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Emission of CO_2, CH_4 and N_2O during Composting of Two-Phase Olive Mill Wastes: A Four-Year Study

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ABSTRACT

A four-year monitoring program was performed to quantify the emission of the three most important greenhouse gases (GHG), CO_2, CH_4 and N_2O, emitted during composting of two-phase olive mill wastes (TPOMW). A composting pile was prepared with typical agro-industrial wastes generated from olive oil mills: TPOMW and olive tree prunings, used as bulking agent, enriched with urea as N source. The study was performed during four consecutive years in order to obtain a wide set of experimental data to evaluate the effect of the variability of the raw material characteristics and the performance of the composting plant on GHG emissions. Biogenic CO_2 was the main gas generated during TPOMW composting. The range of CO_2 emission exhibited low interannual variation regardless the operational conditions at the composting plant. The improvement of the composting operations (better control of pile moisture and improved aeration) reduced the amount of CH_4 and N_2O emitted from the pile by reducing the formation of anaerobic spots within the pile. The control of the CH_4 and N_2O emissions reduced the contribution of TPOMW composting to climate change, expressed as kg of CO_2 equivalents.

Keywords: composting, greenhouse gas, methane, nitrous oxide, manure, olive mill wastes

INTRODUCTION

The interest on the evaluation of the environmental impact of waste management on greenhouse gas emissions (GHG) is growing over the last decades. The most important greenhouse gases generated during organic waste composting are CO_2, CH_4 and N_2O. CO_2 is the most abundant gas generated from organic matter degradation, however, it is not considered for the evaluation of the global warming potential (GWP) since it has a biogenic origin (IPCC 2006). For this reason CH_4 and N_2O have attracted the attention of researchers in recent years since their global warming potential is about 25 and 298 times more potent than carbon dioxide, respectively (over a 100 year period).

The emission of these GHG has been recently reviewed for different composting operations (Brown *et al.* 2008) and different feedstocks such as manures (Kebreab *et al.* 2006) and municipal solid wastes (Lou and Nair 2009). CH_4 is generated in anaerobic spots within the composting matrix even in well managed piles. The high microbial activity characteristic at early stages of the process leads to the decline in O_2 concentration favouring the formation of anaerobic spots where CH_4 can be generated. In the case of N_2O, its generation is commonly associated to nitrification and denitrification process taking place either at the beginning or after the bioxidative phase of composting, when pile temperatures are below 40°C. The characteristics of the raw materials and the composting operations can also affect the generation and diffusion of these gases to the atmosphere (excessive moisture content, poor aeration, physical compaction of the composting materials, etc). For this reason it is important to study the GHG emission for different organic materials in order to understand the mechanisms involved for each particular feedstock and different operational conditions.

Composting of two-phase olive mill wastes (TPOMW), the main by-product generated in olive oil processing industry, represents and important waste management strategy in Mediterranean areas for the recycling of this particular waste (Roig *et al.* 2006). TPOMW are usually treated by windrowing in open air composting pads located close to the olive mills. Despite the increasing number of TPOMW composting plants in operation in Mediterranean areas there is only little information on the GHG emissions from such facilities and the effect of chemical composition of TPOMW on these emissions (Manios *et al.* 2007; Sánchez-Monedero *et al.* 2010). TPOMW is a semisolid waste characterised by a slightly acidic pH, a high organic matter content (mainly lignocellulosic), a relatively low amount of available N and the presence of phenols and fats (Roig *et al.* 2006). The large moisture content and the presence of fats in TPOMW favour the compaction of these wastes and prevent the correct aeration of the pile, increasing CH_4 generation. The slow degradation rate of TPOMW and the limited amount of mineral N reduces the generation of both gases during composting but the addition of different bulking agents and N sources can markedly modify the emission rates (Sánchez-Monedero *et al.* 2010)

The aim of this study was to quantify the emissions of CO_2, CH_4 and N_2O during TPOMW composting during four consecutive years in order to evaluate the effect of the variability in the chemical composition or raw wastes and operational conditions on the emission of the most relevant GHG. The interannual evaluation would also allow to asses the contribution of TPOMW composting operations to climate change by calculating the global warming potential for each year as the sum of the total CO_2, CH_4 and N_2O emissions (expressed as kg of CO_2 equivalents).

MATERIALS AND METHODS

Compost elaboration

Composts were prepared during four consecutive olive oil seasons

Received: 15 March, 2011. Accepted: 6 July, 2011.

Original Research Paper

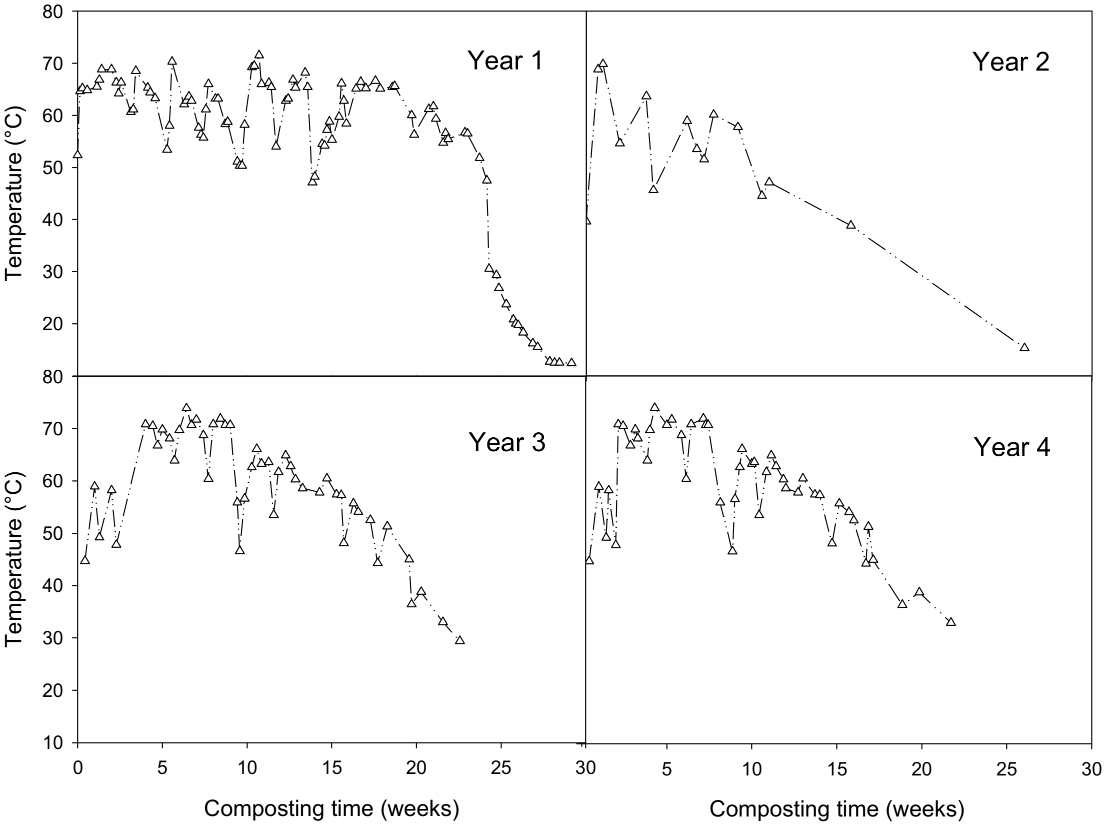

Fig. 1 Temperature evolution of the composting mixture in different production years.

in a pilot-scale composting plant treating the organic wastes obtained from the experimental farm "Venta del Llano" (IFAPA), located in Jaén (Spain). The same composting mixture was prepared every year by mixing fresh two-phase olive mill waste (TPOMW), olive tree prunings and urea. The dry weight proportion of the starting mixture was: TPOMW (30%) + olive tree pruning (70%) + urea (9 kg/tonne). This composting mixture represents one of the most typical compositions being composted in olive oil production areas since it combines the most abundant organic wastes generated in olive oil mills and olive orchards. This mixture was enriched with urea to achieve an initial C/N ratio in the range within 25-30.

About 5000 kg of this mixture was composted by windrowing in a trapezoidal pile (5 m length, 2 m width on the base and 1 m height) as already described for similar composting mixtures (Serramiá *et al.* 2010). The pile was turned every two weeks during the bio-oxidative phase (approximately three months), and the mixture was then allowed to mature over a period of two months. The moisture of the pile was controlled every two weeks and adjusted by adding the necessary amount of water to keep the levels close to 40% during the first two years and between 30 and 40% in years 3 and 4. Pile temperature was measured daily during the bioxidative phase of composting and then every week until the end of the process.

Gas sampling and analysis

A static closed chamber technique was used to measure CO_2, CH_4 and N_2O fluxes from the surface of the composting piles (Sommer *et al.* 2004). Gas samples were taken once a week (during the bio-oxidative phase of composting) and approximately every two weeks (during maturation) from five aluminium closed chambers (volume: 0.010 m^3, area: 0.038 m^2) placed at different locations on the top of the pile. The chamber were pushed 5 cm into the composting pile and air samples were taken within the headspace at 0, 15 and 30 min using disposable syringes. Air samples were transferred to evacuated 5 ml glass vials (Vacutainer®) fitted with butyl rubber stoppers and analysed within 24 hours by gas chromatography (VARIAN CP-4900 Micro-GC). Emission fluxes were calculated by fitting the experimental data to a second-order polynomial equation (gas concentration *vs* time). The flux at time 0 was calculated by taking derivates of the second order polynomial (Hao *et al.* 2001).

The estimation of the global warming potential (GWP) of the composting operation was obtained by multiplying the cumulative mass of CH_4 and N_2O emitted per year by 25 and 298, respectively, in order to obtain the total amount of equivalent CO_2 (IPCC 2006).

Experimental design and statistical analyses

A composting pile was prepared during four consecutive years with the same initial composition (same proportion of organic wastes in the starting mixtures) in order to study the interannual evolution of the process. A GHG monitoring program, consisting of 18 to 20 sampling events, was performed every year during the composting process, as previously described. Gas fluxes were expressed as grams of C or N per unit of area (m^2) and per time (day) and represented the mean and standard error of 5 replicates (n=5).

RESULTS AND DISCUSSION

Performance of the composting process

The general performance of the composting process was evaluated by the evolution of the temperature of the composting piles (**Fig. 1**). The four composting piles exhibited the typical pattern of olive mill waste composting, characterized by a long duration of the process, up to 35-40 weeks, as a consequence of the lignocellulosic composition of the material and the low amount of available N (Cayuela *et al.* 2006; Alburquerque *et al.* 2009). The piles were characterized by long bioxidative phases (around 15 weeks) with high temperatures above 50°C and a maturation phase where pile temperature slowly decreased. The different temperature evolution observed during the four years of the experiment may reflect the variation in the chemical and physicochemical characteristics of the starting materials and also the performance of the composting plant. Since only minor differences in chemical composition of TPOMW and olive tree prunings were expected within olive oil seasons, the composting plant operation can play a key role on the general performance of the process. Composting piles prepared

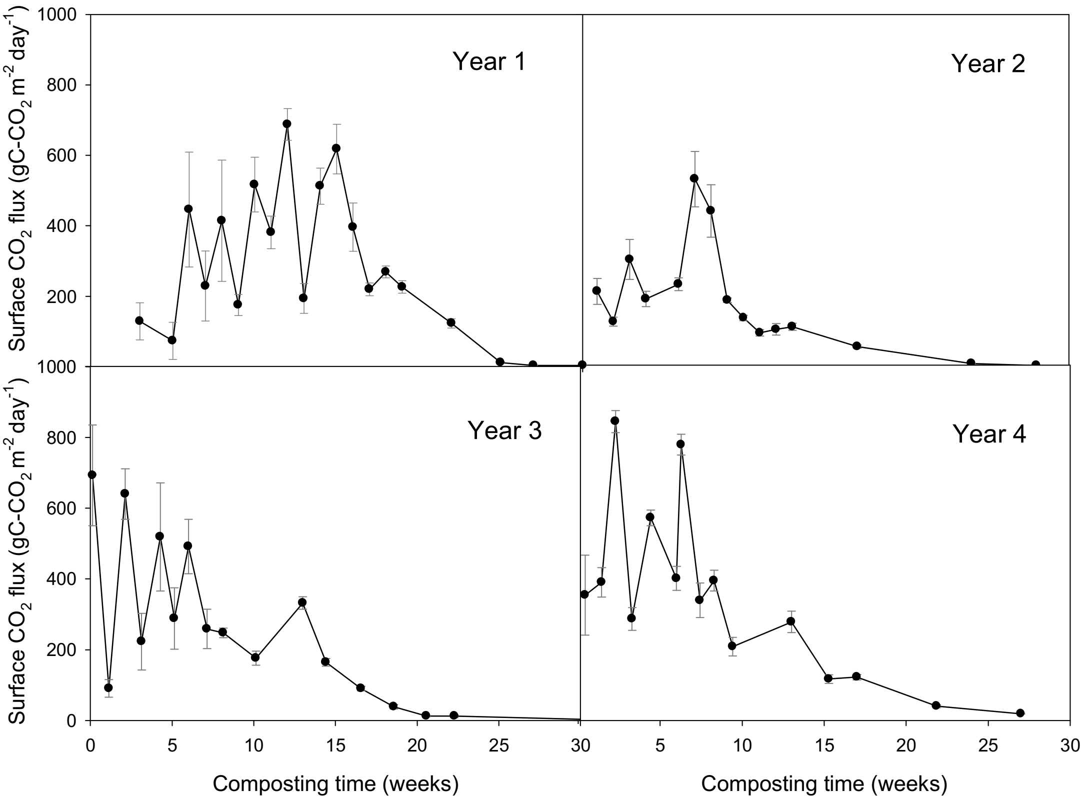

Fig. 2 Surface CO_2 fluxes emitted from the composting pile during different production years. Bars represent the standard error (n = 5).

in years 3 and 4 showed very similar patterns reflecting the experience reached by the composting plant operators in optimizing the composting process (pile size, moisture control, turning skills, etc.). Sánchez-Monedero *et al.* (2010) reported the variation of the typical stabilisation indices during the first year of composting operation.

CO_2, CH_4 and N_2O production during composting

CO_2 was the main gas generated during composting as a consequence of microbial degradation of organic matter. Thus, the emission of CO_2 was closely related to the activity of the composting pile, and both surface CO_2 flux (**Fig. 2**) and temperature (**Fig. 1**) showed parallel trends in the four composting piles. The largest CO_2 fluxes were registered within early stages of the process, during the bioxidative phase whereas a steady decline of CO_2 emission was observed during the maturation phase, corresponding to a decrease in microbial activity. Biweekly turnings during the bioxidative phase originated peaks of CO_2 emissions as a consequence of reactivation of the composting process (homogenisation and water addition). Surface CO_2 fluxes ranged between 200 and 800 gC-CO_2 m^{-2} day^{-1} in the first half of the process and they were reduced to less than 10 gC-CO_2 m^{-2} day^{-1} by the end of maturation. The surface gas fluxes showed a homogeneous distribution along the upper part of the pile surface and showed a relatively low coefficient of variation.

The range of surface CO_2 fluxes only showed little interannual variation reflecting that gas emission was not greatly affected by the origin of the starting materials or the operating conditions at the composting plant. During the first two years of operation, a lag phase was observed in the CO_2 production which was probably caused by the performance of the composting plant and the moisture of the piles (higher during the fist two years of the study). After these initial years, the composting pile showed very similar CO_2 emission patterns as a consequence of the optimisation of the composting operations (turning and moisture) and the training of plant operators.

Despite the prevalent aerobic conditions in composting piles, CH_4 can be generated at early stages of the process when the available O_2 is partially depleted as a consequence of the large microbial activity and high temperatures. Surface CH_4 fluxes were generally lower than 30 gC-CH_4 m^{-2} day^{-1} (**Fig. 3**) during composting, which are below the common surface CH_4 fluxes registered during composting of manures such as beef feedlot and liquid swine, from 20 to 1250 gC-CH_4 m^{-2} day^{-1} (Kebreab *et al.* 2006). During the first two years of operation, large peaks of CH_4 production, up to 170 gC-CH_4 m^{-2} day^{-1}, were registered during the active phase of composting. Contrary to CO_2 emissions, surface CH_4 fluxes showed a large coefficient of variation along the pile surface, reflecting the presence of small anaerobic spots within the pile, rather than a uniform distribution of CH_4 production.

The large interannual variation exhibited by the surface CH_4 fluxes suggests that the emission of CH_4 is likely to be affected by the operational conditions and experience of site workers. In fact, the emission of CH_4 was markedly reduced in the last two years of operation, where the peaks of production were significantly reduced by an improved performance of the process. In the last two years of the experience the moisture levels were kept lower than in previous years 1 and 2 to avoid the compaction of the material. Turnings were also improved in the last to years to allow a better aeration and homogenisation of the composting pile.

The emission of N_2O was generally low (below 200 mgN-N_2O m^{-2} day^{-1}) except in year 2 where two peaks up to 1200 mgN-N_2O m^{-2} day^{-1} were registered at the end of the bioxidative phase (**Fig. 4**), when pile temperature started to decline. Sanchez-Monedero *et al.* (2010) found that N_2O emission was highly dependent on the N source used for olive mill waste composting and they observed that the use of urea could significantly increase the generation of N_2O due to the large availability of mineral N (ammonium and nitrate) in the mixture. The peak observed in year 2 may be related to the limited O_2 availability in the composting mixture, which is a key element controlling the N_2O generation during composting (Czepiel *et al.* 1996), and also to the decrease in pile temperature below 50°C after the 10th week (**Fig. 1**), which allowed the activity of nitrifying bacteria. In

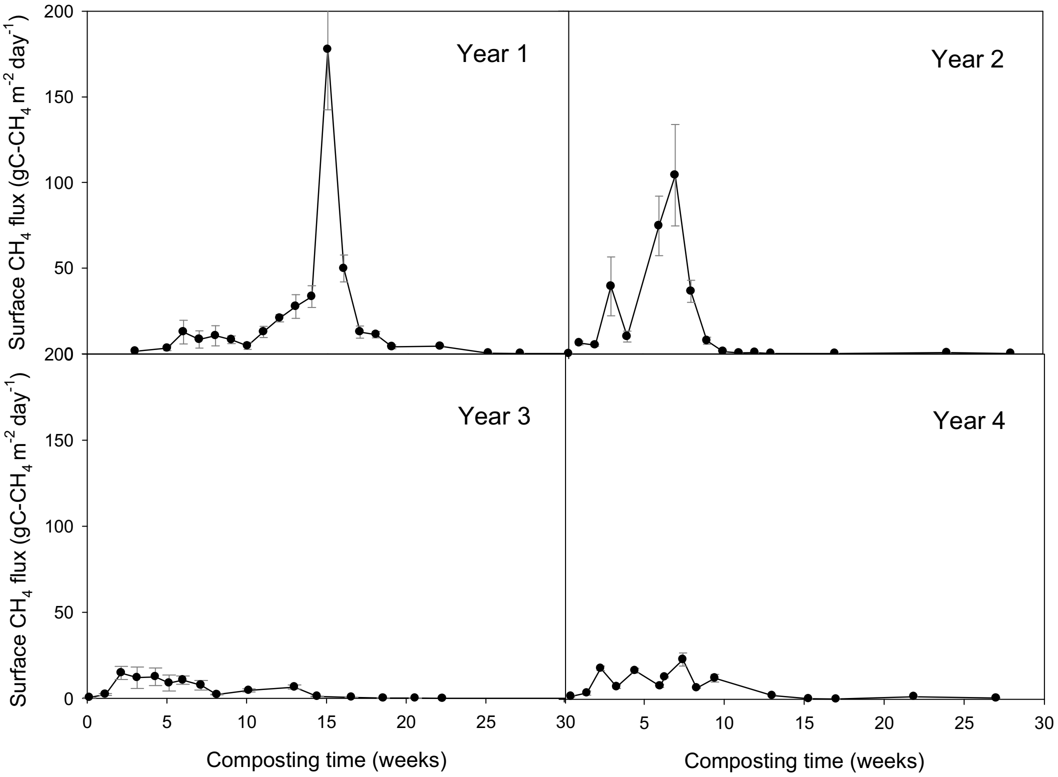

Fig. 3 Surface CH_4 fluxes emitted from the composting pile during different production years. Bars represent the standard error (n = 5).

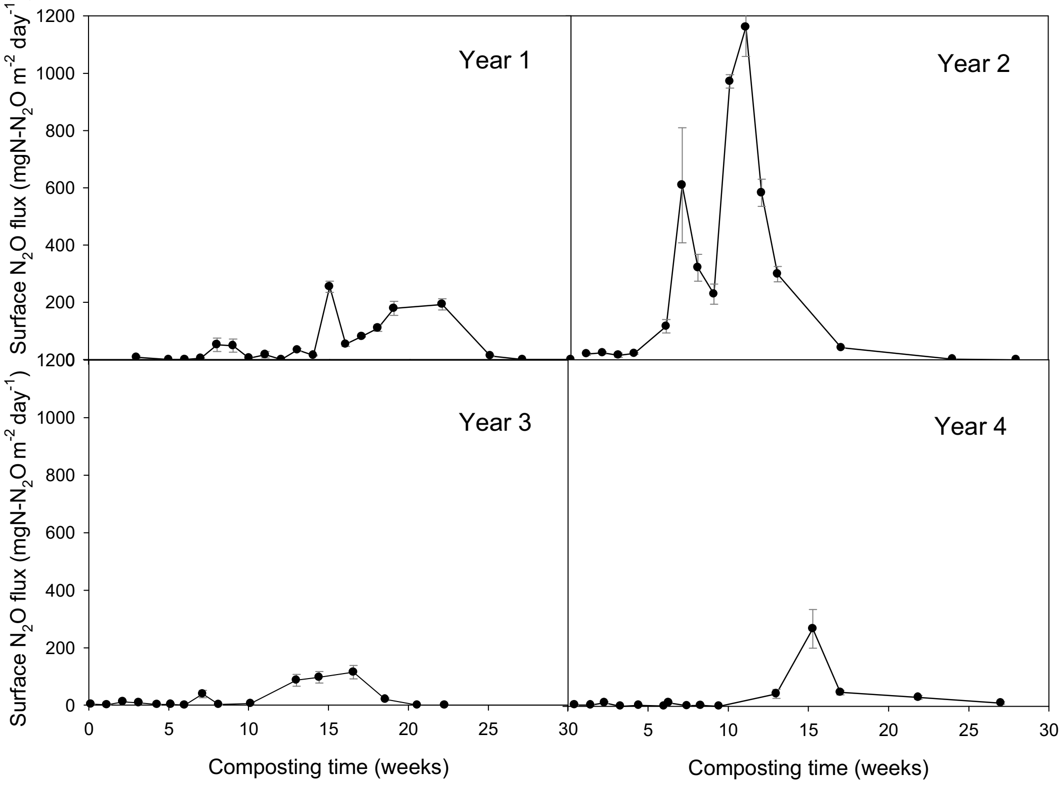

Fig. 4 Surface N_2O fluxes emitted from the composting pile during different production years. Bars represent the standard error (n = 5).

the case of TPOMW composting, Sánchez-Monedero *et al.* (2010) reported that nitrification was thought to be the main source of N_2O emission which was affected by the presence of mineral N and O_2 availability in the pile.

Surface N_2O fluxes exhibited a homogenised distribution along the pile as observed in the low coefficient of variation of the measurements. However, the peaks observed in year 2 were characterised by large standard deviation, as already observed in CH_4 emission. The large spatial variability of N_2O emissions can be due to the presence of anaerobic spots within the pile, with limited O_2 availability, which favours the formation of N_2O by nitrifying bacteria (Béline *et al.* 1999).

Evaluation of GWP of olive mill waste composting

Global warming potential (GWP) was calculated as the sum of total kg of CO_2, CH_4 and N_2O (expressed as kg of CO_2

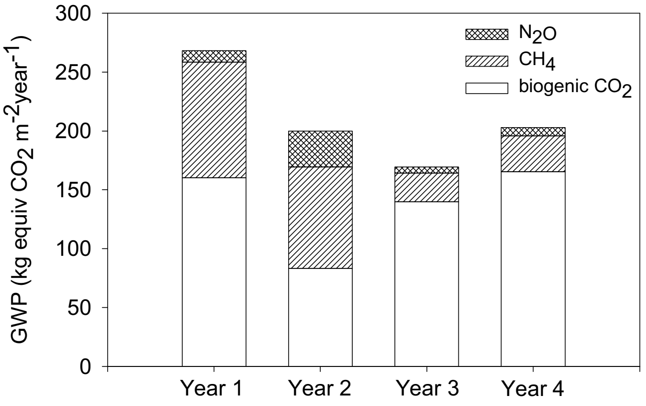

Fig. 5 Global warming potential (GWP) for each year calculated as the sum of total kg of CO_2, CH_4 and N_2O (expressed as kg of CO_2 equivalents) per compost surface unit.

equivalents) per compost surface unit in order to compare the contribution to climate change of each greenhouse gas emitted during composting (**Fig. 5**). During the four campaigns CO_2 represented the gas with the highest contribution, however, according to Smith *et al.* (2001) for biodegradable materials which carbon have been recently absorbed from the atmosphere by photosynthesis: "*... there is no net global warming impact, and no global warming potential is associated with the CO_2 emission, ... These emissions are reported as 'short term CO_2' or 'biogenic CO_2' and are given a global warming potential of zero.*"

The second most important contribution was CH_4, which GWP was substantially high the first two years. Nevertheless, CH_4 emissions were successfully reduced the last two years mainly as a consequence of moisture optimization and the acquired experience of the operators at the composting facility for improved aeration.

N_2O emissions were more difficult to control since N_2O can be produced during both nitrification (aerobic) and denitrification (anaerobic) processes and it seems to be mostly influenced by the original materials used for composting. However, its share to total GWP was always the lowest.

CONCLUSIONS

The levels of CH_4 and N_2O emissions generated during TPOMW composting were relatively low compared to other organic waste such as manures and municipal solid wastes. A proper management of composting operations, in terms of improved aeration and moisture control, can significantly reduce the contribution of TPOMW composting to global change, as far as GHG are concerned.

ACKNOWLEDGEMENTS

The authors wish to thank the Spanish Ministry of Science and Innovation for supporting research project Ref. CTM2009-1473-C02-02 under which this work was financed. Field staff of IFAPA Centro "Venta del Llano" is grateful acknowledged for its assistance in compost preparation.

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