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Ammonia emissions from the composting of different organic wastes

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2 **Dependency on process temperature** 3 4 5 Estel.la Pagans, Raquel Barrena, Xavier Font and Antoni Sánchez\* 6 7 Escola Universitària Politècnica del Medi Ambient 8 Universitat Autònoma de Barcelona 9 Rbla Pompeu Fabra 1 10 08100-Mollet del Vallès (Barcelona), Spain 11 12 \* Corresponding author: Dr. Antoni Sánchez 13 Escola Universitària Politècnica del Medi Ambient 14 Universitat Autònoma de Barcelona 15 Rbla Pompeu Fabra 1 16 08100-Mollet del Vallès (Barcelona), Spain 17 Phone: 34-93-5796784 18 Fax: 34-93-5796785 19 E-mail address: asanchez@eupma.uab.es 20

## **Abstract**

Ammonia emissions were quantified for the laboratory-scale composting of three typical organic wastes with medium nitrogen content: organic fraction of municipal solid wastes, raw sludge and anaerobically digested sludge; and the composting of two wastes with high nitrogen content: animal by-products from slaughterhouses and partially hydrolysed hair from the leather industry. All the wastes were mixed with the proper amount of bulking agent. Ammonia emitted in the composting of the five wastes investigated revealed a strong dependence on temperature, with a distinct pattern found in ammonia emissions for each waste in the thermophilic first stage of composting (exponential increase of ammonia emitted when increasing temperature) than that of the mesophilic final stage (linear increase of ammonia emissions when increasing temperature). As composting needs high temperatures to ensure the sanitisation of compost and ammonia emissions are one of the main environmental impacts associated to composting and responsible for obtaining compost with a low agronomical quality, it is proposed that sanitisation is conducted after the first stage in large-scale composting facilities by a proper temperature control.

### Capsule:

Ammonia emission pattern and correlation with process temperature are presented for the composting process of different organic wastes.

- **Keywords**: Ammonia emissions, Composting, Organic wastes, Process Temperature,
- 24 Sanitisation.

#### 1. Introduction

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(Liang et al., 2000; Sheridan et al., 2002).

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In recent years, composting has been presented as an environmentally friendly and sustainable alternative to manage and recycle organic solid wastes, with the aim of obtaining a quality organic product, known as compost, to be used as organic amendment in agriculture. Composting presents, however, some associated environmental impacts, being the generation of polluted or odorous gaseous emissions one of the major concerns in developed countries (Haug, 1993). Ammonia is one of the main compounds responsible for generation of offensive odours and atmospheric pollution when composting organic wastes with high nitrogen content. Although the detection and recognition thresholds for ammonia are relatively high (17 ppmv and 37 ppmv respectively, Busca and Pistarino, 2003) ammonia gas is the main compound found in exhaust gases from composting, except for carbon dioxide (Beck-Friis et al., 2001), in concentrations well over the threshold limit (Elwell et al., 2002; Hong and Park, 2004). Ammonia gas can cause adverse effects on vegetation and can be converted to N<sub>2</sub>O, a powerful greenhouse gas (Krupa, 2003). Ammonia emissions from several sources such as livestock production (Dore et al., 2004; Scholtens et al., 2004), manure application to soil (Webb, 2001), fertilizer utilization (Sommer et al., 2004) and other industrial sources (Sutton et al., 2000) have been extensively studied. Additionally, ammonia abatement by means of different techniques based on adsorption, absorption and biological processes is also well documented in literature, being biofiltration one of the options more widely reported

1	However, except for animal manures, there is a lack of knowledge about the				
2	ammonia emissions from composting, especially when organic wastes of different				
3	biochemical composition are considered. A few studies conducted on the ammonia				
4	emissions derived from the composting process have concluded that temperature, pH,				
5	and initial ammonium content are the most important parameters affecting the amount				
6	of nitrogen emitted as ammonia, since high temperature and pH favour ammonia				
7	volatilization by displacing $\mathrm{NH_4}^+/\mathrm{NH_3}$ equilibrium to ammonia. Simultaneously, it is				
8	widely reported that high temperature inhibit the nitrification process (Grunditz and				
9	Dalhammar, 2001), and thereby, the possibility for ammonia volatilization is high.				
10	Thus, Beck-Friis et al. (2001) observed that ammonia emissions started when				
11	thermophilic temperatures (> 45°C) and high pH (about 9) coexist in the compost				
12	environment, resulting in a total loss of nitrogen within 24-33% of the initial nitrogen				
13	content. Similarly, Cronje et al. (2002) determined that the nitrogen losses for organic				
14	mixtures with an initial pH $< 6.2$ were below 4% of the initial nitrogen content.				
15	Nevertheless, it must be emphasized that pH control is in practice very difficult during a				
16	composting process, whereas temperature control can be conducted once the sanitisation				
17	requirements are fulfilled (European Commission, 2001; U.S. Environmental Protection				
18	Agency, 1995). In other works, the strategy of using an intermittent aeration are tested				
19	and proved to be effective in decreasing the ammonia emissions (Elwell et al., 2002),				
20	however, this causes an oxygen limitation in the aerobic process and a loss of biological				
21	activity.				
22	The objectives of this work are: i) to determine the ammonia emissions in the				
23	composting process of three typically composted wastes: organic fraction of municipal				
24	solid wastes, dewatered raw sludge and anaerobically digested sludge and two organic				

wastes selected because of its extremely high nitrogen content: animal by-products from slaughterhouses and hydrolysed hair from the leather production industry, ii) to correlate the ammonia emissions with the process temperature, especially the distinction between the mesophilic and thermophilic temperature ranges, which are of crucial interest in the sanitisation of the final compost, iii) to establish a qualitative pattern of temperature control in the composting process in order to minimise the ammonia emissions and therefore, to reduce the environmental impact associated and to improve

8 the agronomical quality of compost.

#### 2. Materials and methods

### 2.1. Composted wastes

Five organic wastes were used in the composting experiments: source-separated organic fraction of municipal solid waste (OFMSW) obtained from the composting plant of Jorba (Barcelona, Spain); dewatered raw sludge (RS) a mixture of primary and activated sludge from the urban wastewater treatment plant of La Garriga (Barcelona, Spain); dewatered anaerobically digested sludge (ADS) from the urban wastewater treatment plant of La Llagosta (Barcelona, Spain); animal by-products (AP) consisting of slaughterhouse wastes composed of rejected pieces of rabbit and chicken (mainly viscera, feather and other organs) obtained from the composting plant of Jorba (Barcelona, Spain); and partially hydrolysed hair (HH) from a factory specialized in leather production from cow skins in Igualada (Barcelona, Spain). Table 1 presents the main initial characteristics of the composted mixtures. OFMSW and AP were

1	composted as they were obtained, since its initial characteristics were appropriate for
2	composting (Table 1). In the case of wastewater sludge (RS and ADS) wood chips from
3	a local carpentry were used as inert bulking agent in a volumetric ratio 1:1 (bulking
4	agent:sludge), which was previously found as optimal for sludge composting (Gea et al.,
5	2003). The main function of bulking agent was to provide an adequate porosity to
6	sludge, and it was not substantially degraded under laboratory composting conditions.
7	HH was mixed with RS (1:1 weight ratio) to act as inoculum in the composting process
8	since in previous experiments with HH alone (data not shown) there was no composting
9	activity probably due to the strong chemical treatment applied to cow skins to remove
10	and hydrolyse hair. This mixture HH:RS were then mixed with wood chips in a

# 2.2. Composting experiments

volumetric ratio 1:1.

All wastes were composted in a 30-L laboratory reactor. A scheme of the composting reactor is shown in Figure 1. Air was supplied to the reactor by a suction-type blower (air flow 5 L min<sup>-1</sup>) to maintain the oxygen content in the composting material over 10%. Oxygen content in the composting material was measured with an oxygen sensor (Sensox, Sensotran, Spain). Ammonia concentrations of the exhaust gas from the composting reactor were measured online by an electrochemical gas sensor (Bionics Instrument Co, Tokyo, Japan). Temperatures of the composting materials were monitored during the composting period using a Pt-100 sensor located at the centre of the composter since the variability of temperature values at different positions of the composter was within the range of 5-10% (Gea et al., 2004). All the values were

1	displayed and recorded with a personal computer every 30 minutes. Moisture content
2	was initially adjusted and maintained between 40-60% during all the experiments
3	(adding tap water when necessary), since it is considered optimal for composting (Haug.
4	1993).
5	Two replications for each waste were conducted. Results presented in this paper
6	correspond to one replication. Differences of ammonia emissions and temperature
7	profiles between composting replications were in the range of 10-20%. Composting
8	experiments were finished when either composting temperature was near ambient
9	temperature ( $< 30$ °C) or ammonia emissions were low ( $< 50$ mg NH <sub>3</sub> m <sup>-3</sup> ).
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11	2.3. Analytical methods
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13	Moisture content, dry matter content, organic matter content, N-Kjeldhal, carbon
14	content, C/N ratio, pH and electrical conductivity were determined according to the
15	standard procedures (U.S. Department of Agriculture and U.S. Composting Council,
16	2001). The composter material was manually homogenized prior to sampling and a
17	representative portion of the material (1 L) was used as sample for analytical
18	determinations.
19	Respiration index was determined using a static respirometer based on the model
20	previously described by Ianotti et al. (1993) and following the modifications and
21	recommendations given by the U.S. Department of Agriculture and U.S. Composting
22	Council (2001). Values of respiration index are expressed as mg of oxygen consumed g
23	<sup>1</sup> organic matter h <sup>-1</sup> and are presented as an average of three replicates.
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#### 3. Results and discussion

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3.1. Composting experiments

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Five organic wastes were composted at laboratory-scale under controlled conditions. The wastes were selected according to the following criterion: a first group of wastes consisting of OFMSW, RS and ADS as the most commonly processed wastes in composting plants, with a medium-to-low nitrogen content from 2 to 3% and a C/N ratio from 10 to 20 (Table 1); and a second group of wastes composed of AP and HH selected by their high nitrogen content (4-7%) and a low C/N ratio (5-10), for which composting appears as a sustainable and environmentally friendly management technology, since nowadays these wastes are being landfilled or incinerated. Temperature profiles for the five wastes composted are presented in Figures 2a to 6a. In the case of OFMSW, RS and ADS the thermophilic range (initial stage) of temperatures was quickly achieved and maintained for 3-5 days (Figures 2a, 3a and 4a, respectively). This period was followed by a mesophilic maturation phase (final stage), which corresponded to a typical composting temperature profile at laboratory scale. In the case of AP and HH, the thermophilic phase was longer (Figures 5a and 6a). This was probably due to the combination of organic compounds found in these wastes, in which organic matter is mainly composed of easily degradable and energetic compounds such as protein and fats, and large amounts of organic nitrogen are available. In any case, it is clear that the characteristics of the wastes composted and the

temperature profiles obtained indicate that composting is a suitable technology to treat

1 these wastes and to recycle them into stabilised and sanitised compost. It is particularly

2 interesting the fact that the initial respiration indices for the wastes considered were in

- 3 the range of active materials and indicated a high aerobic biological activity (California
- 4 Compost Quality Council, 2001). On the other hand, it should be pointed out that
- 5 although sanitisation requirements were not fulfilled at laboratory-scale (European
- 6 Commission, 2001; U.S. Environmental Protection Agency, 1995), it is likely that a
- 7 complete sanitisation may be easily achieved at full-scale (Haug, 1993).

#### 3.2. Ammonia emissions

Ammonia emissions were quantified in the composting of the five wastes studied. Results are shown in Figures 2a to 6a. In all cases, a peak in the ammonia emissions was observed in coincidence with the thermophilic stage. In fact, ammonia emissions have been proposed in some works as an indicator of the biological activity of composting materials with high nitrogen content (Liao et al., 1995). Among the first group of wastes, OFMSW and ADS showed similar concentrations of ammonia in exhaust gases, with maximum values within 500-700 mg NH<sub>3</sub> m<sup>-3</sup>, which are in accordance with other works performed with wastes with similar nitrogen content such as household waste (Beck-Friis et al., 2001) and dairy manure (Hong et al., 2002). On the contrary, ammonia emissions of RS were lower when compared to OFMSW or ADS (Fig. 3a) with maximum emissions in the range of 100 mg NH<sub>3</sub> m<sup>-3</sup>. The reason for this low ammonia emissions was not clear, since temperature profile and the initial characteristics of RS such as nitrogen content, C/N ratio or pH were similar to that of ADS. A possible explanation may be a high initial content of easily biodegradable

1 nitrogen forms in ADS since it is well known that organic nitrogen compounds can be 2 hydrolysed but not consumed in the anaerobic digestion and that there is an important 3 release of ammonia (Nah et al., 2000; Salminen and Rintala, 2002). Therefore, when 4 composting ADS a high emission of ammonia was measured in the initial high 5 temperature phase but little was lost later because the content of ammonium probably 6 corresponded to little digestible organic N. However, in the case of RS ammonia 7 emission was lower during the initial phase with high temperature and the ammonia 8 emission was relatively higher in later phases possibly due to transformation of 9 digestible organic N to ammonium. 10 The second group included AP and HH, wastes with high nitrogen content, low 11 C/N ratio and a relatively high pH (Table 1). As can be seen in Figures 5a and 6a, 12 ammonia emissions were very high, especially at the end of the thermophilic stage of the composting process, in which values of 3000-4000 mg NH<sub>3</sub> m<sup>-3</sup> were detected. 13 14 Table 2 summarise the cumulative ammonia emissions found for each waste composted. 15 These results were higher than those found in the composting of other high-nitrogen 16 wastes, such as fish wastes with an initial nitrogen content of 9.23% and a C/N ratio of 17 7 (Liao et al., 1997) and fish-processing sludge with an initial nitrogen content of 9.3% 18 and a C/N ratio of 4 (Nakasaki et al., 2000). However, it must be pointed that, to our 19 acknowledge, this is the first study on the composting of AP and HH. From our results, 20 it seems clear that an imbalance in the initial C/N ratio provoked a release of high 21 amounts of nitrogen as ammonia, as it has been reported previously (Tiquia and Tam, 22 2000).

### 3.3. Influence of process temperature on ammonia emissions

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Two main factors determine the ammonia emissions in the composting of a given waste: temperature and pH. High temperature affects ammonia volatilization and at a higher pH, non-volatile ammonium ions are converted to the volatile ammonia form. The control of pH in a composting process is only possible by using acid amendments such as bauxite residues (Qiao and Ho, 1997) or even biodegradable plastics (Nakasaki et al., 2000) or by precipitating ammonia into struvite crystals by addition of Mg and P salts (Jeong and Kim, 2001), however, these methods are usually expensive at full-scale or the amendment materials may be not available. Additionally, in previous experiments (data not shown) we observed that the wastes with an initial slightly alkaline pH maintain this value during all the process. This was the case of RS, ADS, AP and HH in which pH values were usually in the range of 7.5-8.5. In the case of OFMSW, with a slightly acidic initial pH, the alkaline range of pH was also achieved in few hours (data not shown). On the contrary, temperature in the composting process can be easily measured on-line and controlled by changing the aeration regime or the turning frequency (Haug, 1993). In Figures 2b to 6b, the ammonia emissions observed in the composting of each waste studied are presented vs. process temperature. Although there was some dispersion in experimental data, it can be observed that during the first thermophilic stage of composting an exponential fit could be positively correlated between temperature and ammonia emissions (significant at 0.05 probability level), whereas the trend during the final stage of composting (mainly mesophilic) was linear (significant at 0.05 probability level). The correlation coefficients for each waste and stage are

1 presented in Table 3. The exponential growth used to describe the ammonia emissions 2 of the initial stage is supported by the fact that the biological activity of the initial 3 mixtures is very high as it is shown by the respiration index values (Table 1). This 4 demonstrates that the material is fully active from the very first moment of the 5 composting process causing a rapid increase of temperature to reach thermophilic 6 values. These results were not observed in RS composting, in which an exponential fit 7 was also suitable for the final stage (although the slope was significantly lower than that 8 of initial stage), and could not be confirmed for the first stage of HH composting, 9 because of a lack of experimental data in this period. Although the effect of compost 10 temperature on ammonia emissions is not clearly understood (Beck-Friis et al., 2001) a 11 possible explanation for the ammonia emissions pattern is that at the initial stage of 12 composting, degradation of large amounts of easily biodegradable organic compounds 13 with high nitrogen content provoke a release of ammonia gas which is exponentially-14 dependent on temperature as it is expected for free soluble ammonia. On the contrary, at 15 the final stage of composting, nitrogen is bound to complex organic molecules and 16 involved in humification processes (Baddi et al., 2004; Paredes et al., 2002), which 17 prevents ammonia release and volatilisation. Moreover, as temperature is an excellent 18 indicator of the biological activity of the composting process (Haug, 1993), the fact that 19 ammonia emissions were exponentially correlated with temperature can be related to an 20 exponential microbial growth in the first stage of composting, which provoked a 21 significant generation of metabolic heat. In relation to this, it should be pointed that 22 although several studies on microbial communities in composting have been carried out 23 and the evolution of specific microorganisms have been presented using several 24 techniques (Gamo and Shoji, 1999; Herrmann and Shann, 1997; Ishii and Takii, 2003;

1 Tang et al., 2004: Tiquia et al., 2002), it is evident that a reliable measure of the total

2 active biomass in a composting process is still lacking. A high percentage of non-

culturable microorganisms and the complexity of applying some advanced

4 microbiological techniques to a composting environment are the main difficulties found

5 in monitoring composting biological profiles. In this context, indirect parameters related

to biological activity such as oxygen uptake rate (Gea et al., 2004) or ammonia

emissions presented in this work can be useful in the monitoring of the global activity of

8 the composting process.

## 3.4. Implications on compost sanitisation

The control of temperature in the composting process has one restriction: the sanitisation of compost. Time-temperature conditions for pathogen inactivation prior to compost application to soil are required by international environmental agencies (European Commission, 2001; U.S. Environmental Protection Agency, 1995), however, the moment of sanitisation is not specified in these international rules. On the other hand, methods for minimizing ammonia evolution in exhaust gas, and thus for maximizing ammonia retention in the compost, would be favourable from the standpoint of preserving the fertilizer element and to avoid atmospheric pollution and odour nuisance. From the results obtained in this study a temperature control at full-scale should be based on maintaining a relatively low temperature during the first stage of the process (50-55°C) where ammonia emissions are low and to fulfil the sanitisation requirements at the final stage of composting after the maximum activity has been

1	achieved. The objective of this control would be to minimise ammonia losses during the
2	composting process to improve the quality of the compost and to reduce treatment costs.
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4	4. Conclusions
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6	A quantitative study on the ammonia emissions produced in the composting of
7	five organic wastes was carried out. The main conclusions are:
8	1) Ammonia emissions exhibited a similar relationship with temperature profile for each
9	waste. However, the total amount of ammonia emitted was as expected directly related
10	to the C/N ratio of the waste.
11	2) Ammonia emissions pattern in the composting of the five wastes investigated
12	strongly depended on process temperature. However, the trend observed in the
13	thermophilic initial stage of composting showed an exponential increase when process
14	temperature increases, whereas a linear correlation was found for ammonia emissions
15	and temperature in the final mesophilic stage of the composting process.
16	3) Sanitisation of the compost is required prior to application and it is achieved by
17	maintained high temperature during a period of time. In large-scale composting
18	operations, sanitisation should be conducted after the initial thermophilic stage. This
19	would reduce ammonia emissions, environmental impact of the composting process and
20	the cost of exhaust gases treatment.
21	4) Optimal temperatures for the composting process in relation to the reduction of

ammonia emissions should be investigated for each waste in order to obtain compost

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with the maximum nitrogen content.

5) Temperature appears to be the most suitable parameter to control ammonia emissions
 when composting of organic wastes. However, the effect of pH on ammonia emissions
 and the combination of temperature-pH effects should be the aim of future studies on
 composting organic wastes.
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# 1 Tables

**Table 1**: Initial characteristics of the different waste mixtures composted. OFMSW:

4 Organic Fraction of Municipal Solid Waste; RS: Raw Sludge; ADS: Anaerobically

5 Digested Sludge; AP: Animal by-Products; HH: Hydrolysed Hair.

Parameter	OFMSW	RS	ADS	AP	НН
Moisture (%)	46.7	61.8	62.4	55.0	55.4
Dry Matter (%)	53.3	38.2	37.6	45.0	44.6
Organic Matter (% dry basis)	67.3	57.7	52.5	69.1	60.1
N-Kjeldhal (% dry basis)	2.2	2.5	2.6	4.3	6.1
N-NH <sub>4</sub> <sup>+</sup> (% dry basis)	0.21	0.47	0.66	0.54	0.30
C/N ratio	17	13	11	8.8	5.4
pН	6.1	7.1	7.6	8.0	8.2
Electrical conductivity (mS cm <sup>-1</sup> )	3.0	1.8	2.1	5.6	3.4
Respiration index (mg O <sub>2</sub> g <sup>-1</sup> organic matter h <sup>-1</sup> )	3.82	6.68	3.74	4.74	3.22

- **Table 2**: Cumulative ammonia emissions for the different waste mixtures composted
- 2 (referred to initial weights of waste, dry matter, organic matter and nitrogen). OFMSW:
- 3 Organic Fraction of Municipal Solid Waste; RS: Raw Sludge; ADS: Anaerobically
- 4 Digested Sludge; AP: Animal by-Products; HH: Hydrolysed Hair.

Organic Waste	OFMSW	RS	ADS	AP	НН
Ammonia emitted (g NH <sub>3</sub> kg <sup>-1</sup> waste)	0.17	0.04	0.23	2.39	10.3
Ammonia emitted (g NH <sub>3</sub> kg <sup>-1</sup> dry matter)	0.32	0.10	0.60	5.30	20.7
Ammonia emitted (g NH <sub>3</sub> kg <sup>-1</sup> organic matter)	0.47	0.17	1.16	7.67	34.4
Ammonia emitted (g NH <sub>3</sub> kg <sup>-1</sup> N)	1.4	0.40	2.3	14.1	33.8

- **Table 3**: Correlations found between ammonia emissions and temperature for the
- 2 different waste mixtures composted. OFMSW: Organic Fraction of Municipal Solid
- 3 Waste; RS: Raw Sludge; ADS: Anaerobically Digested Sludge; AP: Animal by-
- 4 Products; HH: Hydrolysed Hair.

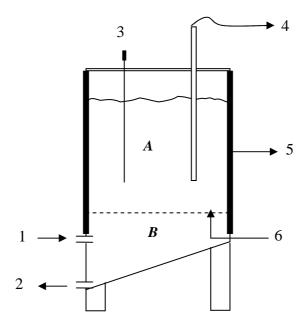
	Correlation ammonia emissions vs. temperature*			
Organic Waste	First stage (thermophilic)	Final stage (mesophilic)		
OEMSW	$NH_3$ emitted = $0.40$ exp $(0.13T)$	$NH_3$ emitted = -122+5.0T		
OFMSW	$(R^2 = 0.96)$	$(R^2=0.94)$		
RS	$NH_3$ emitted = 1.12E-31exp(1.26T)	$NH_3$ emitted = $0.37$ exp $(0.10T)$		
KS	$(R^2=0.89)$	$(R^2=0.92)$		
ADS	$NH_3$ emitted = $0.22$ exp $(0.13T)$	$NH_3$ emitted = $-108+5.6T$		
ADS	$(R^2=0.89)$	$(R^2=0.80)$		
AP	$NH_3$ emitted = $0.10$ exp $(0.15T)$	$NH_3$ emitted = -694+46T		
Ar	$(R^2=0.85)$	$(R^2=0.82)$		
HH	not determined	$NH_3$ emitted = -5270+212T		
1111	not determined	$(R^2=0.85)$		

<sup>9 \*</sup> Temperature in °C and ammonia emissions in mg NH<sub>3</sub> m<sup>-3</sup>

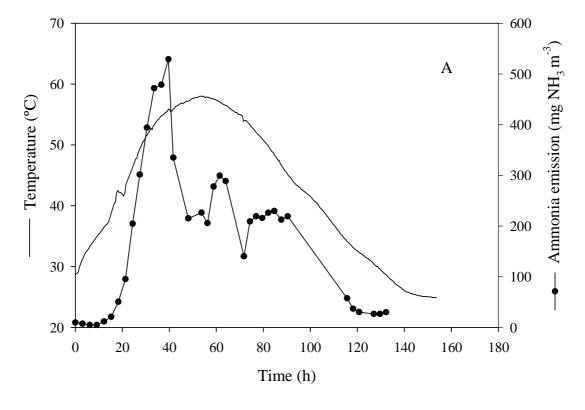
# 1 **Figure Legends** 2 3 Figure 1: Scheme of the composting reactor. 1-Air inlet, 2-Leachates outlet, 3-4 Temperature probe, 4-Air pump to oxygen and ammonia sensors, 5-Thermal insulation, 5 6-Perforated plate, A-Composting volume, B-Leachates collection and air supply. 6 Figure 2: Evolution of ammonia emissions and temperature in the composting of 7 Organic Fraction of Municipal Solid Waste. A) Evolution during composting time. B) 8 Correlation ammonia emissions vs. temperature. 9 Figure 3: Evolution of ammonia emissions and temperature in the composting of Raw 10 Sludge. A) Evolution during composting time. B) Correlation ammonia emissions vs. 11 temperature. 12 Figure 4: Evolution of ammonia emissions and temperature in the composting of 13 Anaerobically Digested Sludge. A) Evolution during composting time. B) Correlation 14 ammonia emissions vs. temperature. 15 Figure 5: Evolution of ammonia emissions and temperature in the composting of 16 Animal by-Products. A) Evolution during composting time. B) Correlation ammonia 17 emissions vs. temperature. 18 **Figure 6**: Evolution of ammonia emissions and temperature in the composting of 19 Hydrolysed Hair. A) Evolution during composting time. B) Correlation ammonia 20 emissions vs. temperature. 21

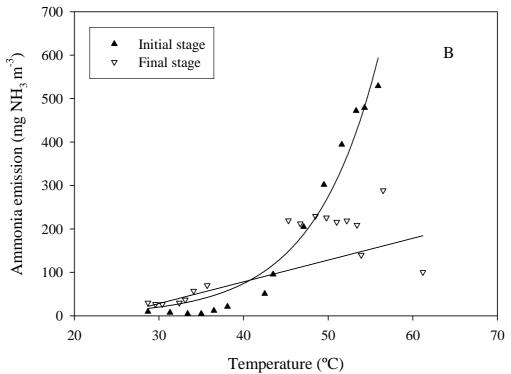
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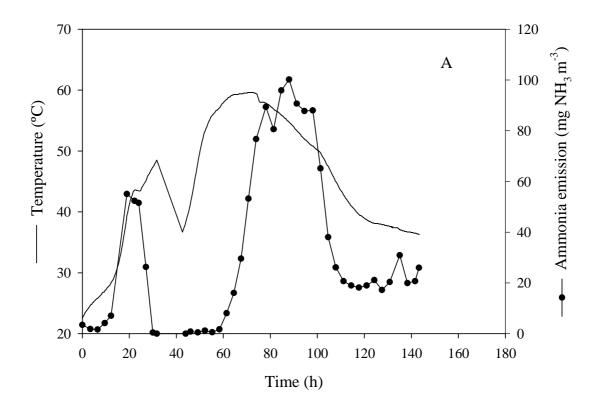
**Figure 1**: Pagans et al.

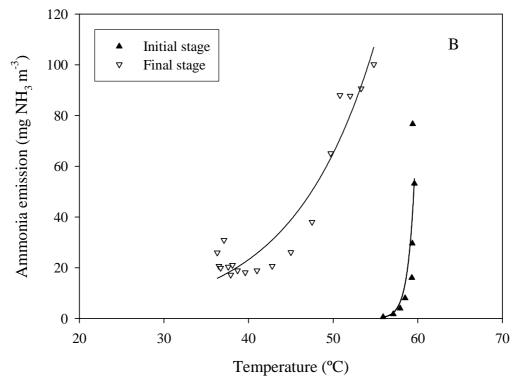


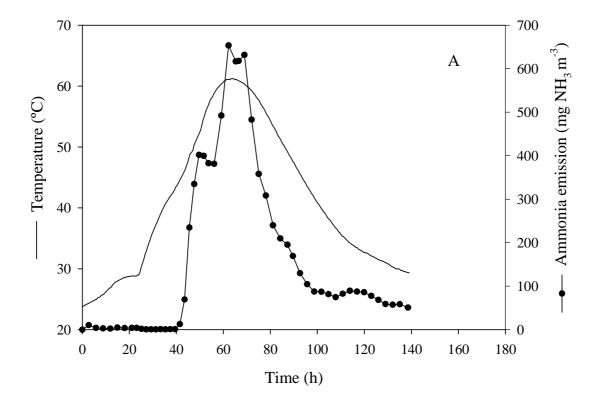
**Figure 2**: Pagans et al.

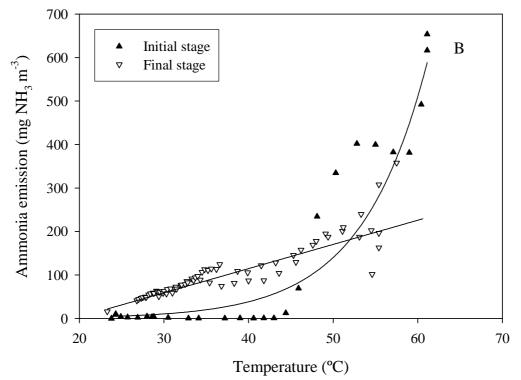


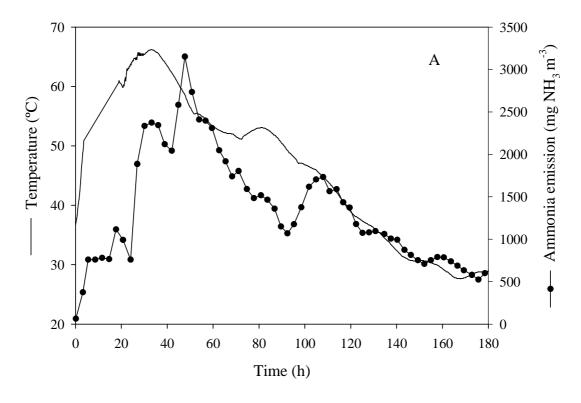


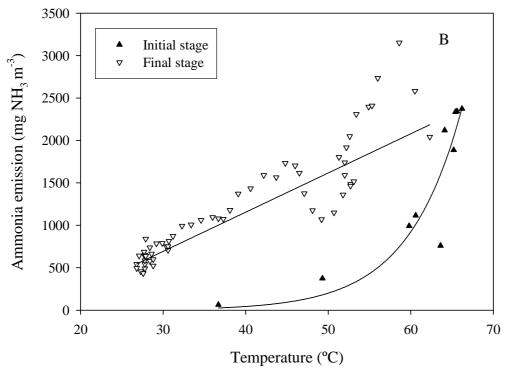












# **Figure 6**: Pagans et al.

