

## Methodology for the assessment of the greenhouse gas impact of wastewater sludge treatment

A. Åkerman, A. Massagué, S. Mehier, E. Senante, I. Escaler, J. L. Martel and J. M. Audic

### ABSTRACT

Nitrous oxide (N<sub>2</sub>O) and methane (CH<sub>4</sub>) emissions from the wastewater sector represent the second largest source of greenhouse gas (GHG) emissions of the waste sector. A protocol has been developed to measure GHG from sewage sludge treatment. GHG measurements were performed on two industrial sewage sludge composting plants. Measurement campaigns showed GHG emissions range between 0.18 to 3.40 kgCH<sub>4</sub>/t dry sludge and 0.74 to 0.48 kg N<sub>2</sub>O/t dry sludge. Converted to CO<sub>2</sub> equivalent to assess climate change impact, emission factors of CH<sub>4</sub> range between 4.5 and 85 kg eq.CO<sub>2</sub>/t dry sludge and N<sub>2</sub>O range between 143 and 221 kg eq.CO<sub>2</sub>/t dry sludge when calculated with global warming potentials (GWP) of 25 and 298, respectively. In terms of climate change impact, N<sub>2</sub>O emissions contribute much more than CH<sub>4</sub> because of its higher GWP. N<sub>2</sub>O is linked to the treatment of nitrogen and produced during the nitrification and denitrification biological processes. Process emissions measured on the two sites were compared with indirect emissions linked to operational energy use.

**Key words** | composting, greenhouse gas, methane, nitrous oxide, sewage sludge

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### INTRODUCTION

The wastewater sector is defined by the US-EPA as one of the larger minor sources of greenhouse gas (GHG) emissions. Nitrous oxide (N<sub>2</sub>O) and methane (CH<sub>4</sub>) emissions from the wastewater sector represent the second largest source of GHG emissions of the waste sector (Bogner *et al.* 2007). An overview study of N<sub>2</sub>O emissions in wastewater and solid waste treatment indicates that the wastewater treatment sector may be the most important operation for managing anthropogenic nitrogen in wastes (Barton & Atwater 2002).

Besides direct GHG emissions, wastewater treatment processes contribute to indirect emissions through the use of fossil energy sources and chemicals. Different national and international accounting tools and methodologies have been developed to quantify GHG emissions from wastewater treatment plants (WWTP). Today large uncertainties still remain with respect to direct emissions, indirect emissions and mitigation potentials for the wastewater sector (Bogner

*et al.* 2007). Hence, in order to reduce these uncertainties, it is important to obtain reliable data to be used in the GHG tools and to better understand the impact of direct emissions, with the aim of optimising wastewater treatment in terms of global GHG impact.

Research has been carried out to assess GHG emissions from wastewater treatment processes (Czepiel *et al.* 1995; Tallec *et al.* 2006, 2008; Foley *et al.* 2010). Recently a methodology has been developed and successfully applied on several WWTPs, enabling *in situ* measurements of direct N<sub>2</sub>O and CH<sub>4</sub> emissions from different wastewater treatment processes (Senante 2008; Vanden Bossche *et al.* 2009).

### GHG emissions and sewage sludge treatment

As for sludge treatment, some studies have been carried out to assess direct emissions from different sewage sludge

treatment processes such as composting (Czepiel *et al.* 1996; Amlinger *et al.* 2008) and incineration (Susuki *et al.* 2003; Gutierrez *et al.* 2005; Svoboda *et al.* 2006). However, GHG emissions from sewage sludge treatment remain in general poorly quantified and no or little work has been performed on an approach to assess the overall GHG impact. Therefore, a comprehensive study has been undertaken with the main objective of developing a methodology to assess the global GHG impact of the whole sludge treatment line.

This paper focuses on measurements performed on sludge composting plants located in France and Spain. First the sampling and analysis methods are described. Then the results from the on-site measurement campaigns and GHG emission factors are discussed.

## METHODS

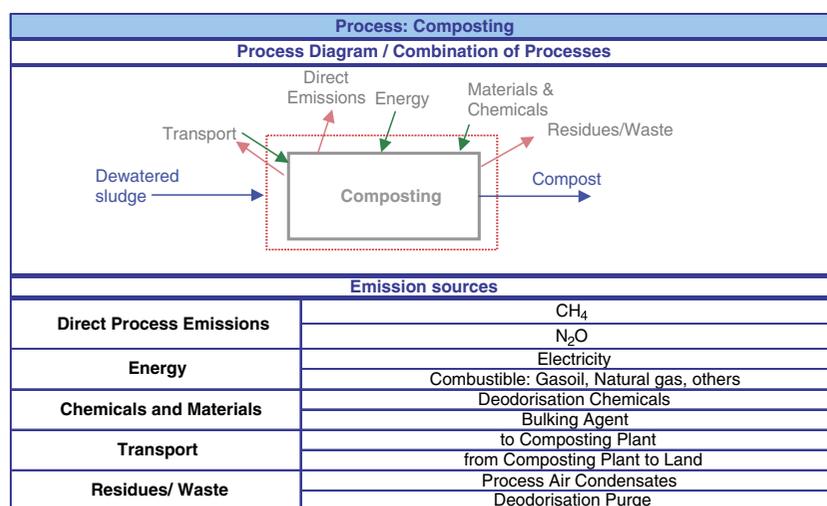
### Goal and scope definition

The current paper aims to assess direct GHG emissions on WWTP from the sewage sludge treatment and provide a methodology to assess GHG emissions in each specific process of sludge treatment. This study has been applied to sewage sludge treatment; that is, biological or thermal treatment. The methodology consists of accounting for indirect emissions linked to operational energy use, chemicals and

material consumption, freight and direct N<sub>2</sub>O and CH<sub>4</sub> process emissions. In order to obtain emission factors for sludge treatment processes, on-site measurement campaigns of N<sub>2</sub>O and CH<sub>4</sub> emissions have been performed. Capital footprint and end-use and/or final destination of treated sludge were excluded from the scope of this study. However it must be kept in mind that, for a more holistic approach, streamline life-cycle activities should also have been quantified; that is, emissions of GHG from sewer collection systems and wastewater treatment plant and all final sludge disposal (agricultural land application, landfilling).

First, each sludge treatment process with its defined boundaries and emission sources were described separately in process boxes as illustrated in Figure 1. Operational data concerning sludge treatment processes were collected on site. Emission factors for electricity use and fuel consumption are those provided by IEA (2009) and ADEME (2007), respectively.

Direct GHG emissions were directly emitted from the process units of the sludge treatment. It was assumed that 100% of the organic carbon in the raw sludge was biogenic. CO<sub>2</sub> emitted from composting is not fossil derived and therefore it was not considered as a GHG emission in accordance with IPCC (2006). CH<sub>4</sub> is linked to the presence of organic matter in anaerobic conditions. N<sub>2</sub>O is linked to the treatment of nitrogen and produced during the nitrification and denitrification of the nitrogen present in the sludge (IPCC 2006).



**Figure 1** | Process diagram and definition of the boundaries for GHG emissions assessment in the case of sewage sludge composting process; the main inputs and outputs of the system are illustrated by arrows; boundaries of the system are illustrated by an interrupted line.

To compare the effect between different gases, their relative global warming potential (GWP) was estimated and referenced to CO<sub>2</sub>. Carbon dioxide equivalency is a quantity that describes, for a given mixture and amount of GHG, the amount of CO<sub>2</sub> that would have the same GWP. The GWP is an estimate of the contribution a given mass of GHG makes to global warming. It depends on its capacity of adsorption of infrared radiation and its atmospheric lifetime. A GWP is calculated over a specific time interval. The GWP of CO<sub>2</sub> is by definition 1. The GWP of CH<sub>4</sub> and N<sub>2</sub>O are 25 and 298, respectively, considering a lifetime of 100 years (IPCC 2007). GHG emissions are expressed in tonnes of carbon dioxide equivalents (t CO<sub>2</sub>eq).

### Operational protocol for sampling and analysis of N<sub>2</sub>O and CH<sub>4</sub> air emissions

The protocol described hereafter was adapted from the protocol developed for on-site GHG emissions measurement of the water line at WWTP (Mehier et al. 2008).

### Sampling methods

Depending on the source of the GHG air emissions to be assessed, they can be either canalised emissions (from a chimney, pipe or exhaust gas stream) or diffuse emissions; that is, surface sources with specific air sampling systems for each type of emission. An area source is defined as a diffuse emission source of a certain surface area. There are two types: 'active' sources and 'passive' sources. Aerated or active sources are those area sources where there is a presence of blown air (regulated by the facility) and with internal gas production, while non-aerated or passive sources have no air-blowing system. The air sampling system differs according to whether the study zones are active or passive.

*Active area sources.* In the case of 'active' sources, the sampling is performed with a flux chimney that enables the isolation of a part of the emitting surface area. The chimney channels the emission enabling the assessment of airflow in order to then calculate the gas flow and thus the flow of N<sub>2</sub>O and CH<sub>4</sub>.

*Passive area sources.* In the case of 'passive' sources the sampling system used is the Odoflux dynamic flow chamber. Flow chambers cover an area to be studied as hermetically as

possible in order to isolate the surface from external conditions. The gases emitted by the isolated surface are collected by a vector gas, which is injected into the chamber.

*Canalised sources.* The channelled air emissions from sludge treatment processes such as composting, drying or incineration are sampled directly with the mobile N<sub>2</sub>O and CH<sub>4</sub> IR analysis system.

### Measurement methods

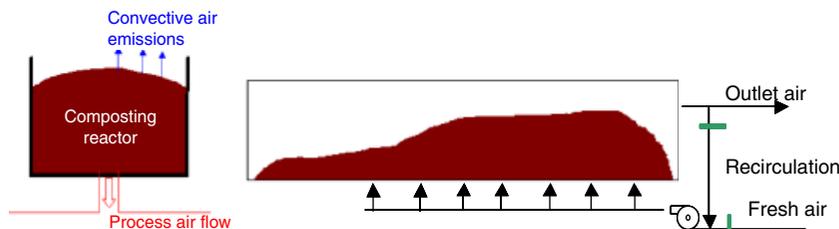
*Analysis of N<sub>2</sub>O and CH<sub>4</sub>.* A mobile analysis system for trace compounds CH<sub>4</sub>/N<sub>2</sub>O is used to continuously analyse the sampled air. The measurement ranges used are, for CH<sub>4</sub>, 0–500 ppm to an accuracy of 5 ppm and, for N<sub>2</sub>O, 0–50 ppm to an accuracy of 0.5 ppm. The measurement gas is sampled at each spur using semi-supple Teflon pipe, 4/6 mm in diameter. This pipe is then connected to a sampling unit which ensures the pre-conditioning of the measurement gas. When the measurement gas leaves the portable sampling unit, it is then connected to the Servomex IR gas analyser. Measurement data is recorded continuously by a numerical recorder. The concentrations measured are expressed in ppm volume/volume.

*Assessment of mass flow of N<sub>2</sub>O and CH<sub>4</sub>.* To calculate the mass flow of N<sub>2</sub>O and CH<sub>4</sub>, the measured concentration in the sampled air in units of ppm is converted into units of mg/m<sup>3</sup> depending on the gas flow and its temperature.

To constitute emission factors, N<sub>2</sub>O and CH<sub>4</sub> emissions can be compared with the content of nitrogen and carbon initially present in the sludge. Hence, quality parameters of the sludge must be analysed during the sampling period. The analysis includes parameters such as dried solids (DS), nitrogen Kjeldahl (NTK) and organic carbon (Corg). Dry matter content and N-Kjeldahl were determined by the methods NF EN 12880 and NF EN 13342, respectively. Organic carbon of input material was calculated from the volatile solids (VS) determined at 550°C in accordance with the NF EN 12879.

## RESULTS AND DISCUSSION

GHG emissions measurement was carried out for two composting sludge treatment processes. Both convective air



**Figure 2** | Air measurements on the two composting plants; convective air (i.e. passive area source gas sampling) and process airflow (i.e. canalised sources gas sampling) of composting plant A (left) and tunnel ventilation system (i.e. canalised source gas sampling) of composting plant B (right).

(plant A) and process air (plant A and B) were analysed (Figure 2).

Composting plant A treats about 42,000 t of dewatered sludge (DM content 16 to 33%) annually, coming from 30 nearby WWTP. The composting process consists of an accelerated aerobic fermentation phase followed by a ventilated maturation phase in a covered and closed building of 8,600 m<sup>2</sup>. The process comprises four main stages:

First, the raw sludge is mixed with new or recycled bulking agents (grinded green waste and wood pallets) in a mixing-loading machine, which has been loaded with a front loader. The volumetric ratio of sludge/bulking agent is 1/3. Second, the mixing-loading machine loads the mix in one of the 20 corridors or one of the six composting boxes. The fermentation lasts 3 weeks, with the pile being turned once a week and removed from one reactor to another. The aeration is continuous (variable flow) and ensured through negative aeration (suction). Third, after the compost mix has been unloaded with the front loader, the fresh compost is screened and the fine fraction is matured inside four positively aerated boxes during one month. The rejects (bulking agents/coarse matter) are recycled at the head of the process. Fourth, the matured compost is stored outside the building on a storage area for at least 2 months.

It should be noted that during the measurement campaigns performed within this study turning was not performed once a week, but occurred only after 14 days.

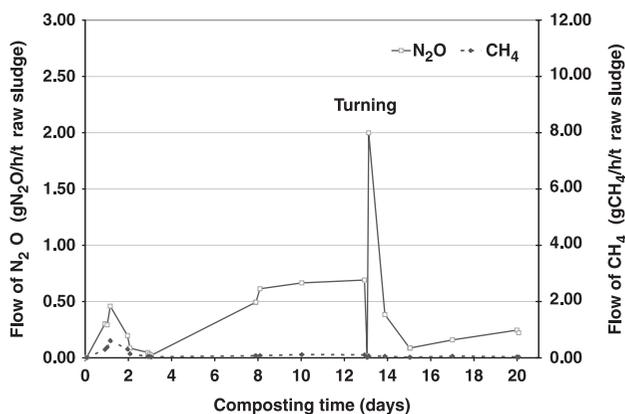
Composting plant B has a treatment capacity of 25,000 t of sludge per year (DM content 18 to 21%). The composting process is performed within six tunnels. The volumetric proportion for bulking agent (i.e. wood chips) is 4/1 and the process is enhanced with the addition of 10% of dried sludge. Each tunnel is loaded with a mix containing 250 t of sludge. Under normal operation, the six tunnels are managed differently: one of them is in loading phase, another in

unloading phase and the other four are in different stages of the fermentation process. The whole process takes from 11 to 14 days and the temperature is controlled in order to maintain optimal conditions. The six tunnels have a ventilation system, which extracts air from each composting tunnel and takes it for odour treatment.

Measurements were performed during five days on different tunnels at different fermentation stages.

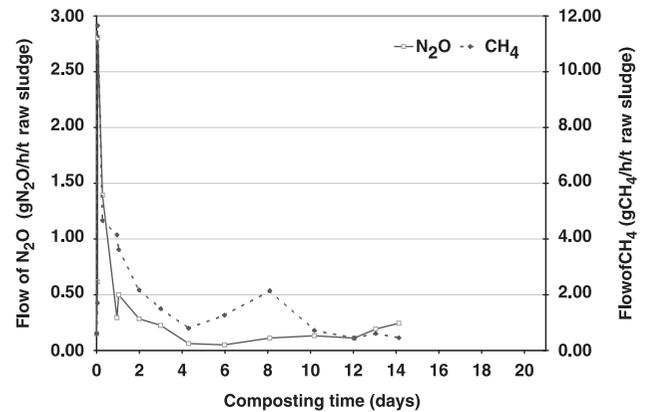
### Plant A

Results from the convective air GHG emissions measurements performed on plant A were found to be very low: 0.02 kg CH<sub>4</sub>/t orgC in sludge and 0.40 kg N<sub>2</sub>O /tNTK giving an emission factor inferior to 0.001% of initial organic C emitted as CH<sub>4</sub> and 0.03% of initial NTK emitted as N<sub>2</sub>O. Hence the convective GHG emissions were considered to be negligible compared with the process air GHG emissions, the results of which are presented hereafter. Figure 3 shows the results from the monitoring of the process air GHG emissions; that is, CH<sub>4</sub> and N<sub>2</sub>O mass flows during



**Figure 3** | Plant A: N<sub>2</sub>O and CH<sub>4</sub> emissions measured during sewage sludge composting (21 days).

the fermentation phase of the composting process on plant A. The following observations can be made: the results show an important peak of  $N_2O$  and  $CH_4$  at the start of the process. He *et al.* (2000) observed a similar pattern when measuring  $N_2O$  and  $CH_4$  from an aerated composting of food waste. This is probably due to the stripping of the gases contained in the sludge – previously generated during the storage and loading – that are stripped when the ventilation system of the composting is started.  $N_2O$  emissions were observed around day 7 to 14 probably linked to nitrification or denitrification processes.  $N_2O$  can be produced either during nitrification or denitrification under specific oxygenation levels. Furthermore a peak of  $N_2O$  emissions around day 13 is clearly linked to the turning of the compost pile. As reported in De Guardia *et al.* (2010) this is often mentioned in the literature, and emissions of  $NH_3$  and  $N_2O$  occurred either at the peaks of oxygen consumption rate or after turning. Fukumoto *et al.* (2003) also observed significant peaks of  $N_2O$  after turning during swine manure composting without forced aeration and concluded that, by turning the pile, nitrate produced in the aerobic portion has an opportunity to move to anoxic parts of the pile where nitrate is denitrified and  $N_2O$  is produced as a by-product. During the fermentation phase of the aerated composting, temperature was maintained above 45°C. Therefore the generation of  $N_2O$  from the nitrification process would not have occurred because temperature above 45°C inhibits specific enzymes responsible for nitrification (Mallard *et al.* 2006). The observed emissions of  $N_2O$  suggest the presence of anoxic microsites in the mix wastewater sludge-bulking agent leading to the formation and emission of  $N_2O$  from the aerated composting system.



**Figure 4** | Plant B, site A:  $N_2O$  and  $CH_4$  emissions measured during sewage sludge composting (14 days).

### Plant B

As for the process air GHG emissions, Figure 4 shows the results from the monitoring of the process air  $CH_4$  and  $N_2O$  mass flows during the whole fermentation phase of the composting process. Results from plant B show an important peak of  $N_2O$  and  $CH_4$  at the start of the process. The same pattern was observed in plant A and linked to the stripping of the gases previously contained in the sludge (i.e. generated during storage and loading) which are stripped when the ventilation system of the composting is started.

$CH_4$  and  $N_2O$  emission factors obtained by the on-site measurements are summarized in Table 1. Results show that 0.04 to 0.7% of the initial organic carbon content is emitted as  $CH_4$  and 0.5 to 1.0% of NTK content is emitted as  $N_2O$ , which is in the order of magnitude of the estimations reviewed in the literature.  $N_2O$  emissions from different kinds of waste have previously been studied. Mallard *et al.* (2006) estimated N- $N_2O$  emissions ranging between 0.1 and 4.0% of the initial nitrogen

**Table 1** |  $N_2O$  and  $CH_4$  emission factors measured on sewage sludge composting plants: plant A (boxes) and plant B (tunnels);  $CH_4$  and  $N_2O$  emissions were converted into eq. $CO_2$  using GWP of 25 and 298, respectively

#### Sewage sludge composting

##### $CH_4$ emission factors

Unit	Plant A	Plant B
kg $CH_4$ /t raw sludge	0.04	0.63
kg $CH_4$ /t dry sludge	0.18	3.40
kg $CH_4$ / t sludge Corg	0.51	8.90
% Corg emitted as C- $CH_4$	0.04%	0.70%
kg eq. $CO_2$ /t raw sludge	1.00	15.7

##### $N_2O$ emission factors

Unit	Plant A	Plant B
kg $N_2O$ /t raw sludge	0.17	0.09
kg $N_2O$ /t dry sludge	0.74	0.48
kg $N_2O$ /t sludge NTK	14.8	7.70
% NTK emitted as N- $N_2O$	1.00%	0.5%
kg eq. $CO_2$ /t raw sludge	50.7	26.8

in waste which indicates a wide variation of  $\text{N}_2\text{O}$  emissions resulting from different waste nature and composting conditions. However, few studies are related to sewage sludge composting. Czepiel *et al.* (1996) quantified 0.7 g  $\text{N}_2\text{O}$ /kg dry sludge which is in the range of the  $\text{N}_2\text{O}$  emissions quantified during the measurement campaigns of both aerated composting processes within the present study (0.48 and 0.74 kg  $\text{N}_2\text{O}$ /t dry sludge). Amlinger *et al.* (2008) observed from two different sewage sludge compositions emission factors of 0.165 and 0.266 kg  $\text{N}_2\text{O}$ /t raw sludge which are slightly higher than those observed in plants A and B (0.170 and 0.090 kg  $\text{N}_2\text{O}$ /t raw sludge, respectively). Nevertheless Amlinger *et al.* (2008) reported specific conditions – that are not comparable to continuously ventilated industrial composting processes – where high unfavourable nitrogen concentrations leading to very low C/N ratios was the main reason why decomposition was considerably inhibited and high  $\text{N}_2\text{O}$  emissions occurred during the composting process.

Process emissions measured on the two sites were compared with indirect emissions linked to operational energy use (electricity and fuel). The results are illustrated in Figure 5, which represents emission factors of each related source: electricity, fuel,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  process emissions.

Based on the two study cases assessed, contribution of  $\text{N}_2\text{O}$  and  $\text{CH}_4$  process emissions to climate change is not negligible regarding GHG emissions linked to energy consumption. As measured and reported within this study,  $\text{N}_2\text{O}$  and  $\text{CH}_4$  emissions vary from 27 to 51 and 1 to 16 kg

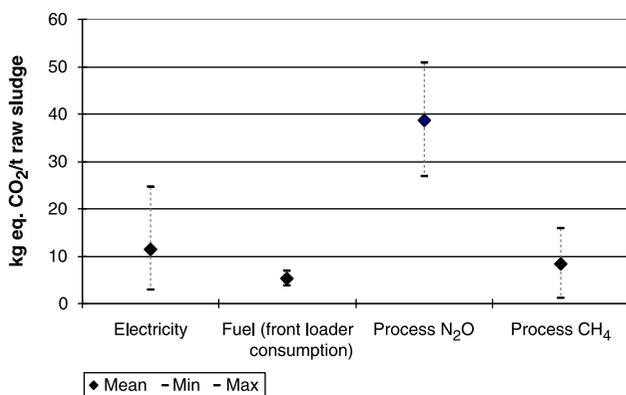
eq. $\text{CO}_2$ /t raw sludge, respectively (Table 1). GHG emissions related to electricity use vary from 3 to 24 kg eq. $\text{CO}_2$ /t raw sludge depending on both the energy consumption and the emission factors used to be converted to GHG emissions. In this study two different emission factors were used: French and Spanish electricity mix whose carbon contents are quite different regarding the different electricity supply mixes (90 and 390 g eq. $\text{CO}_2$ /kWh, respectively). Indeed cumulated process emissions of both  $\text{CH}_4$  and  $\text{N}_2\text{O}$  can generate more impacts on climate change than GHG emissions related to energy consumption. In some other cases GHG emissions related to energy consumption and those linked to process emissions may be in the same order of magnitude.

## CONCLUSIONS

Accounting for direct and indirect GHG emissions, sewage sludge treatment activities contribute to GHG emissions. However, in general, GHG emissions from sewage sludge treatment remain poorly quantified. Indeed, high nitrogen and carbon content of wastewater sludge could lead to  $\text{CH}_4$  or  $\text{N}_2\text{O}$  emissions during biological or thermal processes of wastewater sludge treatment. Regarding the high GWP of  $\text{N}_2\text{O}$  and  $\text{CH}_4$  (298 and 25, respectively), those emissions could appear to be significant.

A methodology was therefore developed to assess GHG emissions of different sludge treatment processes. Indirect emissions linked to operational energy use, chemicals and material consumption and freight and direct  $\text{N}_2\text{O}$  and  $\text{CH}_4$  process emissions are taken into account. In order to obtain emission factors for sludge treatment processes, on-site measurement campaigns of  $\text{N}_2\text{O}$  and  $\text{CH}_4$  emissions were performed. The protocol described in this paper may be used to assess the quantities of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emitted during different sludge treatments. Two sludge composting plants were assessed.

On-site sludge composting measurement campaigns showed GHG emissions ranging between 0.18 and 3.4 kg  $\text{CH}_4$ /t dry sludge and between 0.74 and 0.48 kg  $\text{N}_2\text{O}$ /t dry sludge. Converted to  $\text{CO}_2$  equivalent to assess climate change impact, emission factors of  $\text{CH}_4$  range between 4.5 and 85 kg eq. $\text{CO}_2$ /t dry sludge and for  $\text{N}_2\text{O}$  range between 143 and 221 kg eq. $\text{CO}_2$ /t dry sludge. Regarding direct GHG emissions,  $\text{N}_2\text{O}$  has a much greater impact on climate change



**Figure 5** | Composting GHG emissions related to process and energy (electricity and fuel) emissions;  $\text{EF}_{\text{fuel}} = 2,662$  g  $\text{CO}_2\text{eq}/\text{l}$  fuel,  $\text{EF}_{\text{French electricity}} = 90$  g  $\text{CO}_2\text{eq.}/\text{kWh}$  (French electricity mix) and  $\text{EF}_{\text{Spanish electricity}} = 390$  g  $\text{CO}_2\text{eq.}/\text{kWh}$  (Spanish electricity); emission factors of  $\text{N}_2\text{O}$  and  $\text{CH}_4$  process emissions are those found in this study (i.e.  $\text{CH}_4 = 0.04\text{--}0.63$  kg  $\text{CH}_4$ /t raw sludge and  $\text{N}_2\text{O} = 0.09\text{--}0.17$  kg  $\text{N}_2\text{O}$ /t raw sludge) and calculated with GWP  $\text{N}_2\text{O} = 298$  and GWP  $\text{CH}_4 = 25$ .

than CH<sub>4</sub> because of its higher GWP. N<sub>2</sub>O is linked to the treatment of nitrogen and produced during the nitrification or denitrification biological processes.

Process emissions measured on the two sites were compared with indirect emissions linked to operational energy use. Indeed cumulated process emissions of both CH<sub>4</sub> and N<sub>2</sub>O can generate more impacts on climate change than GHG emissions related to energy consumption. For a more holistic approach, streamline life-cycle activities should also be determined when quantifying a wastewater management strategy impact on GHG emissions.

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