

# GASEOUS EMISSIONS (NH<sub>3</sub>, N<sub>2</sub>O, CH<sub>4</sub> AND CO<sub>2</sub>) FROM A BIOLOGICAL AEROBIC TREATMENT UNIT OF PIG SLURRY

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## ABSTRACT

Biological aerobic treatment of pig slurry is one of the measures to limit land-application of manure. This treatment transforms the nitrogen compounds into N<sub>2</sub> by nitrification and denitrification. However, this treatment may be a source of other gaseous emissions (NH<sub>3</sub>, CH<sub>4</sub>, N<sub>2</sub>O, CO<sub>2</sub>) emitted during the treatment and the following management of the treatment by-products. Within this framework, NH<sub>3</sub>, N<sub>2</sub>O, CH<sub>4</sub> and CO<sub>2</sub> release from a biological aerobic treatment system of pig slurry was investigated under field conditions. Field quantification and integration of the NH<sub>3</sub>, CH<sub>4</sub>, N<sub>2</sub>O, CO<sub>2</sub> emission rates within a model based on the by-product flow enabled us to estimate annual gas flows. The results showed that the emissions of CH<sub>4</sub>, CO<sub>2</sub> and NH<sub>3</sub> were more important for the non-aerated effluents (raw slurry and separated solid fraction). The total gas emissions of NH<sub>3</sub>, CH<sub>4</sub> and CO<sub>2</sub> were close to 480 kgN, 3400 kgC and 12700 kgC per year respectively. The majority of the emissions occurred during the storage of the separated solid fraction (59% of NH<sub>3</sub> and 65% of CO<sub>2</sub>) and the raw slurry (29% of the NH<sub>3</sub> and 54% of CH<sub>4</sub>). N<sub>2</sub>O was only measured above the biological reactor with 2.6 kgN per year.

**Keywords:** ammonia, methane, nitrous oxide, aerobic treatment.

## INTRODUCTION

According to the EU nitrate directive, the high pig production in Brittany (France) implies a slurry treatment for some farms that don't have enough land to respect the level of 170 kg.N.ha<sup>-1</sup>.year<sup>-1</sup>. Among the different treatments the aerobic treatment consists in transforming the slurry nitrogen into gas (N<sub>2</sub>) by a nitrification/denitrification process. However, N<sub>2</sub>O and NH<sub>3</sub> could be emitted during the slurry aeration while the management of the treatment by-products (aerated liquid manure, solid separated fraction, biological sludge and supernatant) can also produce gas emissions (NH<sub>3</sub>, N<sub>2</sub>O, CH<sub>4</sub>, CO<sub>2</sub>). This paper concerns field measurements of NH<sub>3</sub>, N<sub>2</sub>O, CH<sub>4</sub> and CO<sub>2</sub> emissions during aeration of pig slurry and storage of by-products of a biological treatment unit. The integration of the determined gaseous factors to a total model on product flow led to the evaluation of gas flows on the scale of the treatment unit.

## MATERIALS AND METHODS

The study was conducted on a commercial pig operation. The aerobic treatment unit consisted of (i) a raw slurry storage (ii) a solid-liquid separation by centrifuge (iii) a biological aerobic reactor (iv) a aerated slurry decantation-storage (v) a supernatant lagoon and (vi) a biological sludge storage. The gaseous emissions from the aerobic reactor and from the storage of raw slurry, aerated slurry, liquid separated fraction and biological sludge were measured using the technique of the dynamic chamber (Peu, 1999). Gaseous emissions during storage of the solid separated fraction was measured by enclosing the heap in a large polyethylene structure drawn up by the use of a wind motor. The concentrations of CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O were measured either

by infrared detection (analyser URAS 14 and 10) or by FID/ECD gas chromatography (Varian star 3800). The ammonia concentration was determined by using trap bottles filled with sulphuric acid (0.5 N) followed by a distillation and a titration. The gaseous measures were carried out for 4 to 7 weeks, in autumn for the storage of raw slurry and biological sludge, in spring for the biological reactor and the storage of aerated slurry and in winter for the storage of solid separation fraction and the supernatant. In order to estimate annual gaseous flow we developed a mathematical model that integrates the daily variations of volume of pig slurry (Levasseur 1998a, 1998b and 2002) and biological treatment by-products (based on field experimental measures, Beline et al. 2001 and 2003), the livestock production management (number of sows and housing according to the advice of French livestock institutes). The gas emission flows are calculated on a step of daily time by applying the field experimental gaseous factors to the various product storages.

## RESULTS AND DISCUSSION

The results show the impact of the treatment on the characteristics of the by-products compared to the raw slurry (table 1). The level of TAN and organic matter (TVS) of the solid separated fraction, about 4.8 gN.kg<sup>-1</sup> and 270 g.kg<sup>-1</sup> are higher than those of raw slurry while the other biological treatment by-products present less TAN or organic matter.

**Table 1.** Characteristics of pig slurry and biological treatment by-products (Mean values).

Characteristic	Type of manure					
	RS	SSF	LSF	AS	S	BS
Total Ammonium Nitrogen (TAN, gN.kg <sup>-1</sup> )	2.9	4.8	2.3	0.2	0.05	0.2
Total Nitrogen (gN.kg <sup>-1</sup> )	4.1	11	3.1	1.2	0.1	1.7
COD (gO <sub>2</sub> .kg <sup>-1</sup> )	44	353	23	19	2	31
Total Solids (g.kg <sup>-1</sup> )	42	340	19	22	7	34
Total Volatile Solids (TVS, g.kg <sup>-1</sup> )	30	270	11	10	2	14
Total Suspended Solids (g.kg <sup>-1</sup> )	36	-	8	15	1	28

RS: raw slurry, SSF: solid separated fraction, LSF: liquid separated fraction entering the biological reactor, AS: aerated slurry, S: supernatant, LD: liquid decantation fraction, BS: biological sludge.

The gas emission factors of CH<sub>4</sub>, CO<sub>2</sub>, NH<sub>3</sub> and N<sub>2</sub>O are presented in table 2.

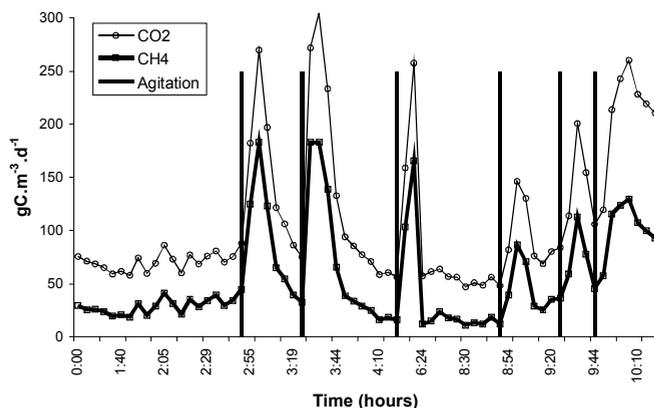
**Table 2.** Gaseous emissions factors (Mean values, ND: no detection).

Gas	Storage/treatment					
	RS	SSF	ABT	AS	S	BS
NH <sub>3</sub>	6.7 gN.m <sup>-2</sup> .d <sup>-1</sup>	28 gN.T <sup>-1</sup> .d <sup>-1</sup>	ND	0.16 gN.m <sup>-2</sup> .d <sup>-1</sup>	0.35 gN.m <sup>-2</sup> .d <sup>-1</sup>	0.3 gN.m <sup>-2</sup> .d <sup>-1</sup>
N <sub>2</sub> O	ND	ND	0.014 gN.m <sup>-3</sup> .d <sup>-1</sup>	ND	ND	ND
CH <sub>4</sub>	49.8 gC.m <sup>-3</sup> .d <sup>-1</sup>	60 gC.T <sup>-1</sup> .d <sup>-1</sup>	0.8 gC.m <sup>-3</sup> .d <sup>-1</sup>	7.6 gC.m <sup>-3</sup> .d <sup>-1</sup>	ND	5.8 gC.m <sup>-3</sup> .d <sup>-1</sup>
CO <sub>2</sub>	41.9 gC.m <sup>-3</sup> .d <sup>-1</sup>	820 gC.T <sup>-1</sup> .d <sup>-1</sup>	11.4 gC.m <sup>-3</sup> .d <sup>-1</sup>	4.8 gC.m <sup>-3</sup> .d <sup>-1</sup>	ND	7.1 gC.m <sup>-3</sup> .d <sup>-1</sup>

RS: raw slurry, SSF: solid separated fraction, ABT: aerobic biological treatment, AS: aerated slurry, S: supernatant, BS: biological sludge.

The CH<sub>4</sub> emissions of pig slurry is higher than the maximum value of 35.8 gCH<sub>4</sub>.m<sup>-3</sup>.d<sup>-1</sup>. cited in previous studies (Martinez et al., 1995, 2003, Husted, 1993, 1994). This value may have been the result of the frequent pit agitation which amplifies the emissions after agitation (Figure 1). The high production of CH<sub>4</sub> and CO<sub>2</sub> of the solid separated fraction is probably due to the

high concentration of water and biodegradable organic matter, two favourable parameters for composting. These results point out the need to a better management of this product for reducing the  $\text{CH}_4$  emissions. The  $\text{CH}_4$  and  $\text{CO}_2$  emissions of the biological sludge were similar to those of aerated slurry. The  $\text{CH}_4$  and  $\text{CO}_2$  emitted by the supernatant lagoon were null probably due to the unfavourable winter temperatures for transforming the low biodegradable organic matter (Safley and Westerman, 1989; Husted, 1993, 1994.)



**Figure 1.** Influence of raw slurry (RS) agitation on gas emission ( $\text{CH}_4$  and  $\text{CO}_2$ ).

The  $\text{NH}_3$  rates of raw slurry were relatively stable ( $6.7 \text{ gN-NH}_3 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ ) despite the TAN variation. This  $\text{NH}_3$  rate was also higher than those cited in the literature (De Bode, 1990; Sommer et al., 1993; Guingand, 2002). As for  $\text{CH}_4$ , the pit agitation was probably the main reason of these considerable emissions. The high  $\text{NH}_3$  emissions of the solid separated fraction ( $28 \text{ gN-NH}_3 \cdot \text{T}^{-1} \cdot \text{d}^{-1}$ ) were related to the high TAN ( $4.8 \text{ gN} \cdot \text{kg}^{-1}$ ) and the high temperature of the heap ( $65^\circ\text{C}$ ). The  $\text{NH}_3$  emitted during the storage of the aerated slurry, biological sludge and the supernatant were low according to their low TAN content.  $\text{N}_2\text{O}$  emissions were only detected during the aeration of separated slurry with a mean value of  $0.014 \text{ gN-N}_2\text{O} \cdot \text{m}^{-3} \cdot \text{d}^{-1}$ . These results are consistent considering previous studies (Beline, 1998, Phillips and al., 1997, Osada and al., 1998) which described that  $\text{N}_2\text{O}$  production takes place mainly during the nitrification/denitrification process requiring aerobic and anaerobic conditions. The emission rates obtained in this study and the variance within the data set are mainly related to the difference of stored product characteristics. Indeed, the manure characteristics are one of the main parameters influencing the methane and ammonia emissions (De Bode, 1990; Husted, 1994; Sharpe, 2002). Under favourable conditions (temperature, pH...), the production of  $\text{CH}_4$  is a function of the content of degradable organic matter while the  $\text{NH}_3$  volatilisation is a function of the TAN. The higher these parameters are, the more important are the emissions of  $\text{CH}_4$  and  $\text{NH}_3$ . The annual emission rates were estimated by using the field-experimental data. We considered only the variation of the daily volume of the raw slurry and the treatment-by-products. The estimates presented in Table 3 concern a 200-sow production unit. These results indicate that the solid fraction storage is the main source of  $\text{NH}_3$  with about 59% of the annual emissions, followed by the raw slurry storage (29%). The raw slurry are also the principal source of  $\text{CH}_4$  with 54% of the annual emission. The gas emissions of the aerated by-product storages were less than for the raw slurry.  $\text{N}_2\text{O}$  was only measured above the biological reactor with  $2.6 \text{ kgN}$  per year.

## CONCLUSIONS

This study confirms the influence of the composition and management of manure on gase-

ous emissions. The higher the ammonium and organic matter level are, the higher are the emissions of  $\text{NH}_3$ ,  $\text{CH}_4$  and  $\text{CO}_2$ . The annual estimates of  $\text{NH}_3$ ,  $\text{CH}_4$  and  $\text{CO}_2$  rate of this aerobic treatment unit were about 480 kgN, 3400 kgC and 12700 kgC respectively. The majority of the  $\text{NH}_3$  and  $\text{CH}_4$  emissions were from the solid separated fraction and the raw slurry. The main source of  $\text{N}_2\text{O}$  was the biological reactor. This study will be completed with a new field experiment to consider the seasonal effect on gas emissions.

**Table 3.** Annual gas estimations (ND: no detection).

Gas	Storage/treatment unit						Total
	RS	SSF	AT	AS	S	BS	
$\text{NH}_3$ kgN.y <sup>-1</sup>	139 (29%)	283 (59%)	ND	1.5 (0.3%)	44 (9%)	12 (3%)	480 (100%)
$\text{N}_2\text{O}$ kgN.y <sup>-1</sup>	ND	ND	2.6 (100%)	ND	ND	ND	2.6 (100%)
$\text{CH}_4$ kgC.y <sup>-1</sup>	1854 (54%)	605 (18%)	143 (4%)	277 (8%)	ND	554 (16%)	3433 (100%)
$\text{CO}_2$ kgC.y <sup>-1</sup>	1560 (12%)	8273 (65%)	2047 (16%)	175 (1.5%)	ND	678 (5.5%)	12732 (100%)

## REFERENCES

- Beline, F. 1998. Etude des transferts d'azote par nitrification/dénitrification ( $\text{N}_2$ ,  $\text{N}_2\text{O}$ ,  $\text{NH}_3$ ) au cours du traitement aérobie et du stockage du lisier de porc. Essais avec 15N. Thèse de Doctorat, Université de Perpignan.
- Beline F., Guiziou F., Peu P., Martínez J. 1999. Quantification des émissions de protoxyde d'azote ( $\text{N}_2\text{O}$ ) au cours du traitement aérobie du lisier de porc. Rapport d'étude CEMAGREF, 21p.
- Beline F., Daumer M.L., Guiziou F., Rapon P. 2001. Bilan de fonctionnement des unités de traitement biologique aérobie du lisier de porcs. Rapport d'Etude Cemagref, 122p.
- Beline F., Guiziou F., Peu P., Daumer M.L. 2002. Bilan environnemental des procédés de traitement biologique des lisiers de porcs. Rapport d'Etude Cemagref/ADEME, 43p.
- Beline F., Daumer M.L., Guiziou F. 2003. Ingénieries EAT, 34: 25-33.
- De Bode M.J.C. 1990. In "Odour and Ammonia Emissions from Livestock Farming", Elsevier Applied Science, London. pp. 59-66.
- Guingand N. 2002. Journées Rech. Porcine en France, 34: 161-166.
- Husted S. 1993. Atmos. Environ., 27A: 1635-1642.
- Husted S. 1994. J. Environ. Qual., 23: 585-592.
- Martínez J., Guiziou F., Peu P., Gueutier V. 2003. Biosys. Eng., 85: 347-354.
- Martínez J., Guiziou F., Gueutier V. 1995. Dossier de l'Environnement de l'INRA 10: 17-22.
- Levasseur P. 1998a. Techni Porc, 21: 17-23.
- Levasseur P. 1998b. Techni Porc, 21: 19-29.
- Levasseur P. 2002. Techni Porc, 25: 19-25.
- Osada T., Rom H. B., Dahl P. 1998. ASAE, 41: 1109-1114.
- Peu P., Beline F., Martínez J. 1999. J. Agric. Engng. Res., 73: 101-104.
- Phillips V.R., Sneath R.W., Williams A.G., Welch S.K., Bergess L.R., Demmers T.G.M., Lynn Short J. 1997. In: *Proceedings of the International Symposium : Ammonia and odour control from animal production facilities*. Voermans J.A.M., Monteny G., Vinkeloord (Eds), October 6-10, The Netherlands, pp 197-208.
- Safley J., L. M., Westerman P. W. 1989. Biol. Wastes, 27: 43-62.
- Sharpe R. R., Harper L. A., Byers F. M. 2002. Agric. Ecosys. Environ., 90: 17-24.
- Sommer S.G., Christensen B.T., Nielson N.E., Schjorring J.K., 1993, J. Agric. Sci., 121: 63-71.
- Zahn J. A., Hatfield J. L., Laird D. A., Hart T. T., Do Y. S., Dispirito A. A. 2001. J. Environ. Qual., 30: 635-647.