

The effect of continuous and intermittent aerobic treatment of pig slurry on nitrous oxide emissions.

L'influence de l'aération continue et séquentielle sur les émissions de protoxyde d'azote au cours du traitement aérobie du lisier de porc.

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Abstract

A laboratory treatment system was designed to study the fate of nitrogen during aerobic treatment of pig slurry. Different aeration processes, continuous and intermittent, were evaluated. For each of the four runs evaluated, the nitrogen mass balance was determined including measurement of the gaseous nitrogen forms (ammonia and nitrous oxide). For continuous aeration, nitrogen removal could rise up to 95% of the total ammoniacal content of the raw slurry (redox control = 0 mV_{Ag/AgCl}) but was less important (45.7%) with redox potential control = -50 mV_{Ag/AgCl} due to oxygen limitation for ammonium oxidation. Concurrently, intermittent aeration with aeration sequences of 9 and 15 hours/day led to a nitrogen removal of about 82% of total ammoniacal nitrogen content of the raw slurry. Between 30-33% of total ammoniacal nitrogen content of the raw slurry was emitted as N₂O during continuous aeration while N₂O emissions could be reduced from 30% of the total ammoniacal nitrogen content of the raw slurry during intermittent aeration (aeration: 2.5 hours, anoxic: 1.5 hours) to 0 using anoxic period longer (aeration: 1.5 hours, anoxic: 2.5 hours).

Résumé

Un pilote de laboratoire a été développé afin d'étudier le devenir de l'azote au cours du traitement aérobie du lisier. Différents procédés aérobies, l'aération en continu et l'aération séquentielle, ont été évalués. Pour chacune des 4 séquences testées, un bilan azoté incluant la mesure des formes azotées gazeuses (ammoniac et protoxyde d'azote) a été effectué. L'aération continue a permis d'obtenir une élimination d'azote de l'ordre de 95% de l'azote ammoniacal du lisier brut (contrôle rédox = 0 mV_{Ag/AgCl}). L'élimination a été moins importante avec un contrôle rédox = -50 mV_{Ag/AgCl} à cause de la limitation en oxygène pour la nitrification. L'aération séquentielle, avec des séquences d'aération de 9 à 15 heures par jour, a permis une élimination d'azote d'environ 82% de l'azote ammoniacal du lisier brut. Environ 30-33% de l'azote ammoniacal du lisier brut a été émis sous forme de N₂O pendant l'aération continue tandis que les émissions de N₂O ont pu être réduites, pendant l'aération séquentielle, de 30% (aération: 2.5 heures, anoxie: 1.5 heures) à 0% en utilisant des périodes anoxiques plus longues.

(aération: 1.5 hours, anoxie: 2.5 hours).

1. Introduction

Aerobic treatment of pig slurry is a source of losses of nitrogen as nitrous oxide (N_2O). N_2O is an important greenhouse gas contributing 260 times more than carbon dioxide on a weight bases¹ and is also implicated in ozone destruction². Anthropogenic N_2O emissions from cultivated soils, waste treatment, industrials processes, ... contribute largely to the increase of atmospheric concentration estimated at a rate of 0.2-0.3% per year¹.

Burton *et al.*³ and Willers *et al.*⁴ observed N_2O emissions during treatment of pig slurry up to 13% of the total nitrogen content of the raw slurry. Also, the increasing of aerobic treatment farm scale units could resulted in an increasing of N_2O emissions. Nevertheless, Osada *et al.*⁵ indicated that it is possible to reduce N_2O emissions during aerobic treatment using intermittent aeration. Indeed, they observed N_2O emissions of about 35% of the total nitrogen content of the influent using continuous aeration and sequential feeding (daily) whereas N_2O emissions represented less than 1% using intermittent aeration.

This paper compares the N_2O emissions from continuous and intermittent aeration of pig slurry in order to determine possible conditions of nitrogen removal without N_2O emissions.

2. Methods and Procedures

2.1. The slurry

Pig slurry was collected from an experimental farm in Brittany (Caulnes, 22). The runs were carried out on the liquid fraction of handled separated (0.63 mm) slurry. The slurry composition varied between runs but was consistent through the duration of each. Its mean composition is given on table 1.

Runs	TN mgN/kg	TAN mgN/kg	TS g/kg	TSS g/kg	VSS g/kg
1	4285 (68)	3153 (78)	32.6 (0.5)	20.1 (0.4)	11.7 (0.3)
2	3544 (19)	2520 (31)	29.0 (0.5)	16.9 (0.2)	9.8 (0.2)
3	2747 (7)	1683 (22)	30.3 (0.2)	20.5 (0.2)	11.6 (0.3)
4	2732 (13)	1692 (6)	30.7 (0.2)	20.8 (0.2)	11.8 (0.1)

Table 1:

Characteristics of the slurry used in each experiment. Value are expressed on a fresh weight bases (Standard deviation shown in paranthese).

2.2. Laboratory treatment system

The laboratory treatment system (figure 1) consisted of a ten litre glass reactor (5 litre working volume), a feed tank (5l) and a discharge tank (5l).

Continuous aeration

The reactor was continuously fed with a peristaltic pump and was discharged every 4 hours with a second peristaltic pump. The slurry was mixed in the reactor by a magnetic stirrer and a flow rate of slurry recirculated from the bottom to the top of the vessel at a flow rate approximately of 0.3 m³/h. This flow of slurry resulted in mixing, aeration and foam control. Redox potential was continuously monitored in the flow of slurry and recorded on a data logger. This data logger could be programmed with a set point for redox potential to enable two solenoid valves to switch and allow the entry of air or di-nitrogen gas into the system in order to control the aeration level. The injection of air or di-nitrogen gas resulted in a constant gaseous flow rate which was quantified by a gas meter.

Intermittent aeration

The reactor was fed at the beginning of each anoxic period with a peristaltic pump and was discharged at the end of each anoxic period with a second peristaltic pump. The slurry was mixed in the reactor by a magnetic stirrer during both periods (aeration and anoxic) and a flow rate of slurry recirculated from the bottom to the top of the vessel at a flow rate approximately of 0.3 m³/h during aeration period. This flow of slurry resulted in mixing, aeration and foam control during aeration period and the cut off of this flow allow anoxic conditions. Continuous injection of air resulted in a constant gaseous flow rate which was quantified by a gas meter.

For both processes, residence time was controlled by fixing the rate of feeding of raw slurry. Gaseous flow rate was monitored continuously for nitrous oxide by non-dispersed infrared analyzer. Ammonia emissions were quantified by recirculating the gas through an acid trap (H₂SO₄, 0.5N).

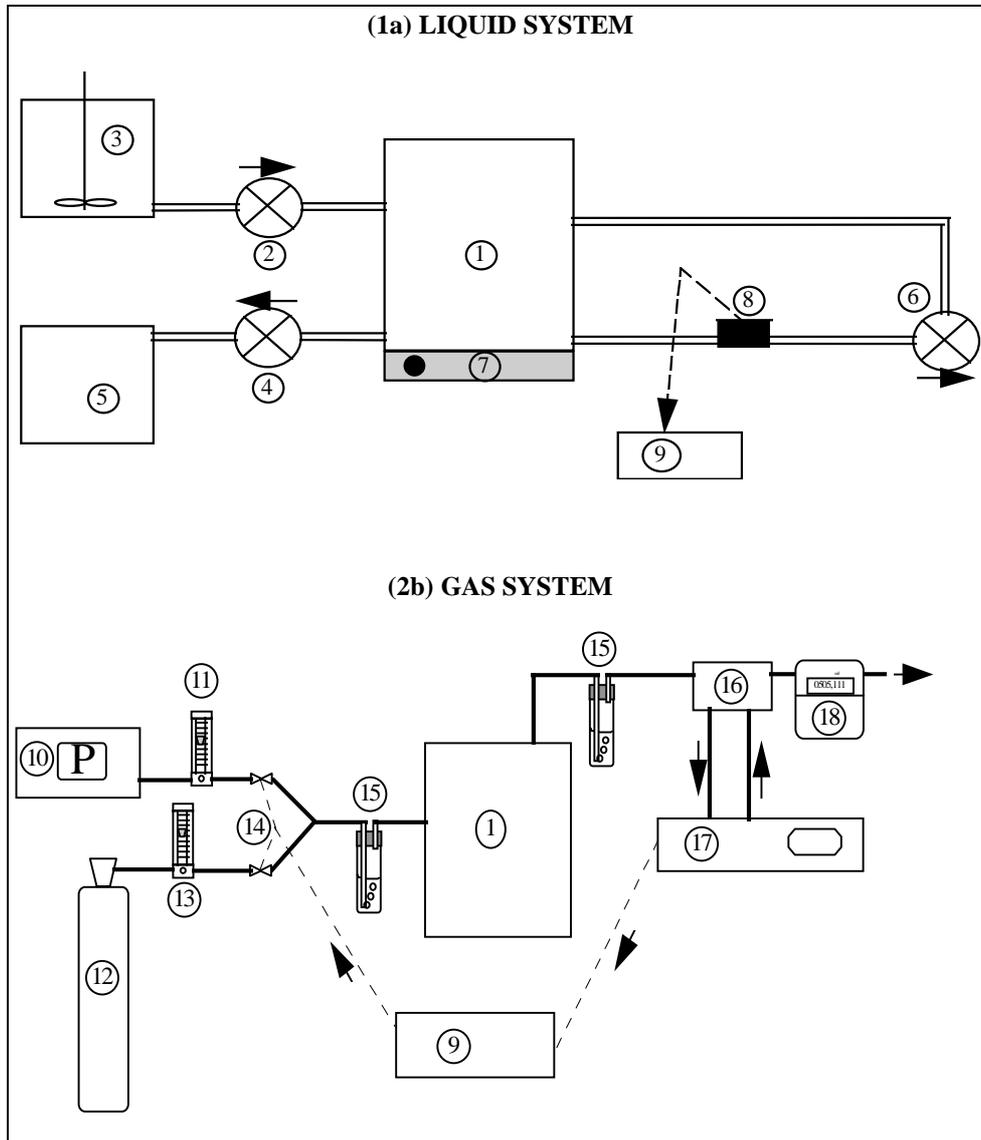


Figure 1

Laboratory treatment system showing : aeration reactor (1), feed slurry peristaltic pump (2), feed slurry vessel (3), discharged slurry peristaltic pump (4), discharged slurry vessel (5), recirculation peristaltic pump (6), magnetic stirrer (7), redox potential sensor (8), data logger (9), air pump (10), gas flow meter (11 and 13), di-nitrogen gas compressor (12), solenoid valves (14), acid trap for ammonia (15), buffer vessel (16), infrared analyzer (17), and gas meter (18).

In each experiment, analyses started after a period equal to three residence times.

Representative samples of raw slurry and treated slurry were then taken daily throughout a two week monitoring period and analyzed for total solids (TS), total suspended solids (TSS), volatile suspended solids (VSS), total nitrogen (TN), total ammoniacal nitrogen (TAN), nitrate and nitrite. The values obtained were averaged over the monitoring period and the values indicated in this paper are the means values of 10-15 individual analyses. All input and output slurry quantities were recorded daily in order to establish mass balances during the monitoring period.

Two continuous aeration runs were carried out with different aeration level in order to evaluate the effect of oxygen dissolved on N₂O emissions. Concurrently, two intermittent aeration runs were performed with different anoxic period times. A total of 4 runs were evaluated with residence time of ca. 5 days; these are summarized in table 2.

Runs	Residence time (days)	Regimes
1	4.6	- Continuous feeding - Sequential discharging (every 4 hours) - Continuous aeration with redox control at 0 mV Ag/AgCl
2	4.8	- Continuous feeding - Sequential discharging (every 4 hours) - Continuous aeration with redox control at -50 mV Ag/AgCl
3	4.5	- Sequential feeding (at the start of each anoxic period) - Sequential discharging (at the end of each anoxic period) - Intermittent aeration with 2.5 hours of aeration and 1.5 hours of anoxic period
4	4.5	- Sequential feeding (at the start of each anoxic period) - Sequential discharging (at the end of each anoxic period) - Intermittent aeration with 1.5 hours of aeration and 2.5 hours of anoxic period

Table 2
Treatment regimes

2.3. Chemical analyses

TAN was analyzed by steam distillation using MgO. Nitrate plus nitrite were determined after reduction with Devarda's alloy, and nitrate by the same method following destruction of nitrite with sulphamic acid. Nitrite was obtained by difference. Samples were digested using the Kjeldahl procedure for raw slurry or Olesen modified procedure⁶ for treated slurry and distilled with NaOH (30%) to determine TN. TS, TSS, VSS were analyzed by standard methods (APHA, 1992⁷). All nitrogen analyses, particularly nitrate and nitrite were made within one hour of sampling. For the other analyses, samples were kept at 4°C (storage < 2 days) or frozen (storage > 2 days).

3. Results and discussions

3.1. Nitrogen transformations

Residues time were closed to 5 days in all treatment, and in this conditions, no transformation in organic nitrogen pool was observed. Thus, only mineral pool is considered and the results of nitrogen behaviour during treatment, expressed as the percentage of the TAN of the raw slurry, are presented in figure 2.

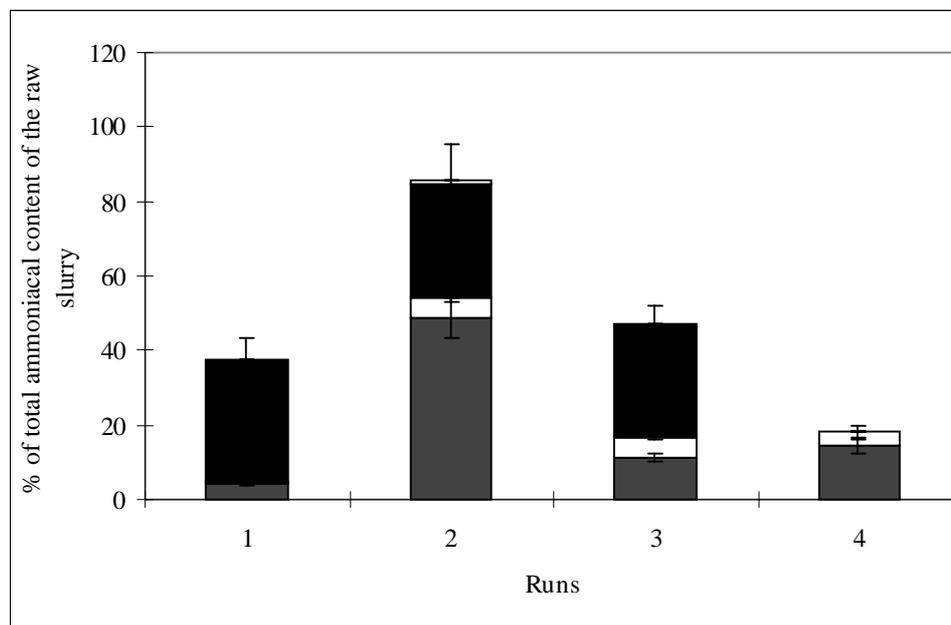


Figure 2
 Nitrogen behaviour during aeration of pig slurry, ammonia, nitrate plus nitrite,
 ■ nitrous oxide and □ ammonia (Vertical line = \pm standard deviation).

We observed an ammoniacal nitrogen oxidation of 95.5% with redox potential controlled to 0 mV_{Ag/AgCl} (run 1) whereas it was only of 51% with redox potential controlled to -50 mV_{Ag/AgCl} (run 2). Further experiments shown an increase of ammonium concentration in the reactor (Fig 3) when redox potential fall from 0 to -50 mV_{Ag/AgCl}. Also, it appears that oxygen supplied become a limiting factor when redox potential fall below 0 mV_{Ag/AgCl}. Concurrently, ammonium oxidation varied between 85.8 and 88.8% using intermittent aeration (Run 3 and 4). Residual ammonium concentrations were, in these cases, partly due to the ammonium supplied by the feeding at the start of the anoxic period.

Low aeration level and intermittent aeration allow denitrification occur^{8,9}. Also, a large part of oxyded ammonium was removed as gas (89 - 95.5%).

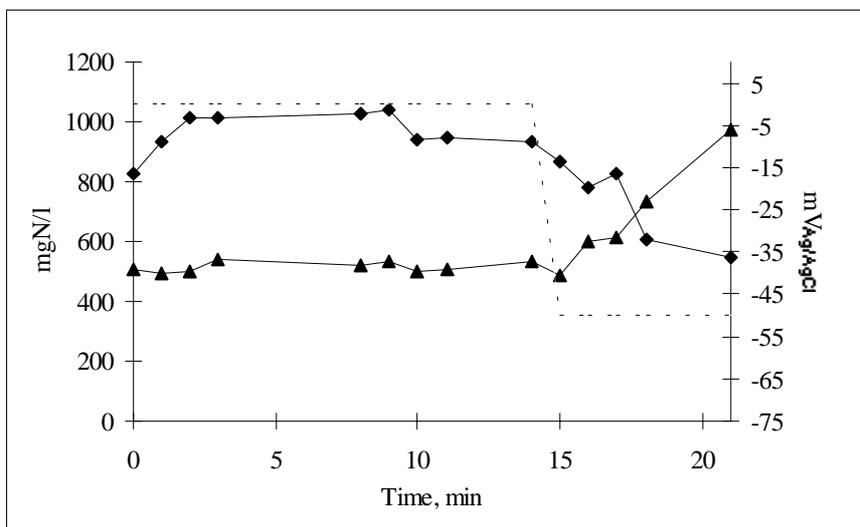


Figure 3
Influence of redox potential on nitrogen transformation during aerobic treatment of pig slurry, ◆ nitrate plus nitrite concentration, ▲ ammonium concentration and — redox potential.

3.2 Gaseous emissions

As observed by Osada *et al.*⁵, the removal of oxidized ammonium led to N₂O emissions in case of continuous aeration. N₂O emissions represented between 30 and 33% of the TAN of the raw slurry. The decrease of redox potential from 0 (run 1) to -50 mV_{Ag/AgCl} (run 2) did not prevent these emissions. Contrary to Osada *et al.*⁵, the use of intermittent aeration (run 3) did not reduce systematically the N₂O emissions. Indeed, the N₂O emissions was, in this run, similar to continuous aeration (30%). Nevertheless, the continuous monitoring of gaseous emissions allowed us to observe that emissions occurred mainly at the start of aeration stage (Fig 4a).

N₂O emissions were assumed to be due to, in this run, the stripping of N₂O produced during incomplete denitrification and trapped in the slurry. Indeed, foam avoided exchange between air and slurry during anoxic stage. Also, an increase of the time of anoxic period (run 4) enabled to a full denitrification into di-nitrogen gas and prevented N₂O emissions (fig 4b).

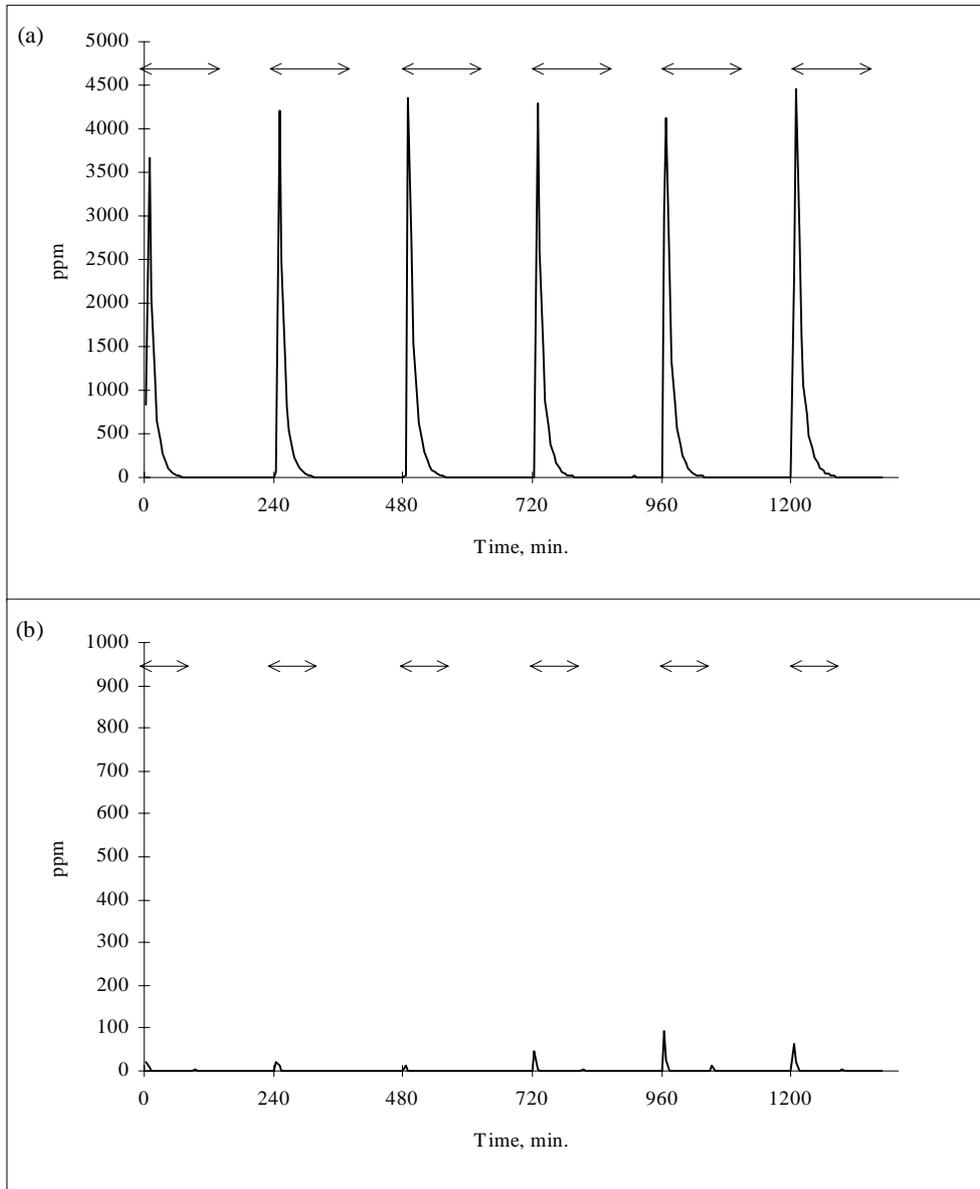


Figure 4
 Continuous emissions of nitrous oxide during intermittent aeration,
 — aeration period. (a: run 3 and b: run 4).

Incomplete oxidation of ammonium in run 2 led to ammonia emission which represented near 1% of total ammoniacal content of the raw slurry. Nevertheless, a full nitrification of ammonium (run 1, 3 and 4) resulted in the low ammonium concentration in the reactor avoiding these emissions.

4. Conclusions

Nitrogen removal between 82 and 95% of the total ammoniacal nitrogen content of the raw slurry could be obtained during aerobic treatment of pig slurry using nitrification and denitrification processes. Continuous aeration with low level of aeration and intermittent aeration allow nitrification and denitrification to occur. Nevertheless, continuous aeration leads systematically to nitrous oxide emissions representing up to 33 % of the total ammoniacal nitrogen content of the raw slurry whereas the use of intermittent aeration can prevent these emissions using a long time of anoxic period allowed complete denitrification. Moreover, ammonia emissions can be avoided by a complete oxidation of ammonium.

Acknowledgment

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