Greenhouse Gas Emissions from Food and Garden Waste Composting

Effects of Management and Process Conditions

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Abstract

Composting is a robust waste treatment technology. Use of finished compost enables plant nutrient recycling, carbon sequestration, soil structure improvement and mineral fertiliser replacement. However, composting also emits greenhouse gases (GHG) such as methane (CH_4) and nitrous oxide (N_2O) with high global warming potential (GWP).

This thesis analysed emissions of CH_4 and N_2O during composting as influenced by management and process conditions and examined how these emissions could be reduced. The GHG emissions from home-scale, large-scale and reactor composting were determined. At small scale, 18 home compost units were analysed over one year. At large scale, the effects of aeration strategies on emissions were studied in 10-20 m long windrow composts. Finally, reactor composting was studied in two sets of experiments under a controlled laboratory environment to investigate the influence of specific process conditions on CH_4 and N_2O emissions.

Methane emissions increased almost 1000-fold when moisture content in the compost substrate increased from about 44% to 66% in the reactor. Moreover, CH_4 emissions increased 100-fold as a result of poor aeration. In home composting CH_4 emissions were low, but increased with temperature, mixing frequency and moisture. In windrow composting, high CH_4 emissions were associated with thermophilic temperatures and large moisture gradients. Moisture content from about 44% to 59% significantly affected N_2O emissions in the reactor studies, with N_2O emissions increasing at lower moisture. The presence of nitrate in the initial substrate resulted in an early N_2O emissions peak in reactor composting. Extended composting period during some reactor runs resulted in higher total GHG emissions due to continued production of both CH_4 and N_2O late in the process, after 50% of initial carbon had been mineralised.

Total direct GHG emissions from home and windrow composting systems, assessed based on their GWP, were similar to or lower than those reported by others, while emissions from most composting reactor runs were lower. To reduce CH_4 emissions, composting at a combination of high moisture and high temperature should be avoided. To reduce N_2O emissions, extremes of moisture content in the compost matrix should be avoided.

Keywords: methane, nitrous oxide, moisture, temperature, aeration

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Dedication

To my family and friends for their love and support.

Посвящается моей семье и друзьям за их любовь и поддержку.

True wisdom comes to each of us
when we realize
how little we understand
about life, ourselves,
and the world around us

Socrates

Contents

List of Publications

This thesis is based on the work contained in the following papers, referred to by Roman numerals in the text:

I Ermolaev, E., Sundberg, C., Pell, M. & Jönsson, H. (2014). Greenhouse gas emissions from home composting in practice. *Bioresource Technology* 151, 174-182.

II Ermolaev, E., Pell, M., Smårs, S., Sundberg, C. & Jönsson, H. (2012). Greenhouse gas emission from covered windrow composting with controlled ventilation. *Waste Management & Research* 30(2), 155-160.

III Ermolaev, E., Jarvis, Å., Sundberg, C., Smårs, S., Pell, M. & Jönsson, H. (2015). Nitrous oxide and methane emissions from food waste composting at different temperatures (manuscript). *Submitted*.

IV Ermolaev, E., Sundberg, C., Pell, M., Smårs, S. & Jönsson, H. (2015). Effects of moisture on emissions of CH_4, N_2O and CO_2 from food and garden waste composting (manuscript).

Papers I and II are reproduced with the permission of the publishers.

The contribution of Evgheni Ermolaev to the papers included in this thesis was as follows:

I Designed the experiment together with the co-authors, carried out the measurements and data analyses together with Master’s students. Wrote the paper with feedback from the co-authors.

II Designed the study together with the co-authors. Performed the field and laboratory measurements with assistance from a Master’s student. Performed the majority of the data analysis for the paper. Wrote the paper with feedback from the co-authors.

III Planned the paper with considerable input from the co-authors. Performed the data analysis. Wrote the paper with feedback from the co-authors.

IV Planned the experiments with input from the co-authors. Designed and changed parts of the reactor system, performed the experiments and measurements. Wrote the paper with feedback from the co-authors.

Abbreviations

FW	Food waste
GC	Gas chromatography
GHG	Greenhouse gas
GWP	Global warming potential
GW	Garden waste
MC	Moisture content
MSW	Municipal solid waste
TS	Total solids
VS	Volatile solids
WW	Wet weight

1 Introduction

1.1 Composting in Sweden and globally

Greenhouse gas (GHG) emissions from the global waste sector are increasing, despite advances made to reduce these in the EU and US (IPCC, 2014). Globally, 67% of municipal solid waste (MSW) is disposed of by open dumping and landfilling (IPCC, 2014), which makes a considerable contribution to global GHG emissions, mainly due to high emissions of methane (CH_4). It has been estimated that only 12% of the 50 Mt of CH_4 produced annually in landfills world-wide is captured, while the remaining fraction is emitted to the atmosphere (Themelis & Ulloa, 2007). Landfilling is now being increasingly replaced by composting, which is considered a better alternative due to its robustness and in its basic form requires simple infrastructure. Moreover, unlike landfilling, composting enables plant nutrient recycling and better control of greenhouse gas (GHG) emissions (USEPA, 2013). Use of finished compost can replace mineral fertilisers in plant production and allows for carbon sequestration. However, in order for composting to remain a beneficial waste treatment alternative, it is important that composting is carried out with minimal GHG emissions, which can be achieved through process optimisation.

Developing economies are currently rapidly increasing their generation of waste, and composting is widely used as one of the treatment options to handle the increasing waste amount. A shift away from landfilling can be expected globally, following the European example (EU 27), where the share of landfilled municipal waste per person and year decreased by 52.5% in the period 1995-2013 (Eurostat, 2014). This is equivalent to the observed decrease from 63.8 to 30.3% in MSW being landfilled. While there is a slow trend towards waste management alternatives with more straightforward possibilities to recover energy, adoption of composting is considered an important step towards reduction of the negative impacts of the waste management sector (IPCC, 2014; Lasaridi, 2009).

Composting is a common method for plant nutrient recycling from green waste, representing over 40% of all waste treated biologically in Sweden in 2013 (Swedish Waste Management, 2014). While the majority of the garden waste in Sweden is composted centrally, the proportion of food waste composted decreased from 34 to 25% during the period 2012-2013 due to a change-over to anaerobic digestion. In the United States, 62% of green waste was composted in 2006, while of all the MSW collected in 2012, nearly 35% was composted (USEPA, 2012; Arsova *et al.*, 2008).

Small-scale home composting is an important waste reduction and sanitisation practice that helps reduce the amount of municipal solid waste (Favoino & Hogg, 2008) and promotes awareness about the amounts of waste generated and sustainable nutrient recycling.

Composting is included in climate-smart sustainable agricultural land management to assist the agricultural sector in adapting to the effects of global climate change (IPCC, 2014). Use of compost improves soil resilience and fertility, without increasing the GHG emissions. Use of compost in climate-smart agriculture allows carbon sequestration, soil organic matter content improvement and fertility maintenance and restoration (McCarthy *et al.*, 2011). It also promotes water conservation due to improved soil structure, as well as reducing direct gaseous emissions from agricultural waste.

1.2 Sources of greenhouse gas emissions in composting

During composting, the organic matter present in the feedstock (material to be composted) is decomposed, forming the finished compost and also gaseous emissions composed mainly of water (H_2O) and carbon dioxide (CO_2), and partly of CH_4 and nitrous oxide (N_2O). In the decomposition process, aerobic microbial mineralisation of organic carbon (C) to CO_2 is the major mechanism for reduction of the waste dry mass. Typically, carbon constitutes 35-50% of the total solids (TS) content of source-separated organic household waste (Sundberg *et al.*, 2011; Eklind *et al.*, 1997) and over 80% of this carbon can be degraded and emitted as CO_2 during composting (Haug, 1993).

The CO_2 emitted from the composting process is considered biogenic, as it originates from organic material recently assimilated from atmospheric CO_2. This is the reason why the majority of reported studies on composting consider the process to be neutral in terms of global warming potential (GWP) (Bernstad & Jansen, 2012). Nevertheless, the CO_2 emitted from composting and other biological waste management studies ought to be reported in order to allow better GHG accounting, even though biogenic CO_2 is excluded when estimating total GWP (Christensen *et al.*, 2009).

1.2.1 Methane emissions

Microbial aerobic decomposition of organic material is the main process driving composting and it requires good aeration. However, parts of the substrate being composted inevitably become anaerobic due to the intense respiration when degrading easily available organic compounds (Beck-Friis *et al.*, 2000; He *et al.*, 2000). Anaerobic zones are created, ranging in size from the central part of smaller compost particles to larger compost aggregates and even portions of the composting pile (Jäckel *et al.*, 2005). The size of the anaerobic zones depends on several factors and process conditions, but is related to greater demand for oxygen (O_2) than can be met by aeration measures such as ventilation and diffusion (Chroni *et al.*, 2009).

Low O_2 conditions stimulate anaerobic degradation pathways such as fermentation and methanogenesis, which ultimately result in production and release of CH_4 and CO_2. Methane is produced by strictly anaerobic methanogenic archaea (Hellmann *et al.*, 1997), but a substantial proportion is aerobically oxidised to CO_2 at the compost surface by methanotrophic bacteria (Jäckel *et al.*, 2005).

Methane is a strong GHG, with a GWP factor of 34 over an assessment period of 100 years. This GWP value means that over the 100-year period, CH_4 gives rise to estimated average potential radiative forcing that is 34-fold that of CO_2, including climate-carbon feedback, which provides a better estimate of the relative impacts of non-CO_2 GHG (IPCC, 2013). Radiative forcing describes the ability of the earth-atmosphere to absorb solar radiation. For composting, different ranges of CH_4 production rates have been reported in the literature, starting from very low, near the detection limit of the equipment, *e.g.* in industrial composting (Martínez-Blanco *et al.*, 2010), to an extreme of 12% of initial carbon as reported for static pile composting (Szanto *et al.*, 2007).

1.2.2 Nitrous oxide emissions

During composting, N_2O is formed in the process of nitrification and denitrification and may also be released from the compost to the ambient air. Ammonium (NH_4^+), produced through mineralisation of organic nitrogen (N) in the feedstock, can be oxidised to nitrate (NO_3^-) in two steps of nitrification. If O_2 becomes limited the NO_3^- can then be reduced in several steps to N_2 via denitrification.

Nitrous oxide is a concern from a climate change point of view as it has a very high GWP, 298 over a 10-year period, and an average lifetime of over 120 years in the atmosphere (IPCC, 2013). During composting, the N_2O emissions are commonly in the range 0.5-2% of initial nitrogen, but can be as high as 10% (Brown *et al.*, 2008; Szanto *et al.*, 2007; Hellebrand & Kalk, 2000; Hellebrand, 1998). Various parameters, including moisture, aeration, temperature and pH, can stimulate N_2O production from nitrification and also from denitrification by shifting the balance towards N_2O from nitrogen gas (N_2) in the final product (Hu *et al.*, 2012; Kim *et al.*, 2010; Liu *et al.*, 2010; Wrage *et al.*, 2001).

The major pathway for nitrogen losses, amounting to 50% of initial nitrogen, is likely to be through emissions of ammonia (NH_3) and N_2 (Eklind *et al.*, 2007; Eklind & Kirchmann, 2000b). Emissions of NH_3 depend on the composition of the compost feedstock and process conditions. If the substrate is rich in protein (organic N) and the temperature, pH and aeration rate are high, then high NH_3 emissions can be expected (Boldrin *et al.*, 2011; Eklind & Kirchmann, 2000b). Emitted NH_3 contributes to eutrophication and acidification and, indirectly, to production of N_2O (Bernstad & Jansen, 2012). It should also be emphasised that all nitrogen losses make the compost product less valuable as a fertiliser, as nitrogen is an essential plant nutrient.

1.3 Aims of the thesis

The overall aim of this thesis was to analyse emissions of the greenhouse gases methane (CH_4) and nitrous oxide (N_2O) from composting of food and garden waste and to provide knowledge on how these emissions can be reduced. Specific aims were to: (1) analyse the effects of scale and management on emissions; (2) investigate and analyse the influence of different process conditions of composting on emissions; and (3) assess the global warming potential of the direct greenhouse gas emissions from composting.

1.4 Structure of the work

This thesis is based on four papers evaluating and analysing emissions of CH_4 and N_2O from composting of food waste with structural amendment of garden waste or other lignocellulosic material as feedstock.

In Papers I and II, emissions from home composting and large-scale composting were examined (Figure 1). In Paper I, various process conditions were measured and analysed in a large number of small-scale home composting units in order to establish which parameters most affected the emissions of CH_4 and N_2O. The study presented in Paper II investigated the emissions dynamics of a large-scale composting system consisting of covered windrows with forced aeration. Overall, Papers I and II examined the integrated effects of process parameters such as aeration, temperature and moisture on emissions of CH_4 and N_2O.

Two laboratory studies (Papers III and IV), allowing more controlled experimental conditions, were used to further investigate the specific influence of composting aeration, temperature and moisture on the emissions of CH_4 and N_2O. The reactor studies in Paper III investigated the effects of temperature and limited aeration and those in Paper IV the effects of moisture on the emissions of CH_4 and N_2O.

The knowledge obtained from the practical composting studies (Papers I and II) contributed to the design and formulation of objectives for the laboratory studies (Papers III and IV) (Figure 1). Remaining questions after the first laboratory study were further investigated in the second study. All four studies contributed to this thesis essay, which combines and interprets the information obtained in a comprehensive analysis of emissions of the greenhouse gases CH_4 and N_2O as influenced by compost management and process conditions.

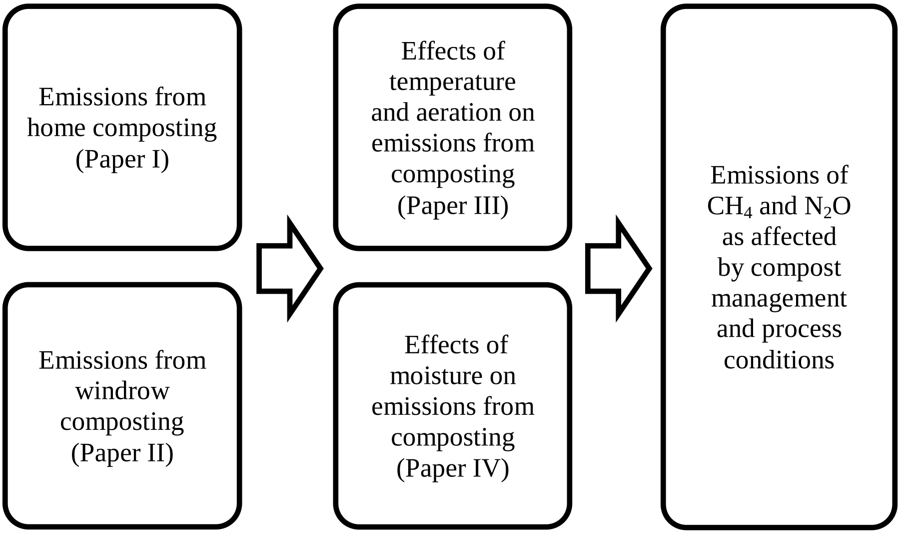

Figure 1. Thesis structure showing the overall development of the thesis based on the contributions from studies performed in Papers I - IV.

2 Emissions of nitrous oxide and methane from composting

2.1 Background

The rates of emission of CH_4 and N_2O during the composting process largely depend on process conditions, such as type and composition of feedstock, moisture and level of aeration. To ensure good aeration in large-scale composts, several techniques are used, including forced ventilation, compost turning and incorporating a structural amendment into the feedstock to provide a porous compost matrix with sufficient free air space. Small-scale composting mostly relies on natural aeration, which makes addition of sufficient structural amendment essential to provide good matrix porosity. Scale and aeration strategy, along with the system type and composted material properties, affect the process performance on multiple levels, including degradation rate, temperature and moisture development (Sundberg & Jönsson, 2005).

The composting process can be performed in continuous, batch or fed-batch operated systems (Haug, 1993). Continuous composting is common in large-scale systems. It is characterised by constant waste addition, which passes through the different phases of the process, and the finished product is produced continuously. Batch process is common in both large and small-scale systems and is started by preparation of a compost mixture, which is then fed into the process without further addition of fresh material until the composting of the whole batch is finished. Household-managed systems, *e.g.* home composts, are usually fed-batch systems, with fresh material added throughout the year and compost bins emptied every one or two years. When the bins are emptied, most or all of the finished compost is removed.

During composting, aerobic respiration is the main microbial process in all process stages and is responsible for mineralisation of available organic material, supplying the microorganisms with energy for growth and other processes. The initial surplus of easily available C leads to faster respiration under aerobic conditions compared with anaerobic conditions. The aerobic degradation generates heat, which typically leads to an increase in process temperature and progression of different phases during the composting process.

In composting, all typical phases of the process successively take place from start and until the finished compost is produced. The initial phase starts when the feedstock is mixed and prepared. In food waste composting, this phase is characterised by lowering of the pH, often to below 6 (Smårs *et al.*, 2002), and mesophilic temperatures. Any easily available organic material, such as monosaccharides, present in the feedstock is quickly consumed, accompanied by rapid consumption of available O_2. The accompanying reduced oxygen availability initiates fermentation and incomplete degradation, leading to formation of organic acids, which drive the pH down.

At the end of the initial phase, the pH starts increasing as the organic acids are consumed and NH_3 is released from mineralisation of proteins and amino acids. The increase in pH and temperature due to the intense microbial activity leads the process to the next stage – the highly active thermophilic composting phase, usually characterised by temperatures in the range 50-70 °C. Composting is typically associated with the presence of this thermophilic phase, which distinguishes it from other aerobic degradation processes found in *e.g.* forest litter degradation. The thermophilic phase is important for fast and efficient waste degradation and sanitisation.

The thermophilic phase ends as the easily available organic sources of energy in the feedstock are depleted, leading to slowing down in activity and process temperature decreasing to the mesophilic range. Next comes the curing phase, which varies in length depending on the requirements for the finished compost product and its future application. At the end of the curing phase, the finished compost product should be stable and, if the thermophilic phase has maintained sufficiently high temperatures for a sufficient period of time, free from pathogens and phytotoxins and hence fit for use as a fertiliser and soil improver (Bernal *et al.*, 2009; Niwagaba *et al.*, 2009).

2.1.1 Processes contributing to CH_4 and N_2O emissions

CH_4 emissions

In the composting process, aerobic respiration is the dominant process responsible for organic carbon turnover. However, anaerobic conditions occurring to different extents within compost lead to the development of anaerobic degradation. Two groups of methanogenic archaea produce CH_4 under strictly anaerobic conditions. Hydrogenotrophic methanogens use hydrogen gas as their energy source and reduce CO_2 to CH_4, while acetotrophic methanogens produce CH_4 from acetate, formate or methanol as energy and carbon sources (Schnurer *et al.*, 1994). The conditions required for methanogenesis are commonly present in the composting process, especially during the thermophilic phase, as oxygen is quickly consumed due to high decomposition rates of the available easily degradable organic material in the compost matrix. Insufficient aeration stimulates anaerobic degradation pathways, causing increased production of organic acids by fermenting microorganisms and CH_4 by methanogens. Side-effects of low oxygen supply include increased formation of malodorous compounds and an overall decrease in waste degradation efficiency (Sundberg *et al.*, 2004; Beck-Friis *et al.*, 2003).

In composts with limited aeration, CH_4 emissions may represent 10% or more of the initial carbon in the feedstock (Brown *et al.*, 2008; Szanto *et al.*, 2007). The rates of CH_4 production decrease rapidly with increasing oxygen supply due to the sensitivity of the methanogens to O_2 (Beck-Friis *et al.*, 2003). Well-aerated composts also contain anaerobic zones of varying size depending on process conditions and therefore a certain amount of CH_4 is always produced and present inside the compost matrix (Beck-Friis *et al.*, 2000; He *et al.*, 2000).

Methanotrophs, *i.e.* aerobic methane-oxidising bacteria, commonly reside in compost surface layers, where the oxygen concentration is higher than inside the compost piles (Brown *et al.*, 2008; Jäckel *et al.*, 2005). The methanotrophs can reduce the amount of CH_4 leaving the system by using it as their energy and carbon source. This activity requires the presence of both CH_4 and O_2 in sufficient concentrations (Scheutz *et al.*, 2009). Turning or mixing of the compost may move the methanotrophs away from the surface layer of the compost. This procedure can disrupt their activity, leading to CH_4 being emitted directly instead of being oxidised to CO_2 (Jäckel *et al.*, 2005). Small-sized compost piles or windrows can be expected to be more efficient in oxidising CH_4 at the surface layer (Wilshusen *et al.*, 2004), because the proportion of compost surface exposed to ambient air in relation to the total volume of compost material is higher than in large piles or windrows of compost.

The efficiency of compost aeration depends largely on the moisture content of the feedstock. A moisture content below the saturation limit allows better diffusion and activity of microbial exoenzymes and more efficient microbial uptake of substrate and exchange of products between microorganisms, and also facilitates free movement of the microorganisms, all of which promote faster and more efficient degradation of the waste material (Zhang *et al.*, 2011a). However, increasing moisture content beyond the optimal range restricts aeration due to water filling the compost pores, thus decreasing both O_2 diffusion and the O_2 concentration in compost particles. Such high moisture levels result in a rapid decline in degradation rate (Richard *et al.*, 2002) and limit aeration, causing an increase in CH_4 emissions (Jiang *et al.*, 2011). Compost matrices containing more structural amendment have better water-holding capacity and can generally support an efficient composting process at higher moisture levels. For composting of the organic fraction of MSW, the optimal moisture range typically lies within 50-70% moisture on a wet weight basis (Richard *et al.*, 2002).

Emissions of CH_4 from composting under both mesophilic and thermophilic temperatures have been reported (Ahn *et al.*, 2011; Andersen *et al.*, 2010a; Szanto *et al.*, 2007). The CH_4 emissions are commonly high when compost reaches the thermophilic phase due to favourable pH and a high rate of degradation. Thermophilic composting performed at temperatures within the range 50-60 °C is reported to produce the highest CH_4 emissions, while temperatures above 65 °C are reported to reduce CH_4 emissions and the overall degradation rate of the compost material (Amlinger *et al.*, 2008; Beck-Friis *et al.*, 2001; Hellmann *et al.*, 1997). In large-scale composting systems, thermophilic temperatures are generally reached quickly and are maintained for longer periods than in small-scale systems (Hermann *et al.*, 2011; Szanto *et al.*, 2007). Maintaining such temperatures for longer than necessary for sanitisation extends the period in which the highest CH_4 emissions occur during composting (Chan *et al.*, 2011; Sundberg & Jönsson, 2008) and stimulates emissions of NH_3 (Beck-Friis *et al.*, 2003).

N_2O emissions

During composting, depending on the technique, a substantial proportion of the available N in the feedstock may be lost due to emissions of N_2, NH_3 and N_2O. Ammonium originating from organic N mineralisation under conditions of high pH and temperature can be emitted directly as NH_3. Part of the NH_3 is nitrified to NO_3^-, which in turn can be denitrified and emitted as N_2. In both nitrification and denitrification processes, N_2O can be formed and emitted (Figure 2).

Nitrification, performed by autotrophic aerobic bacteria, is a two-step process during which NH_3 is first oxidised to nitrite (NO_2^-) by ammonia-oxidising bacteria and then further to NO_3^- by nitrite-oxidising bacteria. In the first step, N_2O can be produced and emitted due to incomplete oxidation of hydroxylamine (Wrage *et al.*, 2001). Another N_2O production route is by so-called nitrifier denitrification (Figure 2), where NO_2^- is reduced to N_2O under limited oxygen conditions, similar to that of the classical denitrification pathway (Kim *et al.*, 2010). Nitrifiers can be active in composts under both mesophilic and thermophilic temperatures (Jarvis *et al.*, 2009; Beck-Friis *et al.*, 2003).

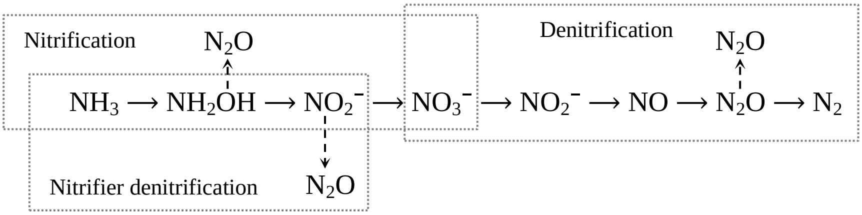

Figure 2. Schematic diagram of N_2O emission pathways during composting.

Heterotrophic denitrification is the anaerobic respiration process during which NO_3^- is reduced to NO_2^- and step-wise further to NO, N_2O and N_2. The ability to perform heterotrophic denitrification is characteristic to most groups of bacteria (Jones *et al.*, 2008). Under conditions of limited oxygen availability and low pH, N_2O can constitute a substantial fraction of the final gaseous product of denitrification due to inhibition of the sensitive nitrous oxide reductase enzyme required for the final reduction to N_2 (de Guardia *et al.*, 2010b; Liu *et al.*, 2010). Furthermore, multiple denitrifiers lack this enzyme altogether, which leads to inability to perform the last step of denitrification, *i.e.* reducing N_2O to N_2, thus resulting in N_2O as the final product (Maeda *et al.*, 2011; Philippot *et al.*, 2011).

The amount of N lost during composting largely depends on the type of feedstock and can represent over 60% or more of the N initially present in the substrate (Eklind & Kirchmann, 2000b). Emissions of NH_3 increase as the concentration of NH_4^+ increases and the pH of the compost rises above 7-8, because more ammonium ions lose protons to hydroxide ions and form gaseous NH_3. The NH_3 emissions are highest during the thermophilic phase of composting due to the high rates of mineralisation and vaporisation (Beck-Friis *et al.*, 2003). Emissions of NH_3 can represent 5-90% of the total gaseous nitrogen losses during composting (Jarvis *et al.*, 2009; Szanto *et al.*, 2007; Hellebrand & Kalk, 2000; Martins & Dewes, 1992). Rates of emissions of nitrogen in other forms can also vary widely depending on the process conditions and the amount of nitrogen initially available in feedstock. Emissions of N_2O in the range 1-10% of initial nitrogen have been reported for composting systems (Jarvis *et al.*, 2009; Szanto *et al.*, 2007). In composts with low NH_3 and N_2O emissions, N_2 can represent up to 90% of the total nitrogen losses (Szanto *et al.*, 2007).

Compost aeration and moisture affect nitrogen transformation dynamics, with large influences on N_2O and NH_3 emissions (Szanto *et al.*, 2007). Anaerobic or low oxygen conditions in the compost, as discussed above, can provoke N_2O production during nitrification of NH_4^+ present in the feedstock or produced by mineralisation of the compost material (Kim *et al.*, 2010). Low oxygen concentration in combination with nitrate availability can stimulate denitrification. However, improved aeration, supplying O_2 during denitrification, can inhibit reduction of N_2O to N_2, resulting in N_2O as the final product of denitrification (Bonin *et al.*, 2002).

A moisture content in the range 40-60%, which is commonly maintained in composting, promotes simultaneous nitrification and denitrification, which may lead to an increase in N_2O emissions (Angnes *et al.*, 2013). Compost moisture lower than the optimal range, on the other hand, can cause higher N_2O emissions due to better access by oxygen to denitrification sites, while higher moisture content stimulates complete denitrification to N_2 (Hwang & Hanaki, 2000).

2.2 Methodology

2.2.1 Home composting set-up (Paper I)

Home composting was investigated during one year in 18 individual home compost units, which were managed independently by the home owners in their backyards. The compost bins ranged in size from 0.2 to 0.4 m^3 (Table 2 in Paper I) and were fed food waste and other material regularly (Paper I). Gas was sampled from the compost bins before the lid was opened. Ambient air samples were collected in the vicinity of the bins as reference. The measurements were performed 13 times, approximately once a month, during the one-year period.

2.2.2 Windrow composting set-up (Paper II)

Large-scale composting was investigated in plastic-liner covered windrows with forced gas evacuation. A mixture of food and garden waste was composted in two sets of experiments, with 10 and 20 m long windrows in experiment 1 and experiment 2, respectively. Each experiment included three windrows following different aeration strategies. Aeration strategies, fan capacities, windrow dimensions and compost mixture are described in detail in Paper II. Gas evacuation pipes were placed on top of the windrows but under the plastic covers, causing vertical forced aeration through the windrows (Figure 3). Gas samples were collected from the exhaust of the evacuation fans, allowing an average sample from the whole windrow to be captured.

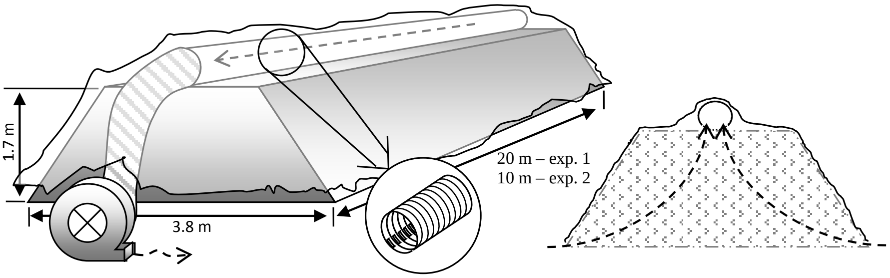

Figure 3. Schematic view of the windrow design, which varied in length between the two experimental set-ups (exp. 1 and exp. 2 in Paper II). Dashed arrows indicate expected airflow within the windrow.

Based on the windrow dimensions and compost matrix density, the amount of waste in the windrows was estimated to be 40 and 16 ton in experiments 1 and 2, respectively. The average aeration rate (Table 1) was estimated based on the measured airflow of the evacuation fans, which had different capacities (Paper II), as well as the frequency of on/off cycles of the fans (Lindström, 2009).

Table 1. *Average aeration rate in large-scale composting in three windrows during two sets of experiments (Paper II)*

Windrow	Aeration strategy	Average aeration rate (m^3 ton^{-1} $hour^{-1}$)	
		Experiment 1	Experiment 2
I	Initially cooled	22	78
V	Varied ventilation	9	17
C	Constantly on	11	28

2.2.3 Reactor composting set-up (Papers III and IV)

An advanced 200-L compost reactor system was used in two sets of reactor experiments to investigate the influence of temperature and levels of oxygen and moisture in the composting process on emissions of CH_4 and N_2O (Papers III and IV). The laboratory reactor allowed automated control of the composting temperature, aeration and oxygen level independently (Smårs *et al.*, 2001). The ventilation of the reactor was designed to ensure fairly constant oxygen levels. This resulted in the CO_2 concentration also being fairly constant at around 5 and 20% for the reactor runs performed at 16 and 1% O_2, respectively.

Before the second set of reactor experiments described in Paper IV, the reactor design used in Paper III was improved to produce a more uniform temperature and moisture distribution.

Gas sampling in both sets of reactor experiments was performed by direct point measurements of gas extracted from the cooling loop of the reactor (Figure 4). In the second set of reactor experiments (Paper IV), a cumulative proportional gas sampling system that collected gas samples proportionally to the volume of the gas leaving the reactor was installed (Figure 4).

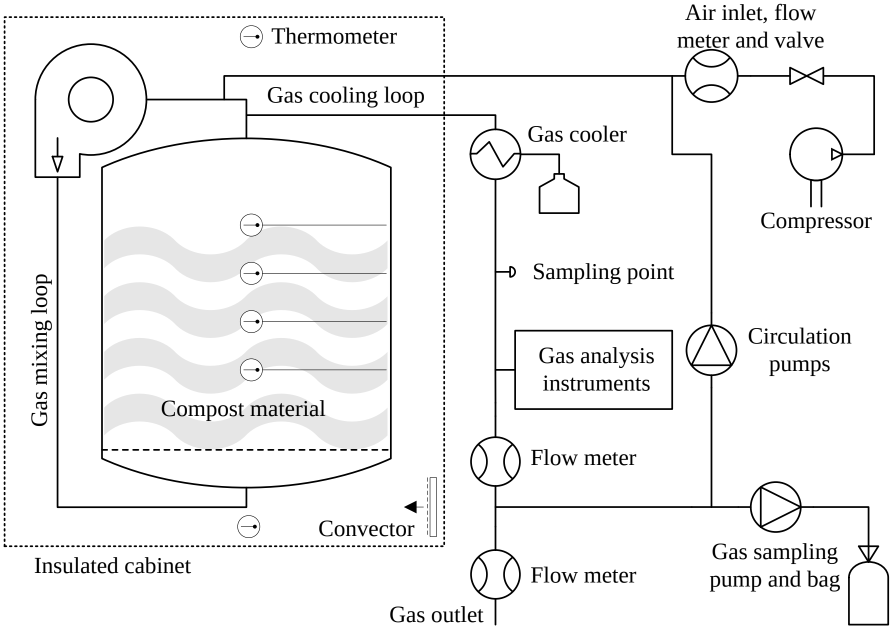

Figure 4. Schematic diagram of the 200-L compost reactor with additional gas sampling system and insulation cabinet used in Paper IV.

2.2.4 Analyses

Gas samples in all experiments were analysed for CO_2, N_2O and CH_4 using gas chromatography (GC) (Papers I-IV). Analytical equipment connected to the reactor allowed online measurement of O_2 and, in most of the experiments, also CO_2 (Papers III and IV).

Samples of compost material were analysed using standard methods for pH, moisture content and volatile solids (Papers I-IV). In some of the experiments the concentrations of NH_4^+ and NO_3^- in the material were also analysed. In all of the reactor experiments, the initial and final total C were analysed and in most of these experiments the total N was also analysed (Papers III and IV).

2.2.5 Ratios of CH_4:CO_2 and N_2O:CO_2 and total emissions

Emissions of CH_4 and N_2O from the composting systems included in this thesis were measured in different experimental set-ups, process stages and environmental conditions. Emissions of CO_2 during composting are a good indicator of the amount of degraded organic carbon, because composting is predominantly aerobic and essentially all the degraded carbon is emitted as CO_2 (Hermann *et al.*, 2011).

In order to provide a good representation of emissions rates of CH_4 and N_2O in relation to the degradation rate in different composting systems and to simplify fair comparisons between these systems, the CH_4:CO_2 and N_2O:CO_2 ratios were calculated and used as a tool in the evaluation. Prior to calculating these ratios, the ambient air concentrations of CH_4, CO_2 and N_2O were subtracted from all measured data, as the purpose of using the ratios was to evaluate the emissions of these gases from the compost process *per se*.

However, in using ratios the overall total emissions can be difficult to estimate if the amount of degraded carbon is not known. This can be circumvented by estimating the total amount of CO_2 emissions. The interpretation of the ratios is complicated if a large part of the decomposition takes place due to anaerobic processes. In such cases, carbon can be produced and released in forms other than CO_2, such as CH_4 or volatile fatty acids, which must be taken into account when estimating the degradation rate.

In the home composting study (Paper I), the minimum, maximum, average and median values for CH_4:CO_2 and N_2O:CO_2 ratios were calculated based on all sampling points in all composts (n=234). In the home composts, the gas flow rates were not known. Therefore, to estimate the total CH_4 and N_2O emissions, the average volume of CO_2 produced was calculated based on estimation of amount of organic carbon in the organic matter and the fraction being degraded. The amount of carbon in the feedstock organics was assumed to be 41% (Sundberg *et al.*, 2011; Eklind *et al.*, 1997). The amount of degraded waste was calculated from the volatile solids (VS) reduction. The average VS in waste in the home composts throughout the study was 74±16% of TS (mean ± standard deviation; n=234), while the VS of the finished compost product sampled from six bins was 59±19% of TS. Calculation of CO_2 yield from degradation of food waste, garden waste and structural amendment was based on degradation of 19.7% of initial VS and was performed as suggested by Ermolaev *et al.* (2011).

In the windrow and reactor experiments (Papers II-IV), the estimation of total emissions was based on the measured gas concentrations and known gas flows through the composting systems. Based on the flows and using direct online measurements of CO_2, the total CO_2 emissions were calculated for the reactor experiments (Papers III and IV). Gas flow rates at the time of sampling together with the concentrations of CO_2, CH_4 and N_2O from the point measurements were used to calculate the emissions of these gases (Papers II-IV). The calculated emissions were then integrated over time using the trapezoidal rule (Holman, 2001).

In the reactor runs presented in Paper IV, the exact concentration of the emissions was known during each period of sampling from the cumulative gas sampling system (described in section 2.2.3 and Paper IV). These concentrations were used to calculate the total emissions using the trapezoidal rule as described above, except that in this case the flow rate was averaged over time of sampling. Calculation of minimum, maximum and median values for the CH_4:CO_2 and N_2O:CO_2 ratios during the reactor runs presented in Paper IV was based on the point measurements, in the same way as for the windrows in Paper II and the reactor runs in Paper III.

2.3 Methane emission patterns

The CH_4 emissions were measured from both small-scale and large-scale composting systems having only few controlled parameters and from laboratory reactor with multiple well-controlled process conditions. The emissions were analysed as ppm concentrations and as CH_4:CO_2 volume ratios in relation to the degradation rate measured as CO_2 emissions.

In the home composting study, the CH_4 emissions were relatively small. The average CH_4 concentration was 28.1 ppm above ambient (Fig. 4 in Paper I). The average CH_4:CO_2 ratio in the gas emitted from the compost bins, with the respective ambient levels of these gases subtracted, was 0.376%, while the median value was 0.037% (Table 2; Table 3 in Paper I).

The home composts displayed very high moisture content, 73% on average (Fig. 3 in Paper I). At such high moisture levels, substantial CH_4 emissions could be expected due to oxygen becoming limiting (Jiang *et al.*, 2011; Amlinger *et al.*, 2008; Richard *et al.*, 2002). However, the CH_4 emissions were lower than those reported in several other home compost studies (Chan *et al.*, 2011; Andersen *et al.*, 2010a; Amlinger *et al.*, 2008) and large-scale systems (Amlinger *et al.*, 2008) with lower moisture levels. The effects of moisture in a more controlled environment were further investigated in Paper IV.

The temperature of the home composting bins was largely influenced by the temperature of the ambient air, as an effect of their small size (Fig. 2 in Paper I) and large surface to volume ratio of the compost material. Average CH_4:CO_2 emissions ratios were lowest in late autumn and winter months, with average values as low as 0.022% during January (Fig. 5 in Paper I). A few home composts displayed temperatures in the thermophilic range and these often had higher CH_4 emissions (Ermolaev *et al.*, 2011). The contributions of several compost bins with high emissions were reflected in the median values being much smaller than the mean values (Table 2).

Table 2. *Emissions ratios of CH_4:CO_2 from the different composting systems in Papers I-IV (P I-IV). The concentrations in ambient air were subtracted from all gas concentrations prior to calculation*

Composting system	CH_4:CO_2 emissions, %			
	Min	Max	Average	Median
Home composting (P I)	<0.001	5.08	0.376	0.037
Windrow composting (P II)	<0.001	7.52	1.01[1]	0.744
Reactor composting (P III)[2]	<0.001	2.35	0.006[3]	–
Reactor composting (P IV)[2]	<0.001	56.4	1.59	–

[1] Average value calculated differently than in Table 6 in Paper I, based on estimated total emissions instead of using average ppm values.

[2] Median values for separate reactor runs are shown in Tables 3 and 4, as composting conditions differed.

[3] Excluding the results from the low oxygen run (55°C-1%).

The CH_4 concentrations in the exhaust gas from the windrow composting in experiments 1 and 2 in Paper II varied from close to 0 to 225 ppm, as observed in windrow C during experiment 1 (Table 1 in Paper II). The emissions of CH_4 started with an initial CH_4:CO_2 ratio of 0.048% on average for the two experiments. At day 12 the emissions ratio peaked at 7.52% during the thermophilic composting phase in windrow V in experiment 2 (Table 2), which corresponded to a CH_4 concentration of 122 ppm (Table 1 in Paper II). The emissions ratio differed between the windrows aerated differently, but the overall average was 1.01% CH_4:CO_2 (Table 2). The average CH_4 emissions were within the range reported for other large-scale and small-scale composting systems (Chan *et al.*, 2011; Hermann *et al.*, 2011; Andersen *et al.*, 2010a; de Guardia *et al.*, 2010a; Amlinger *et al.*, 2008).

When comparing the overall CH_4:CO_2 ratios, the difference in CH_4 emissions dynamics between windrows and experiments was small (Figure 5), which was unexpected, as the aeration rates differed considerably (Table 1). The 20-day experimental period captured only a part of the thermophilic phase, as shown by the temperature still remaining above 55 °C at the end of the experiment (Figure 4 in Paper II).

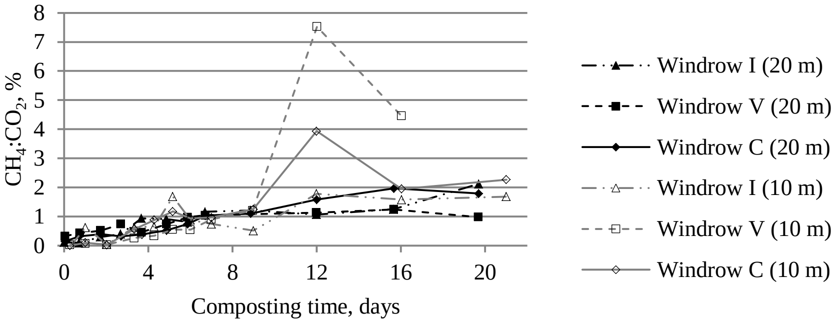

Figure 5. Ratio of $CH_4:CO_2$ in exhaust gas from two compost experiments with 10 and 20 m long windrows, respectively (Paper II). The concentrations in ambient air were subtracted from all gas concentrations prior to ratio calculations.

In the first set of rector experiments examining the effects of temperature and aeration (Paper III) a range of CH_4 concentrations was seen, with peaks from 0.610 to 2020 ppm in the different runs (Table 3). The average $CH_4:CO_2$ ratio was 0.006% for the 16% O_2 runs, while for the 1% O_2 run (55°C-1%) it was 0.133%. The CH_4 peaks normally appeared after the end of the initial low pH phase (Figure 2 in Paper III).

Temperature affected the CH_4 emissions in the first set of reactor experiments (Paper III). During the 67 °C runs, the CH_4 concentrations were low throughout the runs, which resulted in $CH_4:CO_2$ ratios of 0-0.001%, the lowest for all temperatures tested (Table 3). The largest CH_4 peak in the 16% O_2 runs, observed at the mesophilic temperature of 40 °C, was 0.395% $CH_4:CO_2$, which corresponded to a CH_4 concentration of 197 ppm. This peak was wider and appeared later than the peaks in the other runs (Figure 2 in Paper III). The average $CH_4:CO_2$ ratio was 0.029% for the 40 °C run, which was the highest for all 16% O_2 runs presented in Paper III (Table 3).

The run with 1% O_2 and 55 °C (55°C-1%) had the highest CH_4 emissions among the runs in the first set of reactor experiments, most likely due to oxygen limitation. The overall degradation rate in this run was slower than in those at higher O_2 (16%), and the CH_4 peak did not appear until about 20% of the initial carbon had been mineralised (Figure 3 in Paper III).

Table 3. *Concentrations of CH_4 and CH_4:CO_2 ratios during the first set of reactor experiments (Paper III). For designations of runs, see Paper III*

Run[1]	Duration, days	CH_4, ppm			
		Min	Max	Average	Median
40°C	36	0.309	197	40.1	1.95
55°C-a	11	-0.565	11.8	2.86	0.344
55°C-b	8	-1.07	4.27	2.24	2.04
55→30°C	59	-0.720	51.5	6.89	2.23
55°C-1%	16	-0.070	2020	473	3.15
67°C-a	24	0.109	0.610	0.450	0.509
67°C-b	17	-0.938	0.940	-0.192	-0.193
67°C-c	38	0.561	0.848	0.664	0.650

Run[1]	C loss in CO_2-C % initial C	CH_4:CO_2, %			
		Min	Max	Average[2]	Median
40°C	64	0.001	0.395	0.029	0.004
55°C-a	59	<0.001	0.024	0.003	0.001
55°C-b	67	<0.001	0.009	0.005	0.004
55→30°C	67	<0.001	0.103	0.005	0.004
55°C-1%	28	<0.001	1.01	0.133	0.002
67°C-a	56	<0.001	0.001	0.001	0.001
67°C-b	53	<0.001	0.002	<0.001	<0.001
67°C-c	64	0.001	0.002	0.001	0.001

[1] CH_4 emissions not measured for the runs 55°C-e and 55→70°C.

[2] Average ratio calculated from cumulative emissions (m^3/m^3).

In the second set of reactor experiments investigating the effect of moisture (Paper IV), the pattern of CH_4 emissions was similar between the compost runs with different moisture levels. The emissions were low early in the composting process. Production of CH_4 peaked when process pH in the condensate had increased to just above 6 and the temperature had turned thermophilic. The size of the CH_4 peaks in all runs was proportional to the set moisture levels, with CH_4 concentration ranging from 10.1 to 26500 ppm (Table 4). The peak CH_4:CO_2 ratio ranged in value from 0.023% in Run 1 with 44% average moisture content to 56.4% in Run 6 at 66% average moisture. The overall average CH_4:CO_2 ratio of runs at a moisture content of 44% and 66% was 0.007 and 8.59%, respectively.

Table 4. *Concentrations of CH_4 and CH_4:CO_2 ratios during the second set of 20-day reactor experiments with increasing moisture levels (Paper IV). For designations of runs, see Paper IV*

Run	Average moisture, %	CH_4, ppm			
		Min	Max	Average	Median
Run 1, Dry, MC 25-40%	44 ± 11	-0.016	10.1	2.97	1.71
Run 2, Dry, MC 25-40%	43 ± 8	0.083	19.7	8.61	10.4
Run 3, MC 40-50%	48 ± 6	0.095	195	55.0	28.6
Run 4, MC 40-50%	49 ± 6	-0.016	97.8	29.6	24.4
Run 5, Wet, MC 50-60%	59 ± 4	0.022	1670	523	540
Run 6, Wet, MC 60-70%	66 ± 2	0.468	26500	6490	2410

Run	C loss, CO_2-C % initial C	CH_4:CO_2, %			
		Min	Max	Average[1]	Median
Run 1, Dry, MC 25-40%	50	<0.0001	0.023	0.007	0.004
Run 2, Dry, MC 25-40%	45	0.0002	0.041	0.011	0.021
Run 3, MC 40-50%	47	0.0003	0.442	0.115	0.061
Run 4, MC 40-50%	49	<0.0001	0.220	0.043	0.054
Run 5, Wet, MC 50-60%	45	<0.0001	3.76	0.790	1.21
Run 6, Wet, MC 60-70%	40	0.0008	56.4	8.59	7.13

[1] Average ratio calculated from cumulative emissions (m^3/m^3) from continuous sampling system.

The increase in total CH_4 emissions induced by increasing moisture levels was very large. Only a few other studies have reported such a moisture effect, but without reporting the magnitude of the effect (Jiang *et al.*, 2011; Tamura & Osada, 2006). The emissions were almost 1000-fold higher in the 66% moisture Run 6 than in the 44% moisture Run 1 (Figure 5 in Paper IV). The total CH_4 emissions followed the regression model shown in Equation 1 ($P<0.001$; $R^2=0.95$) (Figure S3 in Paper IV):

$$\log_{10} CH_4 = -9.43 + 12.14 \cdot MC \qquad \text{Equation 1}$$

where CH_4 is the total emissions in kg CH_4-C per kg initial C and MC is moisture fraction of the total wet weight.

2.4 Nitrous oxide emission patterns

The N_2O emissions from the different composting systems, representing small-scale, large-scale and laboratory-scale composting, ranged from relatively low in the laboratory reactor experiments to relatively high in the home composts (Table 5). The N_2O:CO_2 ratio was used to compare the N_2O emissions in relation to the degradation rate, measured as CO_2 emissions.

In the home composting study, the N_2O concentration of all 18 compost bins and samplings was on average 5.46 ppm above ambient atmospheric level. The average $N_2O:CO_2$ ratio in the gas in the compost bins after subtraction of ambient levels was 0.147% (Table 5; Table 3 in Paper I). None of the variables tested (Table 1 in Paper I) seemed to affect the N_2O emissions in a significant way and, unlike the CH_4 emissions, there was no clear seasonal pattern in N_2O emissions (Fig. 5 in Paper I). The difference between the average and median $N_2O:CO_2$ ratio (Table 5) suggested that a few events with high emissions influenced the overall average of N_2O emissions. However, analysing the data for these high emissions occasions separately did not provide any further insights into the reasons for the increased emissions (Ermolaev *et al.*, 2011).

Table 5. *Emissions ratios of $N_2O:CO_2$ from the different composting systems in Papers I-IV (P I-IV). The concentrations in ambient air were subtracted from all gas concentrations prior to calculation*

Composting system	$N_2O:CO_2$, %			
	Min	Max	Average	Median
Home composting (P I)	<0.0001	2.41	0.147	0.067
Windrow composting (P II)	<0.0001	0.012	0.003[1]	0.003
Reactor composting (P III) [2]	<0.0001	0.066	0.001	–
Reactor composting (P IV) [2]	<0.0001	0.039	0.004	–

[1] Average value calculated differently than in Table 6 in Paper I, based on estimated total emissions instead of using average ppm values.

[2] Median values for individual reactor runs are shown in Tables 6 and 7, as composting conditions differed.

The relatively high N_2O emissions in the home composting study might partly be explained by the high moisture content of the composts (73% on average). This most likely limited the aeration, promoting denitrification (Angnes *et al.*, 2013), especially in combination with the sub-mesophilic temperatures (Amlinger *et al.*, 2008) observed in most home composts (Fig. 2 in Paper I) and the long compost retention time compared with other systems studied (Papers II-IV). The effect of high moisture was further investigated in the reactor experiment reported in Paper IV and was demonstrated to cause an increase in N_2O emissions (Run 6 in Paper IV).

In the windrow study, the concentration of N_2O in the exhaust gas was higher during experiment 1, with proportionately higher CO_2 concentration, probably due to longer windrows (Table 1 in Paper II). Nevertheless, the N_2O emissions were relatively low in both experiments (Table 5), with an overall average $N_2O:CO_2$ ratio of 0.003% and with very similar emission patterns among the three windrows I, V and C (Figure 6). Windrows C and V demonstrated a peak in $N_2O:CO_2$ on day 12 (Figure 6), even though the actual concentrations of both CO_2 and N_2O in ppm decreased on that day (Figure 2 in Paper II). At the end of the experiments, a tendency for an increase in N_2O:CO_2 ratio was observed in several of the windrows (Figure 5). This indicates that the N_2O emissions could potentially have increased towards the end of the composting period. However, it was not possible to fully conclude whether such an increase occurred, as the composting period in the study was short and included only a part of the thermophilic phase (Figure 4 in Paper II).

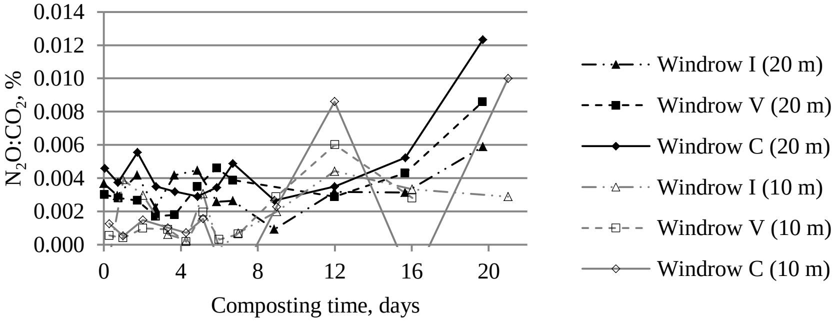

Figure 6. Ratio of N_2O:CO_2 in exhaust gas from two compost experiments with 10 and 20 m long windrows, respectively (Paper II). The concentrations in ambient air were subtracted from all gas concentrations prior to ratio calculations.

In the first set of reactor experiments consisting of 10 runs, the effect of composting temperature and limited aeration on N_2O emissions was evaluated (Paper III). A range of N_2O concentrations was observed, with peaks from 0.524 ppm to 7.13 ppm (Table 6). The N_2O peaks were present both early and late in the process (Figure 4 in Paper III). The early peaks appeared within the first five days from the start of composting and coincided with a reduction in nitrate availability in the initial compost mixture (Figure 5 in Paper III). The composting phase displaying the early peaks was characterised by acidic pH and mesophilic temperatures (Table 1 in Paper III). The later peaks appeared between days 10 and 20, when conditions allowing both nitrification and denitrification had developed (Figure 4 in Paper III).

During the runs at 16% O_2 in the first set of reactor experiments, the average N_2O:CO_2 was 0.001% (Table 5), while it was 0.0003% in the 1% O_2 run (55°C-1%). No clear relationship between composting temperature and N_2O emission could be seen in the different trials (Figure 3 in Paper III). The N_2O:CO_2 ratio in the 67 °C runs was in the same range as that at other temperatures (Table 6), suggesting that nitrification might not have been inhibited by the thermophilic temperature, which is in contrast to findings in some previous studies (Habart *et al.*, 2010; Amlinger *et al.*, 2008) and in agreement with others (Jarvis *et al.*, 2009; Jäckel *et al.*, 2005). The overall N_2O:CO_2 ratio was relatively low, however (Table 5), which might have obscured the possibility to observe any temperature effect.

Table 6. *Concentrations of N_2O and N_2O:CO_2 ratios during the first set of reactor experiments (Paper III). For designations of runs, see Paper III*

Run	Duration, days	N_2O, ppm			
		Min	Max	Average	Median
40°C	36	0.044	0.621	0.254	0.179
55°C-a	11	-0.009	0.524	0.169	0.101
55°C-b	8	-0.023	0.541	0.192	0.105
55→30°C	59	-0.025	7.13	1.23	0.579
55°C-1%	16	-0.285	4.98	0.534	-0.197
55°C-e	16	0.185	1.97	0.708	0.512
55→70°C	16	0.217	5.78	1.92	0.797
67°C-a	24	-0.142	1.00	0.222	0.122
67°C-b	17	0.179	1.75	0.502	0.283
67°C-c	38	0.317	1.32	0.519	0.414

Run	C loss in CO_2-C % initial C	N_2O:CO_2, %			
		Min	Max	Average[1]	Median
40°C	64%	0.0001	0.0012	0.0005	0.0004
55°C-a	59%	<0.0001	0.0657	0.0001	0.0002
55°C-b	67%	<0.0001	0.0011	0.0003	0.0002
55→30°C	67%	<0.0001	0.0143	0.0009	0.0012
55°C-1%	28%	<0.0001	0.0025	0.0003	<0.0001
55°C-e	34%	0.0004	0.0039	0.0017	0.0010
55→70°C	32%	0.0004	0.0116	0.0034	0.0016
67°C-a	56%	<0.0001	0.0020	0.0006	0.0002
67°C-b	53%	0.0004	0.0035	0.0007	0.0006
67°C-c	64%	0.0006	0.0026	0.0008	0.0008

[1] Average ratio calculated from cumulative emissions (m^3/m^3).

Mesophilic temperature at the end (when about 50% initial C had been degraded) of the compost reactor run starting at 55 °C and then downregulated to 30 °C (55→30°C) could have been responsible for the increasing N_2O emissions towards the end of that run (Figure 3 in Paper III). Moreover, CH_4 production was also observed at the end of that run (Figure 2 in Paper III), indicating anaerobic conditions, which could have stimulated denitrification, resulting in the release of N_2O. However, the observed $N_2O:CO_2$ emissions peak of 0.0143% was still low compared with that of 2.41% observed in the home composting study (Table 5 and 6).

During the compost reactor run set to 55 °C and 1% O_2 (55°C-1%), the N_2O emissions were in the same range as during the runs performed at 16% O_2 (Table 6). At first, the emissions dynamics for both O_2 levels were similar, but later in the process during the 55°C-1% run, when about 8% of the initial carbon had been mineralised, only very low concentrations of N_2O were observed (Figure 3 in Paper III). Similar N_2O emission dynamics were observed during Run 6 performed at 66% average moisture in the second set of reactor experiments (Figure 7 in Paper IV).

The runs 55°C-e and 55→70°C had higher N_2O emissions than the other runs in the first set of reactor experiments (Table 6). The increased N_2O emissions during these runs could be due to the difference in source of the material composted compared with the other runs in the first set of reactor experiments (Table 2 in Paper III). These two runs (55°C-e and 55→70°C) also had lower initial moisture content, which was shown to increase the N_2O emissions in the second set of reactor experiments (Paper IV).

In the second set of reactor experiments, investigating the influence of moisture on GHG emissions (Paper IV), a distinct N_2O emissions pattern emerged, displaying an emissions peak early in the process and also a period with elevated emissions later in the composting process (Figure 6 in Paper IV). The peak concentrations of N_2O varied from 5.83 to 61.6 ppm (Table 7), with a general tendency for a larger peak at lower moisture except for Run 6, which had both the highest moisture content (66% on average) and the highest peak concentration of N_2O (Figure 6 in Paper IV). However, the overall emissions dynamics were different during Run 6 than in the runs at other moisture levels (Figure 7 in Paper IV). During Run 6, very little N_2O was produced late in the process, when pH in the condensate had increased above 6 and the temperature was thermophilic. A similar pattern was observed during the low O_2 run (55°C-1%) in the first set of reactor experiments, suggesting that the emissions in both runs were affected by oxygen limitation. Increased CH_4 production in Run 6 confirmed anaerobic conditions in the material.

The N_2O peak occurring early, before the pH increase in the second set of reactor experiments, contributed over half of all N_2O emitted during Runs 3-6 (Figure 7 in Paper IV) and there were indications that it was a result of incomplete denitrification. The role of denitrification was suggested by almost complete reduction of the initially available NO_3^- until day 4 in all reactor runs (Figure 8 in Paper IV). Similar NO_3^- reduction dynamics in the compost material were seen in the runs of the first set of reactor experiments (Figure 5 in Paper III), suggesting that the early N_2O peaks in these runs were also due to incomplete denitrification of nitrate present in the feedstock.

Table 7. *Concentrations of N_2O and $N_2O:CO_2$ ratios during the second set of 20-day reactor experiments with increasing moisture levels (Paper IV). For designations of runs, see Paper IV*

Run	Average moisture, %	N_2O, ppm			
		Min	Max	Average	Median
Run 1, Dry, MC 25-40%	44 ± 11	0.506	28.5	2.77	1.99
Run 2, Dry, MC 25-40%	43 ± 8	0.444	11.4	2.02	1.74
Run 3, MC 40-50%	48 ± 6	0.266	5.83	1.19	1.09
Run 4, MC 40-50%	49 ± 6	0.299	6.70	1.00	0.646
Run 5, Wet, MC 50-60%	59 ± 4	0.135	8.22	0.682	0.231
Run 6, Wet, MC 60-70%	66 ± 2	-0.022	61.6	3.70	0.730

Run	C loss, CO_2-C % initial C	$N_2O:CO_2$, %			
		Min	Max	Average[1]	Median
Run 1, Dry, MC 25-40%	50	0.0013	0.020	0.004	0.005
Run 2, Dry, MC 25-40%	45	0.0010	0.024	0.004	0.003
Run 3, MC 40-50%	47	0.0007	0.014	0.003	0.002
Run 4, MC 40-50%	49	0.0007	0.016	0.002	0.001
Run 5, Wet, MC 50-60%	45	0.0003	0.020	0.001	0.001
Run 6, Wet, MC 60-70%	40	<0.0001	0.039	0.007	0.001

[1] Average ratio calculated from cumulative emissions (m^3/m^3) from continuous sampling system.

The N_2O production continued after the first 5 days in Runs 1-6, but at slower rate than during the initial peak. The combined emissions of N_2O from the initial peak and later production (Figure 7 in Paper IV) resulted in significant differences in the overall $N_2O:CO_2$ ratio between the different runs, from very low (0.001%) during Run 5 to 0.004% in the driest runs (1 and 2) and up to 0.007% in the wettest (Run 6) (Table 7). Regression analysis of N_2O emissions in relation to compost moisture (excluding the outlying results from Run 6) produced the model in Equation 2 ($P=0.04$; $R^2=0.81$):

$$N_2O = 0.003 - 0.005 \cdot MC \qquad \text{Equation 2}$$

where N_2O is the total emissions in kg N_2O-N per kg initial N and MC is moisture fraction of the total wet weight. Average mineralisation of the organic matter for all runs in the second set of reactor experiments was 0.42 kg CO_2-C kg^{-1} initial C (Table 8).

A second regression analysis was performed on the accumulated N_2O after the pH and temperature increase, *i.e.* after about 4 days of composting

(Figures 2 and 3 in Paper IV). This excluded the initial N_2O peak and therefore Run 6 could be included in the regression analysis. The analysis produced a model (Equation 3) with a slightly higher significance level (P=0.02) and a less good fit (R^2=0.66) than when the early peaks were included in the analysis (Equation 2).

$$N_2O_{(late)} = 0.001 - 0.002 \cdot MC \qquad \text{Equation 3}$$

where $N_2O_{(late)}$ is the emissions in kg N_2O-N per kg initial N produced after the temperature increased to 55 °C and MC is moisture fraction of the total wet weight. The lower R^2 value was partly caused by the initial peak and also by the later N_2O emission dynamics of Run 6 being different from the other runs in this set of reactor experiments. Such a difference was also observed in NH_4^+ concentrations in the matrix, which increased during Run 6 and decreased in the other runs (Figure 8 in Paper IV).

The overall N_2O:CO_2 ratios observed in the runs of the second set of reactor experiments (Paper IV) were smaller than the average value observed in the home composting study (Table 5; Table 6 in Paper I) and in other composting systems reported in the literature (Chan *et al.*, 2011; Hermann *et al.*, 2011; Andersen *et al.*, 2010a; de Guardia *et al.*, 2010a; Amlinger *et al.*, 2008). This suggests that additional factors besides substrate moisture content affected the emissions in the different studies presented here.

2.5 Greenhouse gas emissions from composting

The CH_4:CO_2 and N_2O:CO_2 ratios (described in section 2.2.5) were used in evaluating average emissions of CH_4 and N_2O from the different composting systems studied in this thesis (Table 9). The ratios facilitated comparison and gave clear estimates of system performance regarding the emissions of N_2O and CH_4 in relation to the overall activity, *i.e.* the degradation rates of the systems. In order to calculate the total GHG emissions of the different composting systems, the emissions of CH_4 and N_2O are presented in relation to the amount of carbon degraded, which consists of CO_2-C and CH_4-C emissions (Tables 8-11). Values allowing calculation of the emissions per g initial wet weight (WW) and per g initial carbon are also presented. Since the CO_2 emissions from composting are considered to be biogenic, the estimated emission values are given as reference, but were not included in total GWP calculation.

Table 8. *Average emissions of CH_4 and N_2O during composting in the different systems discussed in Papers I-IV (P I-IV) in relation to initial carbon (C) in feedstock and to C emitted as CO_2 and CH_4*

Composting system	Initial TS, g init. solids g^{-1} init. WW	Init. C content, g g^{-1} init. solids	Total CO_2, g CO_2-C kg^{-1} init. C	Total CH_4, g CH_4-C kg^{-1} init. C	Total N_2O, g N_2O-N kg^{-1} init. C	Tot. CH_4, g CH_4-C kg^{-1} C emitted[1]	Tot. N_2O, g N_2O-N kg^{-1} C emitted[1]
Home composting (P I)	0.27	0.47	184	0.692	0.632	3.75	3.42
Covered windrows (P II)	0.45	0.44	209	2.08	0.015	9.87	0.072
Laboratory reactor (P III, 16% O_2)	0.37	0.40	551	0.038	0.010	0.069	0.019
Laboratory reactor (P III, 1% O_2)	0.44	0.44	275	0.365	0.002	1.33	0.006
Laboratory reactor (P IV)	0.45	0.42	460	6.52	0.039	14.0	0.083

[1] Carbon emitted assumed to be sum of total CO_2-C and CH_4-C emissions.

A comparison of the different composting systems studied in Papers I-IV showed that home composting had lower total CH_4 emissions than windrow composting. The lower CH_4:CO_2 ratios observed in home composting (Table 9) were partly due to low degradation rates, estimated to be 184 g CO_2-C kg^{-1} initial C, and partly to low overall emissions of CH_4 (Table 8). The average N_2O emissions from home composts were 3.42 g N_2O-N kg^{-1} C emitted, the highest among all systems studied. This was also evident from the high average N_2O:CO_2 ratio (Table 9). The data on total emissions from the home composts have to be treated with caution, however, due to the very large temporal variation in emissions during the study year and the wide variation in other parameters, both between the compost bins and between the samplings.

Table 9. *Average emissions ratios and total GHG emissions based on GWP using a 100-year time horizon in the different systems discussed in Papers I-IV (P I-IV) in relation to carbon (C) emitted as CO_2 and CH_4*

Composting system	CH_4:CO_2, %	N_2O:CO_2, %	Emissions in g CO_2-eq. kg^{-1} C emitted[1]			
			CO_2	CH_4	N_2O	Total
Home composting (P I)[1]	0.376	0.147	3650	170	1600	1770
Covered windrows (P II)	1.01	0.003	3630	448	33.4	482
Laboratory reactor (P III, 16% O2)	0.006	0.001	3660	3.13	8.86	12.0
Laboratory reactor (P III, 1% O2)	0.133	<0.001	3660	60.2	2.97	63.2
Laboratory reactor (P IV)	1.59	0.004	3610	635	38.9	673

[1] Carbon emitted assumed to be sum of total CO_2-C and CH_4-C emissions.

Based on the regression analysis results, large CH_4 emissions could be expected from home composts displaying high degradation rates and temperatures. In such cases, average concentrations did not accurately represent the whole system and the use of ratios was more beneficial, especially when they were presented with the whole range of values, including minimum, maximum and median (Fig. 5 in Paper I).

In the first set of reactor experiments investigating the influence of compost temperatures and aeration on GHG emissions, the CH_4 emissions during the mesophilic run (40 °C) were 13.0 g CO_2-eq. kg^{-1} C emitted, the highest observed among the 16% O_2 runs (Table 10). From a GWP perspective, the CH_4 emissions in g CO_2-eq. kg^{-1} C emitted were greater than, or approximately equal in magnitude to, the N_2O emissions, except for the 67 °C compost reactor runs (Table 10). The CH_4 emissions were not measured in the 55°C-e and 55→70°C runs, but their emissions of N_2O were high. During the low O_2 run (55°C-1%), total CH_4 emissions were 60.2 g CO_2-eq. kg^{-1} C emitted, which was the highest in this set of compost reactor experiments (Table 10). Comparing this with the emissions during the second set of reactor experiments, however, showed that the increase in CH_4 emissions during high moisture composting was larger than in compost with low O_2 (Table 8).

Table 10. *Total GHG emissions based on GWP using a 100-year time horizon in relation to carbon (C) emitted as CO_2 and CH_4 for the individual compost reactor runs presented in Paper III*

Run	Emissions in g CO_2-eq. kg^{-1} C emitted[1]			
	CO_2	CH_4	N_2O	Total
40°C	3660	13.0	5.32	18.3
55°C-a	3660	1.18	1.34	2.51
55°C-b	3660	1.46	1.76	3.22
55→30°C	3660	2.28	9.95	12.2
55°C-1%	3660	60.2	2.98	63.2
55°C-e [2]	3660	–	18.1	–
55→70°C [2]	3660	–	37.5	–
67°C-a	3660	0.436	6.96	7.40
67°C-b	3660	<0.001	7.82	7.82
67°C-c	3660	0.491	8.91	9.40

[1] Carbon emitted assumed to be sum of total CO_2-C and CH_4-C emissions.

[2] CH_4 emissions not measured.

The emissions of CH_4 were highest from the windrow composting and in the reactor study on the effects of moisture (Table 9). However, the values (Table 6 in Paper I) were within the range reported for other large-scale and small-scale composting systems (Chan *et al.*, 2011; Hermann *et al.*, 2011; Andersen *et al.*, 2010a; de Guardia *et al.*, 2010a; Amlinger *et al.*, 2008). The increased overall average CH_4 emissions during the second set of reactor experiments investigating moisture effects (Table 9) were strongly influenced by the emissions in the wet runs (5 and 6) increasing exponentially (Table 11). This suggests that certain moisture levels during composting could be of high concern for both CH_4 and N_2O emissions. When comparing the CO_2 equivalents of these emissions, both the second set of reactor experiments and the windrow experiments showed that CH_4 was generally more important from the global warming perspective. However, it should be noted that the results presented from the covered windrow composting studies comprised only a part of the thermophilic phase and a different trend in emissions may emerge further into the process.

Table 11. *Total GHG emissions based on GWP using a 100-year time horizon in relation to carbon (C) emitted as CO_2 and CH_4 for the individual compost reactor runs presented in Paper IV*

Run	Emissions in g CO_2-eq. kg^{-1} C emitted[1]			
	CO_2	CH_4	N_2O	Total
Run 1, Dry, MC 25-40%	3660	3.30	48.9	52.2
Run 2, Dry, MC 25-40%	3660	5.16	42.0	47.2
Run 3, MC 40-50%	3660	52.3	34.5	86.8
Run 4, MC 40-50%	3660	19.4	26.1	45.5
Run 5, Wet, MC 50-60%	3640	356	14.0	370
Run 6, Wet, MC 60-70%	3370	3590	70.1	3660

In the home composting study, N_2O was more important from the global warming perspective than CH_4 emissions (Table 11). This contradicts findings in other studies showing that CH_4 is more important (Table 9). This difference could be related to the long curing phase in the home composting study, as the compost bins were emptied only once every 1-2 years. The compost reactor experiments generally had low N_2O emissions and included only the initial and the thermophilic phases of the compost process, with the exception of a few longer runs (Paper III). In spite of this, the average degradation of organics was fairly high, with roughly 50% of initial C emitted as CO_2 in the 16% O_2 runs (Table 8). In general, comparison of the N_2O emissions has to be performed with some caution, as the time of composting differed in the different studies.

3 Process conditions affecting emissions

3.1 Effects of compost parameters on methane emissions

3.1.1 Moisture and degradation

Compost moisture content had a strong effect on emissions of CH_4 and N_2O, as was shown in the home composting and laboratory reactor studies. The most profound effect of high moisture on CH_4 emissions was observed in the second set of reactor experiments (Figure 5 in Paper IV), where total CH_4 production increased exponentially with increasing moisture content. The effect of suboptimal, high compost moisture on CH_4 emissions, as discussed above (section 2.1.1), has commonly been attributed to restricted aeration (Maia *et al.*, 2012; Jiang *et al.*, 2011; Agnew & Leonard, 2003; Richard *et al.*, 2002). However, the scale of the moisture effect and the rate of CH_4 emissions increase observed in Paper IV have not been shown previously.

The magnitude of the moisture effect on CH_4 emissions was also affected by the composting temperature. Such a combined effect was observed when comparing the results of the home composting study (Paper I) with those of the second set of reactor experiments (Paper IV). Both studies included composts with a very high moisture content, but the temperatures and the degradation rates differed.

The effect of moisture on CH_4 emissions in combination with degradation rate was also observed in a controlled study of home composts in Denmark reported by Andersen *et al.* (2010a). The waste added to those home composts had a similar range of moisture and addition rate as that of composts discussed in Paper I, but produced more CH_4. One major difference between the Danish home composting study and that reported in Paper I was the degradation rate, which was more complete in the former and was in the range 63-77% of initial carbon (Andersen *et al.*, 2010a). This was much higher than in Paper I, which was estimated to be 18.4% (Table 8). The higher degradation rate in the Danish compost bins did not cause higher temperatures, however, most likely due to the bins lacking insulation, causing larger heat losses to the environment. When comparing these studies on home composting, it should be noted that the one-year study in Paper I most likely allowed less degradation than the 1.6-year Danish study, which had a dedicated maturation period during the last 7.8 months without feed addition.

The effect of degradation rate on CH_4 emissions during composting at high moisture is of relevance. Higher moisture levels, observed in the home composting study and in the second set of reactor experiments, can cause the aeration to become restricted as degradation rates increase, eventually causing oxygen depletion (Richard *et al.*, 2002; Haug, 1993). Such an effect was observed in the second set of compost reactor experiments, where in the wettest reactor run (Paper IV, Run 6) the CH_4:CO_2 ratio was 8.59%, even though O_2 was set to 16% in the reactor. This CH_4:CO_2 ratio was even higher than that during the previous reactor trial with O_2 set to 1%, which yielded a ratio of 0.133% (Paper III). As the initial moisture in Run 6 in Paper IV was 65%, while it was 56% in the 1% O_2 run in Paper III, this suggests that the CH_4 emissions in this moisture range are more affected by high moisture than by limited aeration alone (Table 9).

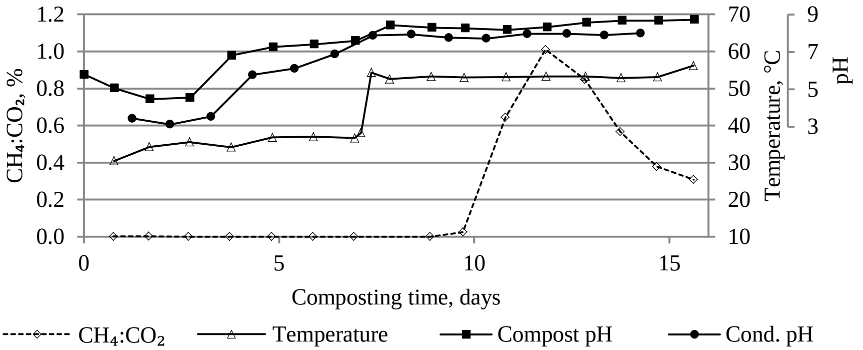

Figure 7. Daily CH_4 emissions presented as CH_4:CO_2 ratios in relation to temperature and material pH in compost reactor run 55°C-1% with limited O_2 conditions (Paper III).

The relatively low decomposition rate, with only 27.5% of initial carbon being mineralised to CO_2, observed during the 1% O_2 reactor run (55°C-1%) in Paper III could explain the lower CH_4:CO_2 than in Papers I, II and IV. The CH_4 production was also much slower until day 10 in that run than in other runs of the second set of reactor experiments (Figure 3 in Paper III). This slow CH_4 production could be due to the slow pH increase, seen up to around day 8, in both condensate and compost (Figure 7). The pH typically increased faster, within 2-5 days, in other compost reactor runs with faster degradation (Papers III-IV).

The high decomposition rate at a similar moisture content, as in reactor runs with average initial moisture of 51 and 60% for the two windrow experiments (Paper II), resulted in an average CH_4:CO_2 ratio of 1.01%, which was higher than in the limited O_2 compost reactor run. The difference in CH_4:CO_2 ratio between the 55°C-1% run (Paper III) and the windrow composting (Paper II) suggests that at higher moisture composting, a high decomposition rate may affect CH_4 emissions even more than the limited aeration alone. Continued CH_4 production, however, could be expected in both the 55°C-1% run and the windrow experiments, as the CH_4 production was sustained in the other, longer runs (Papers III, IV) after the initial CH_4 peak had occurred. The continued production may change the total CH_4 emissions proportions and thus the interpretation of the effects of limited aeration and degradation rates during composting at high moisture contents.

3.1.2 Compost temperature and pH

Production of CH_4 occurs in anaerobic sites in the compost matrix. Methanogens responsible for CH_4 production can be active within certain temperature and pH ranges (Amlinger *et al.*, 2008; Beck-Friis *et al.*, 2003). Methanogens active in mesophilic temperatures are known to be inhibited by acidic pH (Taconi *et al.*, 2008). Such conditions with mesophilic temperatures and low pH existed in the beginning of the compost reactor runs (Papers III and IV) and in this period slow CH_4 production was generally observed.

The first major peak in CH_4 during both sets of reactor experiments (Papers III and IV) usually appeared once the pH had increased above 6-7 units after the initial pH drop. In most of the reactor runs, however, the temperature was allowed to increase simultaneously to thermophilic when the pH rose above 6 in the condensate, which could have favoured the thermophilic methanogens. This made it difficult to separate the effects of temperature and pH on the CH_4 production increase during both sets of reactor experiments. In the 40 °C run of the first set of reactor experiments (Paper III), however, the CH_4 production increase occurred much later than the pH increase, while the temperature was maintained in the mesophilic range. This indicates that in thermophilic compost runs of both sets of reactor experiments, it was not only the pH increase, but also the temperature shift from mesophilic to thermophilic conditions, that stimulated the CH_4 emissions. Thermophilic methanogens, adapted to the high temperatures prevailing during composting, were most likely inoculated with the finished compost added to feedstock mixture or directly from the environment (Thummes *et al.*, 2007), and became active when the temperature reached the thermophilic phase.

When the temperature increased and reached the upper thermophilic range in the 67 °C runs, the CH_4 peak did not appear (Figure 2 in Paper III), most likely due to high temperature inhibition of methanogens (Ho *et al.*, 2014). While the overall degradation rate of the compost material was similar (Table 3 in Paper III), the total CH_4 production in these 67 °C runs was the lowest of all runs in the first set of reactor experiments investigating temperature effects (Figure 3 in Paper III), giving the lowest CH_4:CO_2 ratios for the total emissions.

The significant influence of temperature on CH_4 emissions was shown in the home composting study (Table 4 in Paper I). Relatively low CH_4 emissions were observed, most likely due to the low temperature of the process, despite the high moisture and low content of structural material (Paper I). This could be explained by tolerance of aerobic methanotrophs to submesophilic temperatures and probably inhibition of methanogenic archea by such temperatures (Chaban *et al.*, 2006). This might be part of the reason behind the CH_4:CO_2 ratio decreasing with temperature in seldom mixed composts with undisturbed growth of methanotrophs on the compost surface.

Other factors such as better aeration in home composts due to the smaller distance for oxygen to diffuse inside the compost material and overall slower degradation might also have contributed to lower CH_4 emissions. However, compared with the home composts, the 40 °C run in the first set of reactor experiments (Paper III) produced high CH_4 emissions, in similar conditions to home composting except for mixing frequency and aeration strategy. Such emissions could have occurred in the home composts investigated by Andersen *et al.* (2010a), where degradation was more in line with that observed in the compost reactor run with mesophilic temperature (40 °C run) in the first set of reactor experiments (Paper III), while in our home composting study it was much lower (Paper I). The combination of high moisture, rising temperature and microbial decomposition rate increases the demand for oxygen (Liang *et al.*, 2003). Thus, these factors in combination with reduced aeration due to water-filled pores at higher moisture levels can increase the CH_4 emissions.

3.1.3 Compost management

Feedstock properties are important for the outcome of the composting process. In the home composting study, the added waste material was not controlled and although on average it represented a mixture of food waste, garden waste and structural amendments in wet weight proportions 20:3:1, the materials and proportions added to individual compost bins varied considerably (Fig. 1 in

Paper I). Moreover, the lack of garden waste in the winter months and the higher amounts in autumn resulted in the mixture varying considerably over the year (Fig. 1 in Paper I). The small size of the compost bins caused material accumulation during the winter, due to low compost temperature and suspended degradation. This material accumulated during the winter rapidly decomposed as the ambient temperature increased, seen as a lowering of the CO_2 emissions during the winter and a sharp increase in the spring (Fig. 4 in Paper I). To minimise the heterogeneity and keep the feedstock composition constant, the waste used in the compost reactors was adjusted to resemble the mix used at large scale and was carefully minced and mixed. In large-scale composting, the material mix proportions are usually fairly similar throughout the year.

The limited amount of garden waste added to home composts was expected to provide poor structure and cause increased CH_4 emissions due to poor aeration, but such an effect was not observed. The CH_4 emissions were in the lower part of the range compared with other large-scale and small-scale home composting systems (Table 6 in Paper I). Similarly, regular mixing of the home composts was expected to cause a reduction in CH_4 emissions due to oxygenation, but the opposite effect was observed (Table 4 in Paper I).

Mixing or turning has been shown to reduce (Hrad *et al.*, 2014; Szanto *et al.*, 2007; Beck-Friis *et al.*, 2000) or increase (Ahn *et al.*, 2011; Andersen *et al.*, 2010a) CH_4 emissions from different composting systems, with scale being one deciding factor. In the home compost study in Paper I, increased mixing frequency significantly increased the CH_4 emissions (Table 4 in Paper I). In this and other studies (Ahn *et al.*, 2011; Andersen *et al.*, 2010a), a tendency for mixing of small-scale composts to cause an increase in CH_4 emissions was observed. The increased CH_4 release could also be due to methane-oxidising bacteria residing in the surface layers of the compost matrix being mixed into the deeper layers, disrupting the CH_4 oxidising capacity (Szanto *et al.*, 2007; Jäckel *et al.*, 2005; Beck-Friis *et al.*, 2000). Thus, the CH_4 produced would not be oxidised and CH_4 release would continue until a new methanotrophic population built up at the compost surface.

Another plausible explanation is that CH_4 previously trapped inside the compost matrix is released during mixing and leaks out thereafter due to the creation of new diffusion routes. In the home composting study by Andersen *et al.* (2010a), an additional 8-12% CH_4 was emitted through instant release during compost mixing, compared with the CH_4 emissions during the non-mixing periods of that study. In the home composting study (Paper I), the instant release was not measured. Thus it can be speculated that the observed emissions were lower than the potential emissions had the instantly released CH_4 been included for those bins that were mixed.

After turning, the degradable fractions of the organic matter in the compost matrix become more exposed to microorganisms, causing a degradation peak observed as a peak in CO_2 emissions, *e.g.* as seen in the reactor experiments (Figure 8). Such activity peaks are usually accompanied by a quick increase in temperature and oxygen demand (Szanto *et al.*, 2007; Hao *et al.*, 2001), possibly causing an increase in CH_4 production. Compost material formed aggregates late in the runs in the reactor experiments, which may have contributed to CH_4 emissions due to formation of anaerobic zones inside these aggregates. The formation of aggregates could be related to turning of the reactor and compost matrix degradation. Such degradation is the result of decomposition of lignin and hemicellulose, which is normally observed during composting (Eklind *et al.*, 2007; Tuomela *et al.*, 2000).

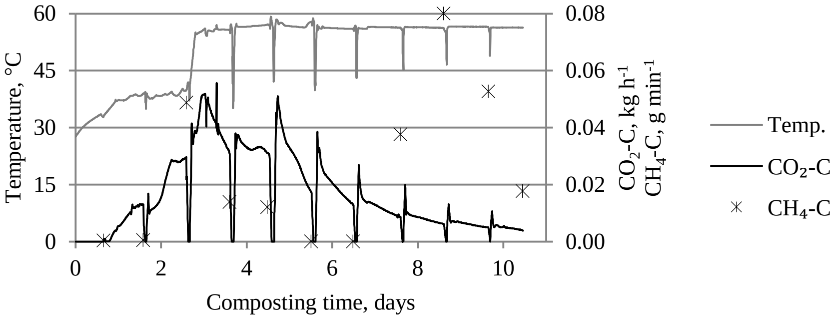

Figure 8. Example of temperature development and emissions of CO_2 and CH_4 during compost run 55°C-a in the first set of reactor experiments (Paper III). Dips in temperature and CO_2 emissions, followed by peaks, represent turning events.

In the home composting study (Paper I), although the regression analysis produced significant models (Tables 4 and 5 in Paper I), these results have to be treated with some caution. The interpretation was complex because during several samplings in many of the compost bins, the CH_4:CO_2 ratio was very low, meaning that the regression analysis results were driven by the few high-emitting composts (Figure 9). Later investigations (Papers III and IV) demonstrated that the effects of process conditions such as moisture content, temperature, mixing frequency and waste addition rate, which were all identified as affecting the CH_4 emissions in the home composting study (Tables 4 and 5 in Paper I) also affected emissions in the reactor experiments and in systems investigated by others (Ahn *et al.*, 2011; Andersen *et al.*, 2010a).

In large-scale systems, a different effect of turning can be expected. Although the effects of mixing small-scale composts also apply to larger systems, as shown by Ahn *et al.* (2011), it can be expected that in large-scale composting the anaerobic zones are much larger and produce considerably more CH_4 (Andersen *et al.*, 2010b). In large systems, a turning event is likely to produce an instant release of CH_4 (Hrad *et al.*, 2014), in the same way as discussed for home composts above. However, that turning event is also likely to cause anaerobic parts of large-scale composts to break up and become exposed to oxygen, which dramatically reduces the methanogenic activity (Hao *et al.*, 2001). Thus for large-scale composts, which are expected to contain extensive anaerobic zones, the positive effects of turning on reduction of CH_4 emissions may outweigh its negative effects, while the opposite seems to be true for small-scale home composts.

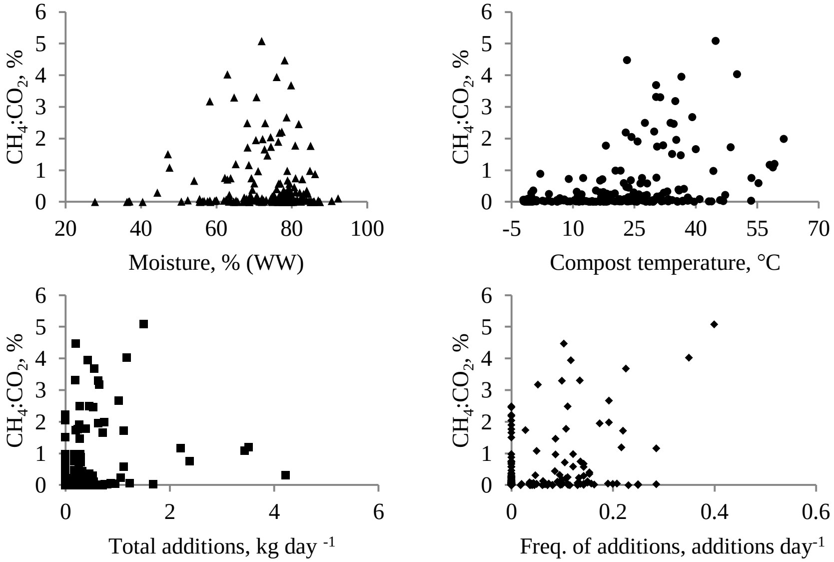

Figure 9. Effects of moisture (▲), temperature in compost (●), total waste addition amounts (■) and mixing frequency (♦) on CH_4:CO_2 emissions ratio for home composts (Paper I). Data points represent values from all compost bins and samplings (n=234).

3.2 Effects of compost parameters on nitrous oxide emissions

3.2.1 Moisture

The N_2O emissions increased during home composting in Paper I, with a combination of mesophilic temperatures and high moisture as the likely cause of these increased emissions. Such conditions presumably stimulated the denitrification process, which combined with reduced aeration resulted in N_2O production (Angnes *et al.*, 2013; Hwang & Hanaki, 2000). The later compost reactor experiments (Papers III, IV) suggested two distinct sources of N_2O; first, an early emissions peak less affected by moisture and more by the combination of NO_3^- and low pH in the substrate. This combination promoted denitrification of the NO_3^- (discussed in section 2.4), which was most likely incomplete due to inhibition by low pH, as discussed in section 2.1.1. The second source of more continuous N_2O emissions occurred during later stages, *i.e.* during the high activity and curing phases. These emissions decreased significantly with increasing moisture content (Paper IV), probably due to complete denitrification at higher moisture levels as discussed in section 2.1.1.

The N_2O emissions dynamics from home composting fitted the suggested explanation on the distinct sources of N_2O provided above. Low NH_3 emissions, below 0.1 ppm in 80% of the measurements in the home composting study, may be explained by the relatively low temperature, mostly below 40 °C (Eklind *et al.*, 2007). Since the average pH in the home composts was 7.2, the low NH_3 emissions suggest that most NH_4^+ either remained in the material or was nitrified to NO_3^-. High moisture content in home composts should allow the NO_3^- produced in nitrification to be denitrified completely to N_2 (de Guardia *et al.*, 2010b). Indeed, the majority of the composts had very high moisture levels and also low N_2O emissions, as shown by the overall median N_2O:CO_2 ratio of 0.067%, *i.e.* less than half the average value of all bins (Table 6 in Paper I). The median value was generally smaller than that reported for other composting systems (Chan *et al.*, 2011; Hermann *et al.*, 2011; Andersen *et al.*, 2010a; Amlinger *et al.*, 2008). During several samplings in some of the home composts, however, the observed high emissions rates were most likely due to recent addition of fresh waste or turning events, which have been reported to stimulate N_2O production (Tsutsui *et al.*, 2013; de Guardia *et al.*, 2010b).

In the compost reactor studies, the N_2O emission patterns were different in two of the runs: the limited aeration run 55°C-1% in Paper III (Figure 3 in Paper III) and the high moisture compost Run 6 in Paper IV (Figure 7 in Paper IV). Between them, these two runs initially showed similarities in N_2O emissions dynamics. Early emissions were high in both cases and probably originated from denitrification, as indicated by the presence of NO_3^- in feedstock and its subsequent concentration decrease (Figure 5 in Paper III; Figure 8 in Paper IV). The denitrification was most likely supported by the high rate of microbial degradation consuming the oxygen (Maeda *et al.*, 2011; Kim *et al.*, 2010). Additional NO_3^- could also have been produced from nitrification of mineralised NH_3. This nitrification could contribute to N_2O emissions (Habart *et al.*, 2010). Later in the process, the N_2O emissions dynamics differed between the two runs (55°C-1% and Run 6).

In the low oxygen run 55°C-1%, N_2O production ceased (Figure 3 in Paper III), most likely due to complete denitrification to N_2 as aeration was restricted. No NH_4^+ or NO_3^- accumulated in the material (Figure 5 in Paper III), which supported this assumption. The late N_2O emissions dynamics in Run 6 were similar to those in other runs in the second set of reactor experiments, suggesting that nitrification continued after the initial N_2O peak, but less actively than in other runs, as evident from accumulation of NH_4^+ in the material (Figure 8 in Paper IV). The lower nitrification rate could be attributed to moisture restricting access by oxygen to the nitrification sites. The overall lower N_2O emissions in Run 6 after the pH and temperature increase compared with the other runs could be due to reduced nitrification rates producing less NO_3^- for denitrification. More complete denitrification leading to N_2 instead of N_2O could also be expected (Angnes *et al.*, 2013; Hwang & Hanaki, 2000) due to the high moisture content in this run.

3.2.2 Composting temperature

Composting under thermophilic temperatures stimulates emissions of NH_3 (Beck-Friis *et al.*, 2003). Increased NH_3 emissions have been discussed as a possibility to reduce the emissions of N_2O from composting through limiting nitrification (Pardo *et al.*, 2015). In large-scale systems, NH_3 can be removed from the compost exhaust gas by *e.g.* acid scrubbing (Webb *et al.*, 2012; Pagans *et al.*, 2007). However, such N losses reduce the fertiliser value of the finished compost. In addition, pollution swapping from N_2O to NH_3 may cause other problems, such as increased N_2O emissions from other parts of the system, *e.g.* the biofilter (Maia *et al.*, 2012), or from the environment. Moreover, if the compost gas is not filtered at all, the NH_3 emitted to the environment causes acidification and eutrophication (IPCC, 2014).

The effects of temperature and aeration on N_2O emissions have been investigated previously (Jarvis *et al.*, 2009; Amlinger *et al.*, 2008; Szanto *et al.*, 2007; Beck-Friis *et al.*, 2001). Mesophilic temperatures are generally discussed as promoting N_2O emissions and thermophilic temperatures as inhibiting them. Nevertheless, due to the complexity of N_2O emissions occurring from multiple processes, there is no universal solution to mitigation of N_2O emissions during composting.

Emissions of N_2O were generally low during the well-controlled compost runs in Paper III (Table 9), which were performed at temperatures ranging from 40 °C and to 67 °C. No clear effect of temperature or aeration on emissions was observed (Figure 3 in Paper III). However, one clear finding in Paper III contradicted previous claims that N_2O emissions are low under thermophilic conditions (Amlinger *et al.*, 2008). In the reactor runs at 67 °C, the total N_2O emissions were similar to those in the run at 40 °C (Table 3 in Paper III).

Home compost temperature was one of the deciding factors for the release of CO_2 and CH_4, which resulted in CH_4:CO_2 ratios which varied seasonally (Figs. 4 and 5 in Paper I). The emissions (N_2O and N_2O:CO_2 ratios) in Paper I were not affected by temperature in the same way, which could be explained by the complexity of the factors leading to N_2O emissions. Such complexity was apparent in the windrow study and the first set of reactor experiments (Papers II, III), but has also been reported in studies by others (Jarvis *et al.*, 2009; Amlinger *et al.*, 2008). The complexity of N_2O emissions, with several possible processes generating N_2O, was most likely the reason why no significant regression equation was found in Paper I and why it was difficult to interpret the influence of temperature on N_2O emissions dynamics in Papers II and III.

3.2.3 Compost management

Emissions data from the two sets of reactor experiments (Papers III and IV) showed no significant correlation between N_2O emissions and initial compost NO_3^- concentration. It was clear, however, that the early N_2O emission peaks were lower when the NO_3^- concentration was low (Figure 10a, group A), while at higher levels of NO_3^- other factors caused the peak emissions to range from low (Figure 10a, group C) to high (Figure 10a, group B). The effect of NO_3^- concentration on N_2O:CO_2 ratio was also tested, but it produced similar results to those shown in Figure 10a. This lack of significant regression suggested that N_2O emission processes were most likely dependent on variables other than the initial content of mineral nitrogen.

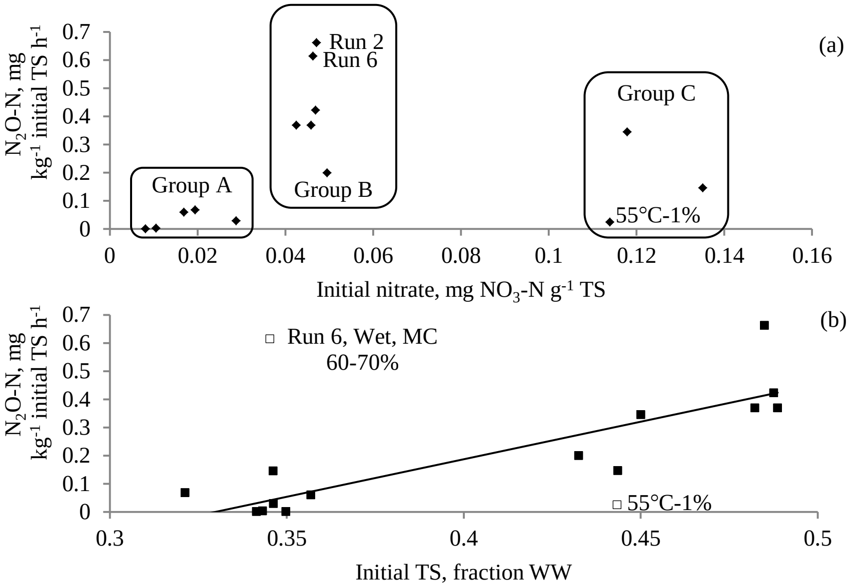

Figure 10. Relationship between the early N_2O emission peaks occurring in the period from start until the pH change in compost reactor runs and the initial NO_3^- concentration (a) or the initial fraction of total solids (b). Groups A and C represent runs in Paper III with low and high initial NO_3^-, respectively; Group B represents runs in Paper IV. Outliers (□) were excluded from regression analysis.

The effect of moisture content on emissions was tested using regression analysis in the same way as above (Figure 10b). The results showed a significant correlation ($P<0.001$) with the increase in early N_2O emissions (Equation 4). A similar finding was also made in Paper IV (Equations 2 and 3). The analysis suggested that the same relationship applies for the majority of reactor runs in both sets of compost reactor experiments (except for the outliers Run 6 and the 55°C-1% run). Lower initial moisture caused higher N_2O emissions during the early (mesophilic) phase of composting, except in the outliers.

$$N_2O = -0.88 + 2.67 \cdot TS \qquad \text{Equation 4}$$

where N_2O is the peak emissions in mg N_2O-N kg^{-1} initial TS h^{-1} and TS is total solids fraction of the total wet weight.

In the windrow experiments (Paper II), the total N_2O emissions were generally low, which was most likely due to the short period of composting preventing observation of any curing phase. The early N_2O emissions observed in the reactor experiments were possibly absent due to low concentration of NO_3^- in the initial waste mix. The NO_3^- concentration was not measured in that study, however, so this hypothesis could not be tested. Both dry and wet zones existed in the compost matrix in the windrow study (Table 2 in Paper II). Applying knowledge from the second set of reactor experiments (Paper IV), it can be hypothesised that the dry zones were mainly responsible for the N_2O emissions, while the CH_4 emissions mainly occurred in the wet zones.

The development of increasing gradients of moisture over composting time (Table 2 in Paper II) was the result of the aeration method in the windrow study. The progressive gradients allowed increasing proportions of dry and wet zones to co-exist in the windrows, which was most likely the cause of the observed gradual increase in both CH_4 and N_2O emissions (Figures 5 and 6). Continued composting of the windrows could have been expected to increase this gradient effect further, as the windrows were not mixed.

The home compost bins that were never mixed displayed a range of N_2O emissions (Figure 11), while the compost bins that were mixed frequently displayed lower N_2O emissions. However, some high emissions peaks were noted in the compost bins with average mixing, while no such peaks were seen in the bins that were mixed most frequently. There was probably a difference in the way the owners mixed the bins, but this information was not collected. Mixing all the material in a pile could be expected to cause an increase in N_2O emissions due to the transfer of NO_3^- produced by nitrification into the interior part of the compost with more anaerobic sites. In these active decomposition sites, the newly available NO_3^- could be taken up by the denitrifiers, possibly leading to an increase in N_2O production and release (Ahn *et al.*, 2011; Hao *et al.*, 2001).

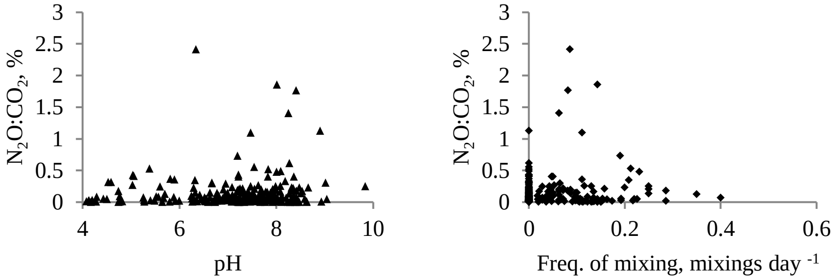

Figure 11. Effects of pH (▲) and mixing frequency (♦) on N_2O:CO_2 emissions ratio from home composts (Paper I). Data points represent values from all compost bins and samplings (n=234).

The addition of fresh waste to an on-going compost process can provide conditions similar to those in the early stages of a newly started composting batch, as observed in the second set of reactor trials (Paper IV). In addition to the effects of turning, as discussed above, and addition of fresh waste, it is also likely that different stages of maturation existed in the compost bins, especially in those with larger volumes of material, and the emissions expected during curing phase could also appear.

4 Global warming potential of composting

4.1 Emissions of nitrous oxide and methane

During composting, different gases are emitted to the atmosphere, primarily H_2O, CO_2, NH_3, CH_4, VOCs and N_2O (Amlinger *et al.*, 2008; Beck-Friis *et al.*, 2003). Excluding H_2O, the major gas emitted during aerobic composting is CO_2, while the proportions of other gases can vary depending on process conditions and process stage, as well as compost mixture properties. The studies presented in the thesis mostly concentrated on emissions of CH_4 and N_2O because of their high GWP, 34-fold and 298-fold more potent than CO_2 over a 100-year horizon, respectively (IPCC, 2013). The emissions of CO_2, which is also a GHG, were presented and discussed in the different studies, but these emissions were considered biogenic and thus not contributing to global warming, as discussed in section 1.2.

Identification of the composting process conditions that mitigate emissions of CH_4 and N_2O was one of the main objectives of the thesis. While the studies performed in Papers I-IV demonstrated that controlling key process parameters such as temperature, moisture and aeration can dramatically reduce the emissions, other studies have investigated how some of these and other parameters affect the emissions (Jiang *et al.*, 2011; Maeda *et al.*, 2011; Zhang *et al.*, 2011b; Amlinger *et al.*, 2008; Eklind *et al.*, 2007; Beck-Friis *et al.*, 2003; Hellmann *et al.*, 1997).

Composting facilities apply different measures to reduce the direct GHG emissions from the composting process and to treat the gases leaving the compost. One difficult parameter to control is the pH, which is often low in incoming food waste due to initial fermentation (Sundberg *et al.*, 2013). Low pH was discussed as one of the causes of increased N_2O emissions during denitrification in Papers III and IV. Temperature and moisture are also parameters that are difficult to control in large-scale composting. Although large-scale facilities can usually control aeration rate and moisture addition, such control is often insufficient to maintain the desired temperature and moisture levels (Sundberg & Jönsson, 2008).

An advantage for central composting facilities in comparison with small-scale composting (*e.g.* home composting) is the possibility to treat the exhaust air from the whole facility. In composting facilities with central ventilation or forced aeration, the exhaust from composting processes can be treated by a biofilter for removing CH_4 (Wilshusen *et al.*, 2004). In such cases, an overall reduction in GWP of 25% can be achieved (Bernstad & Jansen, 2012). On the other hand, the use of biofilters without initially removing NH_3 *e.g.* through NH_3 scrubbing (Busca & Pistarino, 2003), can potentially cause additional N_2O emissions from ammonia oxidation or subsequent nitrification and denitrification (Maia *et al.*, 2012).

4.2 Fate of carbon during and after composting

Waste mixtures intended for composting contain different types of organic molecules, some of which are more easily degradable by microorganisms than others. Compounds with low degradability, such as lignin, cellulose and hemicellulose, degrade slowly or undergo humification (Tuomela *et al.*, 2000) and to a certain extent remain in the finished compost in the form of recalcitrant carbon (Gajalakshmi & Abbasi, 2008). When the finished compost is used as a fertiliser or soil improver, the sequestered carbon can be considered a long-term carbon sink. In general, compost carbon retention in soil depends on the lignin content in the waste and can range from 2 to 16% of the carbon in the applied compost over a 100-year time frame (Hansen *et al.*, 2006).

High temperatures promote lignin and cellulose decomposition during the compost process in alkaline pH conditions (Tuomela *et al.*, 2000). Extended composting and storage further degrade the lignin and reduce the amount of carbon that could potentially be sequestered in soil (Eklind & Kirchmann, 2000a).

In the home composting study (Paper I), the composts were emptied once a year or even less frequently. The finished compost was applied locally in the gardens of the household owners and, according to the interview results (data not shown), partly replaced the otherwise commonly used commercial peat-based soil improver. Based on the assumption that 1 ton compost replaced 285 kg peat using 1:1 volume proportion (Boldrin *et al.*, 2010) and on a survey of compost users, the finished compost product from a central composting facility was estimated to replace 58 kg of peat per ton of compost (Andersen *et al.*, 2010c). An even higher substitution rate can be expected in home composting, where essentially all compost produced will substitute for peat. Peat has an estimated GWP of 970 kg CO_2-eq. per ton of peat used from the whole peat life cycle within 100 years (Boldrin *et al.*, 2009).

During continued composting in the curing phase and lengthy storage of the finished compost, direct emissions of N_2O from denitrification and nitrification can persist and CH_4 emissions can be enhanced by reduced airflow due to compaction caused by decomposition of structural amendment (Hao, 2007). While these emissions are reported to be smaller than those occurring during the active phase of composting (Zeng *et al.*, 2012; Hao, 2007; Cabanas-Vargas & Stentiford, 2006), they can increase considerably if the maturing compost becomes anaerobic or is still rich in nitrogen. When the finished compost is stored before use, additional emissions of both CH_4 and N_2O can be expected, as indicated in different studies (Rodhe *et al.*, 2015b; Clemens *et al.*, 2006). However, these emissions represent only a part of the total GHG emissions from the whole life cycle of waste in a treatment system, including composting (Bernstad & Jansen, 2012), provided that the storage period is not too long. Avoiding a long curing phase and long storage of finished compost can thus lead to a decrease in total direct GHG emissions from composting.

4.3 Biological waste treatment systems

Composting is a robust and relatively simple waste treatment method that does not require comprehensive infrastructure and allows nutrient recycling. Such benefits of composting are especially relevant for developing countries when selecting a waste treatment system (Sundberg & Navia, 2014) and for small-scale systems. There is commonly no energy recovery from composting. If heat is recovered, the facility requires additional infrastructure (Rodhe *et al.*, 2015a). Globally, composting is one of the dominant biological waste treatment alternatives, and is considered a significant improvement over landfilling or dumping (Laurent *et al.*, 2014a).

The life cycle assessment (LCA) methodology is a tool used for comparing waste management systems that are different in treatment principle and in outcomes and products replaced. Using LCA methodology requires a clear description of the systems investigated and the systems boundaries should include all relevant processes and their impacts (Baumann, 2004). LCA studies can help to identify critical points in waste treatment systems and thus give suggestions for improvements. The analysis of different LCA studies performed by Laurent *et al.* (2014a) concluded that LCAs are commonly carried out in Europe, but less frequently performed in developing countries.

This can be a problem, since the studies need to be adapted to local conditions. The majority of LCA studies of waste management systems reflect the local conditions for which they were designed (Bernstad & Jansen, 2012). As in all LCA studies, the local conditions, *e.g.* local energy supply mix, can significantly affect the outcome of the LCA and favour one system over another. Thus LCA results can be difficult to apply to other regions or settings and great caution should be used when doing this.

Waste treatment methods with energy recovery are favoured in Europe. However, in the meta-analysis of different LCA studies on waste treatment performed by Laurent *et al.* (2014a) on composting, thermal treatment or anaerobic digestion for biological waste, there was no clearly preferred waste management system that generally performed better than the others. Performing a meta-analysis LCA allows multiple different treatment systems in other LCA studies to be compared (Morris *et al.*, 2013). Bernstad and Jansen (2012) reviewed 25 studies comparing different treatment alternatives for food waste and found that the results varied greatly depending on the systems analysed and the system boundaries used. The largest differences were in GWP of the systems. In systems including composting, the total GWP emissions ranged from close to 0 to over 800 kg CO_2/tonne of food waste treated. The total GWP of composting increased when the direct process emissions were included, while in anaerobic digestion systems GWP increased when emissions during and after digestate application to soil were included. Another source of emissions in anaerobic digestion that is sometimes underestimated is emissions of CH_4 from biogas leakage during digestion, upgrading and use (Chiew *et al.*, 2015). Incineration systems are affected by the low calorific value of food waste, which can require support fuel (Chaya & Gheewala, 2007). This can reduce the amount of credited recovered energy and result in significant GHG emissions.

Various studies highlight parts of the waste management system involving composting that need to be accounted for when evaluating the emissions from this treatment method. The direct emissions from the composting process represent an important part of the impacts of the whole system (Morris *et al.*, 2013; Martínez-Blanco *et al.*, 2010). Other relevant contributions of the system include emissions from the waste collection chain and emissions during waste handling, compost preparation and management. The emissions from finished compost storage, distribution and application also have to be considered. In addition, the effects after application on the target system (*e.g.* soil) should be investigated.

When composting is investigated, it is common for different parts of the system not to be included in the system boundaries, which can lead to burden shifting between different parts of the life cycle (Laurent *et al.*, 2014b). For instance, it is sometimes assumed that the compost product does not replace any other product (Eriksson *et al.*, 2015), or that direct emissions from the composting process are not included. Luckily, some LCA studies include a more complete process (Chiew *et al.*, 2015; Boldrin *et al.*, 2009; Moller *et al.*, 2009).

Emissions from waste collection represent a small fraction of total process emissions from the whole system and the savings from collection avoidance during home composting depend on the length of avoided transportation and on how the collection is organised. The impact of transportation can be more relevant for remote areas (Martínez-Blanco *et al.*, 2010) and if *e.g.* specially distributed bags are used for the collection. In general, the impact of waste collection can be excluded from comparisons of different treatment methods if the same type of collection is used and the transportation distances are similar (Khoo *et al.*, 2010), for instance when central composting is compared with other waste treatment systems requiring similar collection, such as anaerobic digestion.

Centrally collected waste intended for biological treatment needs to be sorted at source or at the treatment facility, as the fraction to be treated in composting or anaerobic digestion process has to be mostly organic and without contaminants in order to produce a clean treatment product that can be used as bio-fertiliser (Bernstad & Jansen, 2011). The way in which waste is sorted and stored can have a considerable impact on the environmental performance of a system (Rigamonti *et al.*, 2009). For instance production and use of high quality paper bags in food waste collection intended for anaerobic digestion can give a significant environmental impact (Chiew *et al.*, 2015). Transportation and use of high quality materials or products (*e.g.* paper for waste collection bags) for source sorting is not required in home composting (Martínez-Blanco *et al.*, 2010). Moreover, the better quality of source separation and the lack of contaminants in home composting are sometimes considered in LCA studies (Quiros *et al.*, 2014). These categories should be included when comparing other systems with home composting in order to show the avoided impacts in home composting.

This thesis investigated the direct gaseous emissions from the active phase of composting (Papers I-IV) and in part the curing process (Papers I and III). When expressed per ton initial wet waste (WW), the direct GHG emissions ranged from very small, less than 0.1 CO_2-eq. ton^{-1} initial WW, up to relatively large, over 230 CO_2-eq. ton^{-1} initial WW in the wettest compost reactor run (Table 12). In different LCA studies, direct emissions from composting are reported to be one of the main causes of higher total GWP of composting systems in comparison with other waste treatment systems (Spangberg *et al.*, 2013). For instance in the study by Quiros *et al.* (2014), the direct emissions from home composting ranged between 8.3-37.8 and 53-307 CO_2-eq. ton^{-1} initial WW for CH_4 and N_2O, respectively. The results obtained from practical composting in the studies presented here (Papers I and II) had direct emissions in the lower range (Table 12) of values reported by Quiros *et al.* (2014). The emissions were even lower in the reactor studies, highlighting both the uncertainty when evaluating the possible direct emissions from composting and the possibility of emissions reduction through process optimisation.

Table 12. *Greenhouse gas emissions ranges observed in different compost studies (Papers I-IV). The CO_2-equivalents are calculated based on IPCC GWP values over a 100-year lifetime.*

	CO_2-C, % initial C	Emissions in kg CO_2-eq. ton^{-1} initial WW			
		CO_2	CH_4	N_2O	Total
Home composting (Paper I)[1]	4 - 65	16-266	0.776-12.51	7.37-119	8.14-131
Covered windrows (Paper II)	21	144	17.7	1.19	18.9
Laboratory reactor (Paper III, 16% O_2)	32 - 67	173-297	0-0.982	0.094-2.35	0.094-3.33
Laboratory reactor (Paper III, 1% O_2)	28	188	3.09	0.153	3.24
Laboratory reactor (Paper IV, Run 1-5)	45 - 50	294-368	0.329-28.8	1.13-4.89	1.46-33.7
Laboratory reactor (Paper IV, Run 6)	40	213	227	4.42	231

[1] The estimates for minimal and maximal degradation are based on VS from finished compost samples. VS samples which gave negative degradation were excluded. Average degradation in home composting was 18 CO_2-C, as % initial C and the average total GHG was 33 kg CO_2-eq. ton^{-1} initial WW.

Comparing composting with different waste management systems, *e.g.* in the meta-analysis LCA of 82 studies performed by Morris *et al.* (2013) for the whole systems, anaerobic digestion and composting had similar negative GWP values, which means that through use of these waste management systems, due to avoided environmental burdens, the overall avoided emissions from the system were larger than the emissions produced. For composting and anaerobic digestion the GWP was in the range -740 to -60 kg CO_2-eq. ton^{-1} initial WW for anaerobic digestion, while for composting it was in the range -760 to 220 CO_2-eq. ton^{-1} initial WW. Furthermore, home composting had an impact in the range -690 to 290 kg CO_2 eq. per ton waste, while incineration had an impact of 70 CO_2 eq. per ton waste. Another LCA study on home composting, which unlike the study by Morris *et al.* (2013) considered the benefits of use of compost product, had a GWP impact of -203 and -692 kg CO_2 eq. per ton waste for high-emitting and low-emitting composts, respectively (Quiros *et al.*, 2014). These results suggest that composting is a viable waste treatment alternative that can help mitigate GHG emissions. By using compost to replace peat and mineral fertiliser, reducing the direct emissions from the composting process through adjustment of process conditions and promoting home composting, especially in remote areas, the environmental burdens can be lessened.

5 Perspectives and future research

Home composting was investigated under Swedish climate conditions, which meant that sub-mesophilic temperatures were maintained in most of the compost bins studied. This was probably the cause of the relatively low observed CH_4 emissions from home composting. Composting performed in warmer climates is likely to produce higher emissions and should be investigated in future work.

Composting performed in reactor experiments was set-up to have quick start-up, meaning that the temperature increase to thermophilic range happened at the time of the pH rise. During interpretation of the data obtained, it became evident that both the pH rise and the temperature increase could have influenced CH_4 emissions, as indicated by the 40 °C run having different emissions dynamics compared to other runs. A new set of composting experiments where the change in temperature is not coupled to pH change could help clarify the influence of these process conditions on emissions.

The initial peak in N_2O emissions in many reactor runs was correlated with moisture content and was caused by the presence of nitrate in the initial compost substrate. Additional experiments to test the effects of greater ranges of nitrate concentrations on early N_2O emissions are needed to investigate how to achieve composting with minimal GHG emissions in conditions when it is not possible to minimise the amount of nitrate in the initial substrate.

In windrow composting, the increasing moisture gradients in the material were associated with increasing emissions of both CH_4 and N_2O. Further investigation of the influence of these moisture gradients could establish the mechanism behind this effect and whether compost mixing could reduce it without causing additional N_2O emissions due to supply of nitrate to zones with active denitrification.

Moisture was found to be an important parameter affecting emissions of both CH_4 and N_2O and, except in home composting, CH_4 emissions were the most important GHG in total GWP comparisons. A composting experiment could be set up to test whether GHG emissions can be reduced by dynamic moisture adjustment, allowing low moisture during the active phase when CH_4 emissions can be expected and higher moisture later in the process, to keep emissions of both CH_4 and N_2O low.

In the reactor studies and in home composting, persistent emissions occurred during the ongoing composting and maturation process. Thus, composting and maturation should not be allowed to proceed for longer than necessary. However, long-term maturation and storage are often required and thus more studies should be performed on long-lasting composting processes to investigate the influence of process conditions on emissions during the maturation and curing phase, as relevant information is currently scarce. More studies are also needed to investigate the influence of compost moisture and aeration, and of other parameters such as compost matrix structure, in the curing and maturation phase of composting.

Further investigations of compost produced in home and central composting are needed to establish whether the finished compost is applied to soil, whether it replaces fossil peat and fertiliser and the impacts of such application.

The studies in this thesis mainly concentrated on direct GHG emissions. An investigation using a systems approach would be beneficial to identify the adjustments to process conditions that could produce the best outcomes in terms of reducing the negative impacts of the whole composting system. An LCA study comparing central and home composting with other alternatives available in the region should be performed. A sensitivity analysis in such a study, with more detailed information about the effects of different parameters on GHG emissions from composting, obtained from reactor experiments, could be used to evaluate whether process adjustments to reduce direct GHG emissions have a sizable effect on the performance of the whole system.

6 Conclusions

Emissions of the greenhouse gases (GHG) methane (CH_4) and nitrous oxide (N_2O) can have a considerable environmental impact when composting is performed under inappropriate process conditions. The studies included in this thesis demonstrated that moisture content was a crucial parameter significantly affecting emissions of both CH_4 and N_2O in composting.

The emissions of CH_4 in composting were strongly affected by compost moisture content and aeration. Increasing moisture content from on average 44% to 66% moisture in laboratory compost reactor runs led to 1000-fold higher CH_4 emissions with the total CH_4 emissions increasing from on average 3 to 3590 g CO_2-eq. per kg C degraded, respectively. In compost aerated at low oxygen (1%) concentration, the CH_4 emissions increased 100-fold compared with otherwise similar systems with 16% oxygen concentration. A combination of high degradation rate with mesophilic temperature resulted in considerable CH_4 emissions in compost reactor experiments, while composting at 67 °C produced the least CH_4 emissions. In most compost reactor experiments, there was an initial delay in peak CH_4 emissions until the pH of the compost increased above 6-7. In home composting at sub-mesophilic temperatures, however, in spite of high moisture content at lower temperatures the CH_4 emissions were low, especially when there was no or infrequent turning and when the degradation rate was low.

The emissions of N_2O were also affected by moisture, with decreasing moisture giving increasing emissions. The total N_2O emissions were reduced at higher moisture contents within the range 43-59% in reactor experiments. With considerably less nitrate available in the substrate, the early N_2O emissions occurring prior to the thermophilic phase in compost reactor were considerably smaller. The compost reactor run with a very high average moisture content of 66% produced the highest N_2O emissions of all compost reactor experiments, largely due to the initial peak in N_2O emissions. There was no clear effect of composting temperature in the range 40-67 °C on N_2O emissions. The N_2O emissions from home composting were high.

Of all the systems investigated, home composts had the highest overall GHG emissions when expressed as g CO_2-eq. per kg C degraded. However, the average emissions were affected by a few high-emission events and the overall median emission values were considerably smaller, demonstrating potential for reduction of direct emissions from this system.

The investigated large-scale windrow composting was characterised by thermophilic temperatures, which occurred in combination with large moisture gradients, meaning that the windrows simultaneously contained different volumes of compost with high and low moisture content. High moisture and thermophilic conditions were shown to cause high CH_4 emissions in the reactor study. Thermophilic temperature and low moisture were shown to provoke an increase in N_2O emissions in the reactor studies. Thus, large moisture gradient in the compost seems to be a risk factor for high GHG emissions.

Extended periods of composting resulted in higher total GHG emissions during some reactor runs, due to continued production of both CH_4 and N_2O late in the process, even after 50% of initial carbon had been mineralised.

Total direct GHG emissions from the practical composting systems studied were in the lower range compared with those reported by others. The total GHG emissions from home composting were 1770 g CO_2-eq. per kg C degraded mostly due to N_2O emissions, while from windrow composting the total emissions were 482 g CO_2-eq. per kg C degraded mostly due to CH_4 emissions. The emissions were even lower in many reactor experiments, highlighting both the uncertainty when evaluating the possible direct emissions from composting and the possibility of emissions reduction through process optimisation. Moreover, the use of finished compost has potential to further reduce the total global warming potential of composting, as carbon is sequestered and soil fertility increased.

The results in this thesis indicate that in order to maintain low greenhouse gas emissions from food and garden waste composting, it is important to minimise the amount of nitrate in the initial compost substrate, to avoid having too high a moisture content during the active composting thermophilic phase and to prevent the compost drying out at the end of active composting phase and during maturation, unnecessarily prolonging the composting and maturation period. It is also important that the compost is used as a soil improver and to replace mineral fertiliser.

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