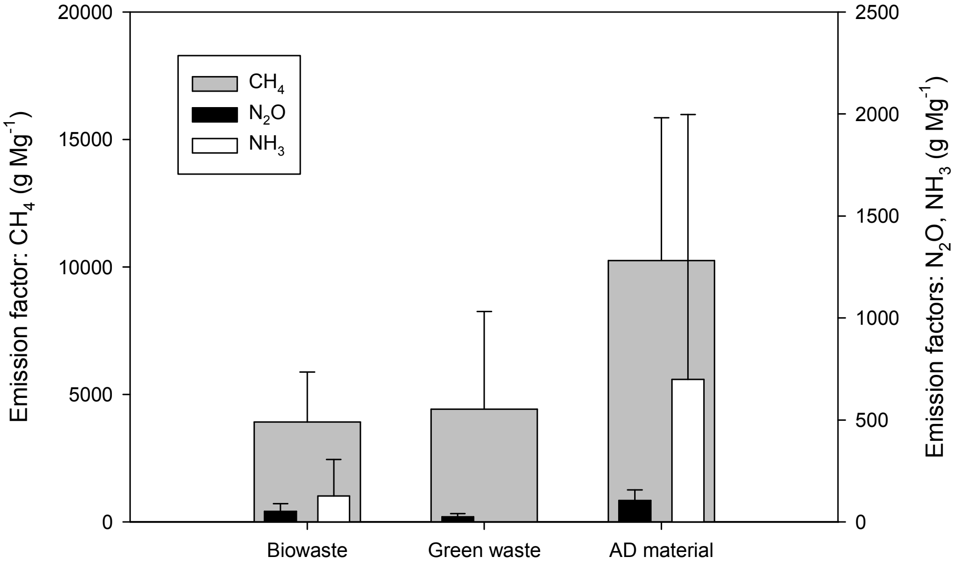

Figure 1.20: Mean CH_4, N_2O and NH_3 emission factors g (Mg waste)$^{-1}$ from the composting of biowaste, green waste and AD material; white bars: emission factor of NH_3; black bar: emission factor of N_2O, refer to the right axis; grey bar: emission factor of CH_4 refers to the left axis. Error bars: standard deviation (n= 4 for biowaste, n=2 for green waste and n=3 for AD material)

1.7 Conclusions

The composting of biowaste, green waste and AD material is a source of greenhouse gases. CH_4 is the major contributor to overall CO_2 emissions, whereas NH_3 emissions are insignificant. N_2O emissions contribute between 1 and 32% of total emissions.

Pore space concentration of CH_4 correlated with its emissions in small triangle windrows.

Analysing O_2 concentration alone is not enough to determine whether the windrow is aerobic or anaerobic. The CO_2/CH_4 ratio is an important factor in determining whether the windrows are anaerobic or aerobic.

GHG emissions varied at different plants, indicating that how they are managed is important in reducing GHG emissions during the composting process. Active aeration and frequent turning can reduce emissions of CH_4 significantly, but they may increase emissions of N_2O and NH_3. In order to reduce GHGs, small windrows with high turning frequencies and a short composting duration would result in the lowest emissions.

In Germany it is estimated that 4 million Mg biowaste and 6.5 million Mg of green waste could be treated by composting. Based on these results, biowaste and green waste composting would result in a 0.13% share of national emissions of N_2O and a 1.74% share of national emissions of CH_4.

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2. GREENHOUSE GAS EMISSIONS FROM ANAEROBIC DIGESTION PLANTS IN GERMANY

Abstract

This study investigated emissions of CH_4, N_2O and NH_3 from nine anaerobic digestion plants that treat biowaste. The treatment is in form of mechanical pre-treatment, anaerobic digestion followed by a composting with or without intensive aeration. The exhaust gases from the mechanical and anaerobic steps are treated by biofilters. The emission sources at the plants consisted of biofilters, combined heat and power units (CHP), liquid digestate treatment systems (LTS) and open composting windrows of the solid digestate. In average, the biofilters removed 30% of total organic carbon (TOC), 50% of non methane volatile organic carbon (NMVOC) and 51% NH_3, whereas N_2O concentrations increased by 26%. For CH_4 the biofilters had only a small removal effect (6%). Overall, the emission factors were 1.2-16kg (Mg biowaste)$^{-1}$ for CH_4, 9-172g (Mg biowaste)$^{-1}$ for N_2O and 41-6,031g (Mg biowaste)$^{-1}$ for NH_3. Open composting windrows of solid digestate resulted in high emissions of CH_4 and N_2O. Intensive aeration of the solid digestate could reduce greenhouse gas emissions.

2.1 Introduction

Anaerobic digestion for treatment of biowaste is rapidly gaining interest in Germany (Mata-Alvarez *et al.*, 2000; Fricke *et al.*, 2005). The treatment is essentially based on the activities of microorganisms that transform organic substances into biogas (Appels *et al.*, 2008). Biogas is used as renewable energy source, and nutrients in the residue can be recovered in agriculture as fertilizer or soil conditioner (Møller *et al.*, 2009). In addition, AD of biowaste is attracting attention as an effective method to reduce GHG emissions according to Kyoto protocol (Møller *et al.*, 2009). According to the life cycle analysis (LCA), AD results in negative GHG emissions. The total greenhouse gas (GHG) emissions for AD can reduce up to one tonne CO_2 equivalent/ Mg separated organic waste (Sanscartier *et al.*, 2011).

Actually, many studies have been conducted to show the benefits of AD treatment, for instance the works of Bockreis and Steinberg (2005), Fricke *et al.,* (2005), Zupančiča, (2008) and Møller *et al.,* (2009). However, there is still missing an overall evaluation of GHG emissions during treatment. For example, the GHG emissions associated to the pre-treatment and post-treatment of AD were often excluded in previous studies. In fact, AD plants may have fugitive emissions of CH_4, N_2O and NH_3.

The aim of the study was to investigate emission factors of CH_4, N_2O and NH_3 g (Mg biowaste)$^{-1}$ and to compare emission sources in the plants. Additionally, the efficiency of biofilters was taken into account. Nine operating AD plants, two wet digestion plants, four dry digestion plants and three solid digestion plants, were evaluated.

2.2 Hypotheses/ Objectives

2.2.1 Hypotheses

It is hypothesised that anaerobic digestion plants contribute significantly GHG emissions.

2.2.2 Objectives

The objectives were

- to quantify emission factors of CH_4, N_2O and NH_3 g (Mg biowaste)$^{-1}$.
- to compare different emission sources such as CHP, biofilter, composting and the liquid treatment system.
- to evaluate the biofilter's efficiency.

2.3 General background

2.3.1 GHG emissions in Germany

Overall, GHG emissions in Germany have been significantly reduced since 1990 (Figure 2.1). Total GHG emissions in Germany were 920 million tonnes of CO_2 equivalents in 2009 accounting 2.5% to the overall emissions in the world. Germany has a plan to reduce GHG emissions until the year 2020. The aim of this plan is to reduce GHG emissions to 749 million tonnes CO_2 equivalents which will be around 30% below the level in 1990 (UBA, 2009). For waste management, the aims are to reduce mass and volume of waste that enters the landfills to avoid emissions of landfill gases.

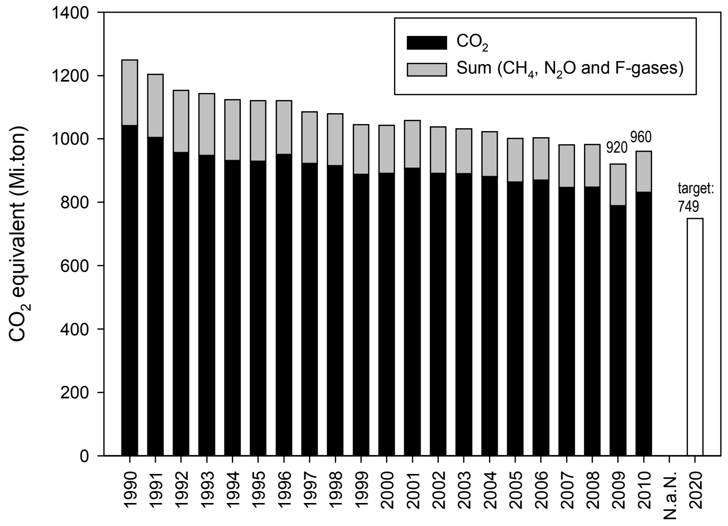

Figure 2.1: GHG emissions in Germany from 1990 to 2010 (UBA, 2011)

2.3.2 Biowaste generation

In Germany, the generation of biowaste doubled from 6.5 million tonnes in 1996 to 13 million tonnes in 2009. Generally, biowaste consists of animal manure, agricultural waste, wood processing waste, sewage sludge, garden waste, organic household waste and food waste. In fact, organic household waste and garden waste contribute significantly to the total waste in Germany. There were 4 million tonnes of garden waste and 3.8 million tonnes of organic household waste collected in 2009, which was equivalent to 31% and 29% of the total biowaste respectively (UBA, 2011) (Figure 2.2).

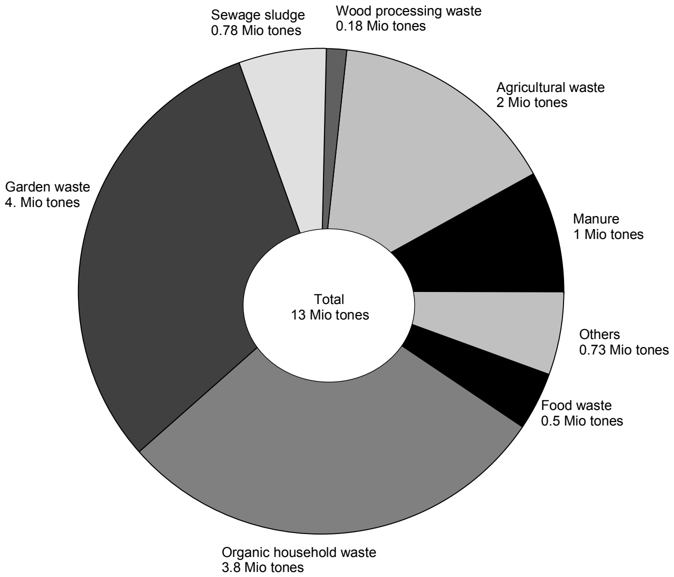

Figure 2.2: Summary of the statistics collected biowaste in 2009 (UBA, 2011)

2.3.3 Treatment of biowaste

Organic waste is collected separately and treated biologically either by composting or by anaerobic digestion (Weiland, 2000; Clemens and Cuhls, 2003). Since the anaerobic digestion plants contribute to prevent GHG emissions from fossil fuels combustion due to energy recovery from biogas (Møller *et al.,* 2009), many different anaerobic digestion types are applied in Germany. They vary in fermenter systems, material and digestion conditions. Overall, there are 85 full-scale anaerobic treatment plants in Germany with capacities from 5.000 to 150.000 Mg year^{-1} in Germany (Weiland, 2000; Puchelt, 2000 and UBA, 2009).

2.3.4 Anaerobic digestion process

Anaerobic digestion is a complex biological process in which the organic material is decomposed in the absence of oxygen. Biogas and digestate are produced (Duerr *et al.*, 2007). Typical biogas contains 55-65 vol% CH_4, 30-40 vol% CO_2 and small amounts of H_2S and NH_3 (lower than 1%). The production of these trace gases in the biogas depends on the sulphur and nitrogen content in the substrate (Duerr *et al.,* 2007; Appels *et al.,* 2008; Papport, 2008; Li *et al.,* 2010). The anaerobic digestion of organic material basically consists of following steps: hydrolysis, acidogenesis, acetogenesis and methanogenesis as shown in the Figure 2.3. The hydrolysis step breaks down high molecular compounds (e.g. lipid, protein) to soluble substances such as sugar, amino acids and fatty acid. The second step is acidogenesis, where volatile fatty acid (VFA), acetic acid, H_2 and CO_2 are produced. During methanogenesis, VFA are converted into acetic acid along with H_2 and CO_2. CH_4 and CO_2 are formed by conversion of acetate into CH_4 and CO_2 or by a reaction of H_2 and CO_2 to produce CH_4 (Appels *et al.,* 2008). The optimal conditions for anaerobic digestion are a neutral pH and a constant temperature either mesophilic (30-40^{o}C) or thermophilic (50-60^{o}C) (Papport, 2008).

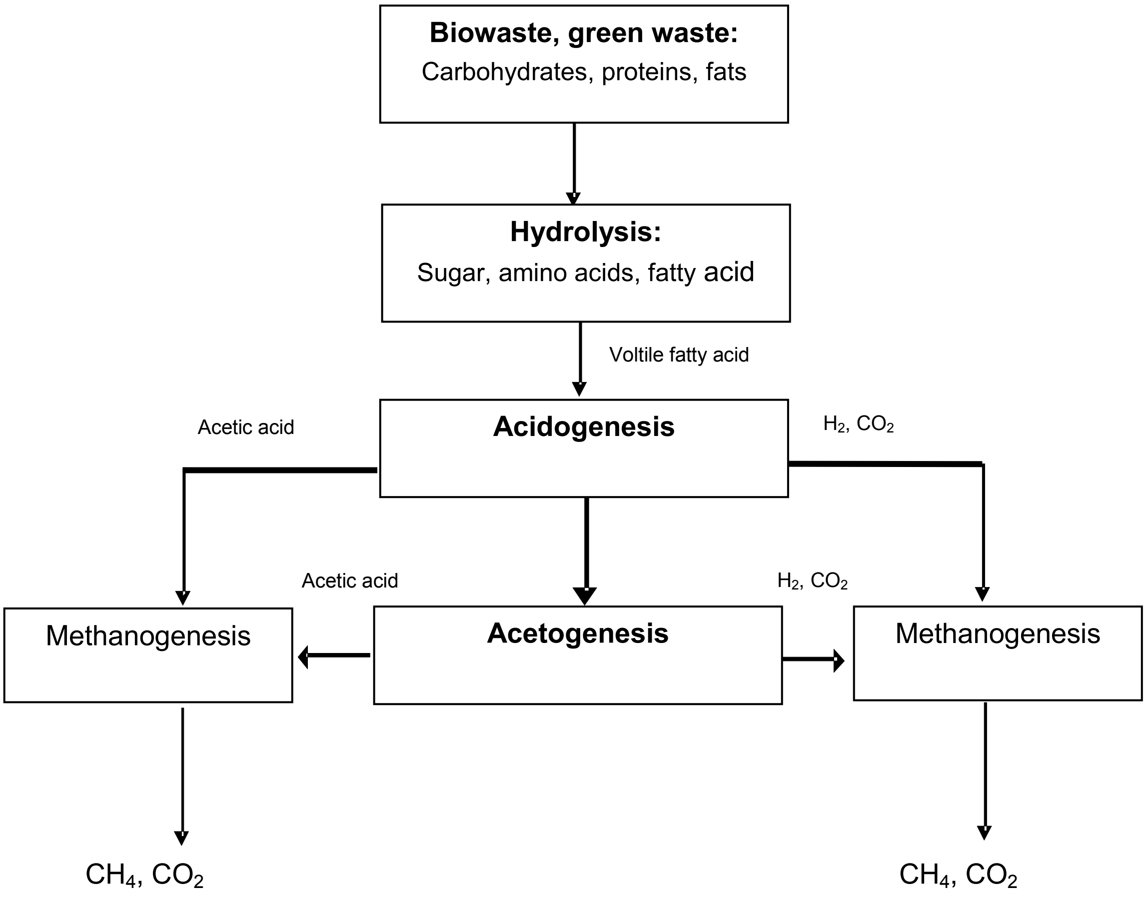

Figure 2.3: Main steps and pathways of the degradation of organic material through anaerobic digestion process (Appels et al., 2008)

2.3.5 Overview of an anaerobic digestion plant

Anaerobic digestion plants combine mechanical and biological processes. The mechanical process removes inert components such as iron, timber and plastic, which disturb the biological process. The biological process includes anaerobic fermentation or combined anaerobic fermentation and aerobic composting. The largest advantage of anaerobic digestion is energy recovery. Biogas collected from anaerobic fermenter is used as a substitute for fossil fuels to produce electricity and heat (Soyez, 2002). Digestate is treated by composting with or without intensive aeration.

There are two typical fermenters: continuous and batch fermenters. The continuous fermenter is quasi-continuously stirred and is suitable for substrate with a high moisture content (>85%) (Duerr *et al.,* 2007). Fresh substrate is fed to the fermenter in regular intervals and the same amount of digestate is removed. Due to the continuous feeding, the operation and biogas production are stable. Batch fermenters are used for substrate with a lower moisture content (<85%) (Li *et al.*, 2010). During the anaerobic period, leachate is pumped back from a percolator to the batch fermenter to inoculate a new batch. When the digestion is finished, the fermenter is opened for unloading the digestate and reloading with new substrate by front end loader (Figure 2.4) (Yoshida *et al.*, 2011). The large advantages of the batch fermenter are the low cost investment and the simplicity of the technology (VDI 3745, 2005).

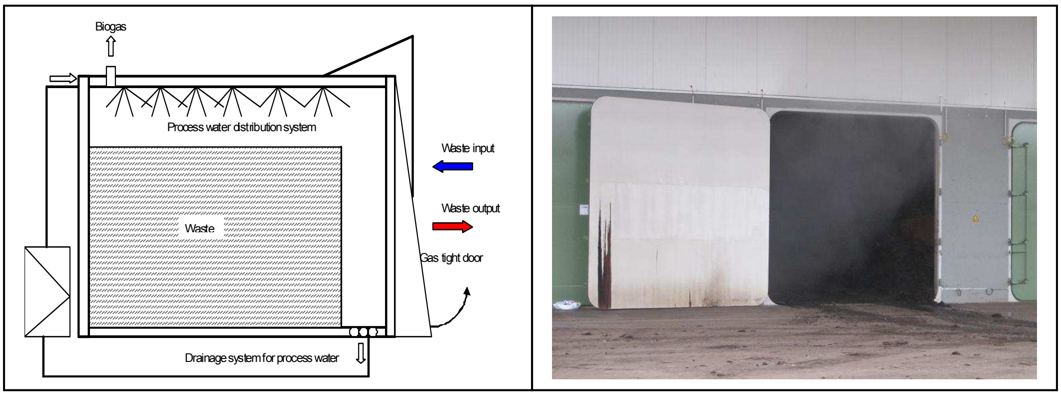

Figure 2.4: Left: Schematic of batch fermenter. Right: A fermenter is opened for unloading digestate

2.3.6 Types of anaerobic digestion

Wet anaerobic digestion can be employed to material with less than 15% of total solids (TS) (VDI 3475, 2005). The input material is mixed with liquid digestate and pumped into a fermenter. After digestion, the digestate is separated to a solid and a liquid fraction, e.g. by a screw press. A part of the liquid digestate is reused as inoculate. The solid fraction is mixed with green waste and treated aerobically to produce compost.

Dry anaerobic digestion can be used if the TS of the input material varies from 15-30% (VDI 3475, 2005, Bolzonella *et al.,* 2003). Fermenters are operated in batch or continuous fermentation in thermophilic conditions (ca. 55^{o}C). After digestion, the digestate is also separated into a solid and a liquid fraction.

Solid anaerobic digestion can be applied for input material with a TS >30% (VDI 3475, 2005; Li *et al.,* 2010). The fermenters are usually rectangular concrete fermenters. The floors of the fermenters are perforated for leachate collection.

They are operated in batch fermentation at mesophilic conditions (37-42°C). After the fermenter is filled, it is closed by air tight doors. Because of the rather low water content, the digestate is composted directly.

2.3.7 Emission sources

2.3.7.1 Encapsulated emission sources

In the receiving hall, biowaste is delivered and placed on a floor before feeding to the mechanical processing. The incoming biowaste is stored in the receiving hall from a few hours to several days before it is fed into a fermenter. Thus, emissions may take place from the degradation of biowaste in the receiving hall.

In the pre-treatment hall, technical equipment is installed to remove ferrous metals as well as plastic. Then, the waste is shredded and transported by conveyor belts into a screen. The oversize material such as big parts of the trees is crushed in a shredder. After pre-treatment, the digestible organic substances are fed to the fermenters.

The anaerobic degradation is carried out inside the fermenters. Fugitive GHG emissions at batch fermenters may occur when fermenters are opened for refilled.

After digestion, the effluent of the fermenters is separated. The separators are either screw press or decanters which divide the digestate into a solid and a liquid fraction. The solid fraction is composted in composting tunnels or in open windrows. The liquid fraction is recirculated to the fermenter. Excess liquid is treated by conventional waste water treatment processes before being discharged to the sewer system.

In compost halls, digestate is aerated and stabilized (Figure 2.5). Exhaust air is usually collected and treated by a biofilter.

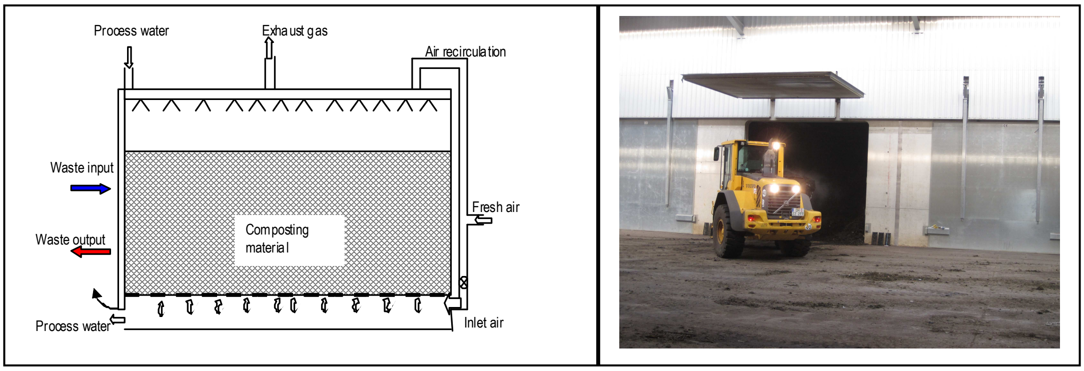

Figure 2.5: Left: Schematic of intensive composting tunnel. Right: A front end loader is unloading material at a composting tunnel

2.3.7.2 Open emission sources

Biofilter: exhaust gases from the encapsulated parts of AD plants are treated by biofilters before they are released into the atmosphere. In a biofilter, waste gas passes biofilter material (e.g. wood chip and root bark) and organic compounds are degraded by microorganisms (Hort *et al.*, 2009). The performance of a biofilter depends on the composition of the exhaust gas, packing material, nutrient supply, temperature, pH, pressure drop and residence time (Deshusses *et al.*, 1999).

Combined heat and power units (CHP): CHPs consist of a combustion engine and a generator. Biogas is used to generate electricity and heat in these combustion engines. Electricity produced is fed into the local grid, whereas heat is usually used to maintain temperature in the fermenters. During the combustion process, CH_4 is burned to generate electricity. Some unburned CH_4 is emitted into the atmosphere. By this way, CHP contributes to GHG emissions (Møller *et al.*, 2009).

Liquid treatment system (LTS): in wet anaerobic digestion plants, additional water is needed to guarantee mixing and pumping of the material. After digestion, there is excess liquid digestate that needs to be treated before it is discharged into the environment. The liquid treatment system includes nitrification and denitrification tanks. During waste water treatment, N_2O may be released (Clemens and Ahlgrimm, 2001; Paul, 2009 and Sánchez-Monedero *et al.,* 2010).

Composting windrows: solid digestate is mixed with green waste and used for composting. The material is composted either in aerated composting tunnels or in static windrows.

2.4 Materials and Methods

Nine German AD plants were investigated in the study. The detailed inventories of the AD plants are listed in Table 2.1.

Table 2.1: Processing parameters, emission sources and measured points of the anaerobic digestion plants

Greenhouse gas emissions from the anaerobic digestion plants										
Anaerobic digestion Types		Wet digestion TS<15%		Dry digestion TS: 15-30%				Solid digestion TS>30%		
Fermenter Feeding		Continuous	Continuous	Continuous	Continuous	Continuous	Continuous	Batch method	Batch method	Batch method
		PLANT 1	PLANT 2	PLANT 3	PLANT 4	PLANT 5	PLANT 6	PLANT 7	PLANT 8	PLANT 9
Pre-treatment		Machenical	Machenical	Machenical	Machenical	Machenical	Machenical	Machenical	Machenical	Machenical
Digestion										
DM content	**Input**	-	-	-	ca. 30% DM	ca. 30% DM	15-45% DM	-	-	-
	Output	-	4% DM	-	-	-	-	-	-	-
Temperature range		mesophilic	thermophilic	thermophilic	thermophilic	thermophilic	thermophilic	mesophilic	mesophilic	mesophilic
Hydraulic Retention time		-	20 days	15-30 days	15-20 days	15-20 days	21 days	28 days	21 days	21 days
AD material treatment										
Separation		Centrifuge	Decanter	Belt dryer	Press	Press	Press	-	-	-
		↓	↓		↓	↓	↓	↓	↓	↓
AD composting		Tabular	Tabular	No treatment	Tunnel	Tunnel	Tunnel	Tabular aerated	Tunnel	Triangle windrows
Measured points										
Biofilter		x	x	x	x	x	X	x	x	x
Combined heat and power units		▨	▨	x	x	▨	▨	x	▨	▨
Open windrow		x	x							x
Liquid treatment		x								
Receiving hall			x	x	x					
Re-treatment hall				x						
Closed composting chamber					x					
Fermenter			x					x		
Separator							X			
Belt dryer				x						
Liquid tank from separator							X			
Storage tank						x	X			
Solid storage		x								

▨ : emission sources

x : emission sources measured

2.5 Sites descriptions of AD plants

2.5.1 PLANT 1

The plant is designed to process 30,500 Mg biowaste per year. All of the incoming biowaste enters the anaerobic digestion system. The separated biowaste is unloaded into an underground bunker. The biowaste is then transported to a drum mill by a conveyer band for crushing and from there to a hydrolysis pulper. The crushed biowaste is suspended in process water in the hydrolysis pulper. The floating fraction and the heavy fraction are removed. A centrifuge is then used to separate the liquid and solid fraction. The liquid fraction is pumped directly to a fermenter, while the solid fraction is mixed with process water and then pumped into a hydrolysis tank with a retention time of 4 days. The fermenter is capable to handle waste with 10% total solid (TS) at mesophilic temperatures. The liquid fraction from the hydrolysis tank is also pumped into the fermenter. After anaerobic digestion, the digestate is dewatered by a second centrifuge. The solid digestate is mixed with green waste and used for composting. The liquid is treated in nitrification and denitrification tanks. Gases emitted from the receiving hall, the pre-treatment hall and the solid digestate storage hall are treated by a capsuled biofilter (Figure 2.6).

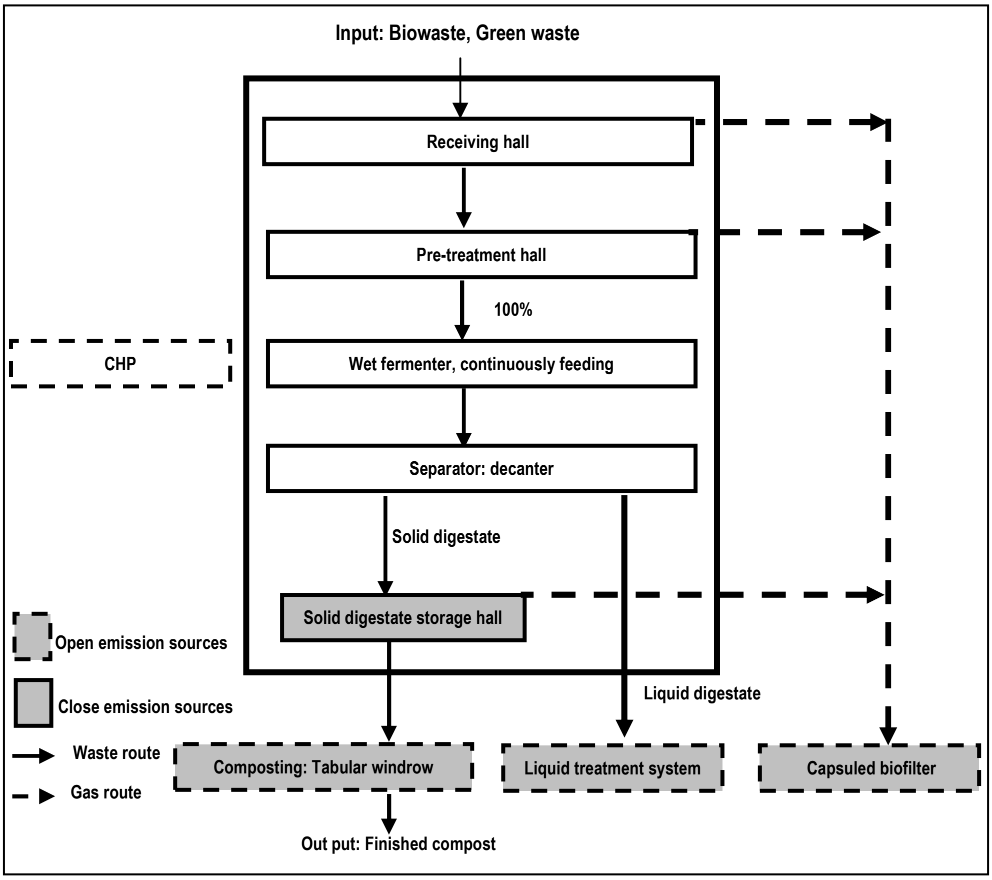

Figure 2.6: Schematic waste and gas flow diagram and measuring points (marked in grey) of plant 1

2.5.2 PLANT 2

The total capacity of the plant is 21,000 Mg year^{-1}. All of the incoming biowaste enters the continuous anaerobic digestion system. The plant consists of one wet fermenter. The biowaste is first shredded and mixed with process water. The mixture is then pumped into a vertical fermenter, where temperature is maintained at thermophilic conditions (55°C). The substrate is stirred continuously. The hydraulic retention time is about 20 days. Feed is added to the fermenter at regular intervals. The digestate is separated into a solid and a liquid fraction by a decanter. The liquid fraction is re-circulated into the process. The excess liquid is transported to farms and used as liquid fertilizer. The solid digested material is further matured by being placed on a tabular windrow for about 20 weeks. Gases that emit from the receiving and the pre-treatment halls are treated by an open biofilter (Figure 2.7).

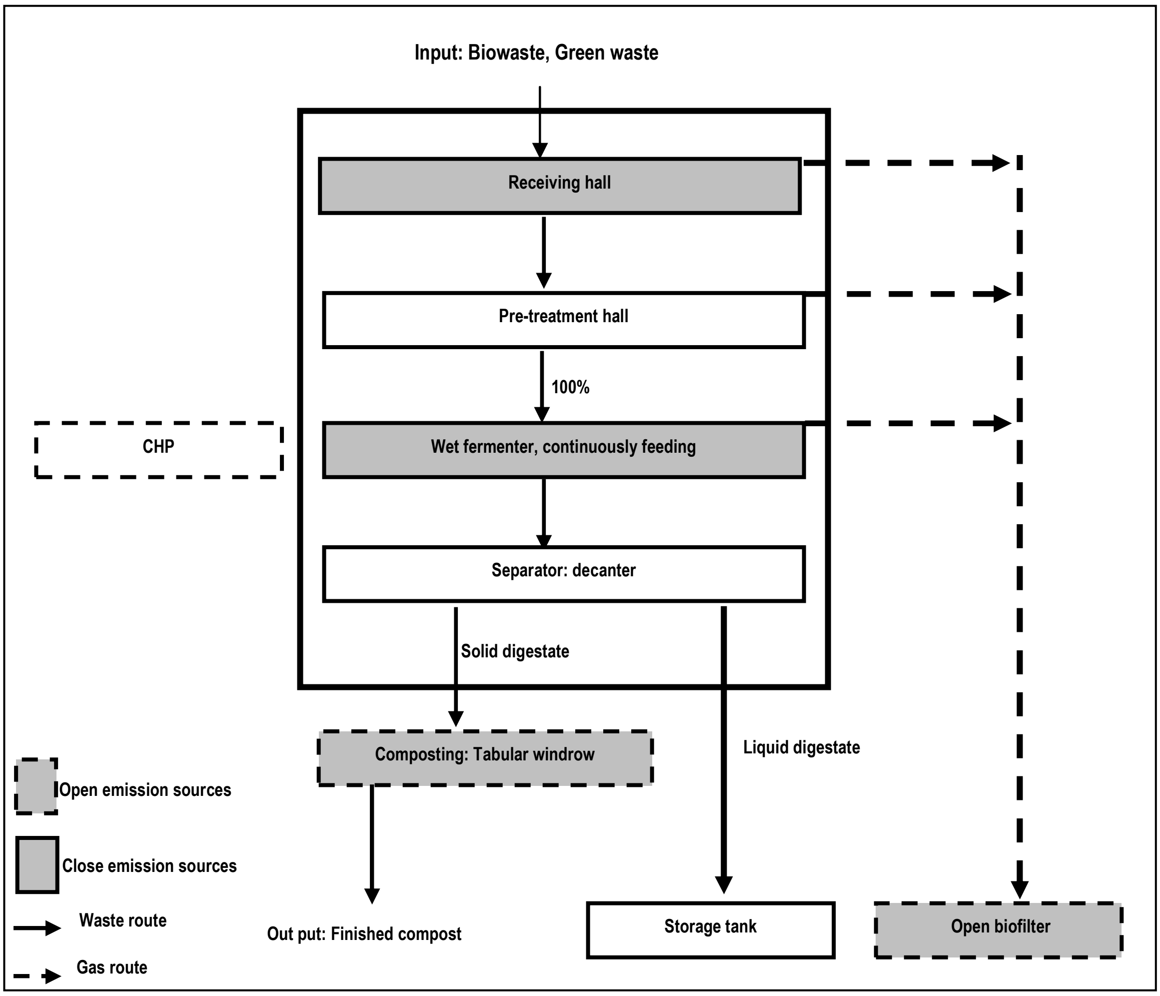

Figure 2.7: Schematic waste and gas flow diagram and measuring points (marked in grey) of plant 2

2.5.3 PLANT 3

The total capacity of the plant is 32,000 Mg year^{-1}. After pre-treatment, all of the shredded biowaste is digested in the anaerobic digestion fermenters. The site consists of one vertical fermenter. The fermenter has a cylindrical shape with a volume of 2,700m^3. It is a single-stage anaerobic digestion fermenter that operates at thermophilic temperatures with a retention time of 21 days. Material is fed into the top of the fermenter and moves downward to the conical bottom where digestate is removed by an auger. A part of the digestate is pumped back to the mixing pump where it is mixed with fresh waste as inoculate. The waste in the fermenter is neither mixed nor heated. The accumulated digested solids at the bottom of the fermenter are regularly removed. The digestate is directly dried in a belt dryer with temperatures about 1000°C by combustion of oil and biogas. Dry digestate is transported to a composting plant. Biogas is stored and subsequently burned in combined heat and power engines. Gases produced from the belt dryer, the receiving and the pre-treatment halls are collected and routed through a biofilter (Figure 2.8).

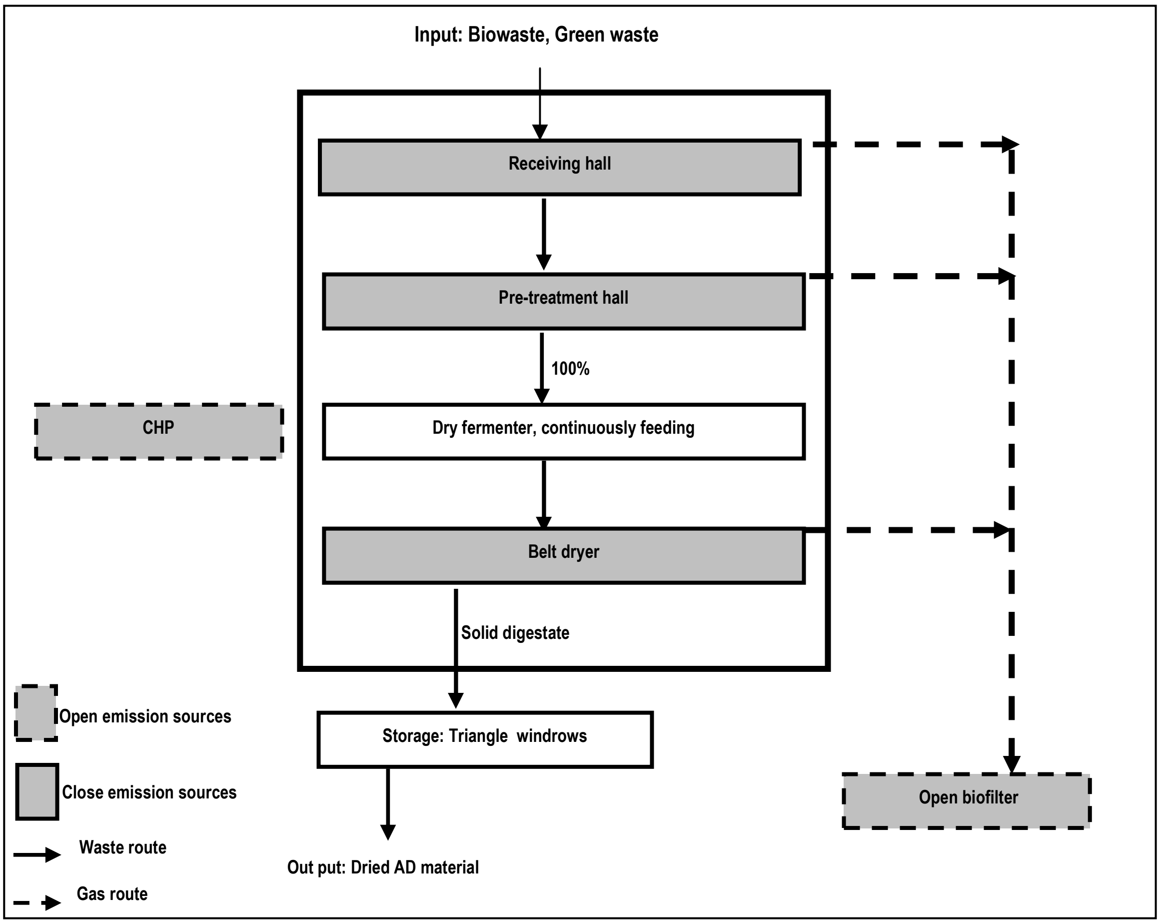

Figure 2.8: Schematic waste and gas flow diagram and measuring points (marked in grey) of plant 3

2.5.4 PLANT 4

The total capacity of the plant is 30,000 Mg year^{-1}. Only 50% of incoming biowaste enters the anaerobic digestion fermenter. The rest of the waste is mixed with digestate for a composting process. Unlike the other single-stage dry fermenters, the plant 4 uses a horizontal plug flow fermenter with an internal agitator to mix the waste. The fermenter is operated with a retention time of 14 days under thermophilic conditions. In average, the temperatures are about 57°C. After digestion, the digestate is separated into a solid and a liquid fraction by a screw press. The solid fraction is aerated in 10-12 days in an intensive composting tunnel without turning. Gases emitted from the receiving hall and the pre-treatment hall as well as from the intensive composting tunnels are treated by a capsuled biofilter (Figure 2.9).

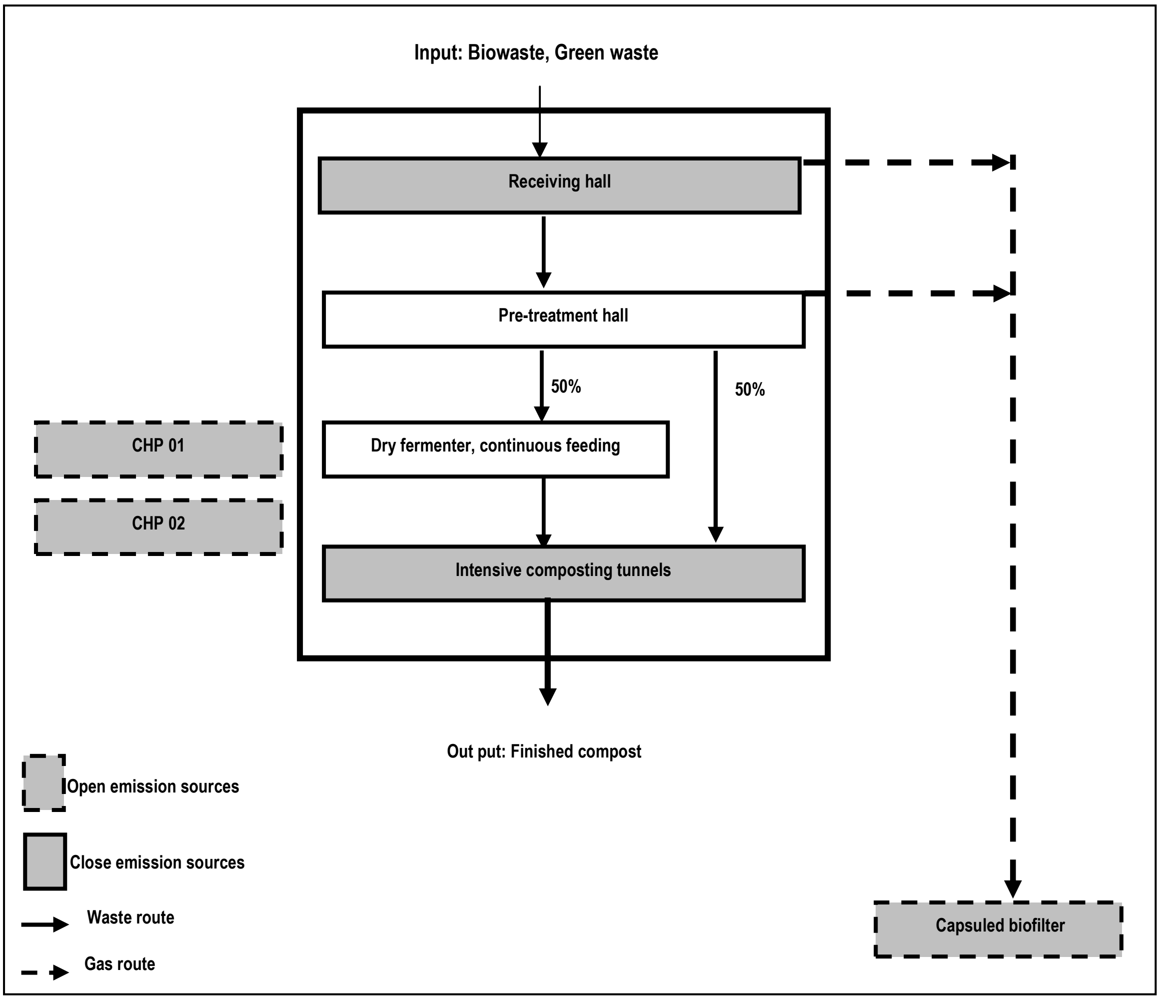

Figure 2.9: Schematic waste and gas flow diagram and measuring points (marked in grey) of plant 4

2.5.5 PLANT 5

The total capacity of the plant 5 is 29,500 Mg year^{-1}. About 62% of the incoming biowaste is digested in the fermenter. Similar to plant 4, the plant has only one horizontal fermenter with a slowly rotating agitator that transports the material from the inlet to the outlet. The digestate and percolate are mixed with fresh material as inoculums before the fresh material enters the fermenter. The waste is fed to the fermenter at regular intervals and removes an equal amount of digestate. The fermenter operates at thermophilic conditions (55^{o}C) with a retention time of about 14 days. After digestion, the digestate is separated in a solid and a liquid fraction by a screw press. The solid digestate is then mixed with green waste and aerated for two days. Then, the mixture is composted in an intensive composting tunnel for 10 days without turning. Two open biofilters clean the exhaust air from the composting tunnels and the fermenter separately (Figure 2.10).

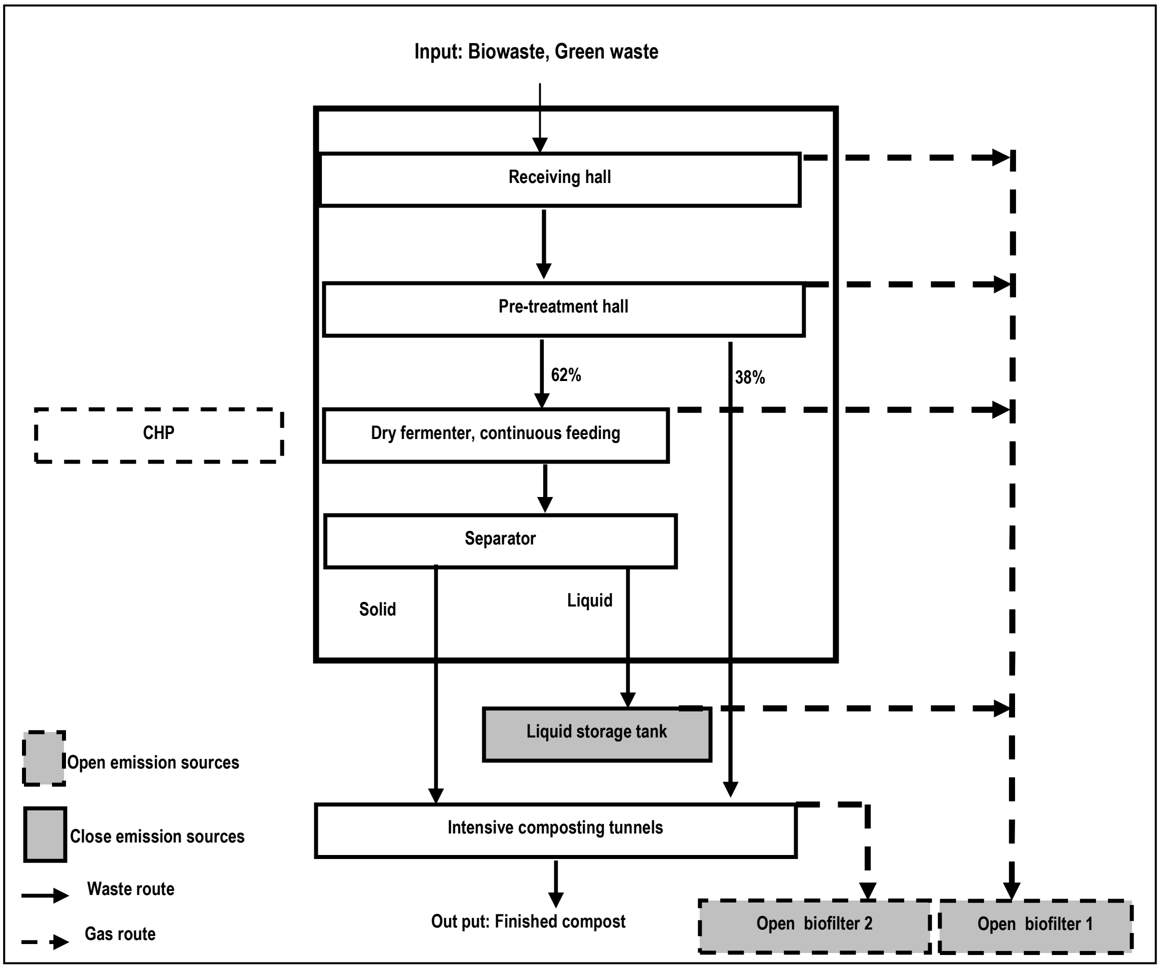

Figure 2.10: Schematic waste and gas flow diagram and measuring points (marked in grey) of plant 5

2.5.6 PLANT 6

The total capacity of the plant is 23,000 Mg year^{-1}. About 75% of the incoming waste enters the fermenter (volume of 1600m^3). Similar to plant 5, biowaste from the pre-treatment hall is transported via a conveyor belt to a horizontal plug-flow fermenter with internal rotors for mixing. The process is operated at thermophilic conditions. At the end of the fermenter, digestate is directly discharged from the fermenter to a screw press to separate the liquid and the solid digestate. The liquid digestate is stored in a small tank and subsequently pumped to a big storage tank. The solid digestate is further treated in intensive composting tunnels for 21 days without turning. Gases that emit from the screw press, the small and big liquid storage tanks are routed through an acid scrubber before they are treated by a capsuled biofilter (Figure 2.11).

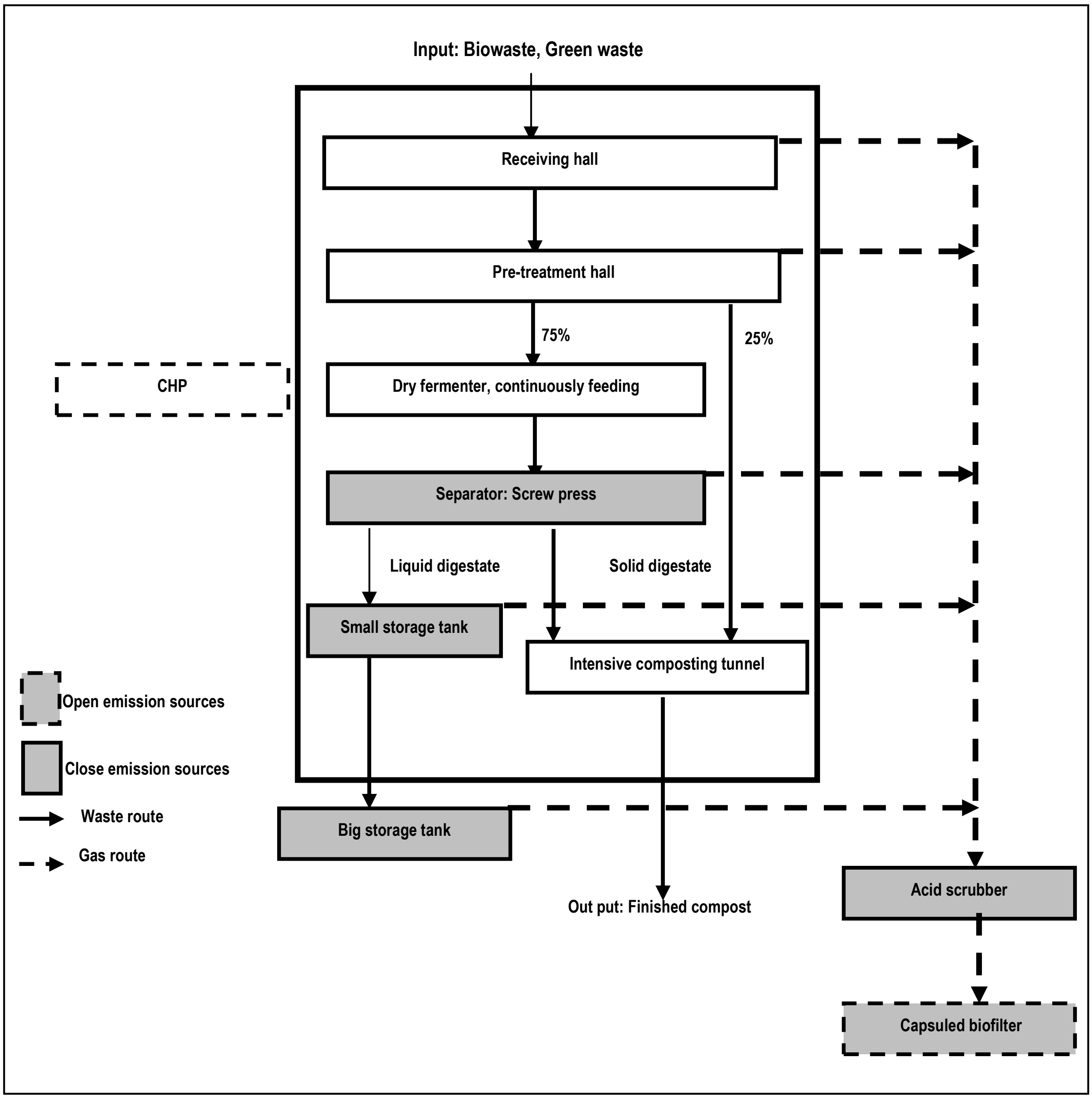

Figure 2.11: Schematic waste and gas flow diagram and measuring points (marked in grey) of plant 6

2.5.7 PLANT 7

The total capacity of the plant is 38,000 Mg year^{-1}. About 48% of the total waste enters the anaerobic digestion system. About 52% of the waste is treated by composting. The plant consists of six boxes for solid fermentation. The pre-treated waste is mixed in a 1:1 ratio with the digestate as inoculum. The mixture is filled into the fermenter by front end loader. The material in the fermenter is periodically sprayed with percolate that is collected below the fermenters. The fermentation process lasts about 4 weeks. Temperatures range from 37 to 39°C. At the end of the digestion, the fermenter is opened. Then, half of the material is transported to an intensive composting hall. The second half is used to mix with the fresh input material and refilled the fermenter. Biowaste and digestate from the anaerobic digestion are mixed and distributed on a tabular windrow in the hall. Shredded trees and branches are also added as bulking agent. The digestate is continuously aerated. Once a week, the windrow is turned and piled up again by an automatic turning equipment. The composting process lasts 8 weeks. The exhaust air from the composting, the receiving hall and the pre-treatment hall is treated by a biofilter (Figure 2.12).

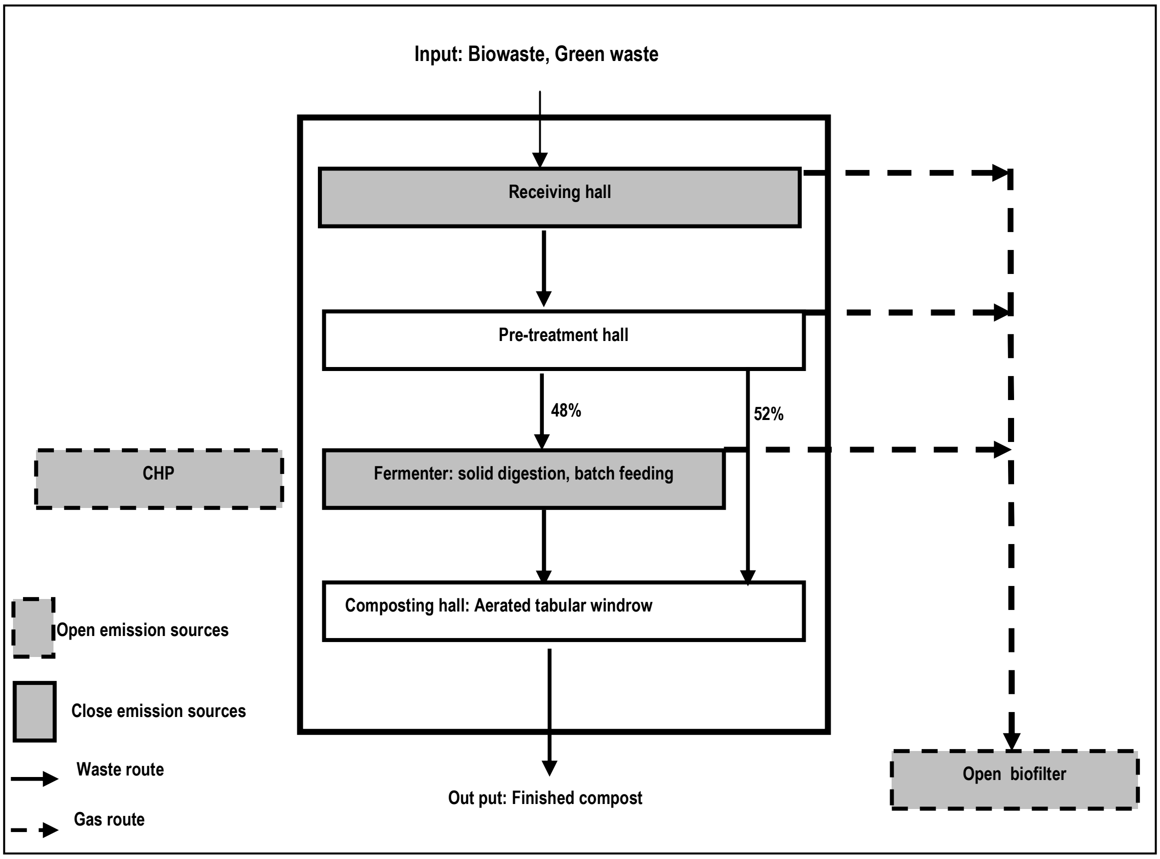

Figure 2.12: Schematic waste and gas flow diagram and measuring points (marked in grey) of plant 7

2.5.8 PLANT 8

The total capacity of the plant is 85,000 Mg year^{-1}. Only about 30% of the waste is treated by anaerobic digestion. About 70% of the waste is treated in a composting tunnel. The plant consists of 8 batch fermenters. Each fermenter treats 150 to 190 Mg waste as a batch. The fermenters are operated with a retention time of 21 days under mesophilic conditions (40-42°C). Afterwards, the digestate is aerated in intensive composting tunnels for 18 days. During the composting process, the digestate is turned once. Then, the digestate is piled up in small triangle windrows placed on a floor with an aeration system and is composted for another 16 days. Exhaust air from the composting tunnels, the receiving hall and the pre-treatment hall are treated by a biofilter. The biofilter consists of two segments (each 25m x 26m) with a total area of 1.300m^2 (Figure 2.13).

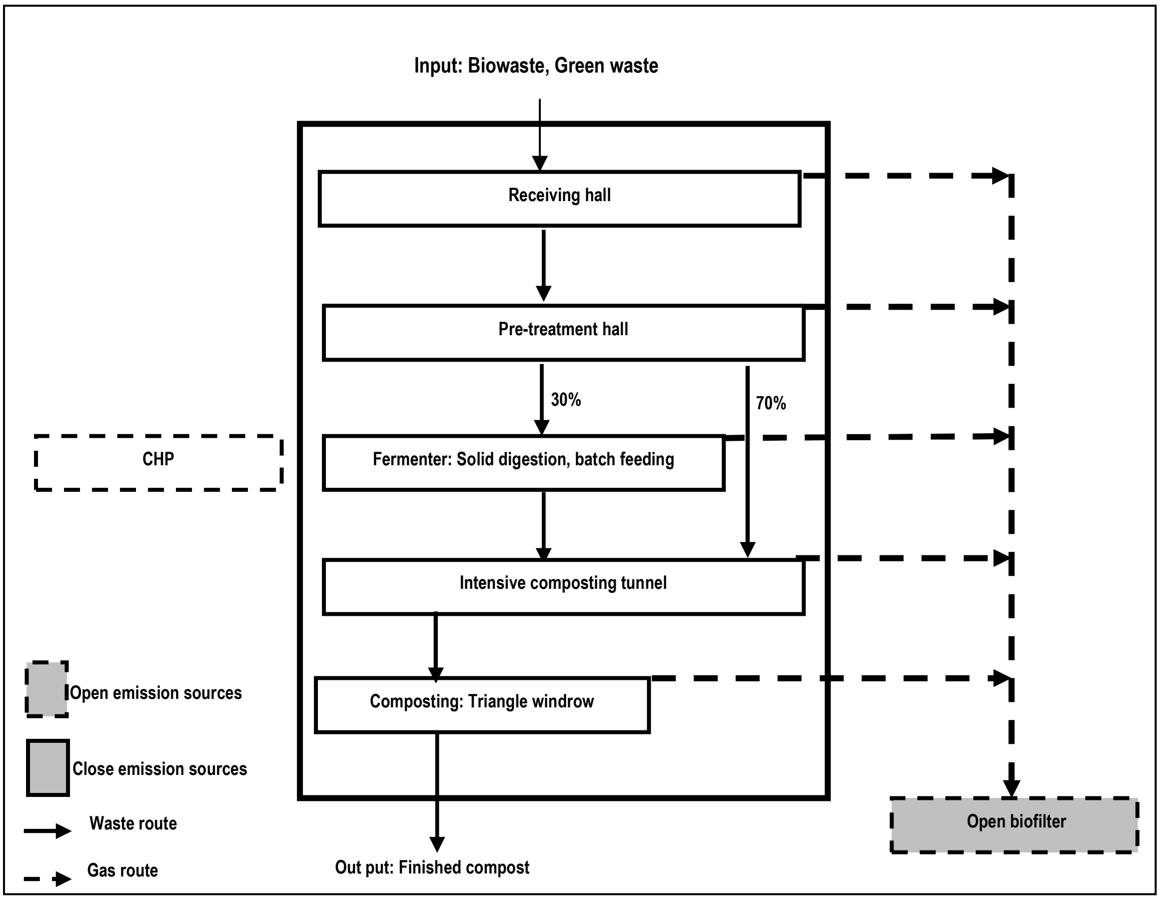

Figure 2.13: Schematic waste and gas flow diagram and measuring points (marked in grey) of plant 8

2.5.9 PLANT 9

The total capacity of the plant is 12,000 Mg year^{-1}. All of the coming waste enters the anaerobic digestion system. The plant consists of 5 garages. Similar to the plant 8, the fermenter is also filled by front end loader. Each fermenter treats up to 260 Mg waste. The fermenters are operated with a retention time of 21 days under mesophilic conditions (40-42°C). When the anaerobic process is stopped, the fermenter is opened and the digestate is unloaded by front end loader. A part of digestate is mixed with fresh waste in the receiving hall before it is refilled back to the fermenter. Gases emitted from the receiving hall are routed through two container biofilters. The digestate is piled up in triangle windrows (height: 2.5m) covered by semi-permeable membrane sheets and aerated for 4 weeks. Thereafter, the digestate is mixed with green waste and piled up to smaller triangle windrows (height: 1.2m) for 60-70 days. The small triangle windrows are turned once a week (Figure 2.14).

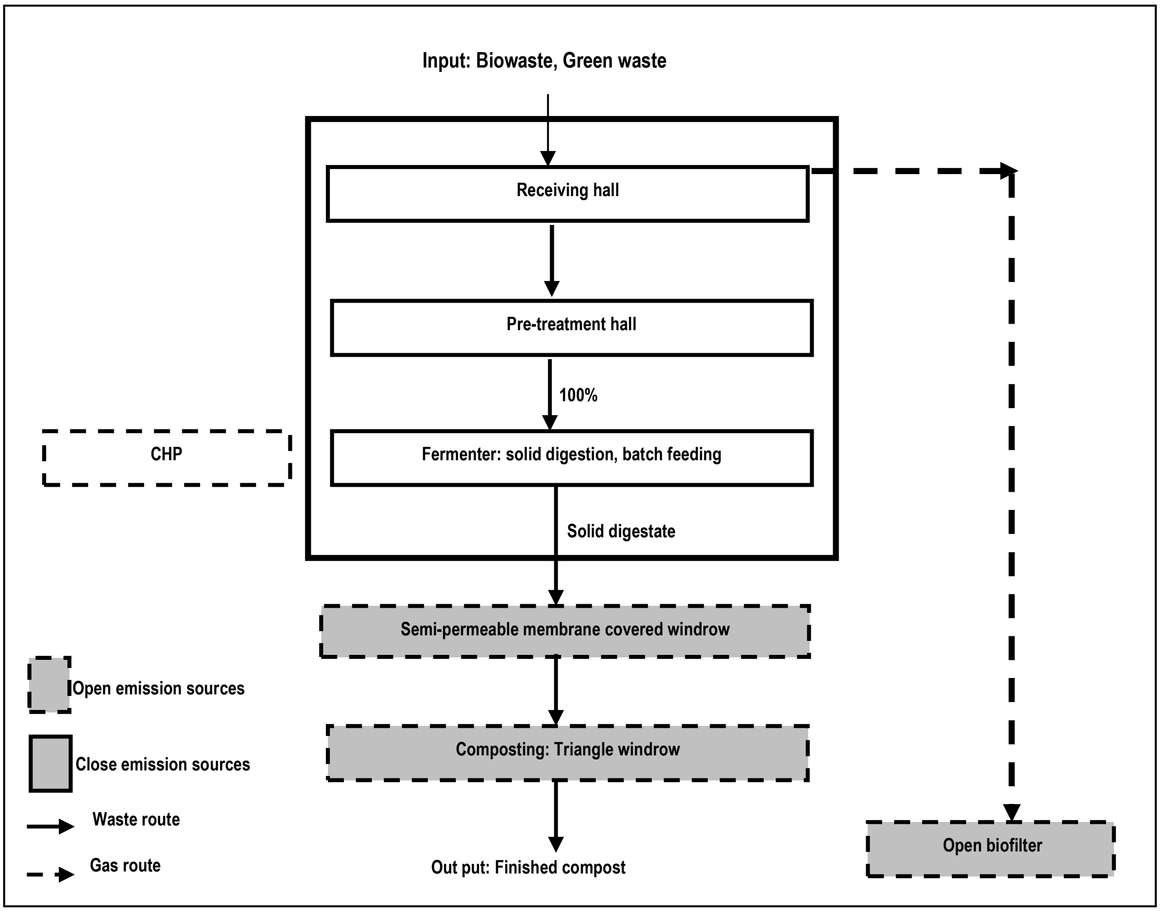

Figure 2.14: Schematic waste and gas flow diagram and measuring points (marked in grey) of plant 9

2.6 Measured locations and emission determinations

2.6.1 Biofilter

The gas before and after biofilter was analysed at each plant for 1 week. At capsuled biofilters the treated air left the biofilter in a chimney. Here the gases were measured (biofilters at plants 1, 4, 6 and 9). At open biofilter (at plants 2, 3, 5, 7 and 8), 16m^2 of the biofilter (4x4m) was covered by a thin foil. Concentrations of the treated gases were measured under the foil (Figure 2.15). Continuously monitored parameters included TOC, CH_4 and N_2O. TOC was measured by flame ionisation detector

(Bernath Atomic 3006) while CH_4 and N_2O were measured by an infrared gas analyser (ABB) (compare chapter 1). Gas concentrations in the treated and untreated exhaust air were recorded every minute. To control the accuracy of the infrared gas analyser, exhaust gases were sampled manually by evacuated headspace vials and subsequently analysed on CH_4 and N_2O by GC (ECD/FID) in the laboratory. A manual discontinuous analysis was applied for NH_3 measurement: NH_3 was extracted from the waste gas stream by absorbing it in sulfuric acid and subsequently measured colorimetrically in the laboratory (compare chapter 1). NH_3 samples of treated and untreated gases were collected twice. Air fluxes to the biofilter were measured by an anemometer (testo 435) or micromanometer (Müller Instruments EPM-300-BA, Germany). It was assumed that the volumes of treated and untreated air were the same.

Figure 2.15: Left: Sampling of gases at before biofilter and measuring equipments. Right: Foil covers on a biofilter for CH_4, N_2O and NH_3 concentration measurements

2.6.2 Open composting windrows

The measurements at open composting windrows were described in the chapter 1 (materials and methods).

2.6.3 Other measuring points (e.g. CHP, receiving and pre-treatment hall)

Other emission sources were point sources with preinstalled sampling points. For one hour the TOC concentrations were recorded every minute by FID. In parallel, gas samples were taken regularly using evacuated headspace vials for CH_4 and N_2O. For

NH_3, samples were taken by absorbing it in sulfuric acid solution. Air fluxes were also determined by measuring velocity (m/s) and cross section area (m^2).

2.6.4 Calculations of emissions factors for anaerobic digestion plants

The emission factors of CH_4, N_2O and NH_3 g (Mg biowaste)$^{-1}$ were calculated using the aeration rates and concentrations of gases. The emission rates and emission factors for each gas were calculated using the following formula:

$$E_{MF} = \frac{E \times Q}{1000} \qquad (\text{g h}^{-1})$$

$$E_f = \frac{(E_{MF} \times 24 \times 7)}{M_w} \qquad \text{g (Mg waste)}^{-1}$$

With

E: concentration (mg x m^{-3})

Q: air flow (m^3 x h^{-1})

E_{MF}: emission mass flow (g x h^{-1})

M_w: total mass of incoming waste (Mg per week)

E_f: emission factor g (Mg waste)$^{-1}$

The emissions were calculated in form of CO_2 equivalent according to IPCC (2007). N_2O and CH_4 are potential GHG with respective global warming potentials 298 and 25 times higher than that of CO_2 respectively (IPCC, 2007). Additionally, it was assumed that the CO_2 equivalent of NH_3 is 2.98 (Cuhls, 2011) and (Wulf, 2002).

$$E_{fCO_2 equivalent} = \sum (E_{fCH_4} \times 25 + E_{fN_2O} \times 298 + E_{fNH_3} \times 2.98)$$

Overall GHG emissions from AD plants were calculated by the sum of emissions of CH_4, N_2O and NH_3 from open emission sources such as biofilter, CHP, open composting windrows and liquid digestate treatment systems. Emissions from machinery and energy used in the plants were not considered in the calculations.

$$E_{plant1} = \sum (E_{BF} + E_{OW} + E_{LTS})$$

$$E_{plant2} = \sum (E_{BF} + E_{OW})$$

$$E_{plant3} = \sum (E_{BF} + E_{CHP})$$

$$E_{plant4} = \sum (E_{BF} + E_{CHP})$$

$$E_{plant5} = \sum (E_{BF})$$

$$E_{plant6} = \sum (E_{BF})$$

$$E_{plant7} = \sum (E_{BF} + E_{CHP})$$

$$E_{plant8} = \sum (E_{BF})$$

$$E_{plant9} = \sum (E_{BF} + E_{OW})$$

With

E_{plant}: overall emission factor of an AD plant

E_{BF}: emission factor of biofilter

E_{OW}: emissions factor of open windrows

E_{LTS}: emission factor of liquid treatment system

E_{CHP}: emission factor of CHP

2.7 Results and Discussion

2.7.1 The effects of biofilters on the removal of TOC, NMVOC, CH_4, NH_3 and N_2O in waste air at AD plants

The concentrations at the inlet and outlet of the biofilters in form of TOC, NMVOC, CH_4, NH_3 and N_2O are shown in the Figure 2.16. The concentrations of NMVOC were calculated by subtracting TOC and CH_4-C. Inlet gas contained an average of 151mg/m^3 TOC in the range of 26-333 mg/m^3, an average of 151mg/m^3 NH_3 in the range of 2.4-1,704mg/m^3 and in average of 2.3mg/m^3 N_2O in the range of 0.8-6mg/m^3 and in average of 168mg/m^3 CH_4 in the range of 13-380 mg/m^3 respectively. The results are in line with those found by Amlinger *et al.,* (2008) and Clemens and Cuhls (2003).

Biofilters reduced TOC, NMVOC and NH_3 but were a source for N_2O. Additionally, biofilters reduced CH_4 only slightly. Mean relative reductions were 30, 6, 50 and 51% for TOC, CH_4, NMVOC and NH_3 respectively, whereas N_2O concentrations were 26% higher in treated air. The increase of N_2O may be explained by the fact that NH_3 is converted to N_2O by nitrification due to continuous aerobic conditions in the biofilters (Melse and Van der Werf, 2005). According to previous studies, around one third NH_3 that enters biofilters can be transformed and released as N_2O (Trimborn, 2003).

Similarly, Clemens and Cuhls (2003) reported that 26% of NH_3 was transformed into N_2O in biofilters.

The reduction of TOC, CH_4 and NH_3 were lower than in previous studies (Table 2.2). Ojstrsek and Fakin (2009) found that TOC removal efficiency of biofilters varied from 31 to 75%. Similarly, Soyez (2002) found that 50% of TOC was removed in the biofilter. With regard to other literature, CH_4 was reduced by 15% (Amlinger *et al.*, 2008). NH_3 was removed in biofilters by more than 90% (Soyez, 2002; Chen *et al.*, 2005; Hort *et al.,* 2009; Ryu *et al.*, 2011), whereas Akdeniz (2012) found that the reduction efficiency of NH_3 were from 53 to 64% at full-scale biofilters.

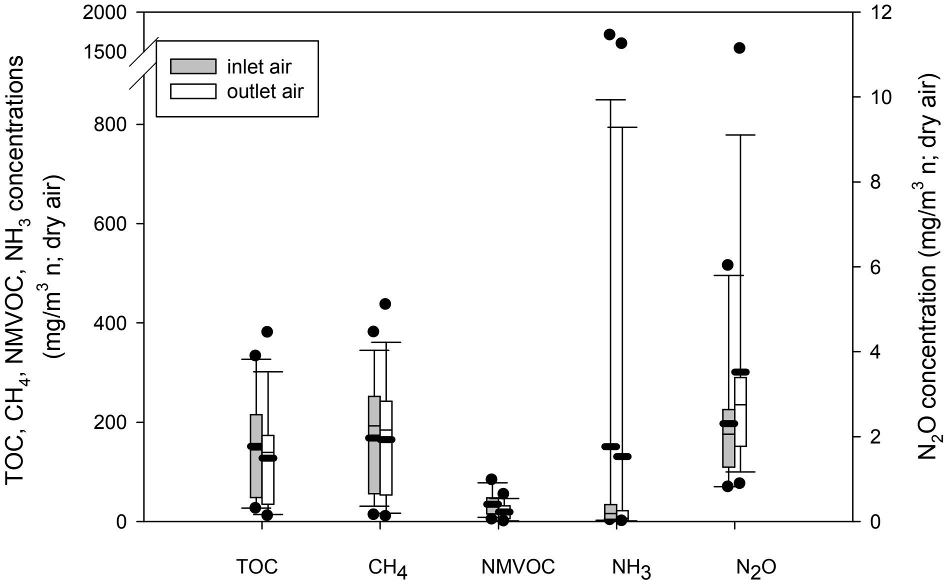

Figure 2.16: Box plots (n=15) show mean gas concentration values (mg/m^3) of treated and untreated air. Box indicates 25 and 75% percentile; - minimum and maximum of total organic carbon (TOC), methane (CH_4), non-methane volatile organic carbon (NMVOC), ammonia (NH_3) and nitrous oxide (N_2O)

Table 2.2: Comparison of removal efficiency of biofilters: this study and data from literature

Authors	**Removal efficiency of biofilter (%)**				
	TOC	**CH_4^***	**NMVOC**	**NH_3^***	**N_2O^***
This study	11 to 70	1 to 25	11 to 100	6.4 to 94	0.1 to -500
(Soyez, 2002)	50	-	83	90	-
(Akdeniz, 2012)	-	-	-	53 to 64	-29.2

(Amlinger *et al.*, 2008)	-	15	-	-	-
(Ryu *et al.*, 2011)	95 to 99	-	-	92	-
(Lopez *et al.*, 2011)	90	-	-	-	-
(Hort *et al.*, 2009)	-	-	-	94	-
(Chen *et al.*, 2005)	-	-	-	97 to 99	-
(Ojstrsek and Fakin, 2009)	31 to 75	-	-	-	-
(Schlegelmilch *et al.*, 2005)	-	-	-	100	-
(Clemens and Cuhls, 2003)	-	-	-	13 to 89	-
(Trimborn, *et al.,* 2003)	32 to 78	-7 to 26	75 to 100	-35 to 91	-9 to -116

- no data

* Negative values mean a production in the biofilter

2.7.2 Acid scrubber for NH_3 removal

The inlet concentrations of TOC, CH_4, NH_3 and N_2O were 215.4, 227.2, 23.5 and 2.4mg m^{-3} respectively (Figure 2.17). Removal efficiency of the acid scrubber was 48% for NH_3. Mean concentrations of TOC, CH_4 and N_2O were not significant different before and after the acid scrubber.

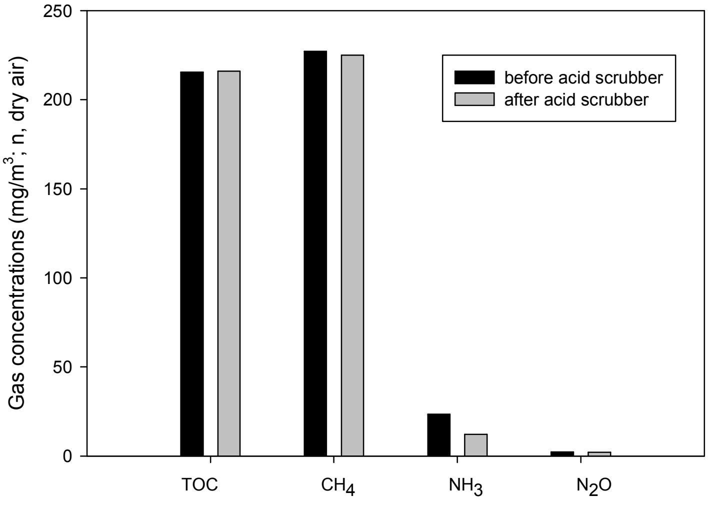

Figure 2.17: Gas concentrations before and after acid scrubber at the AD plant 6

2.7.3 Emission factors of CH_4, N_2O and NH_3 from open emission sources in AD plants

The emission factors of CH_4 varied from 16 to 819g (Mg biowaste)$^{-1}$ for liquid treatment system (LTS), from 50 to 1,500g (Mg biowaste)$^{-1}$ for CHP, and from 0.4 to 15.4kg (Mg biowaste)$^{-1}$ for open windrows (Figure 2.18). Liquid digestate still contains potential to form CH_4 (Bockreis and Steinberg, 2005). Thus, CH_4 emissions still occur in treatment systems of liquid digestate. Biogas produced at the AD plants is burned in CHPs to produce electricity and heat. Since the combustion process is not 100%, some CH_4 escapes unburned into the atmosphere. By this way, CHP contributes to CH_4 emissions.

The emission factors of N_2O were in the range of 1.22 to 37.57g (Mg biowaste)$^{-1}$ for LTS, 0.1 to 2.7g (Mg biowaste)$^{-1}$ for CHP, and 56 to 201g (Mg biowaste)$^{-1}$ for open windrows. The emissions of N_2O at the CHP were insignificant, while the N_2O emissions from LTS and open windrows need to be considered. The results are in line with the findings of (Clemens and Cuhls, 2003) and (Møller *et al.,* 2009).

The emission factors of NH_3 were in the range of 0.1 to 0.16g (Mg biowaste)$^{-1}$ for LTS, 0.03 to 1.16g (Mg biowaste)$^{-1}$ for CHP and 65 to 3,327g (Mg biowaste)$^{-1}$ for open windrows. The emissions of NH_3 from the LTS and CHP were low, while open windrows had high emissions of NH_3.

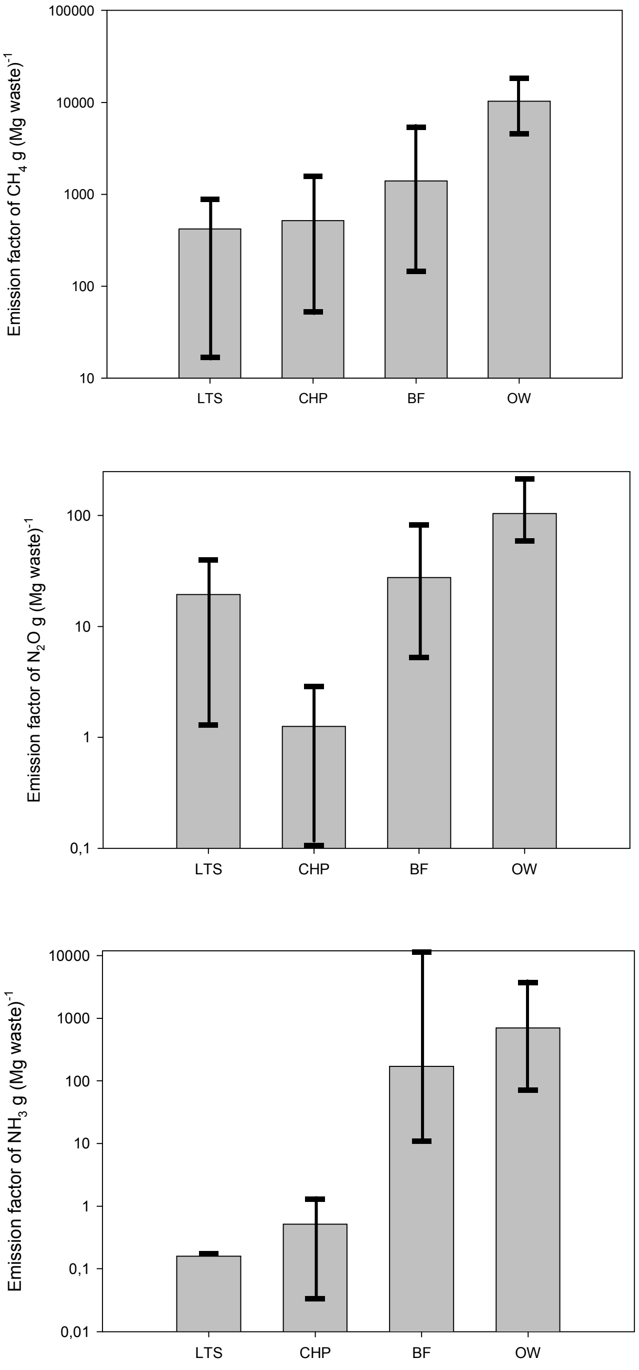

Figure 2.18: Emission factors of CH_4, N_2O and NH_3 from emission sources in AD plants. Error bars show min and max values. Liquid treatment system (LTS) (n=2), Combined heat and power units (CHP) (n=6), biofilter (n=15), open windrow (n=3)

2.7.4 Emissions factors of CH_4, N_2O and NH_3 at the AD plants

The CH_4 emission factors from AD plants were from 1,246 to 16,567g (Mg biowaste)$^{-1}$ (Figure 2.19). The median CH_4 emission factor was 3,828g (Mg biowaste)$^{-1}$. The plants 1, 2 and 9 with open composting windrows showed highest CH_4 emissions. The CH_4 emission factors from composting windrows were 16,567, 8,122 and 10,788g (Mg biowaste)$^{-1}$ which contributed relatively to 95%, 73% and 96% and of the total CH_4 emissions at the plants 1, 2 and 9 respectively.

Emission factors of CH_4 from CHPs were measured only in the plants 3, 4 and 7. CH_4 emission factors from CHPs varied from 52 to 2,040g (Mg biowaste)$^{-1}$. The results were higher than a previous study: Møller *et al.,* (2009) reported that the emission factors of CH_4 from CHP ranged from 16 to 819g (Mg biowaste)$^{-1}$.

The emission factors of CH_4 from biofilters varied from 236 to 5,237g (Mg biowaste)$^{-1}$. The results are in line with the findings of Clemens and Cuhls (2003) and Møller *et al.,* (2009) but comparatively higher than the results of Soyez (2002), who found that the emission factors of CH_4 were about 100g (Mg waste)$^{-1}$.

The emissions from open composting windrows (plants 1, 2 and 9) were higher than the composting process indoors (biofilters at plants 4, 5, 6, 7 and 8) most likely because the composting process indoors is actively aerated. Adequate air supply keeps of the windrow aerobic and suppresses the CH_4 formation. The anaerobic conditions in open composting windrows are most likely because of the insufficient O_2 supply.

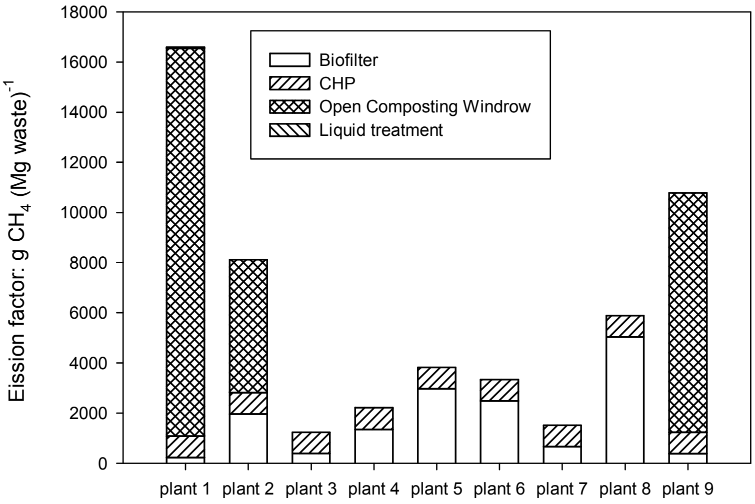

Figure 2.19: Median emission factors of CH_4 g (Mg biowaste)$^{-1}$ from different AD plants. CHP emission data are mean values derived fromplants 3, 4 and 7

The emission factors of N_2O were in a range of 9-172g (Mg biowaste)$^{-1}$ (Figure 2.20). The median N_2O emission factor was 64g (Mg biowaste)$^{-1}$. N_2O emissions from open composting windrows contributed significantly to the total N_2O emissions of the AD plants. The contributions of open composting windrows to the total N_2O emissions were 76%, 80% and 94% at the plants 1, 2 and 9 respectively. The N_2O emissions from CHPs were from 0.5 to 5g N_2O (Mg biowaste)$^{-1}$ and contributed only 2-7% to the total N_2O emissions. The N_2O emission factors from biofilters ranged from 6.7 to 78g (Mg biowaste)$^{-1}$. The N_2O emissions from LTS contributed in the range of 2-27% of the total N_2O emissions.

As for CH_4 emissions, N_2O emissions from the open windrows (plants 1, 2 and 9) were higher than the composting process indoors (biofilters at plants 4, 5, 6, 7 and 8). The relative high N_2O production from open windrows may be related to long treatment duration. The N_2O emissions increase with the compost age (Beck-Friss *et al.,* 2000). Other studies found that N_2O was emitted during the last stage of composting, when temperatures were low (He *et al.,* 2001 and Hellmann, 1997).

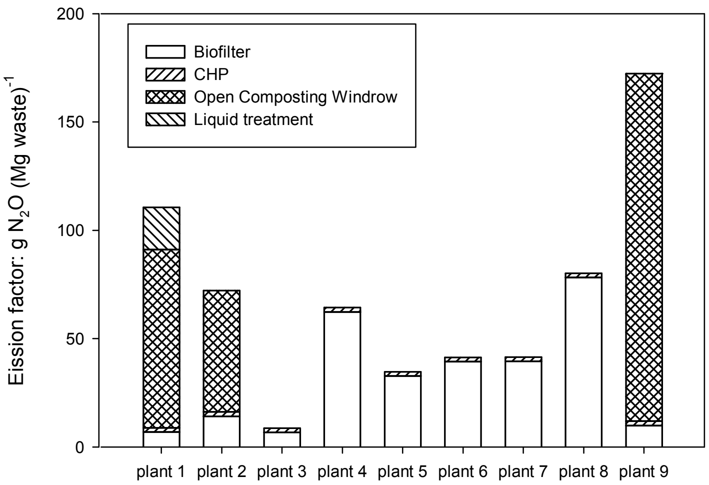

Figure 2.20: Median emission factor of N_2O g (Mg biowaste)$^{-1}$ from different AD plants. CHP emission data are mean values derived fromplants 3, 4 and 7

The emission factors of NH_3 were in the range of 41-6,031g (Mg biowaste)$^{-1}$ (Figure 2.21). The median NH_3 emission factor was 101g NH_3 (Mg biowaste)$^{-1}$. Open composting windrows contributed 91%, 86% and 99% to the total NH_3 emissions at the plants 1, 2 and 9 respectively. High NH_3 emissions at the plant 3 were due to conversion of NH_4^+ in digestate to NH_3 at a high pH and a high temperature in the belt dryer.

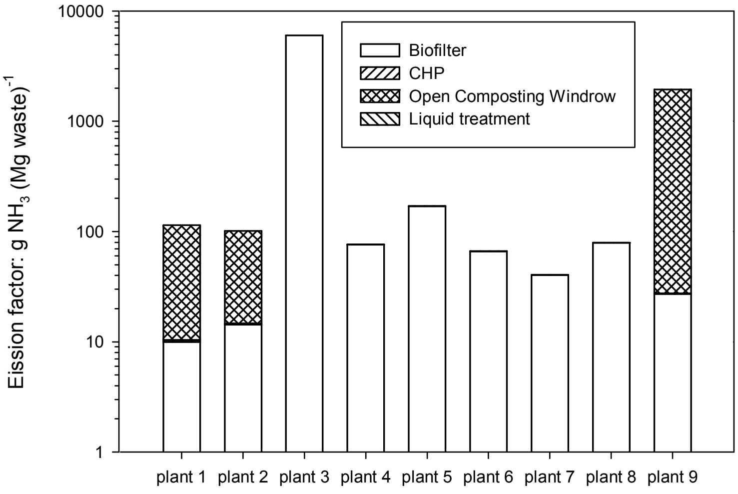

Figure 2.21: Median emission factors of NH_3 g (Mg biowaste)$^{-1}$ from different AD plants. CHP emission data are mean values derived fromplants 3, 4 and 7

2.7.5 The contribution of CH_4, N_2O and NH_3 from AD plants to global warming potential

The emission factors were transferred into CO_2 equivalents according to IPCC (2007). The overall CO_2 emissions were in a range from 50 to 457kg (Mg biowaste)$^{-1}$. The CH_4 emissions from AD plants were more important than the emissions from N_2O and NH_3 (Figure 2.22A). The emissions of CH_4 accounted from 36-92% while the emission of N_2O and NH_3 contributed from 6.9-30% and from 0.08-58% respectively to the overall CO_2 emissions. The median CO_2 equivalent emission was 107kg CO_2 (Mg biowaste)$^{-1}$. The results were in line with a previous study. Møller *et al.,* (2009) reported that an AD plant contributed up to 111kg CO_2 equivalent (Mg waste)$^{-1}$. The AD plants with open composting windrows (1, 2 and 9) showed higher CO_2 equivalent emissions than the AD plant without open composting windrows.

Figure 2.22B shows the net total of CO_2 equivalent from different emission sources at AD plants. The open composting system resulted in high GHG emissions accounting from 73 to 96% to the total emissions at plants 1, 2 and 9. CHP contributed from 5 to 50% to the total emissions at plants 3, 4 and 7. The liquid treatment system resulted in insignificant (3.7%) to the total CO_2 equivalent emissions at plant 1.

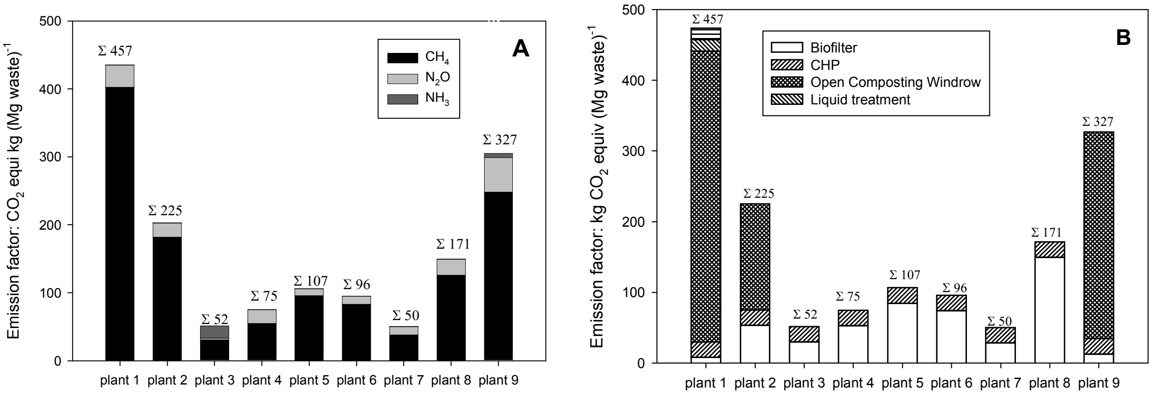

Figure 2.22: Left: The contribution of CH_4, N_2O and NH_3 in form of CO_2 equivalent emissions at AD plants. Right: The contribution of different emission sources at AD plants. CHP emission data are mean values derived fromplants 3, 4 and 7

2.7.6 Some specific findings in AD plants

In plant 3, the belt dryer contributed 99.7% to the overall NH_3 emissions, 50% to the total CH_4 emissions and 79% to the overall N_2O emissions (Figure 2.23). Pre-treatment activities contributed only 0.15%, 38% and 16% to the total NH_3, CH_4 and N_2O emissions respectively. The receiving process contributed 0.15%, 12% and 5% to the overall NH_3, CH_4 and N_2O emissions. The emissions of GHGs from the belt dryer were significant higher than from the receiving and pre-treatment processes. NH_4^+ in digestate was transferred to NH_3 under high temperature (1000°C) in the belt dryer.

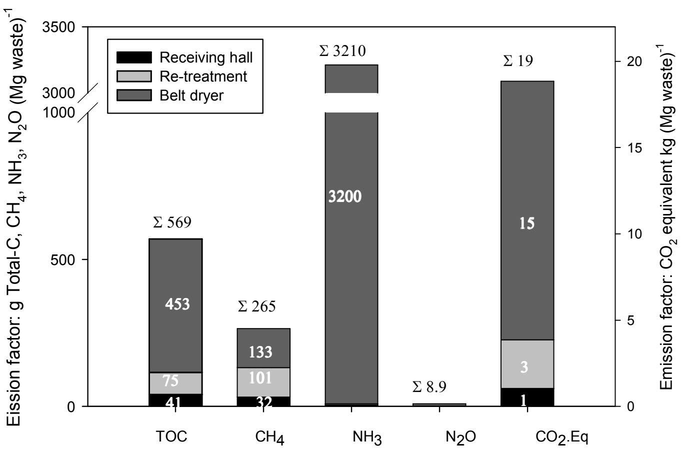

Figure 2.23: Contribution of encapsulated emission sources to TOC, CH_4, NH_3, N_2O (left axis) and CO_2 equivalent (right axis) in the plant 3

In plant 6, a storage tank of liquid digestate contributed more than 90% to the total emissions of TOC and CH_4. Additionally it contributed 80% to the overall CO_2 emissions (Figure 2.24). The high emissions of TOC and CH_4 from this small storage tank could be explained by the fact that the small storage tank received liquid digestate from the separator directly. Liquid digestate still has a high CH_4 potential (Kaparaju and Rintala, 2007) and the material was still warm. The anaerobic process continued in the liquid digestate and led to high CH_4 emissions.

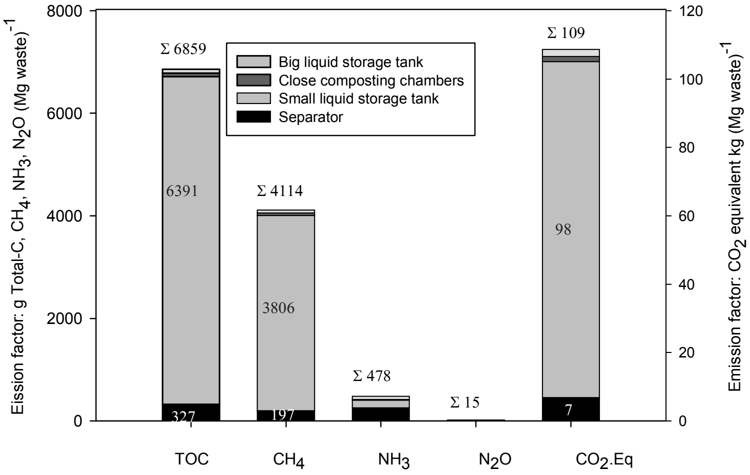

Figure 2.24: Contributions of encapsulated emission sources to TOC, CH_4, NH_3, and N_2O (left axis) and CO_2 equivalent (right axis) in the plant 6

2.8 Conclusions

Anaerobic digestion plants are a source of GHG emissions. Emission sources are biofilter, open windrows, CHP, liquid digestate treatment system and open composting windrows of residues from aerobic digestion. Especially, open windrows have adverse impacts on environment. Inside the AD plants, the emissions at the receiving and pre-treatment processes play less important roles, whereas the separation of digestate into a solid and a liquid phase results in high GHG emissions.

Based on the results, the emissions factors were 3,828g (Mg waste)$^{-1}$ for CH_4 (96 kg CO_2 equivalent) and 64g (Mg waste)$^{-1}$ for N_2O (19 kg CO_2 equivalent). In Germany, ca. 10.5 million tonnes biowaste are produced per year. If all biowaste would be treated by AD combined with post-treatment of solid digestate, they would result in a contribution of 0.31% for N_2O and 1.83% for CH_4 to the overall national GHG emissions (base: 2012).

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3. SUMMARY OF GHG EMISSIONS FROM COMPOSTING AND AD PLANTS

3.1 Summary of GHG emissions from composting and AD plants

3.1.1 CH_4 emissions

Irrespective of the type of anaerobic digestion procedures (e.g. wet or dry AD, one or two stages), AD showed the lowest CH_4 emissions 950g (Mg biowaste)$^{-1}$ compared with composting 4,060g (Mg biowaste)$^{-1}$, AD combined with tunnel composting 3,000g (Mg biowaste)$^{-1}$and AD combined with open composting windrow 11,000g (Mg biowaste)$^{-1}$ (Figure 3.1). Post-treatment of solid digestate is the main source of CH_4 emissions in an AD plant. In particular, open windrows contributed significantly to CH_4 emissions.

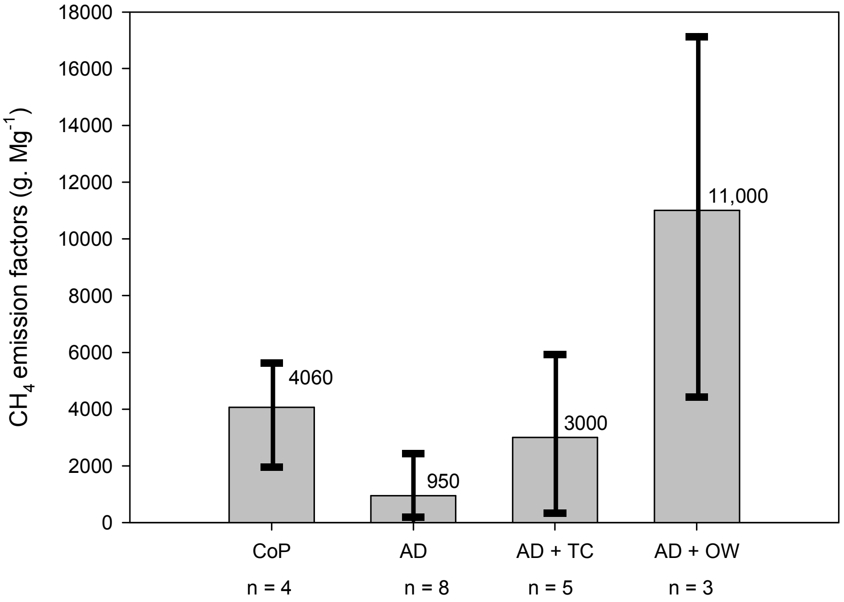


Figure 3.1: Mean CH_4 emission factors (g Mg^{-1}) from different treatments: composting (CoP), anaerobic digestion (AD), anaerobic digestion with intensive tunnel composting (AD + TC) and anaerobic digestion with open windrow composting (AD + OW). Bars show minimum and maximum values, n: number of plants

3.1.2 N_2O emissions

AD had lower N_2O emissions than composting, but AD with post-treatment of solid digestate resulted in higher N_2O emissions than composting. The emission factors of

N_2O were 55g (Mg biowaste)$^{-1}$ for composting, 13g (Mg biowaste)$^{-1}$ for AD, 72g (Mg biowaste)$^{-1}$ for AD with tunnel composting and 120g (Mg biowaste)$^{-1}$ for AD with open windrows (Figure 3.2).

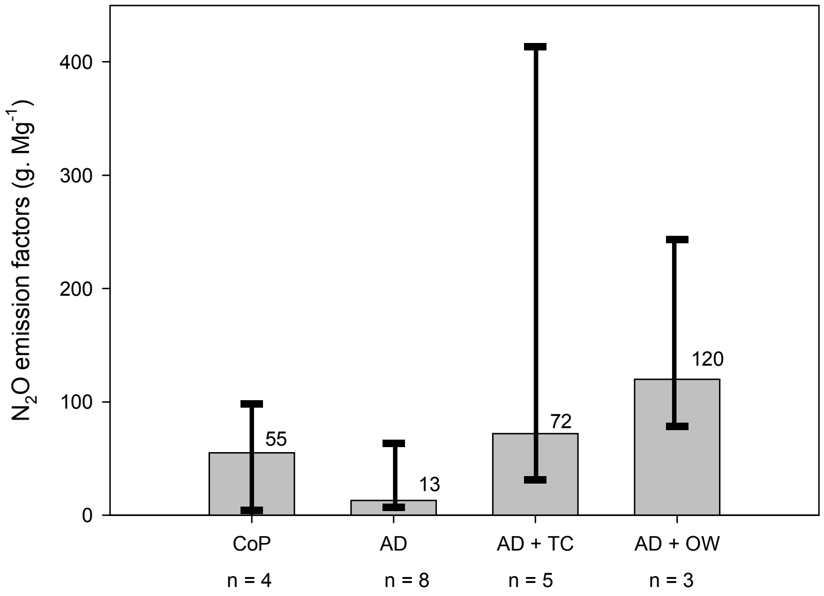


Figure 3.2: Mean N_2O emission factors (g Mg^{-1}) from different treatments: composting (CoP), anaerobic digestion (AD), anaerobic digestion with intensive tunnel composting (AD + TC) and anaerobic digestion with open windrow composting (AD + OW). Bars show minimum and maximum values, n: number of plants

3.1.3 NH_3 emissions

AD with open windrows resulted in highest NH_3 emissions. The emissions factors of NH_3 were 157g (Mg biowaste)$^{-1}$ for composting, 24g (Mg biowaste)$^{-1}$ for AD, 130g (Mg biowaste)$^{-1}$ for AD with tunnel composting and 720g (Mg biowaste)$^{-1}$ for AD with open windrows (Figure 3.3).

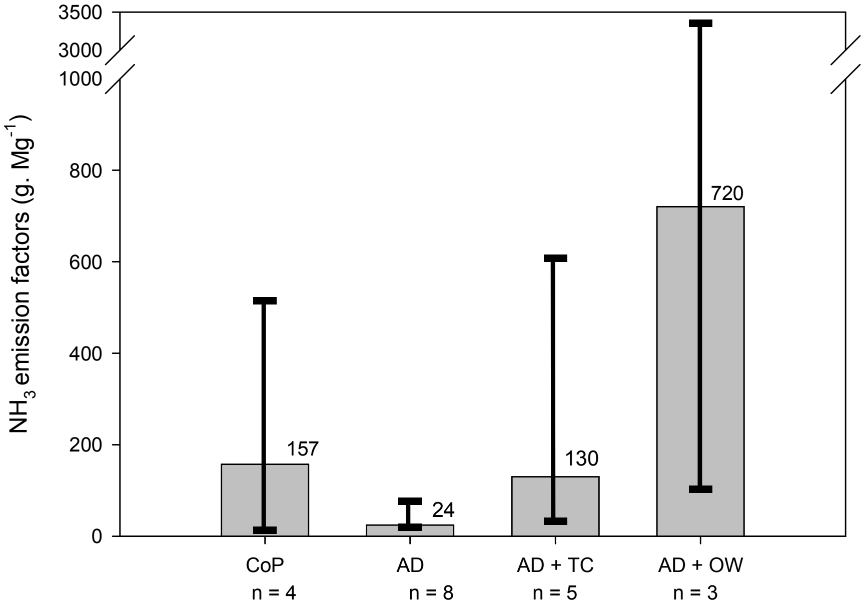


Figure 3.3: Mean NH_3 emission factors (g Mg^{-1}) from different treatments: composting (CoP), anaerobic digestion (AD), anaerobic digestion with intensive tunnel composting (AD + TC) and anaerobic digestion with open windrow composting (AD + OW). Bars show minimum and maximum values, n: number of plants

3.2 The purification efficiency of biofilters

Biofilters showed only a small influence on CH_4 emission reduction (6%) (Figure 3.4), whereas they were a source of N_2O emissions (from 0.1 to 500% N_2O was generated in the biofilters). Biofilters removed significantly TOC (30%), NMVOC (50%) and NH_3 (51%).

The purification efficiency of the analysed biofilters differed. According to IPCC (2007), the emission factors were transferred into CO_2 equivalents. Five of fifteen investigated biofilters resulted in higher CO_2 equivalents Mg^{-1} in the exhaust gas as compared to the untreated gas. These five biofilters increased the overall CO_2 emissions from 8 to 16% due to additional N_2O production. The other ten biofilters showed a positive CO_2 equivalents balance and reduced GHG by 0.02 to 21%.

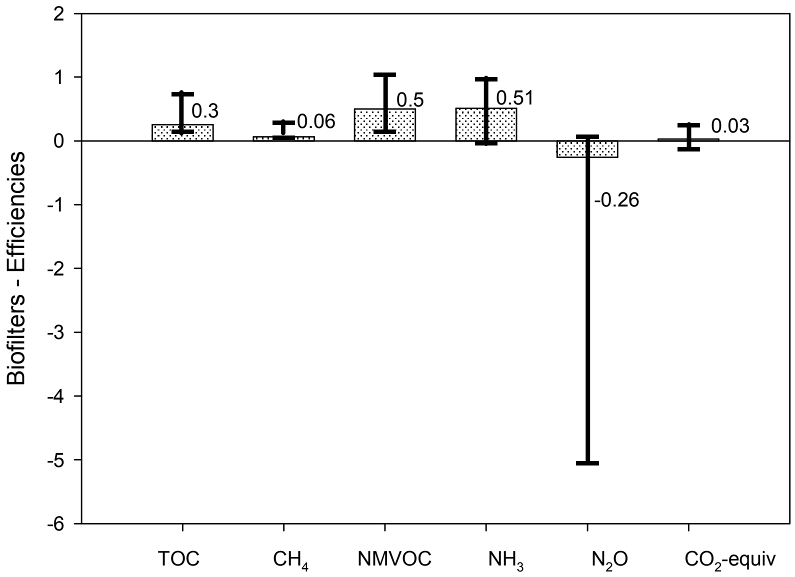


Figure 3.4: Mean biofilter's efficiencies (n=15) in AD plants. Bars show indicates minimum and maximum values of total organic carbon (TOC), methane (CH_4), non-methane volatile organic carbon (NMVOC), ammonia (NH_3) and nitrous oxide (N_2O)

3.3 Overall CO_2 emissions

In comparison to composting, AD had lower GHG emissions. However, AD with post-treatment of digestate results in higher GHG emissions than composting. The overall CO_2 equivalents were 118kg Mg^{-1} for composting, 76kg Mg^{-1} for AD, 97kg Mg^{-1} for AD with tunnel composting and 506kg Mg^{-1} for AD with open windrows (Figure 3.5 and Table 3.1). The post-treatment of solid digestate is therefore recognized as an important source of GHG emissions.

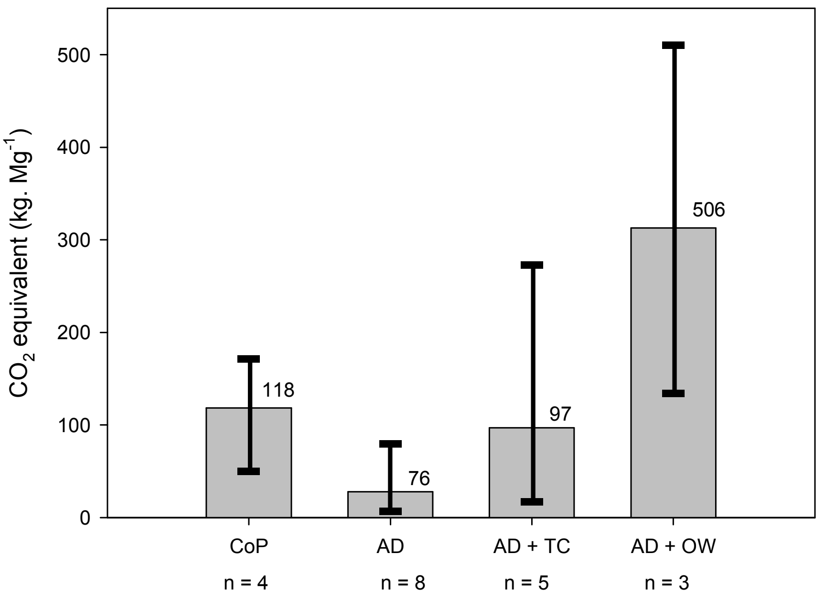


Figure 3.5: Mean CO_2 equivalent (kg Mg^{-1}) from different treatments: composting (CoP), anaerobic digestion (AD), anaerobic digestion with intensive tunnel composting (AD + TC) and anaerobic digestion with open windrow composting (AD + OW). Bars show minimum and maximum values, n: number of plants

Table 3.1: Emissions from different treatment technologies of biowaste

Emissions	Composting	AD	AD + CT	AD + OW
CH_4 (g Mg^{-1})	4,060	950	3,000	11,000
N_2O (g Mg^{-1})	55	13	72	120
NH_3 (g Mg^{-1})	157	24	130	720
CO_2 equivalent (kg Mg^{-1})	118	76	97	506

4. REDUCTION AND AVOIDANCE GHG EMISSIONS AT COMPOSTING AND AD PLANTS

4.1 Composting plants

To reduce GHG emissions from composting windrows, it is recommended to provide enough oxygen during the whole composting process. The composting windrows should be small and contain a high proportion of bulking agents. The windrows need to be turned frequently or aerated actively. Even if high O_2 concentration would result in higher NH_3 and N_2O emissions, the overall GHG emissions from composting is still low due to low CH_4 emissions. Furthermore, a short composting duration would decrease GHG emissions additionally because N_2O tended to be emitted at the end of the composting process.

4.2 AD plants

All three types of AD digestion including wet, dry and solid digestions have their own specific advantageous and disadvantageous (Table 4.1). For example, it is important to take into consideration fugitive emissions, when a batch fermenter is opened to unload digestate and reload new material. Solid batch digestion can be realized with low investments and low technology requirements, whereas wet digestion requires a larger investment and the process is more complex. But, continuous digestions are more ecologically advantageous. In summary, it is affirmed that wet continuous digestion is appropriate for AD of biowaste with regard to GHG emissions.

Table 4.1: Advantageous and disadvantageous of anaerobic digestion alternatives

	Wet continuous digestion	**Dry continuous digestion**	**Solid batch digestion**
Complex	+++	++	+
Investment cost	+++	++	+
Biogas production	+++	+++	+
Min – Max of overall CO_2 emissions (g Mg^{-1})*	8-53	30-85	13-149

+ low; ++ medium; +++high

*Overall emissions from biofilters (see Figure 2.22). Plant 1 and 2: wet continuous digestion; plant 3, 4, 5 and 6: dry continuous digestion; plant 7, 8 and 9: solid batch digestion.

4.3 GHG emission mitigation options for post-treatment of digestate

The post-treatment of digestate results in high GHG emissions at the investigated AD plants. AD without post-treatment of digestate is favoured as it shows the lowest GHG emissions. It can therefore be recommended that the digestate is transported directly to nearby farms and used as fertilizer. It needs ca. 30 tons digestate for one ha grass or arable land (Wulf, *et al.,* 2002). Therefore, the land is covered by a layer of only 3 mm digestate. After application the GHG emissions are reduced significantly due to the reduction of CH_4 emissions.

Potentially there are great GHG emissions benefits if biowaste is treated by wet anaerobic digestion. Wet digestion results in liquid digestate only, which should be directly applied without being composted. If anaerobic digestion was performed at mesophilic condition, sanitation of digestate may be required before it is field applied.

If a separation of liquid and solid digestate is necessary, the post treatment of the solid material should show only small GHG emissions. For this purpose it is suggested:

- The digestate should be mixed with bulking material.
- The mixture (ca. 50cm high) is aerated in small triangular windrows (maximum height: 0.5m) for about 2 days. It is assumed that this treatment will create an aerobic environment in the windrow and stops methane formation.
- The material is piled up to triangular windrows with a height of ca 1.5m. Then the windrows are aerated for minimum two weeks.

5. GENERAL CONCLUSIONS

In conclusion, both composting and AD entail GHG emissions. CH_4 and N_2O are generated significantly and are the major contributors to the emissions. AD treatment alone shows the lowest GHG emissions in comparison to composting, AD with tunnel composting and AD with open windrows composting. Active aerated windrows (tunnel composting) showed lower emissions than open windrows.

GHG emissions during post treatment of AD material can be limited by intensive aeration.

In general, it was shown that environmental loads from composting of biowaste were lower than AD combined with post-treatments of digestate. However, AD generates biogas, which is used as a renewable energy source. In connection with the energy recovery and rather low GHG emissions, it is reasonable to believe that wet AD has a high potential to treat biowaste sustainably.

GHG emissions calculations in the study were linked directly to aerobic and anaerobic activities at the composting and AD plants. The data on fuel or electricity consumptions and emissions from the application on land of digestate are missing. Additionally, GHG emission savings by using biogas as a renewable energy source were not calculated. Thus, the overall emissions of the composting and anaerobic digestion plants should be studied in further researches.

6. APPENDIX

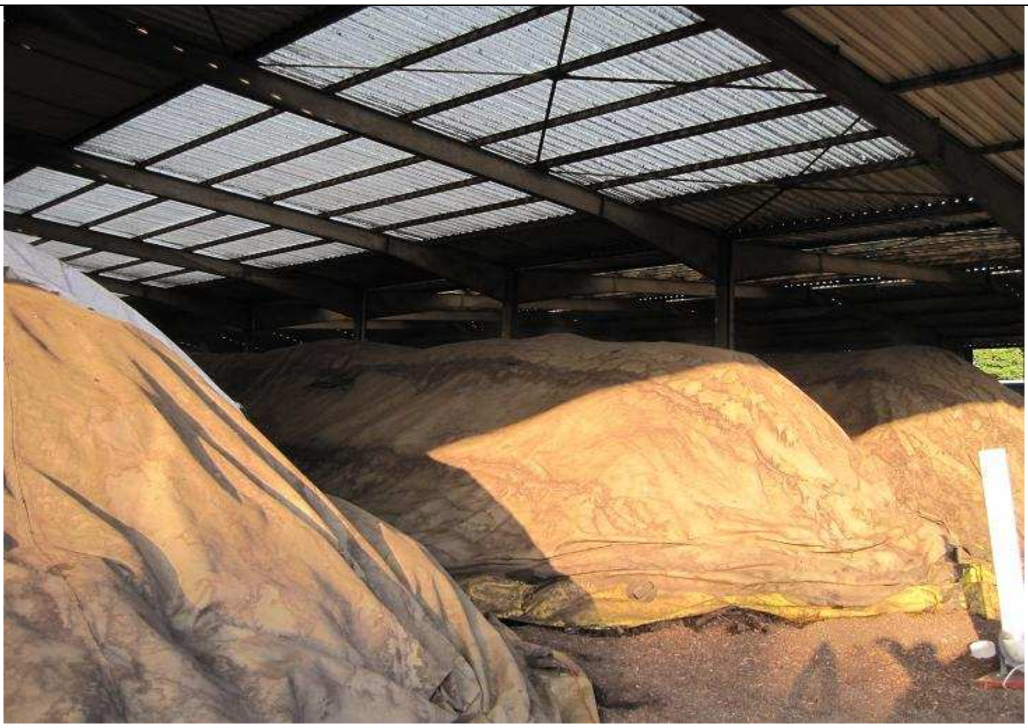
Appendix 1: Equiments in measurement

Appendix 2: NH_3 sampling at biofilter (unfiltered air)

Appendix 3: Triangle windrows

Appendix 4: Trapezoidal windrow

Appendix 5: Tabular windrow



Appendix 6: Active aerated windrows with cover sheets

Appendix 7: Continuous fermenter

Appendix 8: Batch fermenter

Appendix 9: Processing parameters of different AD plants

Anaerobic digestion plants									
DM content	**Wet digestion DM<15%**		**Dry digestion DM. ca: 15-30%**				**Solid digestion DM>30%**		
Fermenter feeding	**Continuous feeding**		**Continuous feeding**				**Batch feeding**		
	PLANT 1	**PLANT 2**	**PLANT 3**	**PLANT 4**	**PLANT 5**	**PLANT 6**	**PLANT 7**	**PLANT 8**	**PLANT 9**
Total capacity (Mg/y):	30500	2100	32000	30000	29500	23000	38000	85000	12000
Digestion (%):	100%	100%	100%	50%	62%	75%	48%	28%	100%
Products	Compost	Compost	Compost	Compost	Compost and effluent	Compost and effluent	Compost	Compost	Compost
Operation	1997	2008	2004	1999	1999 intensive composting 2007 digestion step	2002	1993 Composting 2009 digestion	1996 2007 rebuilt	2010
Processes	AD + Composting	AD + Composting	AD	AD + Composting	AD + Composting	AD + Composting	AD + Composting	AD + Composting	AD + Composting
Fermenter system:	Wet digestion	Wet digestion	Dry digestion	Dry digestion	Dry digestion	Dry digestion	Solid digestion	Solid digestion	Solid digestion
No of Fermenter:	1	1	1	18	1	1	6	8	5
Volume m^3:	-	2700, 4% DM	2400	1300	1600	1500	-	8 x ca 150-190Mg	260
Feeding:	continuous	continuous	continuous	discontinuous	continuous	continuous	discontinuous	discontinuous	discontinuous
Temperature:	Mesophilic (37 °C)	Thermophilic (55°C)	Thermophilic (55°C)	Thermophilic (57°C)	Thermophilic (55°C)	Thermophilic	mesophilic (38°C)	mesophilic (41°C)	mesophilic (41°C)
Retention time:	-	20 days	21 days	14 days	14 days	21 days	28 days	21 days	21-23 days
Separation	Centrifuge	Decanter	Drying band	Aeration	Aeration: 2 Tunnels, 2 days	Press	Aeration: not in operation	-	Aeration
Composting1	Tabular windrow without aeration	Tabular windrow without aeration	-	Intensive Tunnel, close, 10-12 days	Intensive tunnel, Closed 10 days, 8 tunnels	Intensive tunnel, closed, 21 days,	Intensive tunnel, closed, 56 days,	Intensive tunnel, closed, 2 x9 days, active aeration	Intensive tunnel, windrows, under semi permeable membrane, pressure
Turning:	1 time	1 time		active aeration without turning	pressure aeration without turning	active aeration without turning	Turning 1/week	Turning 1 time	aeration without turning
Composting 2								Windrows 2 x8 days	Triangle windrows, open 60-70 days without aeration
Turning:								Turning 1 time	turning:1/week
Gas treatment									
Volume(m^3):	16000-19000	15500	25000-28000	23000	14000 and 20000	22000-22000	100000	110000	2700-2700
Acid absortion:	-	-	Not operation	-		acid absortion	-	acid absorbsion	-
Measurement time	08.2010 12.2010	08.2010 01.2011	07.2010 10.2010	07.2010 -	11.2010 -	05.2010 02.2011	06.2010 12.2010	05.2010 -	09.2010 03.2011

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