

# Chapter 2

## Greenhouse Gas from Organic Waste

### Composting: Emissions and Measurement

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**Abstract** There is actually common consensus to use biological technologies for the treatment of organic wastes. For instance composting involving the aerobic biological stabilization of organic wastes is gaining popularity. The amount of materials and the variety of wastes composted is increasing fast. However

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composting is a process emitting gases some of which being greenhouse gases (GHG) that favour global warming. In particular carbon dioxide ( $\text{CO}_2$ ), methane ( $\text{CH}_4$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ) are responsible for the global warming potential of composting. A part of these gases can be abated by low-cost biological technologies such as biofiltration. This review compiles all the points related to the emission of GHG from composting processes, from detection and measurement to minimization and abatement. We focus on measurements of GHG to obtain reliable emission factors for designing composting technologies. This will help to compare waste treatment options based on integrated tools such as Life Cycle Assessment (LCA). A chapter discusses C and N dynamics in the compost, and implications on emitted C and N gases. Finally we review the best available practices to minimize the GHG emissions from composting. We also present the final treatment of composting gases.

**Keywords** Composting · Anaerobic digestion · Greenhouse gas (GHG) · Environmental impact · Life cycle assessment (LCA) · Carbon dioxide · Methane · Nitrous oxide · Volatile organic compounds (VOCs) · Biofiltration

## 2.1 Introduction

The sustainable use of resources and wastes, including waste minimisation and valorisation, is a common objective of the plans, directives and rules published in the last few decades. One example is the Sixth Programme of Community Action in the field of Environment (“Environment 2010: the future is in our hands”) published by the European Union for the period 2001–2012 (European Union 2008a). The Sixth Program of Action includes the implementation of seven thematic strategies and, among them, specifically waste prevention and recycling, with the objective to reduce the negative environmental impacts during the whole life cycle of wastes, from their production to their elimination, including their recycling. One of the results of all these legislation efforts was the publication of the Waste Framework Directive in 2008. This Directive considers waste not only as a potential source of pollution, but also as a resource that can be used. Specifically, in the case of biodegradable wastes, the Directive 1999/31/CE on landfilling of wastes encourages the diversion of these wastes to other treatment technologies involving the recycling and energy recovery from wastes, where composting will have a great importance (European Union 2008b; Commission of the European Communities 2008). Nevertheless, the environmental impact assessment during the whole life cycle of wastes lacks of data obtained directly at full-scale waste treatment facilities operating in different locations, thus limiting the quality and reliability of these analyses necessary for the decision-making process.

A direct consequence of the above mentioned plans and directives has been the proliferation of a large number of new waste treatment plants installed in Europe

and all over the world in the last years, as well as the modification and adaptation of the existing ones. In particular, composting and anaerobic digestion are the more widely accepted processes for organic waste treatment. Composting plants are typically operated either in piles or tunnels, whereas anaerobic digestion can take place either in wet or dry digesters, typically followed by composting of the digested sludge with the aim of ensuring its sanitation and stabilization (Ponsá et al. 2008). The anaerobic decomposition process that is carried out in anaerobic digestion facilities allows energy recovery from wastes in the form of biogas. All these treatments also allow the valorisation of wastes by their use in agriculture or as soil organic amendments.

Waste treatment facilities can be the origin of public complaints, most of them associated to annoyances caused by odour emissions generated during the process. Biological treatment plants are a clear example of this problem. Odours generated from this type of treatment plants are mainly associated to the emissions of volatile organic compounds (terpenes, alcohols, ketones, sulphur compounds, amines, etc.) and ammonia (Goldstein 2002; Komilis et al. 2004). Some of the annoyances caused by these emissions are often magnified because of the lack of real data from operating plants that would contribute to have an objective and scientific base to analyse these problems. Such lack of data also represents a problem for the design of mitigation measures such as the use of biofilters. In addition to this, greenhouse gases (GHG) emission inventories evidence the increase in the amount of these compounds that are emitted from waste treatment facilities. Emission of  $\text{CO}_2$ ,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  are the main responsible of this increase (Colón et al. 2012).

Emissions generated in waste treatment plants, in particular those based on biological treatments, are related to the type of technology, the type of wastes treated and the operational conditions of the plant. For this reason, it is very important to relate the emissions to the performance of the biological treatment plants and also to the wastes being treated, since each treatment technology and waste will give rise to different end products quality and organic matter stabilisation degrees. The use of respirometric indices to monitor the stability of the organic matter has been one of the main research topics in the last years (Barrena et al. 2005; Barrena et al. 2006; Barrena et al. 2009a; Barrena et al. 2009b; Ponsá et al. 2008).

Although ammonia is not considered a GHG, its emissions during composting are usually studied because it causes acid rain and from the point of view of the conservation of nitrogen in the end-product because of the potential use of compost in agriculture as organic fertiliser, as well as for the determination of the efficiency of the systems for gas emission treatment, such as scrubbers and biofilters. Ammonia emissions are affected by the C/N ratio of the initial composting mixture, by the temperature reached during the process and by the aeration (Pagans et al. 2006b; Raviv et al. 2002; Sánchez-Monedero et al. 2001). Biofilters have shown to be an efficient equipment for the reduction of ammonia emissions in enclosed waste treatment plants (Hong and Park 2004; Pagans et al. 2006b), although for long periods ammonia tends to reduce the efficiency of this technology (Baquerizo et al. 2005).

An important part of the published literature in the field of gaseous emissions is related to odours, mainly by means of dynamic olfactometry, in both composting plants and mechanical-biological treatment plants (MBT). As already mentioned, a number of laboratory-scale experiments have been performed with the aim of determining the compounds that more significantly contribute to odour pollution. Thus, Goldstein (2002) identified terpenes, alcohols, aldehydes, fatty acids, ammonia and a range of sulphur compounds as the main responsible of odour emissions at composting plants. Other authors have studied the effect of some operational conditions, such as ventilation and turning, in these emissions (Szanto et al. 2007). Gage (2003) proposed a number of managing practices aimed at reducing the annoyances generated by odour emissions; for instance, preparation of an optimal initial mixture and the maintenance of high levels of porosity to assure aerobic conditions in the pile (Ruggieri et al. 2009). Enclosing the composting operations and the use of biofilters are among the main mitigation strategies for both odours and GHG.

The importance of GHG emissions generated during the biological treatment of wastes has been also stated by several authors. CO<sub>2</sub> emissions coming from biological process are not considered to contribute to global warming since this carbon has a biogenic origin, i.e., this carbon has been previously fixed biologically. Regarding other gases, He et al. (2001) measured the emissions of N<sub>2</sub>O and CH<sub>4</sub> during the composting of food wastes under laboratory conditions in a closed system with forced aeration. Although generated in small amounts, N<sub>2</sub>O and CH<sub>4</sub> have a great contribution to global warming since they have a warming potential 25 (CH<sub>4</sub>) and 296 (N<sub>2</sub>O) times higher than that of CO<sub>2</sub>.

There are some scientific publications that provide gaseous emissions data generated during the biological treatment of organic wastes, mainly for manures and sewage sludge. However, the number of published papers dealing with municipal solid wastes is scarce (Colón et al. 2012). The works carried out by Eitzer (1995) and Staley et al. (2006) are very important for the characterization of the emissions generated during the biological treatment of wastes and the identification of specific compounds. In 1995, Eitzer performed a comprehensive characterisation of the volatile organic compounds (VOC) generated in composting plants treating municipal solid wastes and its possible relation to the process performance. On the other hand, Staley et al. (2006) studied the VOC emissions originated during the aerobic treatment of wastes and also during the anaerobic biodegradation process. These works highlighted the importance that forced aeration, used in the biological processes, had on the total emissions (Delgado-Rodríguez et al. 2012). Terpenes and ketones are shown to be the most abundant compounds. These experimental works were performed under laboratory conditions, which would limit their extrapolation to full-scale plants. Pagans et al. (2006a) also evaluated the effect of the type of waste (industrial and municipal solid waste) on VOC emissions under laboratory conditions. Komilis et al. (2004) identified the main VOC emitted during composting of pruning residues (mainly terpenes, alkyl benzenes, ketones and

alkanes) and also during composting of food wastes (sulphides, organic acids and alcohols), as well as the stages of the process that generated the highest emissions (thermophilic phase).

The determination of emission factors for different wastes and different treatment technologies will be a useful tool for the calculation of global emissions at facilities operating with a technology already studied in other treatment plants. Emission factors for VOC, NH<sub>3</sub> or GHG are usually expressed per ton of treated waste or per amount of obtained compost (Amlinger et al. 2008).

Sampling and measurement protocols for the determination of emissions have been also studied (Sironi and Botta 2001). Even though there are several published papers about this topic (Sommer et al. 2004), there is a lack of information on the measurement of emissions from surface sources, in both non-aerated (composting piles with natural aeration by convection) and those with a common source that will be later spread in an outlet surface (biofilters).

The main factors controlling a composting process are those characteristics of an aerobic biological process such as oxygen concentration, temperature, moisture, pH and C/N ratio. The optimum values for the C/N ratio range from 15 to 30, even though it is possible that composting takes place in a wider range of values (Haug 1993). For this reason, adjusting the optimum C/N ratio of the starting mixture is recommended. The use of different organic wastes or some selected additives could also be satisfactory (Charest and Beauchamp 2002). Nevertheless, the amounts of carbon and nitrogen used for the calculations should be referred to the amounts that are ready available for the microorganisms when considering the C/N ratio as a parameter to be optimised (Puyuelo et al. 2011). This specific point is very important for the potential practical implications in the preparation of starting composting mixtures. In relation to pH, recent studies have demonstrated its effect on the emissions of odours (Sundberg et al. 2013).

In this context, respirometric methodologies have been shown to be suitable and reliable for the determination of the amount of biodegradable organic matter in wastes of different origin and characteristics. There are two types of respirometric analysis for this purpose: dynamic and static determinations, being the dynamic methods the most widely accepted and recommended (Adani et al. 2004; Barrena et al. 2006; Gea et al. 2004). The measurement of the CO<sub>2</sub> produced during the respirometric test is also used as a measurement of the biodegradability of the organic matter (Cooper 2004) and, consequently, of the biodegradable organic C.

Other researchers have worked on the emissions generated during the composting process of agricultural wastes (Komilis et al. 2004; Cayuela et al. 2006; Mondini et al. 2006; Mondini et al. 2007; Sánchez-Monedero et al. 2008; Szanto et al. 2007). In the USA, other studies are focused on VOC and NH<sub>3</sub> emissions during the composting of biowaste (Büyüksönmez and Evans 2007).

This review is a compilation of the different works dealing with the measurement, detection, minimization and treatment of the GHG emitted during the composting process of a wide variety of organic wastes.

## 2.2 Composting

### 2.2.1 *The Specific Role of Composting in Greenhouse Gas (GHG) Emissions*

Composting is an environmentally friendly waste treatment process where organic matter is biologically degraded. Although the benefits of composting are evident, GHG can be generated and emitted to the atmosphere during this process contributing to global warming.

In this context, composting of organic waste contributes (composting process) and avoids (compost application) at the same time to GHG emissions. GHG are released from composting facilities due to degradation of organic matter and the use of electricity and fuels in management waste operations. The use of compost in agriculture has a positive effect in GHG emissions since its application as an organic amendment provokes that carbon stays bound to soil, although the content of other nutrients (N, P, etc.) is typically low. GHG emissions from composting processes depend on the waste type and composition, the technology systems used (static and dynamic process, open and closed systems, presence or not of gas treatment units) and the final use of compost.

Benefits of compost application have to be assessed together with a real knowledge about the amount of GHG such as  $N_2O$  and  $CH_4$  generated during the composting process. The relation of GHG with some operational conditions and the technology used must be also considered. Data on GHG emissions from full-scale composting facilities are necessary to improve the knowledge about the contribution to the composting in GHG emissions. In the last years, there has been an increase in the number of scientific publications studying GHG emissions during composting (Amlinger et al. 2008; Boldrin et al. 2009; Sánchez-Monedero et al. 2010; Cayuela et al. 2012; Colón et al. 2012; Deportes 2012).

GHG emissions from composting processes are highly dependent on the waste type and composition. The composition and characteristics of the feedstock are key parameters for the design and operation of the composting facilities and for the final quality of the compost (Haug 1993).

Wastes with a low C/N ratio and high water content have a great potential for generating GHG emissions both during the storage and the composting process. In fact, wastes lacking of nutrients, porosity and structure, or presenting low biodegradability can hamper the correct evolution of the process, increasing the GHG emission. In order to minimize these emissions, optimal conditions for the initial mixture are required.

For some wastes, pretreatment operations before composting are required. This is the case of municipal solid wastes, especially when a source-separation system is not implemented. The production of high-quality compost from MSW may require a lot of energy because of the use of heavy machinery that makes GHG emissions unavoidable (Lou and Nair 2009). Other materials, such biosolids or manure, have a poor structure and an excess of water content and require the use of a bulking agent.

Grinding and mixing this bulking agent are operations that require energy that again contribute to GHG emissions.

Composting technologies can be open and closed systems. In open systems, composting is performed in facilities where, in general, gaseous emissions are neither collected nor treated. However, when the composting process takes place in an enclosed system usually the exhaust gases are treated. As expected, concentrations of GHG reported in facilities when the gas treatment systems are well-implemented were lower (Colón et al. 2012) than those of open systems. Effects of forced aeration and turning in GHG emissions have been also studied. Szanto et al. (2007) observed lower  $N_2O$  and  $CH_4$  emissions in turned piles than in static systems. They related these emissions to the prevalence of anaerobic regions in the static systems, as other similar studies (Parkinson et al. 2004). Amlinger et al. (2008) proposed that high aeration and effective stripping of  $NH_3$  during the early stages of composting can reduce  $N_2O$  formation. Ermolaev et al. (2012) studied the effects of different aeration and temperature settings on the emission of  $CH_4$ ,  $N_2O$  and  $CO_2$  during windrow composting with forced aeration following three different control strategies. However, they found that the emissions of  $CH_4$  and  $NO_2$  were low regardless the amount of ventilation. The oxygen concentration, temperature profile and moisture content are factors controlling GHG emissions. Nowadays, in the composting field, the technology that allows the control of these parameters is available.

Regarding  $CO_2$ , its emissions in composting derived from the organic matter biodegradation are not taken into account in their contribution to global warming since this carbon has a biogenic origin. The  $CO_2$  that contributes to GHG emissions is generated by composting facilities as a result of operational activities. In composting, the main GHG that can contribute to global warming are  $CH_4$  and  $N_2O$ . Both are related to a lack of oxygen during the composing process and consequently they depend on the management of the composting process (Cayuela et al. 2012; Colón et al. 2012). These gases, although they are generated in small amounts, have a great contribution to global warming since they have a warming potential of 25 ( $CH_4$ ) and 296 ( $N_2O$ ) times higher than that of  $CO_2$ .

Several authors reported that even in well-aerated process  $CH_4$  was emitted (He et al. 2000; Clemens and Cuhls 2003) while Beck-Friis et al. (2000) observed a rapid decrease when the oxygen supply was increased. The production of  $N_2O$  can be due to an incomplete ammonium oxidation or incomplete denitrification (Beck-Friis et al. 2000). Emissions of  $N_2O$  have been reported at different stages of the process. Some authors reported high emissions at the beginning of composting (He et al. 2000; Parkinson et al. 2004). Other studies reported the production of  $N_2O$  during the mesophilic and maturation phases (Beck-Friis et al. 2000; Hao et al. 2004) when the readily available carbon sources has been depleted (He et al. 2000). According to Cayuela et al. (2012),  $N_2O$  formation will be hampered if there are conditions to inhibit nitrification (such as low available  $NH_4^+$  in the pile or high pH). Beck-Friis et al. (2000) and Fukumoto et al. (2003) related  $N_2O$  emissions to the temperature of the process and  $CH_4$  emissions to the size of the pile (both works were performed at full-scale, using windrows and forced aeration systems, respectively), the structure



of the material and the time of the process. Higher emissions were measured in larger piles, with a poor structure and longer composting times. Monitoring of CH<sub>4</sub> emissions showed a large experimental fluctuation in all works.

Several authors have reported the GHG emissions generated during the biological treatment of several typologies of wastes. Most of them were calculated from laboratory and pilot scale processes, although interesting data at industrial scale have been also reported (Boldrin et al. 2009; Colón et al. 2012; Ermolaev et al. 2012). There are an important number of studies that quantify CH<sub>4</sub> and N<sub>2</sub>O emissions from animal manures (Fukumoto et al. 2003; Hao et al. 2004; Szanto et al. 2007). However, less published works dealing with municipal solid wastes can be found, and even less works studying the GHG emissions of different composting systems have been published.

Colón et al. (2012) evaluated four different full-scale facilities treating the source-separated organic fraction of municipal solid wastes (OFMSW). They reported a range of CH<sub>4</sub> and N<sub>2</sub>O emissions between 0.34 and 4.37 kg CH<sub>4</sub> Mg OFMSW<sup>-1</sup> and 0.035 and 0.251 kg CH<sub>4</sub> Mg OFMSW<sup>-1</sup>, respectively. Regarding CH<sub>4</sub>, the highest values were found in facilities without gas treatment units. Also, Boldrin et al. (2009) presented a study where several technologies for municipal solid waste treatment were evaluated. They reported CH<sub>4</sub> and N<sub>2</sub>O emissions ranging from 0.02 to 1.8 kg CH<sub>4</sub> Mg OFMSW<sup>-1</sup> and 0.0075 and 0.252 kg CH<sub>4</sub> Mg OFMSW<sup>-1</sup>.

As previously commented, although ammonia is not considered a GHG, its emission during composting plays an important role. Ammonia emissions are affected by the C/N ratio of the initial composting mixture, by the temperature reached during the process and the aeration (Pagans et al. 2006b). High loads of ammonia can reduce the optimal use of the biofilter system in enclosed facilities (Amlinger et al. 2008). Moreover the conservation of nitrogen in the end-product improves compost use in agriculture as organic fertiliser. Consequently, from a global warming point of view, less use of chemical fertilizers will be required (Favoino and Hogg 2008).

In the role played by composting in GHG emissions it is important to bear in mind the role of compost as an end-product. The use of compost as an organic amendment can contribute to mitigate GHG in several forms.

Compost utilization can reduce the need of chemical fertilisers and pesticides, which implies the reduction of GHG emissions associated with their production and application. Also, a positive effect in soil structure is produced with compost application by improving tillage and workability. Improved structure of soils associated with the application of organic matter can help to reduce requirements for water irrigation in periods of drought and to increase the potential of soils to retain moisture (Favoino and Hogg 2008).

One of the aspects associated with compost utilization that more attention has received in the last years is the potential for sequestration of carbon in agricultural soils (Mondini et al. 2007; Favoino and Hogg 2008). By applying compost, biogenic carbon is held in soils for a period of time before carbon is released, increasing carbon uptake and storage within the plant and removing CO<sub>2</sub> from the atmosphere.



## ***2.2.2 GHG Emitted During Composting and Their Relationship to C and N Dynamics***

Microbial transformations involved in the formation of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  in composting piles are similar to those taking place in other environments such as soil, water bodies, wastewater treatment plants, etc. However, the microbial gas production and the final emission to the atmosphere will be affected by the particular environmental conditions of composting piles (such as temperatures up to  $70^\circ\text{C}$ , high organic matter content, easily available organic compounds, rich and active microbial population and limited amount of oxygen, etc.) and composting management operations (turning, watering, pile size and geometry, etc.). All these variables represent a characteristic environment affecting not only the microbial gas production in the pile, but also its transport within the pile and the final emission to the atmosphere. In the following sections the impact of the C and N dynamics on GHG emissions during composting will be also discussed.

### **2.2.2.1 Carbon Dioxide ( $\text{CO}_2$ )**

As previously mentioned, there are two main sources of  $\text{CO}_2$  emissions from composting facilities, biogenic and non-biogenic  $\text{CO}_2$ . Biogenic  $\text{CO}_2$  emissions derive from the biological degradation of the organic matter, mostly as a consequence of aerobic decomposition and, to a lesser extent, from anaerobic processes or the oxidation of  $\text{CH}_4$  by aerobic methanotrophic bacteria. This emission accounts for the highest amount of gas generated during the process, since between 40 and 70% of the original organic matter can be degraded during composting (Haug 1993). However, the global warming potential of these emissions are not taken into account in the environmental impact of composting operations since this biological  $\text{CO}_2$  is considered to be carbon neutral (IPCC 2006). The exclusion from the inventories has reduced the number of papers studying  $\text{CO}_2$  emissions and this gas is only studied from the point of view of establishing mass balances of composting operations (Boldrin et al. 2009) or as an index of the overall microbial activity of the pile, reflecting the progress of the process (Hobson et al. 2005; Sánchez-Monedero et al. 2010) and the evaluation of the stability of the end-product (Barrena et al. 2006).

Non-biogenic  $\text{CO}_2$  from composting includes the emissions associated to energy and fuel consumption in the composting facility. These emissions are dependent on the technology of the plant and the machinery used such as shredders, front-loaders, turning equipment, screenings, and other processing activities. These emissions are beyond the scope of this review, but updated information can be found elsewhere (Boldrin et al. 2009; Scheutz et al. 2009; Lou and Nair 2009; Brown et al. 2008).

### 2.2.2.2 Methane (CH<sub>4</sub>)

Methane emissions derived from organic waste composting have attracted the attention of researchers as a considerable contributor to global warming since this greenhouse gas has a global warming potential 25 times greater than carbon dioxide over a time horizon of 100 years (IPCC 2006).

The optimum growing conditions for methanogenic bacteria are a lack of oxygen (strict anaerobic microorganisms), a redox potential below  $-200$  mV, neutral pH and the presence of nutrients and substrates rich in organic matter (Kebreab et al. 2006). These conditions can be temporally found at the early stages of the composting process, where large amounts of nutrients and available sources of organic compounds stimulate microbial growth, depleting the oxygen levels in the pile. Accordingly, most of CH<sub>4</sub> emissions have been recorded during the initial weeks of the process, at the beginning of the thermophilic phase (Beck-Friis et al. 2000; Sánchez-Monedero et al. 2010). The high temperatures reached at the beginning of the process reduce oxygen solubility (Pel et al. 1997), facilitating the creation of anaerobic spots within the pile. However, there are other variables such as high concentration of ammonia, which may inhibit the activity of methanogens at pH > 9 (Kebreab et al. 2006), or the presence of electron acceptors such as sulphates, which reduce their activity by competition with sulphate reducing bacteria (Hao et al. 2005). Sánchez-Monedero et al. (2010) reported that the high ammonia levels generated by the hydrolysis of urea, used as nitrogen source, inhibited the production of CH<sub>4</sub> in olive mill waste composting piles.

The emission of CH<sub>4</sub> from composting piles is governed by the biological activity of the pile (Hao et al. 2001) and also by other factors affecting gas transport from the anaerobic spots to the pile surface, such as gas diffusion within the pile and the presence of methanotrophic bacteria. Methanotrophs are aerobic microorganisms colonising the surroundings of anaerobic zones and pile surface, which are able to oxidise between 46 and 98% of the CH<sub>4</sub> generated in the pile (Jäckel et al. 2005). Methanotrophic bacteria also play an important role in the production and consumption of other relevant GHG emitted during composting, such as N<sub>2</sub>O and CO (Topp and Hanson 1991). Sánchez-Monedero et al. (2011) performed a 4-year interannual evaluation of the GHG emissions from a composting plant treating olive mill wastes and found a reduction of CH<sub>4</sub> emissions associated to the improvement of the management of the composting plants (watering and turning frequencies).

Kebreab et al. (2006) and Brown et al. (2008) reviewed the topic of GHG emissions from livestock and composting operations and they highlighted the importance of the composting feedstock, the height and shape of the pile, the control of moisture content and turning frequency as the main factors governing CH<sub>4</sub> emissions during the process, since these variables will affect both the oxygen availability and gas diffusion in the composting pile. The presence of manure can also increase the methane emissions due to the incorporation of anaerobic microorganisms, as

observed by He et al. (2000) and Sánchez-Monedero et al. (2010) in composting piles treating food and olive mill wastes, respectively.

### 2.2.2.3 Nitrous Oxide (N<sub>2</sub>O)

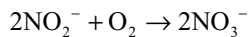
There is an increasing awareness about the emission of N<sub>2</sub>O from composting operations due to the high global warming potential of this gas (296 times higher than that of CO<sub>2</sub> over a 100 year horizon, IPCC 2006) and its impact on the ozone layer (Smith et al. 2010). Despite the relatively small amounts of N<sub>2</sub>O released during composting, its contribution to the global N<sub>2</sub>O budget in waste management or live-stock agriculture cannot be discarded due to the impact of composting operations treating manures or other N-rich organic wastes (de Klein et al. 2010).

The biological production of N<sub>2</sub>O during composting is a complex process since there are different microbial pathways involved in the formation of N<sub>2</sub>O (nitrification, nitrifier denitrification and denitrification among others), which may simultaneously occur at different locations within the pile (Czepiel et al. 1996; Kebreab et al. 2006; Maeda et al. 2011). For this reason, the identification of N<sub>2</sub>O sources as well as the microorganisms involved in these processes still remains a key research topic (Maeda et al. 2011).

Nitrification is one of the main microbial processes leading to the emission of N<sub>2</sub>O during composting. Aerobic nitrification involves the initial transformation of ammonia to nitrite by different genera of ammonia-oxidising bacteria (AOB), such as *Nitrosomonas* and *Nitrososporas*, according to the following equation:



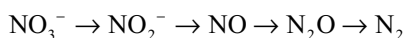
and the oxidation of nitrite to nitrate by nitrite-oxidising bacteria (NOB), such as *Nitrobacter* (Kowalchuk et al. 1999; Maeda et al. 2010):



NH<sub>4</sub><sup>+</sup> is the main precursor of nitrification. NH<sub>4</sub><sup>+</sup> is generated by ammonification of OM at early stages of the process (Sánchez-Monedero et al. 2001). Typical alkaline pHs found in composting matrices favour the transformation of this soluble NH<sub>4</sub><sup>+</sup> into NH<sub>3</sub>, which is then initially oxidised by AOB into NO<sub>2</sub><sup>-</sup> and then transformed to NO<sub>3</sub><sup>-</sup> by nitrite-oxidising bacteria. N<sub>2</sub>O is produced during the initial step of the oxidation of NH<sub>4</sub><sup>+</sup>, as an intermediate between NH<sub>2</sub>OH and NO<sub>2</sub><sup>-</sup> (Czepiel et al. 1996). Ammonia-oxidising *archaea* (AOA) have been recently suggested to be actively involved in nitrification in composting piles, but the contribution of AOA to the total amount of N<sub>2</sub>O still remains unclear (Yamamoto et al. 2010; Zeng et al. 2012).

Denitrification has traditionally represented the main source of N<sub>2</sub>O, especially in the case of manures (Kebreab et al. 2006). Denitrification is an anoxic process

carried out by denitrifiers, which are heterotrophic microorganisms that can use  $\text{NO}_3^-$  as the electron acceptor, causing the reduction of  $\text{NO}_3^-$  to  $\text{N}_2$  according to the following steps:



In absence of  $\text{O}_2$ ,  $\text{NO}_3^-$  is reduced to  $\text{N}_2$  without appreciable  $\text{N}_2\text{O}$  production, but  $\text{N}_2\text{O}$  production can increase as the concentration of  $\text{O}_2$  increases in the pile (Czepiel et al. 1996). In this case, nitrifier denitrification (denitrification coupled to an incomplete nitrification at low  $\text{O}_2$  concentrations) can be the responsible of the generation of  $\text{N}_2\text{O}$  during the initial step of ammonia oxidation and also as a consequence of  $\text{NO}_2^-$  reduction. This mechanism has been studied in agricultural soils (Wrage et al. 2001), but there is only limited information during composting (He et al. 2001; Hobson et al. 2005). Fukumoto and Inubushi (2009) observed that the addition of NOB reduced the emission of  $\text{N}_2\text{O}$  during composting of pig manure, suggesting that the accumulation of  $\text{NO}_2^-$  in the pile could be a significant source of  $\text{N}_2\text{O}$ , due to the reduction of  $\text{NO}_2^-$  to  $\text{N}_2\text{O}$  (under limited  $\text{O}_2$  conditions) rather than the final oxidation to  $\text{NO}_3^-$  (with no  $\text{O}_2$  limitation). Under these conditions, when available C was depleted, nitrifier denitrification would be the main mechanisms leading to  $\text{N}_2\text{O}$  emissions, as observed by He et al. (2000), who found an increase in the  $\text{N}_2\text{O}$  emission when the ratio between water-soluble C and dissolved N was lower than 5.

Nitrifiers and denitrifiers show their optimal growth under different environmental conditions. Nitrifiers require aerobic conditions, mesophilic temperatures (below 40 °C), pH values above 5 and the presence of  $\text{NH}_4^+$ , whereas denitrifiers need anaerobic conditions, or at least low  $\text{O}_2$  concentration, the presence of sources of available C and the presence of  $\text{NO}_3^-$ ,  $\text{NO}_2^-$  or NO as electron acceptors (Kebreab et al. 2006). Due to the heterogeneity of the composting materials, both environmental conditions (aerobic and anaerobic zones) can coexist simultaneously in the composting mass, since different oxygen concentration gradients are created along the pile (Beck-Friis et al. 2000; Hao et al. 2001). Denitrifiers may colonise the inner part of the pile whereas nitrifiers, which require oxygen concentrations in the range within 1 and 10% (Béline et al. 1999), may colonise the aerobic pile surface. The relative contribution of nitrifiers and denitrifiers to the  $\text{N}_2\text{O}$  emission is governed by the oxygen concentration and moisture of the pile (Hwang and Hanaki 2000). These authors reported that denitrification was the main source of  $\text{N}_2\text{O}$  at moisture levels between 40–60% and oxygen concentrations around 10%, whereas nitrification became more dominant at higher oxygen concentrations.

Similarly to those of  $\text{CH}_4$ ,  $\text{N}_2\text{O}$  emissions can be affected not only by the biological activity of the composting mixture, but also by the N availability and gas diffusion within the pile (Hao et al. 2001). Several authors reported peak  $\text{N}_2\text{O}$  emissions either at early stages of the process or after the thermophilic phase of composting, when the environmental conditions of the pile (temperatures below 40 °C) favour the growth of nitrifying bacteria (He et al. 2001; Kebreab et al. 2006; Sánchez-Monedero et al. 2010). Once  $\text{NO}_3^-$  has been generated, the mixing of the

composting matrix facilitates the transport of nitrates from the surface to the interior of the pile where they can be reduced to  $N_2$  and  $N_2O$  by denitrifiers. The use of urea as N source can enhance  $N_2O$  emissions up to levels similar to those found in N-rich manure heaps due to the increase of available N from the hydrolysis of urea (Sánchez-Monedero et al. 2010). Vermicomposting also increases the  $N_2O$  emissions by stimulating denitrification and nitrification processes, due to the increase of N availability and the transport of N facilitated by the activity of earthworms (Frederickson and Howell 2003; Hobson et al. 2005).

Gas exchange within the pile also plays an important role since the generation of  $N_2O$  by both nitrifiers and denitrifiers is enhanced at low oxygen concentrations (Czepiel et al. 1996).  $N_2$  is obtained as the final product of denitrification in absence of  $O_2$ , but significant amounts of  $N_2O$  are generated as the concentration of  $O_2$  increases in the pile. In addition, pure cultures of *Nitrosomona* bacteria responsible of the initial step of ammonia oxidation have been shown to significantly increase the production of  $N_2O$  under limited oxygen conditions (Goreau et al. 1980). Since these factors are highly dependent on the composting material and the process performance, the specific characteristics of the starting materials will determine the environmental conditions for N transformation during composting.

#### 2.2.2.4 Other Relevant Greenhouse Gases

There are other gases generated in small amounts during organic waste composting that have been studied due to their impact on global warming. Carbon monoxide (CO) and nitrogen oxides different than  $N_2O$  ( $NO_x$ ) have small direct global warming potential, but they both lead to indirect radiative effects by increasing  $CH_4$  lifetime and elevating concentrations of tropospheric  $O_3$  (IPCC 2006). The calculation of their contribution to global warming is subject to large uncertainties due to the short lifetime and reactivity of these gases in the atmosphere. According to IPCC (2006) the global warming potential, over a 100-year horizon, is likely to be 1–3 for CO, and in the order of 5 for surface  $NO_x$  emissions.

The emission of CO occurs during the aerobic decomposition of the organic wastes during composting by a mixture of physical processes and biological activity (Hellebrand and Halk 2001; Hellebrand and Shade 2008). These authors found the maximum CO-flux rates at the beginning of the composting process, probably due to physicochemical generation, and then the levels decreased during periods of high biological activity, reflecting the temperature dependence of CO emissions and also the impact of oxygen availability and the oxidation to  $CO_2$ . CO emissions only represent a minor GHG source in green waste and livestock waste (Hellebrand and Shade 2008) and in urban wastes, where CO-C emissions varied from 0.07 to 0.13 kg  $Mg^{-1}$  of wet feedstock, which represents approximately about 0.04–0.08 % of the total C emitted (Andersen et al. 2010a, b). CO emissions have been also investigated as a potential health risk to workers in enclosed facilities treating municipal solid wastes (Phillip et al. 2011).

From the two gases composing  $\text{NO}_x$  ( $\text{NO} + \text{NO}_2$ ), only  $\text{NO}$  is generated during composting, either as by-product or intermediate of microbial nitrification and denitrification (Del Prado et al. 2006; Hao et al. 2001). Fukumoto et al. (2011) studied the  $\text{NO}$  emissions from swine manure composting and observed a similar trend to that of  $\text{N}_2\text{O}$ , characterised by a peak after the thermophilic phase of composting (coinciding with the activity of nitrifiers) and a decreasing trend towards the end of the process. Total  $\text{NO}$  emissions only represented one-tenth of the magnitude of  $\text{N}_2\text{O}$  emission, approximately 3% of total N losses.

### ***2.2.3 Greenhouse Gas Production for Different Typologies of Organic Wastes***

There is a wide range of organic wastes that can be used as composting substrates such as manures, municipal solid wastes, garden and yard wastes, agricultural crop residues, sewage sludge and other industrial sludge, etc. The characteristics of these starting materials will affect the physicochemical properties of the pile and, consequently, will govern the microbial processes leading to the formation of GHG and also their diffusion and transport within the pile. As already discussed in the description of the main pathways of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  generation, the main variables affecting GHG emissions are the moisture content and porosity, which control the oxygen availability and gas diffusion, and the presence of nutrients and organic compounds to be used as substrates for the microorganisms participating in gas production. The composting technology used for the aeration (forced aeration or windrowing), the size of the piles and pile temperature also represent key variables affecting GHG generation and emission.

#### **2.2.3.1 Manures**

Manures represent one of the most important and studied substrates for composting (Kebreab et al. 2006). Manures are N-rich organic materials characterised by high moisture contents that make them to be considered as wet feedstock for composting (Haug 1993). The treatment of manures through composting permits the reduction of volume and moisture, their sanitisation and organic matter stabilisation, giving rise to a valuable end-product that can be safely used in agriculture. However, manure characteristics favour GHG emission during composting. The large amounts of easily available N compounds enhance the microbial activity of the pile and can serve as substrates for the nitrification and denitrification processes leading to the emission of  $\text{N}_2\text{O}$ . Furthermore, high moisture together with enhanced microbial activity at early stages of the process can lead to the creation of anaerobic spots for the formation of  $\text{CH}_4$ .

A summary of the amounts of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  generated during manure composting is shown in Table 2.1. The amounts of  $\text{CH}_4$  emitted during composting are

**Table 2.1** Summary of CH<sub>4</sub> and N<sub>2</sub>O emissions reported in the literature for different typologies of wastes and composting technologies

Feedstock	Bulking agent	System	N <sub>2</sub> O	CH <sub>4</sub>	Reference
<i>Manures</i>					
Swine	Cardboard	Windrow	0.1% of initial N	–	Kuroda et al. 1996
Swine	Bedding (straw)	Static	9.8% of initial N	12.6% of initial OM	Szanto et al. 2007
		Windrow	0.5% of initial N	0.5% of initial OM	
Swine	Sawdust	Static	3.7–4.7% of initial N	1.0–1.9 kg CH <sub>4</sub> ton <sup>-1</sup>	Fukumoto et al. 2003
Swine	Sawdust	Static	3.0–9.3% of initial N	–	Fukumoto and Inubushi 2009
Swine	Barley straw	–	0.058 kg N <sub>2</sub> O-N ton <sup>-1</sup>	0.19 kg CH <sub>4</sub> -C ton <sup>-1</sup>	Sommer and Moller 2000
Swine	Sawdust	Static	1.6 µg m <sup>-2</sup> s <sup>-1</sup>	5.2 µg m <sup>-2</sup> s <sup>-1</sup>	Park et al. 2011
		Windrow	7.9 µg m <sup>-2</sup> s <sup>-1</sup>	7.5 µg m <sup>-2</sup> s <sup>-1</sup>	
Cattle	Bedding (straw)	Static	0.11 kg N <sub>2</sub> O-N ton <sup>-1</sup>	6.3 kg CH <sub>4</sub> -C ton <sup>-1</sup>	Hao et al. 2001
		Windrow	0.19 kg N <sub>2</sub> O-N ton <sup>-1</sup>	8.1 kg CH <sub>4</sub> -C ton <sup>-1</sup>	
Cattle	Bedding (straw)	Windrow	0.077 kg N <sub>2</sub> O-N ton <sup>-1</sup>	8.92 kg CH <sub>4</sub> -C ton <sup>-1</sup>	Hao et al. 2004
	Bedding (wood chips)		0.084 kg N <sub>2</sub> O-N ton <sup>-1</sup>	8.93 kg CH <sub>4</sub> -C ton <sup>-1</sup>	
Cattle (dairy)	House wrap	Static	0.370 kg N <sub>2</sub> O-N ton <sup>-1</sup>	1.14 kg CH <sub>4</sub> -C ton <sup>-1</sup>	Pattey et al. 2005
Cattle (beef)			0.103 kg N <sub>2</sub> O-N ton <sup>-1</sup>	0.11 kg CH <sub>4</sub> -C ton <sup>-1</sup>	
Cattle (dairy)	Bedding (straw)	Static	0.046 kg N <sub>2</sub> O-N ton <sup>-1</sup>	–	El Kader et al. 2007
	Bedding (straw)	Windrow	0.070 kg N <sub>2</sub> O-N ton <sup>-1</sup>	–	
Turkey	Wood shaving & straw	Static	0.091 kg N <sub>2</sub> O-N ton <sup>-1</sup>	–	
Cattle and horse (50:50)	Bedding (hay)	Windrow	0.32 kg N <sub>2</sub> O-N ton <sup>-1</sup>	–	Czepiel et al. 1996
Cattle (dairy)	Bedding	Windrow	0.90 g N <sub>2</sub> O m <sup>-2</sup> d <sup>-1</sup>	13.5 g CH <sub>4</sub> m <sup>-2</sup> d <sup>-1</sup>	Leytem et al. 2011
<i>MSW</i>					
Food waste	Biochip	Static	1.36 kg N <sub>2</sub> O-N ton <sup>-1</sup>	–	He et al. 2001
MSW	–	Static	0–0.24 kg N <sub>2</sub> O-N ton <sup>-1</sup>	4.5–9 kg CH <sub>4</sub> -C ton <sup>-1</sup>	Clemens and Cuhls 2003
Separated organic household waste		Static	0.02–0.11 kg N <sub>2</sub> O-N ton <sup>-1</sup>	0.04–0.8 kg CH <sub>4</sub> -C ton <sup>-1</sup>	Amlinger et al. 2008



Table 2.1 (continued)

Feedstock	Bulking agent	System	N <sub>2</sub> O	CH <sub>4</sub>	Reference
Organic fraction source-separated MSW	Wood chips or pruning wastes	Windrow In vessel	0.048 kg N <sub>2</sub> O-N ton <sup>-1</sup>	0.26 kg CH <sub>4</sub> -C ton <sup>-1</sup>	Colón et al. 2012
		Confined windrow	0.048 kg N <sub>2</sub> O-N ton <sup>-1</sup>	1.26 kg CH <sub>4</sub> -C ton <sup>-1</sup>	
		Turned windrow	0.160 kg N <sub>2</sub> O-N ton <sup>-1</sup>	3.28 kg CH <sub>4</sub> -C ton <sup>-1</sup>	
		Home composting	0.430 kg N <sub>2</sub> O-N ton <sup>-1</sup>	0.12 kg CH <sub>4</sub> -C ton <sup>-1</sup>	
Source-separated MSW	Yard waste	Windrow	261 mg N <sub>2</sub> O-N m <sup>-2</sup> d <sup>-1</sup>	35 g CH <sub>4</sub> m <sup>-2</sup> d <sup>-1</sup>	Beck-Friis et al. 2000
<i>Other wastes</i>					
Cattle mortalities	Barley straw	windrow	0.97% of initial N	0.55% of initial C	Hao et al. 2009
	Cattle manure & barley straw	windrow	0.85% of initial N	0.13% of initial C	
Animal meals	Straw & cotton	windrow	0.07–0.11 % of initial N	–	Cayuela et al. 2012
Biosolids	Wood ash	static	0.32 kg N <sub>2</sub> O-N ton <sup>-1</sup>	–	Czepiel et al. 1996
Grass	Soil	static	0.054 kg N <sub>2</sub> O-N ton <sup>-1</sup>	5 kg CH <sub>4</sub> -C ton <sup>-1</sup>	Hellebrand 1998
Hens mortalities	–	windrow	0.003–0.004 kg N <sub>2</sub> O-N ton <sup>-1</sup>	52–120 kg CH <sub>4</sub> ton <sup>-1</sup>	Dong et al. 2011
Garden waste	–	windrow	0.05 kg N <sub>2</sub> O-N ton <sup>-1</sup>	1.9 kg CH <sub>4</sub> -C ton <sup>-1</sup>	Andersen et al. 2010a
Olive mill waste	Diverse manures & olive pruning	windrow	0.01–3.36 g N-N <sub>2</sub> O m <sup>-2</sup> d <sup>-1</sup>	1–147 g CH <sub>4</sub> -C m <sup>-2</sup> d <sup>-1</sup>	Sánchez-Monedero et al. 2010
<i>Default emissions factors for CH<sub>4</sub> and N<sub>2</sub>O emissions from waste composting from IPCC 2006</i>					
On a dry basis			0.6 g (0.2–1.6) N <sub>2</sub> O per kg waste treated	10 g (0.08–20) CH <sub>4</sub> per kg waste treated	IPCC 2006
On a wet basis			0.3 g (0.06–0.6) N <sub>2</sub> O per kg waste treated	4 g (0.03–8) CH <sub>4</sub> per kg waste treated	

within 0.1 and 8.93 kg of  $\text{CH}_4$  per ton of treated manure. This wide range may be affected by the pre-treatment of manure prior to composting (manure storage can represent an important source of  $\text{CH}_4$ ) and also by the aeration system, windrow vs. forced aeration (agitation favours  $\text{CH}_4$  emissions) (Kebreab et al. 2006). The levels of  $\text{N}_2\text{O}$  emitted from manure composting piles varied from 0.046 up to 0.370 kg  $\text{N}_2\text{O-N}$  per ton of treated manure depending on the composting system. Aerated static piles usually increase the emissions of  $\text{N}_2\text{O}$  by preventing ammonia losses, which can be later oxidised to nitrates generating  $\text{N}_2\text{O}$ . The emission of  $\text{N}_2\text{O-N}$  from manure composting can represent up to 9.8% of the initial N. These experimental results have been used by IPCC (2006) to propose default emission factors of 4 kg  $\text{CH}_4 \text{ ton}^{-1}$  and 0.3 kg  $\text{N}_2\text{O ton}^{-1}$  (Table 2.1) from the biological treatment of organic wastes (for different types of feedstock and composting operations).

### 2.2.3.2 Municipal Solid Wastes (MSW)

Municipal solid wastes also represent a major source of organic wastes suitable for composting. This group includes not only mixed MSW, but also other materials such as the organic fraction of the source separated MSW, garden and yard wastes, food wastes, etc. This type of composting substrates is characterised by lower organic matter, nitrogen and moisture content than manures. For this reason the impact on GHG emissions is expected to be different, since lower amounts of organic C and N in the feedstock would lead to reduced GHG emissions (Brown et al. 2008; Büyüksönmez 2012).

Amounts of  $\text{CH}_4$  emitted during MSW composting varied from 0.12 up to 9 kg  $\text{CH}_4$  per ton of treated waste (Table 2.1). This large variability in gaseous emissions reflects the impact of the feedstock, the composting system and the efficiency of the composting facility on GHG emissions (Colón et al. 2012). The levels of  $\text{N}_2\text{O}$  emitted from MSW composting ranged from 0 to 0.430 kg  $\text{N}_2\text{O-N}$  per ton of treated waste, which represents values generally lower than those registered from the biological degradation of manure. In the case of MSW, where most of the composting piles are operated with little amounts of water, the small amount of  $\text{CH}_4$  generated in the pile is most likely oxidised when it reaches the aerobic surface, considering  $\text{CH}_4$  emissions to be essentially zero from a practical point of view, as far as life cycle assessments are concerned (US EPA 2006).

### 2.2.3.3 Other Organic Wastes

Table 2.1 shows the  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions for a range of organic wastes used as feedstock for composting. The impact of the different wastes will depend on their physical-chemical composition. Organic wastes such as biosolids, characterised by high N and moisture contents are expected to have a similar behaviour than manures, whereas other wastes such as cattle and hens mortalities or olive mill wastes can have different behaviour depending on their physical-chemical characteristics.

Sánchez-Monedero et al. (2010) studied GHG emissions from composting piles prepared with olive mill wastes and different N sources and bulking agent observing that the peculiar characteristics of these wastes, characterised by a low degradation rate and low N levels, reduced the emission of GHG.

## **2.3 Methodologies to Determine GHG Emissions in Composting Processes**

### ***2.3.1 Closed and Open Chambers***

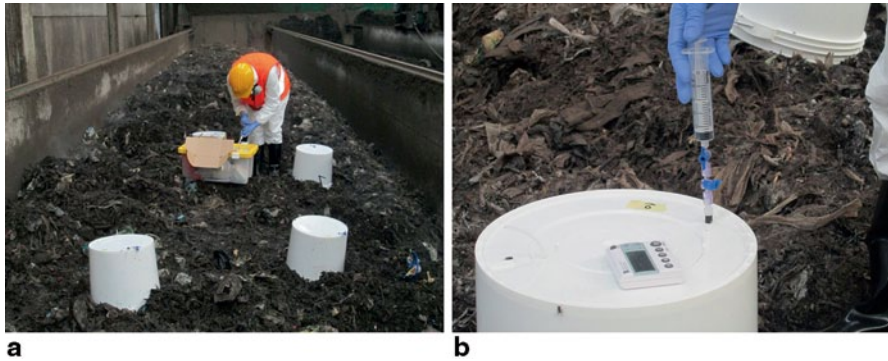
The most widely used method to sample GHG emitted from composting piles is based on the use of chambers. This technique is based on enclosures, generally inverted boxes or cylinders of known dimensions, that are placed over a part of the pile surface or encompasses all the composting pile and the measurement of the concentration of gases emitted from the composting material by several instrumental techniques.

This section describes the two main versions of this technique, namely closed and open chambers, and discusses the advantages and disadvantages related to each of them.

#### **2.3.1.1 Closed Chambers**

Closed chambers involve the sealing of the compost surface (or the entire pile) with an enclosure of known dimensions for determined periods. This method has been successfully used for measuring surface GHG emissions in piles prepared with manure (Hao et al. 2001), source-separated organic household wastes (Beck-Friis et al. 2000), lignocellulosic wastes (Andersen et al. 2010a; Cayuela et al. 2012) and olive mill wastes (Sánchez-Monedero et al. 2010). There are several kinds of chambers varying on shape, materials and dimensions. It is recommended that flux chambers should be fabricated of non-reactive materials (stainless steel, aluminium, PVC, polypropylene, polyethylene, or plexiglas) and the material should be white or coated with reflective material (mylar or painted) (Parkin and Venterea 2010). The most widely used shapes are cylinders or boxes (parallelepipeds) and the volume of the chamber may vary from 10 to 400 l.

When operating in this mode, gases are sampled at intervals during the closing period, which duration varies depending also from the instrumental technique used to measure the gas concentration. In the specific literature, this time ranges from few minutes to 30 min, with the latest as the most frequently used enclosure time with a sampling frequency of every 10 or 15 min. Gas analysis can be performed on site or off-site. In the first case air samples from the chamber are pumped through the measurement cell of the instrument (IRGA, Photoacoustic analyzer, FTIR) and



**Fig. 2.1** Example of greenhouse gas (GHG) sampling using vacutainers: **a** pile sampling; **b** gas sampling for analysis

then back into the chamber to avoid pressure changes. This chamber design is called dynamic closed chamber and represents a variation of the closed chamber method.

In the second case, few millilitres of air are withdrawn from the chamber headspace through a septum or a sampling port fitted into the chamber top. Samples are stored in syringes, vials, vacutainers, Tedlar bags or metal tubes and then analyzed in the laboratory, usually by gas chromatography (Fig. 2.1). Operations can be facilitated by automation of chamber operation and sampling.

Gas fluxes are calculated from concentrations by assuming a steady state gradient in the underlying windrows. Emission fluxes are determined by fitting the experimental data to a second-order polynomial equation (gas concentration vs. time). The flux at time 0 is calculated by taking derivatives of the second-order polynomial (Hao et al. 2001).

The advantage of the closed chamber method is that it is an easy and cheap technique that does not require sophisticated instruments. Further strengths of closed chambers are the versatility, as their design and deployment protocol can be adapted to a wide range of situations, and the capability to measure very low gas fluxes. On the other hand this method suffers for several disadvantages and limitations. The first limitation is due to the fact that the chamber may induce pressure gradients between compost pore space and chamber headspace resulting in convective transfer and biased flux estimates (Rochette 2011). This problem can be overcome by inserting a properly designed vent tube on the chamber to equalize barometric pressure inside and outside the chamber. For instance, Sommer et al. (2004) inserted a vent tube with an internal diameter of 1.6 mm and a length of 17.4 cm. However, care need to be taken as wind passing over the vent tube can cause a continue depressurization of the chamber due to the Venturi effect, thus resulting in much larger measurement errors than could be found with a chamber without vent.

Another problem of emission measurements from compost with closed chambers is the variation in the rate of diffusion of gas from compost to the headspace of the chamber. In undisturbed composts, diffusion is driven by a very large difference in concentration between low values in the atmosphere and high values in the

compost (one to two orders of magnitude higher). The increase in concentration in the headspace of the closed chamber, especially for that concerning CO<sub>2</sub> in the bio-oxidative phase of the composting process, may decrease the rate of diffusion of gases resulting in an underestimation of compost emission. However, there are studies demonstrating that the critical CO<sub>2</sub> concentrations in the chamber affecting the rate of CO<sub>2</sub> diffusion ranged from 1000 to 1500 ppm<sub>v</sub> (Bekku et al. 1997). Therefore care should be taken in order to utilize enclosure times that does not allow for the build-up of CO<sub>2</sub> concentrations leading to a decrease of the rate of diffusion.

A further problem is related to the fact that the chamber may lead to a rise in the temperature causing a perturbation in the rate of diffusion of the gases. This limitation can be overcome, at least partially, by constructing the chamber with insulated or reflective material and limiting the enclosure time.

Finally, gas fluxes can be affected by the height of the chamber. High chambers decrease the error in headspace volume determination and problems associated with high headspace gas concentration (e.g., leakage; feedback on gas flux), but on the other side high chambers may not allow adequate mixing of headspace air (Rochette 2011). To overcome this problem, some authors used a fan positioned within the chamber (Czepiel et al. 1996), but other authors do not agree with this solution, as it has been observed that fans can induce pressure perturbations within the chamber (Parkin and Venterea 2010).

### 2.3.1.2 Open Chambers

The open chamber method still includes the presence of an enclosure, but the main difference with the closed chamber is that it involves a continuous flow, through the chamber, of outside air (Ahn et al. 2011). Calculation of the flux is related to the difference in gas concentration between the incoming and out coming air.

Conversely to closed chambers, which are generally used to measure flux from limited surfaces of the composting materials, open chambers are usually designed as large-scale chambers that entirely encompass the compost piles and therefore allow to capture the whole flux of gases generated by the compost. Large-scale chambers have been utilized by Ahn et al. (2011), Amlinger et al. (2008), Fukumoto et al. (2003) and Park et al. (2011).

Open chambers systems are generally coupled with portable automatic gas analyzers and therefore they present the capacity to measure emissions on site; this allows for timely variations in the sampling strategy in order to capture increased gas fluxes due to changes in management operations and/or in environmental conditions.

On the other side, open chambers are more expensive and require higher technical skills to be operated. Moreover, these systems present the limitation that the accuracy of the measure strongly depends on the rate of air flux and consequently are greatly affected by variations in the environmental conditions. Therefore, they need a strict flow control and a continuous correction for changes in temperature and atmospheric pressure.

### 2.3.1.3 Chamber Methods and GHG Emissions from Composting Processes at Plant Scale

Chamber used in composting processes at laboratory and pilot-scale may be constructed in such a way that they cover all the composting material and consequently they can account for the total flux of gases generated by the compost. This is not possible in composting process at full-scale. A potential problem with the application of flux chambers for this purpose is that the large spatial and temporal variations in gas emissions increase the difficulty in estimating the whole-plant emissions. Investigations performed on composting processes at full-scale showed an extremely high spatial and temporal variability in GHG flux dynamics and this make particularly difficult to accurately estimate emissions using chamber methods (Andersen et al. 2010a).

Nevertheless, Amlinger et al. (2008) stated that a manual discontinuous analysis of  $N_2O$  and  $CH_4$  with closed chambers from single air samples is acceptable for measurements over short-term durations. However, it is extremely relevant to consider the fact that gaseous emissions in windrow composting tend to be concentrated in the windrow top, the so-called chimney effect. The chimney effect was thoroughly investigated by Andersen et al. (2010a) by placing small chambers at nine different locations across the section of a windrow. The highest fluxes were observed at monitoring points located near the windrow top and nearly all other points showed insignificant gas fluxes. The investigation indicates that most (>85%) of the gases vented through a narrow (1 m wide), chimney-like area in the top of the windrow. Andersen et al. (2010a) also found the same flux pattern for all gases measured at different points across the windrow section. Their results are contradicted by those of Sommer et al. (2004), who found that a significant amount of  $CH_4$  and  $CO_2$  were emitted from the top, while  $N_2O$  was emitted preferably from the sides of the pile, indicating that the spatial emission patterns of the three gases were not related. Such evidences highlight the fact that the sampling strategies need to be carefully planned in terms of chamber size, position and sampling frequencies in order to capture emissions from all areas and relevant phases of the process exhibiting different emission behaviour.

In conclusion, results from several studies indicated the need to further validate the chamber flux technique for estimation of GHG emissions from composting plants (Amlinger et al. 2008; Andersen et al. 2010a).

### 2.3.2 Other Specific Methodologies

There are other methodologies reported in literature that have been used to determine GHG emission rates.

The methodology proposed by Cadena et al. (2009) is based on direct sampling and simultaneous measurement of airflow rate. This methodology has been applied for plants where the composting process takes place in enclosed facilities with air collection and treatment, where it is supposed that all emissions are finally released



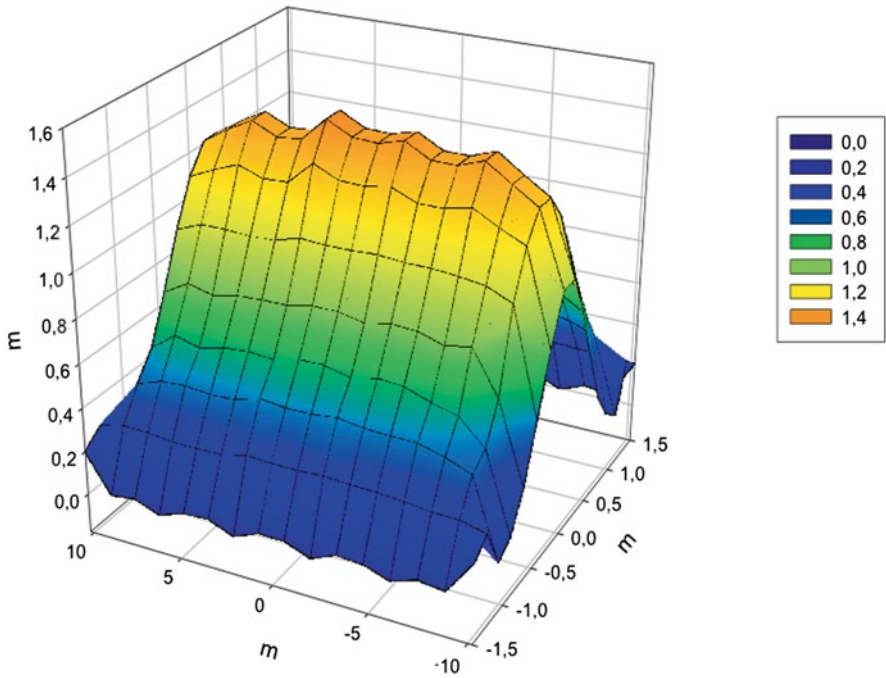


**Fig. 2.2** Image of fictitious partition of a biofilter surface to undertake the measurement of air velocity and gaseous emissions

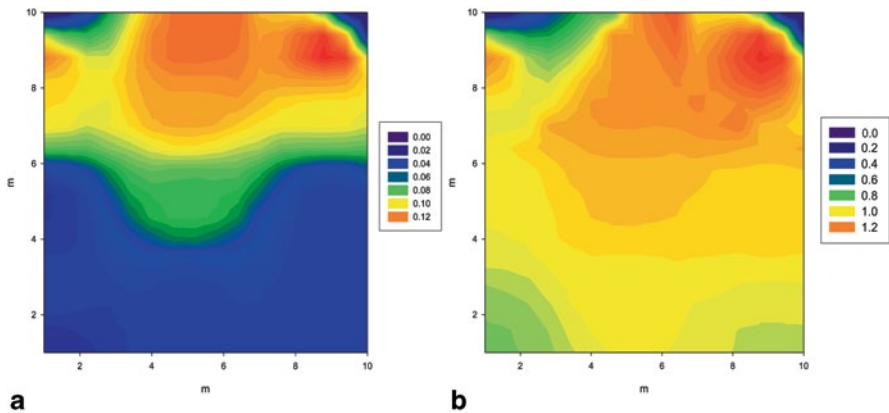
through a biofilter, and for plants using open air composting processes (i.e. turned or aerated windrows). This methodology is based on measuring the dimensions of the emitting surface (windrow or biofilter) and gas sampling and airflow rate measurement in different points of this surface (Fig. 2.2 and 2.3). When outlet air velocity is under the anemometer detection limit, a Venturi tube can be used to accelerate air velocity (Veeken et al. 2002). The product of compound concentration ( $\text{mg m}^{-3}$ ) and air velocity ( $\text{m s}^{-1}$ ) results in the mass flow of a given compound released per surface area unit ( $\text{mg s}^{-1} \text{m}^{-2}$ ). Thus, multiplying the pollutant mass flow per area unit by the corresponding total surface results in the compound mass flow. Then, the sum of the different quantities obtained corresponds to the total mass flow of a pollutant ( $\text{g s}^{-1}$ ) (Fig. 2.4). Measures of gaseous emissions must be repeated at different days during the composting process to determine the evolution of the emission of each compound. The periodicity of sampling must be established as a function of plant operation and the development of the composting process. Pollutant mass flows obtained for each sampling day are represented versus process time. The area below the curve obtained corresponds to the total mass of a given pollutant emitted throughout the composting process analysed.

The Funnel method proposed by Andersen et al. (2010a) is based on the use of a funnel shape instrument made of aluminium. In fact, it can be considered a modification of the chamber methods. The instrument is placed upside-down on the windrow to let the gases escape through the pipe of the funnel. Measurement of air velocity and gas sampling are performed in the pipe of the funnel. The funnel covers  $1 \text{ m}^2$  of the windrow top and the flow mass of the contaminant is calculated multiplying the contaminant concentration by the air velocity. The obtained flow is corrected with the ratio between the pipe area and the funnel area.





**Fig. 2.3** Example of a threedimensional representation of a composting pile



**Fig. 2.4** Image of gaseous emissions from a superior view of a biofilter. Axes correspond to the width and length of the biofilter (in m) and colours to the intensity of gas (in concentration): **a** ammonia; **b** total volatile organic compounds (VOC)

The Dynamic Plume method was designed for measuring emissions in landfills (Galle et al. 2001) and it has also been applied to manure storage emission in farms (Skiba et al. 2006). However, Andersen et al. (2010a) applied the dynamic plume method to a composting facility. This method was proven to be the most effective

when compared to the Funnel and the Dynamic Chamber methods. The method is based on the release of a tracer gas ( $\text{SF}_6$ , CO or  $\text{N}_2\text{O}$ , depending on the target gas) and its measure in the downwind plume. According to this, the dynamic plume method should be applied to the determination of the whole emissions of the studied facility. The ratio of the emission rates of tracer gas and contaminant are the same as the ratio of the concentrations of the tracer and the contaminant measured downwind, allowing the calculation of the contaminant emission rate. Measures upwind must be also performed to determine background concentrations.

Mathematical models such the Integrated Horizontal Flux (IHF) method or the backward Lagrangian stochastic (bLS) dispersion technique have been also used to measure emissions of GHG in composting piles (Sommer et al. 2004; Leytem et al. 2011). IHF method is based on a number of measurements along the vertical (downwind and upwind of the emission point) of contaminant concentration and windspeed (Wilson et al. 1983). bLS method allows estimating the emissions within the downwind plume from measurements of wind speed and contaminant concentration at specific heights and distances downwind (Flesch et al. 1995).

Finally, some optical methods developed for natural gas and other very specific fields have been applied to the measurement of fugitive emission of VOCs. Although not extended in the waste area of study, they are used in some cases when the precision need is high (e.g. Differential Absorption Lidar: DIAL) and they have been reported in some recent works (Steffens et al. 2009).

### 2.3.3 *Specific Analytical Methods and Sensors*

Gas chromatography (GC) has been commonly used to determine  $\text{N}_2\text{O}$ ,  $\text{CH}_4$  and  $\text{CO}_2$  concentrations in gaseous emissions from biological treatment processes. Different detectors, separation columns and operating conditions have been proposed.

Methane concentration is determined by GC using a Flame Ionization Detector (FID) and  $\text{CO}_2$  concentration by means of a Thermal Conductivity Detector (TCD). Sommer et al. (2004) determined  $\text{CH}_4$  and  $\text{CO}_2$  simultaneously using TCD in series with a FID and a  $2\text{ m} \times 3\text{ mm}$  SS Poropack QS 80/100 pre-column followed by a  $0.5\text{ m} \times 3\text{ mm}$  SS Poropack N 80/100 column. Carrier gas was helium at  $30\text{ ml min}^{-1}$  flow rate being temperatures of the column oven, TCD and FID, 55, 130 and  $230\text{ }^\circ\text{C}$  respectively. Martínez-Blanco et al. (2010) used a HP-Plot Q column for  $\text{CH}_4$  ensuring a detection limit of  $1\text{ ppm}_v$ . The gas chromatography operation conditions were as follows: oven temperature,  $60\text{ }^\circ\text{C}$ , injector temperature,  $240\text{ }^\circ\text{C}$ , FID temperature,  $250\text{ }^\circ\text{C}$ ; carrier gas,  $\text{N}_2$ . The same gas detectors were also used by Börjesson and Svenson (1997) when measuring gaseous emissions from a landfill and by Hobson et al. (2005) in  $\text{CH}_4$  determination in the final phase of household waste composting. Methods for  $\text{CO}_2$  and  $\text{CH}_4$  in biogas samples require different sensibility because of the concentration range in which both compounds are found in biogas (Ward et al. 2011).

N<sub>2</sub>O was measured by GC using an Electron capture detector (ECD). Czepiel et al. (1996) dried gas samples across CaSO<sub>4</sub> before being injected into the GC injection loop. These authors used a 2 m × 3 mm Porapak Q column with a mixture of Ar (95 %) and CH<sub>4</sub> (5 %) as carrier gas. ECD calibration was performed over the range of 310 ppb<sub>v</sub> to 100 ppm<sub>v</sub> using N<sub>2</sub>O in N<sub>2</sub> standard gases. He et al. (2001) used the same column and carrier gas, but preceded by gaseous samples cleaning-up across two glass made columns packed with magnesium perchlorate and ASCARITE II (Thomas Scientific) to remove moisture and CO<sub>2</sub>, respectively Carrier gas flow rate was of 40 ml min<sup>-1</sup> and temperatures of the detector and oven were 340 and 80°C respectively. Sommer et al. (2004) also used a pre-column (0.5 m × 3 mm SS Poropak N 80/100) and a 2 m × 3 mm SS HayeSep D 80/100 column with a Ar (90 %)-CH<sub>4</sub> (10 %) mixture as carrier gas at 30 ml min<sup>-1</sup>. Temperature of column oven and detector were 55 and 330°C, respectively. Colón et al. (2012) used a HP-Plot Q column (30 m × 0.53 mm × 40 μm) with N<sub>2</sub> as carrier gas being operation temperatures: 60°C (column oven), 120°C (injector) and 345°C (detector). Detection limit was established in 50 ppb<sub>v</sub>.

Some authors simultaneously determine the concentrations of CO<sub>2</sub>, CH<sub>4</sub> and NO<sub>2</sub>. Mondini et al. (2010) when measuring soil respiration used a micro-GC especially designed for continuous gas analysis. The GC was equipped with two capillary columns, PoraPlot Q (fused silica, 10 m length, 0.25 mm ID, 8 μm Df) and Molsieve (fused silica, 10 m length, 0.32 mm ID, 30 μm Df), in which head pressure and temperature could be electronically programmed. The measure of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O was accomplished by means of a TCD detector. The concentration operating range is from 1 ppm<sub>v</sub> to 100 %, with a linear dynamic range of 10<sup>6</sup>. Operative conditions of micro-GC were: 30 s sampling time, 30 ms injection time, 120 s running time, 40 and 60°C column temperature for PoraPlot Q and Molsieve, respectively, 30°C injector temperature. The chromatograph was calibrated by injecting a mixture of pure standard gases of CO<sub>2</sub>, N<sub>2</sub>O and CH<sub>4</sub> at a concentration of 5000, 50 and 1000 ppm<sub>v</sub>, respectively. The detection limits of the system for CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O were 2, 4 and 1 ppm<sub>v</sub>, respectively. Cayuela et al. (2012) used the same procedure in the determination of GHG emissions during composting of lignocellulosic residues.

In addition to GC determination, other techniques have been presented, most of them with the advantage of being able to determine gas concentration on field without gaseous samples capture, transport and storage.

The photoacoustic field multi gas monitor has been widely used to determine GHG concentration. Andersen et al. (2010a) performed CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O and CO determination in gaseous emissions from home composting of organic household waste using an INNOVA 1312 model (Lumasense Technologies A/S, 2750 Ballerup, Denmark). These authors state that this instrument measures real-time concentrations, provides high accuracy over a broad concentration range and only one calibration is necessary per year (calibration is performed by the manufacturer). The equipment requires a water filter to ensure that no moisture was transferred to the measuring chamber. Calibration ranges were 1.5–10,000 ppm<sub>v</sub> for CO<sub>2</sub>, 0.4–20,000 ppm<sub>v</sub> for CH<sub>4</sub> and 0.03–50 ppm<sub>v</sub> for N<sub>2</sub>O (range for CO was 0.2–50 ppm<sub>v</sub>).

Also Fukumoto et al. (2003), Tamura and Osada (2006) and Ahn et al. (2011) used the same technique when measuring the presence of these compounds in gaseous emissions from swine manure, dairy manure and farm manure composting respectively.

Photoacoustic spectroscopy is the measurement of the effect of adsorbed electromagnetic energy on matter by means of acoustic detection. Laser-based photoacoustic detectors are able to monitor trace gases concentrations under atmospheric conditions with sensitivity of orders of magnitude better than conventional scientific instrumentation in non-invasively and on-line way under dynamic conditions (Harren et al. 2000).

CO<sub>2</sub> has also been measured in gaseous emissions from full-scale composting plants using an infrared sensor (Abd et al. 2007). FTIR (Fourier Transform Infrared) absorption spectroscopy was also used by Andersen et al. (2010b) to measure GHG emission from windrow composting of garden waste including particular gases such as N<sub>2</sub>O, CH<sub>4</sub> or CO. Manios et al. (2007) used a mobile gas analyzer to measure the volumetric composition of CO<sub>2</sub> and CH<sub>4</sub> in gaseous emissions from olive oil mill sludge composting in windrow piles. The equipment (GA2000, Geotechnical Instruments) was an infrared gas analyzer able to detect simultaneously CO<sub>2</sub>, CH<sub>4</sub>, O<sub>2</sub>, CO and H<sub>2</sub>S, originally used for landfill gas composition determination. This equipment is actually replaced by the GA5000 portable gas analyzer. Although not a GHG, VOCs have recently studied by on-line monitoring, in a clear advance with respect to current techniques (Shen et al. 2012).

## 2.4 Reduction of GHG Emitted from Composting

### 2.4.1 *Best Practices for the Minimization of GHG Emissions*

GHG emissions from composting can be minimized through diverse actions undertaken from different points of view: the material to be composted and the process performance.

#### 2.4.1.1 Feedstock and Initial Mixture

The effect of the composition of the mixture of wastes to be composted is critical in the amount and type of emissions derived from the process. High moisture content and high bulk density has been related to higher GHG emissions. An excess of water reduces free air space (FAS) and creates anaerobic sites where methane can be formed (Tamura and Osada 2006). A correct level of FAS ensures the proper aeration of the composting material both in forced and natural aerated systems and prevents anaerobiosis (Ruggieri et al. 2009).

The biochemical composition of the material to be composted also plays an important role on gaseous emissions, especially the C/N ratio. However, the bioavailability of these nutrients determines the carbon and nitrogen dynamics along the process and the derived emissions (Cayuela et al. 2012). Consequently, the C/N ratio assessment should be based on the biodegradable content (Puyuelo et al. 2011). Co-composting of complementary wastes to obtain a balanced initial mixture with a balanced porosity and biodegradable C/N ratio should significantly reduce the GHG emissions of the subsequent composting process.

#### 2.4.1.2 Composting Process

The composting process can be undertaken in different industrial systems. A general classification is made as open and closed systems. Contrary to open systems, closed systems present the collection of the exhaust gases to a gas treatment system.

Closed systems include closed reactors such as rotatory drums and composting tunnels, but also confined piles (with textile cover) or composting piles inside closed buildings with a gas management system. Plants with gas treatment systems present much lower environmental impact because process emissions are not released to the atmosphere (Colón et al. 2012). Discussion on how to treat GHG emissions is presented below. In this sense and according to published conclusions (Colón et al. 2012), a first technical recommendation to minimize GHG emissions would be to include gas treatment systems wherever possible.

Another important process parameter to consider is process temperature. Higher temperatures enhance volatile compounds volatilization resulting in higher gaseous emissions (Pagans et al. 2006b; Cayuela et al. 2012).

Open systems as static piles, turned piles and aerated windrows at open air have been studied to better understand gaseous emissions dynamics related to aeration strategies: airflow and pile turning. Different authors have highlighted the importance of airflow in gaseous emissions from composting. It is considered that a high airflow increases oxygen availability, avoiding anaerobic pockets and consequent methane formation, and dilutes gaseous emissions. On the negative side, a high airflow strips gaseous compounds present in the composting mass facilitating their volatilization (Pagans et al. 2006a). Jiang et al. (2011) reported that an increase in the aeration rate reduced methane emissions, but increased  $\text{NH}_3$  and  $\text{N}_2\text{O}$  emissions. Pile turning enhances the composting process by providing matrix homogenization (moisture and microorganisms redistribution) and particle size reduction. It also provides punctual oxygenation of the solid material and compaction correction. From a biodegradation point of view, turning is recommended to enhance the process. However, pile turning has been shown to have a negative effect on gaseous emissions, including GHG (Colón et al. 2012). The turning itself releases the entrapped gases within the pile. Ahn et al. (2011) reported that  $\text{CO}_2$ ,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  fluxes increased after turning due to greater gas diffusion rates resulting from porosity increased after turning. They recommend avoiding pile turning in the first stage of the process if the oxygen concentration and temperature of the pile are in an appropriate range. In a second stage, when oxygen levels within the pile increase

the formed methane is oxidized to  $\text{CO}_2$ . These authors suggest considering a turning plan to minimize  $\text{CH}_4$  emissions and maximize  $\text{CH}_4$  oxidation within the pile. Park et al. (2011) also reported higher emissions in turned systems than in aerated systems. When considering methane and nitrous oxide as  $\text{CO}_2$  equivalents, the non-aerated system provided the higher process emissions, followed by the turned system, the system aerated by natural convection and finally the forced aerated system, which presented the lowest process emissions. However, as pointed by the authors, when approaching the problem from an overall impact assessment, the energy consumed to aerate the pile contributes to total  $\text{CO}_2$  non-biogenic emissions. The operational activities can contribute to GHG of composting process more than the decomposition process itself (Lou and Nair 2011).

LCA tools impute the impact of both process emissions and emissions related to energy consumption (operational activities, aeration, turning and mass displacement within the plant) to assess the comparison of different waste management systems. In this sense, turned pile composting systems resulted in an overall higher impact than aerated systems (confined aerated windrows and tunnel) because of fuel consumption and turning that implies the above mentioned increase in gas emissions (Colón et al. 2012; Kong et al. 2012).

#### **2.4.1.3 Final Recommendations to Minimize GHG Emissions**

From the text above it can be stated that a critical point for the success of the composting process with minimal gaseous emissions is the disposal of the material in piles with a suitable size and porosity to favor homogeneous oxygen distribution. In non-aerated systems, this would enhance natural convection. In aerated systems it is recommended to adjust forced aeration to ensure aerobic conditions without providing air in excess. High air flows beyond oxygen needs can be justified to avoid the emissions increase due to high temperatures. To overcome these problems, new advanced controllers have been proposed to base the aeration on the oxygen uptake rate measured on-line (Puyuelo et al. 2010).

Besides the physical structure of the matrix, the mixture to be composted should present appropriate moisture content and a balanced biodegradable C/N ratio.

Despite of whether the composting system is open or closed, the operational activities that imply electricity or fuel consumption must be optimized to reduce the overall environmental impact of the process.

Finally, gas treatment (by biofiltration or other technologies) is recommended when possible as the final solution to minimize gaseous emissions to the atmosphere.

#### **2.4.2 Treatment of GHG Emissions**

A variety of technologies are available nowadays for treating emissions from composting processes. Selection of the best available technologies depends essentially

on the composition and gas flow rate to be treated. Amongst such technologies, chemical scrubbing combining acidic plus caustic scrubbers coupled to biological processes such as biofilters are the most common technologies installed in full-scale facilities (Artola et al. 2009). However, current reactors design and operation focuses on treatment of VOCs and ammonia as main pollutants contained in composting emissions while low attention has been paid to GHG treatment. In any case, biological systems still are the preferred alternative from an economical and environmental point of view for GHG removal since the low concentrations of GHG in composting emissions make existing physical-chemical technologies non-viable from an economical point of view.

Acidic scrubbers preceding biofilters are installed to reduce the large ammonia loads often generated during composting. Otherwise, ammonia may inhibit AOB and/or NOB that, concurrently, would hinder the performance of the biofilters (Gabriel et al. 2007). Caustic scrubbers are often installed to remove acid gases such as hydrogen sulfide and to absorb highly soluble VOCs emitted such as alcohols. Biofilters have demonstrated to work well as end-of-pipe systems to treat a variety of odorant compounds found in off-gases from composting systems.

Design and operating conditions of chemical scrubbers and biofilters do not provide suitable conditions for the treatment of GHG. Dimensionless gas-liquid Henry coefficients for  $N_2O$ , NO,  $CH_4$  and CO of 1.7, 21.5, 29.2 and 43.1 (Sander 1999), respectively, indicate that GHG are sparingly soluble in water. Except for  $N_2O$ , which can be considered as moderately soluble in water, large gas contact times in the treatment system are required to solubilise significant amounts of NO,  $CH_4$  and CO which, consequently, leads to large reactor volumes and investment costs. In addition, the relatively low concentrations of GHG in the gas phase provide low driving force for GHG mass transfer from the gas to the liquid/biofilm phase. Chemical scrubbers generally operate at gas contact times below 2–3 s and retain large amounts of water within the packed bed to facilitate the absorption of soluble compounds (Gabriel and Deshusses 2003). Instead, biofilters are generally operated at gas contact times between 20 and 40 s for the treatment of composting off-gases with low to no external supply of water to improve sorption of poorly soluble compounds (Gabriel et al. 2007; Pagans et al. 2006a). Altogether leads to reduced elimination capacities of GHG in both systems in conventional chemical scrubbers and biofilters.

A short number of references exist about GHG treatment capacities in biofilters from composting emissions, even if several references exist about  $CH_4$  removal by biofiltration. The latter has been addressed by several authors and shown as an effective technology for biofiltration of landfill biogas or gaseous emissions from the piggery industry (Nikiema et al. 2007; Girard et al. 2012). Moderate-to-large  $CH_4$  concentrations of such gases are partly responsible for such effectiveness and treatment capacities. In composting facilities with biofilters, where much lower methane concentrations are found, removal efficiencies between 33 and 100% have been reported for  $CH_4$  (Boldrin et al. 2009).

In the case of  $N_2O$  emissions, Amlinger et al. (2008) reported that additional  $N_2O$  may be synthesized during the oxidation of  $NH_3$ . Also, Maia et al. (2012) found a



clear correlation between the  $\text{NH}_3$  load and the  $\text{N}_2\text{O}$  production in a compost biofilter demonstrating that  $\text{NH}_3$  removal was a trigger for  $\text{N}_2\text{O}$  production. Clemens and Cuhls (2003) studied the emission of direct and indirect greenhouse gases in a MBT facility. They also found that biofilters had no net effect on  $\text{CH}_4$  and approximately 26% of the  $\text{NH}_3$  that was removed in the biofilter was transformed into  $\text{N}_2\text{O}$  when  $\text{NH}_3$  was the exclusive nitrogen source due to nitrifier denitrification.

Regarding NO emissions, Barnes et al. (1995) showed that removal efficiencies up to 90% can be achieved in a compost biofilter for NO concentrations of 500 ppm<sub>v</sub> at a gas contact time of 60 s if an external carbon and energy source were added. Similarly, Yang et al. (2007) found that NO concentrations in the range of 200 to 500 ppm<sub>v</sub> can be treated in aerobic and anoxic biofilters with a strong influence of the  $\text{O}_2$  percentage on NO removal. Even if hardly difficult to implement in composting facilities, anoxic conditions were reported to almost double NO removal compared to aerobic biofilters.

It is interesting to notice that almost no studies exist concerning CO biofiltration. Prado et al. (2008) reported CO removal efficiencies higher than 80% for low (40 ppm<sub>v</sub>) CO concentrations from synthetic-resin producing industries in a biofilter operated at above 30 s gas contact time. Further optimization showed that a maximum elimination capacity of 33 g CO m<sup>-3</sup> h<sup>-1</sup> could be obtained with a mixture of lava rock and peat as packing material with more than 85% removal efficiency at gas contact times of 3 min or more suggesting that biofiltration offers potential for the biological removal of CO from polluted gas streams (Jin et al. 2009).

Reported data on GHG removal in chemical scrubbers is inexistent. However, one can infer from biofilters design, operating conditions and performance that GHG removal efficiencies in chemical scrubbers are probably very close to zero mostly due to the extremely reduced gas contact time of the gas in the scrubber coupled to the reduced solubility of most GHG.

Most of the research efforts on biological processes for GHG removal have been directed towards the use of existing bioreactor configurations (bioscrubbers, biotrickling filters or biofilters) while improving methane solubility using other solvents different to water. As reviewed by Muñoz et al. (2007), two-phase partitioning bioreactors (TPPBs) provide a non-aqueous phase (e.g. hexadecane, silicone oil) to an aqueous phase that contains the microorganisms responsible for degrading  $\text{CH}_4$ . Larger  $\text{CH}_4$  mass transfer coefficients are encountered in TPPBs compared to conventional biofilters. Thus, improved solubilisation of hydrophobic compounds and, concomitantly,  $\text{CH}_4$  elimination capacities are found. Rocha-Rios et al. (2009) reported increases of 131 and 41% in the specific and volumetric  $\text{CH}_4$  elimination capacity, respectively, in a biotrickling filter when silicone oil was added compared to the elimination capacities without silicone oil addition. However, silicon oil is expensive and difficult to manipulate which may hinder its use in full-scale systems. Alternatively, non-ionic surfactants do not pose the abovementioned problems and have shown to improve  $\text{CH}_4$  elimination capacities in biofilters, even if some growth problems may exist leading to decreased biomass accumulation in the packed bed due to their detergent character (Ramirez et al. 2012). Similarly, ionic liquids have shown to largely improve non-methane-VOCs absorption in biological

reactors without much toxicological issues (Quijano et al. 2010; Darracq et al. 2012). Such ionic liquids can be specifically designed based on the characteristics of the gaseous compound to be selectively separated (Carvalho and Coutinho 2011), which provides potential application for improving  $\text{CH}_4$  absorption in biofilters and biotrickling filters.

Overall, there are a number of opportunities to improve GHG removal by means of biological reactors. While  $\text{CH}_4$ , CO and NO can be treated to a certain extent in conventional biofilters already installed in full-scale composting facilities,  $\text{N}_2\text{O}$  has been shown to be generated rather than removed in biofiltration systems. Thus, research efforts should be directed towards reducing  $\text{N}_2\text{O}$  generation during the composting process and improving biofiltration conditions to reduce its production. Also, proper characterization of current biofiltration systems installed in composting facilities in terms of GHG treatment capacities is necessary to gain specific knowledge. Finally, design and operating conditions of end-of-pipe systems should not be only based on odours and ammonia removal, but also GHG loads should be taken into consideration.

## 2.5 Conclusions

GHG from composting are an important issue for research and for improvement in real-scale composting facilities. From this review, it is evident that now GHG can be accounted, measured and properly characterized. However, it is clear that the disparities of emissions factors for the different GHG that can be found in scientific literature are due to several factors:

1. The diversity of wastes and technologies used for the composting of organic wastes.
2. The absence of a consensus in the way to measure GHG, especially in open systems, where the flow is not controlled.
3. There is wide margin to minimize the GHG emissions from composting, by changing or updating the current facilities and by improving the performance of the treatment technologies.
4. The beneficial uses of compost must be also investigated, since it is not clear if the GHG emitted during the process are compensated by this compost utilization in the long term.
5. From a Life Cycle Assessment perspective, it is necessary to have experimental data both on the GHG emissions and the efficiency of the process, to have a fair evaluation of the environmental impacts of composting.

Further research is necessary to solve these limitations and to provide reliable emissions factors for composting processes and, in general, for any biological technology for waste treatment.

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