

SLURRY TREATMENT WITH FOOD INDUSTRY WASTES FOR REDUCING METHANE, NITROUS OXIDE AND AMMONIA EMISSIONS

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ABSTRACT

Livestock manure is the main source of ammonia (NH₃) emissions and an important source of greenhouse gases (GHG) especially methane (CH₄) and nitrous oxide (N₂O). Ammonia emissions contribute to eutrophication and acidification of water, soils and ecosystems. The greenhouse gases contribute to the global warming. These gaseous emissions can be reduced by controlling the pH-value of manure. Acidifying the manure can reduce CH₄, N₂O and NH₃ emissions. Inorganic acids are feasible, but have several disadvantages, e.g. soil contaminants. The use of organic acids is an efficient but expensive method; therefore using the food industry wastes which already contain organic acids is highly feasible. The objective of this research is to investigate the effects food industry wastes as manure additives on the emissions of CH₄, N₂O and NH₃. Dairy cattle manure was treated with food industry wastes (whey and waste of citrus and orange juices industry) and the gas flux of CH₄, N₂O and NH₃ were quantified using a specially designed gas detection system which consists of several flasks and a multi-gas monitor. The results showed that the gas fluxes emitted from manure treated with waste of citrus juice industry were 13, 0.219 and 2.523 g m⁻² day⁻¹ for CH₄, N₂O and NH₃ respectively. The gas fluxes emitted from manure treated with waste of orange juice industry were 13.58, 0.223 and 2.581 g m⁻² day⁻¹ for CH₄, N₂O and NH₃ respectively. The gas fluxes emitted from manure treated with whey were 17.45, 0.279 and 3.063 g m⁻² day⁻¹ for CH₄, N₂O and NH₃ respectively. The gas fluxes emitted from the control sample (mixture of manure and water) were 58.21, 0.347 and 18.9 g m⁻² day⁻¹ for CH₄, N₂O and NH₃ respectively.

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Consequently, manure treatment with food industry wastes enabled high reduction rates of gaseous emissions from manure.

Keywords: *Manure management, slurry treatment, food industry wastes, emissions abatement techniques, emission factors, greenhouse gases, ammonia, methane, nitrous oxide.*

INTRODUCTION

The Kyoto Protocol, which is a protocol to the United Nations Framework Convention on Climate Change (UNFCCC), aimed at achieving the stabilization of greenhouse gas concentrations in the atmosphere at a level that would prevent dangerous anthropogenic interference with the climate system. Under this protocol, several countries committed themselves to a reduction of the greenhouse gases. Furthermore, the Gothenburg Protocol is a multi-pollutant protocol designed to reduce acidification, eutrophication and ground-level ozone by setting emissions ceilings for several pollutants, where ammonia is one of them. On the other side, the agriculture (with its two main sectors: plant and animal production) is one of the main sources of greenhouse gases emissions and the main source of ammonia emissions. Thus, reducing greenhouse gases and ammonia emissions from the agricultural sector is crucial.

Enteric fermentation and manure management account for 35% to 40% of the total anthropogenic methane (CH₄) emissions and 80% of CH₄ release from agriculture (FAO, 2006). Livestock activities contribute with 65% of the global anthropogenic nitrous oxide (N₂O) emissions and account for 75% to 80% of the emission from agriculture (FAO, 2006). CH₄ and N₂O are greenhouse gases (GHG) with global warming potentials of 23 and 296 times that of CO₂, respectively (IPCC, 2007). About 94% of global anthropogenic emissions of ammonia (NH₃) to the atmosphere originate from the agricultural sector of which close to 64% is associated with livestock management (FAO, 2006). Excessive levels of NH₃ emissions contribute to eutrophication and acidification of water, soils and ecosystems (Schuurkes and Mosello, 1988). In addition to the global warming potential of the greenhouse gases, ammonia emissions

contribute to global warming when the ammonia is converted to nitrous oxide (*Berg, 1999; Sommer et al., 2000*).

Manure is animal excreta (feces and urine) collected from animal buildings. Whereas slurry is a mixture of scraped animal excreta and flushing water and is collected from animal buildings. Hence, slurry is a mixture of manure and water. On the other hand, litter is animal excreta and bedding material collected from animal buildings (*Samer, 2011a*). Liquid manure storage facilities are sources of gaseous emissions of NH_3 and greenhouse gases especially CH_4 and N_2O . Additives can reduce gaseous emissions from swine waste lagoons and pits. The additives have the potential to reduce methane emissions from anaerobic swine lagoons (*Shah and Kolar, 2012*). Different materials for covering liquid manure storage facilities have been investigated and are in use for mitigating odor and ammonia emissions (*Sommer et al., 1993; Williams, 2003*). These materials abate also methane and nitrous oxide emissions. Different materials for covering liquid manure storage facilities to reduce gaseous emissions were investigated on laboratory scale: perlite, lightweight expanded clay aggregate and chopped straw - both individually and combined with lactic acid or saccharose, respectively (*Berg et al., 2006a*). Manipulating the balance between ammonia and ammonium by lowering the pH-value of slurry is another measure to reduce emissions (*Stevens et al., 1989; Oenema and Velthof, 1993; Hendriks and Vrieling, 1997; Kroodsmas and Ogink, 1997; Martinez et al., 1997; Beck and Burton, 1998; Pedersen, 2003*).

Ammonia and methane emissions can be controlled by pH-value. Manipulating the pH-value of slurry has an effect on the balance between ammonia and ammonium. The pH-values of untreated slurries range between 7 up to 8 usually. Lowering the pH reduces the gaseous emission. From former investigations it is known that a slurry pH around 5.5 can reduce ammonia emission by 80 to 90% (*Al-Kanani et al., 1992; Berg et al., 2006a,b; Husted et al., 1991; Li et al., 2006; Pain et al., 1990; Stevens et al., 1989*). A slurry pH below 4.5 nearly avoids ammonia emission (*Hartung and Phillips, 1994*). The pH-value influences the activities of microorganisms. Higher methane production occurs, when the pH-value is between 6 and 7 (*Lay et al., 1997*). A slurry pH below 6 is nec-

essary to reduce methane emission and below 5 impede methane formation (*Berg et al., 2006a,b*).

Whereas the use of inorganic acids has several disadvantages, using organic acids is a promising possibility to reduce methane, nitrous oxide and ammonia emissions (*Berg and Hoernig, 1997; Berg and Pazsiczki, 2003; Berg, 2003*). *Wheeler et al. (2011a)* stated that amendments can be practical and cost-effective for reducing NH_3 and GHG emissions from dairy manure. They found that amendment products that act as microbial digest, oxidizing agent, masking agent or adsorbent significantly can reduce NH_3 by more than 10%. Whereas, microbial digest/enzymes with nitrogen substrate appeared effective in reducing CH_4 fluxes. For both CH_4 and CO_2 fluxes, aging the manure slurry for 30 days can significantly reduce gas production. *Reinhardt-Hanisch (2008)* mentioned that the implementation of urease inhibitors is effective in reducing ammonia emissions from cattle and pig slurry. *Wheeler et al. (2011b)* stated that some amendments reduced odor emission depending on the storage period. Previous studies have evaluated additives for reducing gaseous emissions from manure in laboratory using glass jars or plexiglass tanks and a multi-gas monitor (*Wheeler et al., 2011c; Reinhardt-Hanisch, 2008; Berg et al., 2006b*).

Mathematical models and computer programs were developed to be implemented in constructing manure tanks and manure handling systems (*Samer et al., 2011a, 2008a*) as well as biogas plants (*Samer, 2010*), whereas the location of such systems in the farm vicinity was specified to be downwind to avoid gas transmission to the different farm facilities with a specified minimum distance between the farm and any adjacent residential communities, roads and ecosystems (*Samer et al., 2008b*). On the other hand, manure pits for temporary manure storage in livestock buildings form another effective source of gaseous emissions as shown in the different emissions inventories (*Samer, 2013a*). Airflow profiles affect the gas emission rates which increase with the increasing air volumetric flow rates and air velocities, where free air streams allow more gas release through convection mass transfer (*Samer et al., 2011a; Samer, 2012*). Additionally, gaseous emissions increase with increasing temperatures (*Samer et al., 2012, 2011b; Samer, 2011b*). The implemen-

tation of proper waste management which is safe to the surroundings fulfils the green building specifications (*Samer, 2013b*).

The hypothesis of this study was: treating manure with acidic liquid biowastes (e.g. wastes of citrus, orange and milk industries) will reduce gaseous emissions, where the organic acids in the acidic liquid biowastes will reduce the pH of manure which consequently mitigates gaseous emissions. Eventually, this process is an integrated waste management of both manure and acidic liquid biowastes. Therefore, the objective of this study was to investigate the possibility of reducing gas emissions (CH_4 , N_2O and NH_3) from dairy manure by adding low-pH biowastes (whey and waste of citrus and orange juices industry).

MATERIALS AND METHODS

1. Collecting manure

The fresh raw manure was collected randomly from dairy cows (female, 3 years, 580 Kg) holding pen unit.

2. Manure analyses

The pH and the temperature were measured using a pH meter (Jenway 3520, Staffordshire, UK). Total solids (TS), volatile solid (VS) and ash (Table 1) were determined using the standard methods (*EPA, METHOD 1684, 2001*) using muffle furnace (Ney Tech, Vulcan D-550, York, USA).

Table (1): Chemical composition of fresh manure.

Parameter	Value
TS (%)	16.92
VS (%)	12.97
Ash (%)	4.02
Organic carbon (% from VS)	44.63
Total Nitrogen	1.6
C:N ratio	25:1
pH	7.13

3. Slurry preparation

The collected raw manure was homogenized by propeller mixer for 30 minutes. Afterwards, manure was poured in a tank and then whey was

added and mixed with the dairy manure. Similarly, these procedures were conducted to mix manure with waste of citrus or orange juices industry.

4. Experiments

The experiments were carried out through two phases: preparatory experiments and laboratory experiments which were the main experiments.

4.1. Preparatory experiments

Pre-experiments were carried out to ascertain the relationship between the amount of the added biowastes and the pH-value. For this purpose different biowaste: manure ratio (1:4, 1:3, 1:2 and 1:1) were investigated. The pH-values were measured over a period of 24 hours. Samples of dairy manure were mixed with whey, waste of citrus or orange juices industry with different ratios. Whey collected from milk industries was a mixture of whey sucralose and whey protein beverages (pH = 3.9). On the other hand, waste of citrus juice industry contained sodium citrate and sodium acetate (pH = 3.6). Similarly, waste of orange juice industry contained sodium citrate and sodium acetate but with different acidity (pH = 3.75).

The pH and temperature of the mixture were measured using a pH/ION-Meter. This procedure allowed the determination of the best treatment specifications (biowaste:manure ratio, and pH of the mixture), where this procedure should be conducted with any new additive and before the main experiments. All preparatory experiments were carried out under the same laboratory conditions (i.e. lab temperature of 25°C approximately), respectively.

4.2. Main experiments

4.2.1. Experimental set up

A batch aerobic system was designed and implemented in this study. The main experiment tools consist of: flask, temperature control and gas measurement system. A 2.2-liter ($\varnothing = 18$ cm) wide neck reaction Pyrex flask (Scilabware, FR2LF, Staffordshire, UK) was used and plugged (just during the measurements) with tightly Teflon cap equipped and gas outlet connected to the multi-gas monitor (Fig.1).

The temperature was controlled using a thermostatic water bath (Raypa, BAD-12, Barcelona, Spain) and maintained at $38 \pm 0.3^\circ\text{C}$ in order to simulate the toughest weather conditions as the gas emissions increase with

the increasing temperatures. The gas concentrations of methane (CH_4), nitrous oxide (N_2O) and ammonia (NH_3) were measured using an infra-red photo-acoustic analyzer (INNOVA 1412, Innova AirTech Instruments, Ballerup, Denmark) once a day.

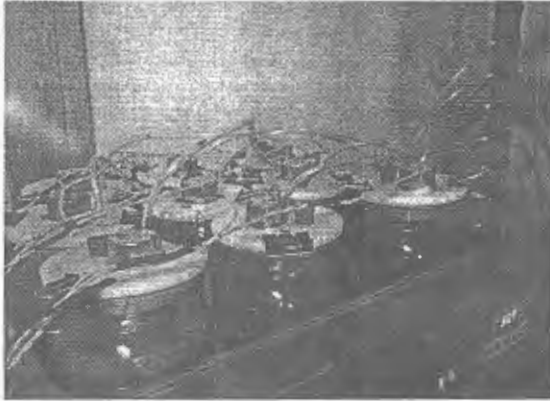


Figure (1): The experimental set-up.

4.2.2. Experimental design

A series of laboratory experiments using 2200 ml flasks were performed in batch operation mode to investigate the influence of acidic liquid bio-wastes on CH_4 , N_2O and NH_3 emissions. Each flask was fed with 800 g of fresh dairy manure (M) mixed with 800 ml acidic liquid biowaste (B): as either whey (P), which is Treatment 1 with 3 replicates; waste of citrus juice industry (C), which is Treatment 2 with 3 replicates; or waste of orange juice industry (O), which is Treatment 3 with 3 replicates. The control sample was a mixture of dairy manure and water (W) which is Treatment 4. The diameter of the sample surface area, after the flasks were filled, was equal to 13.7 cm approximately. Tables 2 and 3 present the initial composition of all flasks used in the study.

One of the most important factors affecting the emission flow rates is the temperature. The optimum temperature for most of involved bacteria groups is the range of 35-40°C. Therefore, 38°C was selected as optimum temperature of bacterial activities as well as to simulate the toughest climatic conditions in summer where the gas emissions increase dramatically.

Table (2): Experimental design of the treatments and their replicates.

Treatment	Replicate	Flask	M:W:P:C:O	M	W	B	Total Volume	Total Mass
				g	ml	ml	ml	g
Treatment 1	Replicate 1	F1	1:0:1:0:0	800	0	800	1867	1867.24
	Replicate 2	F2	1:0:1:0:0	800	0	800	1867	1867.20
	Replicate 3	F3	1:0:1:0:0	800	0	800	1867	1867.27
Treatment 2	Replicate 1	F4	1:0:0:1:0	800	0	800	1867	1867.53
	Replicate 2	F5	1:0:0:1:0	800	0	800	1867	1867.55
	Replicate 3	F6	1:0:0:1:0	800	0	800	1867	1867.61
Treatment 3	Replicate 1	F7	1:0:0:0:1	800	0	800	1867	1867.65
	Replicate 2	F8	1:0:0:0:1	800	0	800	1867	1867.61
	Replicate 3	F9	1:0:0:0:1	800	0	800	1867	1867.72
Treatment 4 (Control)	-	F10	1:1:0:0:0	800	800	0	1867	1867.12

Table (3): Initial chemical composition of the treatments and their replicates.

Treatment	Replicate	Flask	TS	VS	Ash	Organic carbon	C:N ratio	pH Value
			%	%	%	% from VS		
Treatment 1	Replicate 1	F1	8.73	6.18	2.55	44.80	24.6:1	5.34
	Replicate 2	F2	8.65	5.99	2.66	44.76	24.8:1	5.36
	Replicate 3	F3	8.72	6.10	2.62	44.78	24.7:1	5.35
Treatment 2	Replicate 1	F4	8.42	6.53	1.89	44.91	25.2:1	4.87
	Replicate 2	F5	7.95	6.38	1.57	44.84	25.1:1	4.91
	Replicate 3	F6	8.49	6.41	2.08	44.87	25.1:1	4.85
Treatment 3	Replicate 1	F7	8.51	6.58	1.93	45.15	25.3:1	4.97
	Replicate 2	F8	8.43	6.42	2.01	45.18	25.1:1	5.02
	Replicate 3	F9	8.54	6.45	2.09	45.23	25.2:1	4.99
Treatment 4 (Control)	-	F10	7.92	5.89	2.03	44.63	25.0:1	7.13

4.2.3. General procedures

The investigations were carried out in the laboratory. The treatments were replicated 3 times. Dairy manure was stored in containers/flasks with a capacity of 2.2 l each. The dry matter content of the manure was

16.9% and was reduced to 10% when treated with acidic liquid biowastes having high water contents of at least 95%. The storage period was 180 days. Manure was treated with whey, where this was the first treatment. The second manure treatment was with waste of citrus juice industry. The third manure treatment was with waste of orange juice industry. Hence, three treatments, three replicates each, and one control were adopted. Therefore, ten flasks were used. The slurry (manure treated with acidic liquid biowastes having high water contents) was stored in open flasks which were closed and ventilated only during measurement, i.e. ventilated chamber method. In other words, the flasks were open permanently, closed and ventilated only during the measurement of headspace gas concentrations which is the dynamic chamber method. The ventilation rate during measurements was adjusted so that the air in the headspace always was changed one time per minute.

The capacity is 2200 ml for each flask each. At the beginning of the investigations the headspace in each flask was about 333 ml for the control and all treated samples. During the investigation period, the headspace was increasing because of the evaporation. The ventilation rate during the measurements was adjusted accordingly so that the air in the headspace always was changed one time per minute.

The Total Kjeldahl Nitrogen (TKN) and Total Ammoniacal Nitrogen (TAN) were measured in slurry by chemical analysis (according to DIN EN 25663 and DIN 38406-E5-2, respectively) at the beginning and the end of the investigation period. The room temperature was continuously measured throughout the experiments. However, all flasks were controlled using the water bath system to a constant temperature of $38.0 \pm 0.3^\circ\text{C}$. The pH-value of slurry was daily measured 2 cm below the surface and 2 cm above the base (in the sediments) of the slurry flask. The airflow rates were measured through flasks' headspace using airflow meters.

The results of the different treatments and their replicates were compared with each other and with the control sample, i.e. the untreated slurry. The statistical analysis was carried out using SAS v.9.2 (SAS Institute, Inc., Cary, N.C.). The comparisons of the mean values were made by using the t-test and the Wilcoxon test at the 0.05 probability level.

RESULTS AND DISCUSSION

1. Preparatory experiments

The results of the pre-experiments showed that the pH-values subjected to the biowaste: manure ratio of 1:4, 1:3, 1:2 and 1:1, were 6, 5, 4.5 and 4 for waste of citrus juice industry; 6.4, 5.3, 4.7 and 4.2 for waste of orange juice industry; and 6.9, 5.8, 5 and 4.6 for whey, respectively. This procedure allowed the determination of the best treatment specifications (biowaste: manure ratio, and pH of the mixture), where these preparatory experiments recommended a mixture ratio of 1:1 with a significant difference at the 0.05 probability level.

2. Main experiments

The trend of the main experiments is quantifying the gas concentrations from the different treatments using the gas detection system, measuring the pH-values of the different treatments and their replicates and then comparing the results to determine which treatment is the most effective in reducing the emissions of CH₄, N₂O and NH₃.

2.1. Post-treatment manure properties

The results of manure analysis showed that the manure chemical composition have been significantly changed in comparison to the initial chemical composition. For instance, the C:N ratio had a special case where adding whey (Treatment 1) to manure decreased the ratio (Table 3) where the added whey contains protein which increased the N content and consequently decreased the C:N ratio. Table 3 showed that manure treatment with waste of citrus juice was the most effective in decreasing the pH-value of the resulting slurry. Although manure treatment with whey reduced significantly the pH, but its effect was lower than the waste of citrus juice. The final chemical composition of the treatments and their replicates, presented in Table 4, showed that the pH-value slightly increased in comparison to the initial pH-values presented in Table 3. This can be explained by the continuous biochemical reactions as well as the biological activities in slurry which continued during the investigation period (180 days), where these results led to the anticipation that the daily gas concentrations would slightly increase with progress of the investigation.

Table (4): Final chemical composition of the treatments and their replicates.

Treatment	Replicate	Flask	TS	VS	Ash	Organic carbon	C:N ratio	pH Value
			%	%	%	% from VS		
Treatment 1	Replicate 1	F1	5.55	3.77	1.78	43.90	25.6:1	5.79
	Replicate 2	F2	5.48	3.44	2.04	43.81	25.9:1	5.71
	Replicate 3	F3	5.29	3.79	1.50	43.89	25.8:1	5.68
Treatment 2	Replicate 1	F4	5.13	4.01	1.12	44.55	25.5:1	5.31
	Replicate 2	F5	4.83	3.92	0.91	44.58	25.4:1	5.25
	Replicate 3	F6	5.23	3.96	1.27	44.63	25.5:1	5.21
Treatment 3	Replicate 1	F7	5.51	4.11	1.4	44.21	26.0:1	5.40
	Replicate 2	F8	5.21	3.94	1.27	44.14	25.8:1	5.45
	Replicate 3	F9	5.15	3.99	1.16	44.17	26.0:1	5.51
Treatment 4	-	F10	6.01	3.12	2.89	43.03	26.9:1	7.35

2.2. The pH-values

The mean values over the whole investigation period show the differences in reducing the manure pH (Table 5), where each flask was observed 180 times (i.e. daily over a period of 180 days). Furthermore, studying the course measurements of the pH-values over the investigation period provides a complete evaluation. Figure 2 shows the results of the course measurements of the pH-values from the different treatments. The pH-values increased slightly through the 180 days of investigations because of the continuous microbial activities. All treatments with whey and waste of citrus and orange juices industry were able to reduce manure pH. However, waste of citrus juice industry was the most effective additive in reducing pH of the treated manure with an average pH-value of 4.94. Additionally, waste of orange juice industry showed similar effect on manure pH, with an average value of 5.08. Although, the differences between both additives were insignificant (difference at the 0.05 probability level), waste of citrus juice industry was able to achieve the lowest manure pH compared with other treatments (Table 5). Whey was able to reduce manure pH achieving an average pH-value of 5.53 which is higher than the other additives, where the differences to the other additives were significant at the 0.05 probability level. The control (mixture of manure and water) had a mean pH-value of 7.25.

Table (5): Measurements of pH-values of the investigated samples.

Treatment	Replicate	Flask	pH-Value	
			Mean	Standard Deviation
Treatment 1: manure treated with whey	Replicate 1	F1	5.55 A ^a	0.32
	Replicate 2	F2	5.52 A	0.41
	Replicate 3	F3	5.51 A	0.35
Treatment 2: manure treated with waste of citrus juice industry	Replicate 1	F4	4.95 B	0.33
	Replicate 2	F5	4.94 B	0.37
	Replicate 3	F6	4.92 B	0.29
Treatment 3: manure treated with waste of orange juice industry	Replicate 1	F7	5.05 B	0.36
	Replicate 2	F8	5.09 B	0.31
	Replicate 3	F9	5.10 B	0.28
Treatment 4: control, a mixture of dairy manure and water	-	F10	7.25 C	0.19

^a Values with different letters have a significant difference at the 0.05 Probability level.

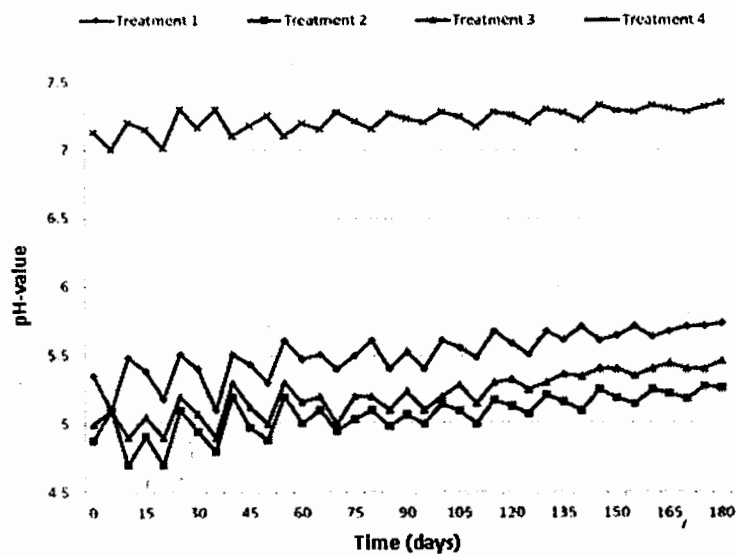


Figure (2): Results of course measurements of the pH-values form the different treatments.

The temperatures of the different flasks were kept constant to $38 \pm 0.3^\circ\text{C}$ using a water bath instrumentation system, therefore the temperatures of the samples were similar and did not have an effect on the measured values. The pH-values in sediments of the different flasks were average of 0.4 (treated) and 0.25 (control) lower than near the slurry surface (significant at the 0.05 probability level).

2.3.1. Methane emissions

Manure treatment with waste of citrus juice industry reduced CH_4 concentrations to average $17.3 \text{ mg CH}_4/\text{m}^3$ (Table 6) which corresponds to a gas flux of $13 \text{ g/m}^2 \text{ day}$ with a mean reduction rate of 77.7%, where this treatment reduced the pH-value to less than 5. Similarly, manure treatment with waste of orange juice industry reduced CH_4 concentrations to average $18 \text{ mg CH}_4/\text{m}^3$ which corresponds to a gas flux of $13.58 \text{ g/m}^2 \text{ day}$ with a mean reduction rate of 76.7%, where this treatment reduced the pH-value to slightly above than 5. The differences between both treatments were insignificant at the 0.05 probability level. However, manure treatment with whey was less effective and reduced CH_4 concentrations to average $23.2 \text{ mg CH}_4 \text{ m}^{-3}$ which corresponds to a gas flux of $17.45 \text{ g/m}^2 \text{ day}$ with significant differences to both other treatments at the 0.05 probability level, where the mean reduction rate was about 70%.

Table (6): Measurements of methane concentrations of the investigated samples.

Treatment	Replicate	Flask	Methane Concentration (mg CH_4/m^3)		Number of Observations
			Mean	Standard Deviation	
Treatment 1: manure treated with whey	Replicate 1	F1	23.4 A ^a	39.2	180
	Replicate 2	F2	23.1 A	39.0	180
	Replicate 3	F3	23.0 A	38.9	180
Treatment 2: manure treated with waste of citrus juice industry	Replicate 1	F4	17.5 B	19.4	180
	Replicate 2	F5	17.2 B	19.2	180
	Replicate 3	F6	17.1 B	19.0	180
Treatment 3: manure treated with waste of orange juice industry	Replicate 1	F7	17.9 B	19.9	180
	Replicate 2	F8	18.0 B	19.7	180
	Replicate 3	F9	18.2 B	19.5	180
Treatment 4: control, a mixture of dairy manure and water	-	F10	77.3 C	129	180

^a Values with different letters have a significant difference at the 0.05 probability level.

The measured CH₄ concentrations in the control sample (mixture of dairy manure and water) were average of 77.3 mg CH₄/m³ which corresponds to a gas flux of 58.21 g/m² day. On the other hand, methane emissions - from the same sample- varied widely during the whole period of investigation.

2.3.2. Nitrous oxide emissions

Manure treatment with waste of citrus juice industry reduced N₂O concentrations to average 0.291 mg N₂O/m³ (Table 7) which corresponds to a gas flux of 0.219 g/m² day with a mean reduction rate of 36.9%, where this treatment reduced the pH-value to less than 5. Similarly, manure treatment with waste of orange juice industry reduced N₂O concentrations to average 0.296 mg N₂O/m³ which corresponds to a gas flux of 0.223 g/m² day with a mean reduction rate of 35.9%, where this treatment reduced the pH-value to slightly above than 5. The differences between both treatments were insignificant at the 0.05 probability level. However, manure treatment with whey was less effective and reduced N₂O concentrations to average 0.371 mg N₂O/m³ which corresponds to a gas flux of 0.279 g/m² day with significant differences to both other treatments at the 0.05 probability level, where the mean reduction rate was about 19.5% only.

Table (7): Measurements of nitrous oxide concentrations of the investigated samples.

Treatment	Replicate	Flask	Nitrous Oxide Concentration (mg N ₂ O m ⁻³)		Number of Observations
			Mean	Standard Deviation	
Treatment 1: manure treated with whey	Replicate 1	F1	0.369 A ^a	0.552	180
	Replicate 2	F2	0.371 A	0.549	180
	Replicate 3	F3	0.373 A	0.551	180
Treatment 2: manure treated with waste of citrus juice industry	Replicate 1	F4	0.290 B	0.511	180
	Replicate 2	F5	0.291 B	0.517	180
	Replicate 3	F6	0.292 B	0.513	180
Treatment 3: manure treated with waste of orange juice industry	Replicate 1	F7	0.293 B	0.520	180
	Replicate 2	F8	0.295 B	0.519	180
	Replicate 3	F9	0.299 B	0.522	180
Treatment 4: control, a mixture of dairy manure and water	-	F10	0.461 C	0.652	180

^a Values with different letters have a significant difference at the 0.05 probability level.

The measured N_2O concentrations in the control sample (mixture of dairy manure and water) were average of $0.461 \text{ mg } N_2O/m^3$ which corresponds to a gas flux of $0.347 \text{ g}/m^2 \text{ day}$. On the other hand, nitrous oxide emissions -from the same sample- varied widely during the whole period of investigation.

2.3.3. Ammonia emissions

Manure treatment with waste of citrus juice industry reduced NH_3 concentrations to average $3.35 \text{ mg } NH_3/m^3$ (Table 8) which corresponds to a gas flux of $2.523 \text{ g}/m^2 \text{ day}$ with a mean reduction rate of 86.7%, where this treatment reduced the pH-value to less than 5. Similarly, manure treatment with waste of orange juice industry reduced NH_3 concentrations to average $3.43 \text{ mg } NH_3/m^3$ which corresponds to a gas flux of $2.581 \text{ g}/m^2 \text{ day}$ with a mean reduction rate of 86.3%, where this treatment reduced the pH-value to slightly above than 5. The differences between both treatments were insignificant at the 0.05 probability level. However, manure treatment with whey was less effective and reduced NH_3 concentrations to average $4.1 \text{ mg } NH_3/m^3$ which corresponds to a gas flux of $3.063 \text{ g}/m^2 \text{ day}$ with significant differences to both other treatments at the 0.05 probability level, where the mean reduction rate was about 83.8%. The measured NH_3 concentrations in the control sample (mixture of dairy manure and water) were average of $25.1 \text{ mg } NH_3/m^3$ which corresponds to a gas flux of $18.9 \text{ g}/m^2 \text{ day}$. On the other hand, ammonia emissions -from the same sample- varied widely during the whole period of investigation. Table 9 shows the Total Ammoniacal Nitrogen (TAN) and the Total Kjeldahl Nitrogen (TKN) of all slurry samples after storage, and the calculated absolute Total Nitrogen Losses (ATNL) of the slurry samples during storage on the basis of TKN before and after storage. The chemical analyses of the TAN and TKN (Table 9) confirmed the results of the measurements of gaseous nitrogen emissions. Initially, slurry (mixture of manure and water) had a TAN content of 3.16 g N/kg fresh mass and a TKN content of 5.06 g N/kg fresh mass. The absolute nitrogen losses were calculated based on the TKN content. These values correspond to the measured total gaseous nitrogen emissions.

Table (8): Measurements of ammonia concentrations of the investigated samples.

Treatment	Replicate	Flask	Ammonia Concentration (mg NH ₃ m ⁻³)		Number of Observations
			Mean	Standard Deviation	
			Treatment 1: manure treated with whey	Replicate 1	
	Replicate 2	F2	4.12 A	5.29	180
	Replicate 3	F3	4.07 A	5.18	180
Treatment 2: manure treated with waste of citrus juice industry	Replicate 1	F4	3.35 B	4.77	180
	Replicate 2	F5	3.37 B	4.39	180
	Replicate 3	F6	3.33 B	4.51	180
Treatment 3: manure treated with waste of orange juice industry	Replicate 1	F7	3.42 B	4.98	180
	Replicate 2	F8	3.41 B	4.95	180
	Replicate 3	F9	3.45 B	4.87	180
Treatment 4: control, a mixture of dairy manure and water	-	F10	25.1 C	5.88	180

^a Values with different letters have a significant difference at the 0.05 probability level.

3. Global warming potential

The global warming potentials (GWP) of the different gases should be taken into consideration when evaluating the effects of greenhouse gas emissions from manure storage facilities. According to the *IPCC (2007)*, the global warming potentials of CH₄ and N₂O are 23 and 296 times that of CO₂, respectively. Regarding NH₃, it is estimated that 1% of the deposited nitrogen emits as N₂O (*Houghton et al., 1997*). Considering these different GWPs and the measured emission rates, CH₄ is the predominant greenhouse gas emitted from liquid manure storage facilities (Fig. 3). The highest N₂O emission occurred when manure surfaces become encrusted. Ammonia has barely an effect on global warming. However, ammonia emissions contribute to global warming when the ammonia is converted to nitrous oxide. Anyway, ammonia is a main source for eutrophication and acidification of water, soils and ecosystems.

Manure treatment with acidic liquid biowastes (waste of citrus, orange and milk industries) which contain organic acids, reduces the manure pH which inhibits the bacterial activities where most of the bacterial activi-

ties impede at a pH-value equal to 4.5 and, consequently, mitigates the gaseous emissions from manure.

Table (9): Total Ammoniacal Nitrogen (TAN), Total Kjeldahl Nitrogen (TKN) and Total Nitrogen Losses (ATNL).

Treatment	Replicate	Flask	TAN (g N/kg fresh mass)	TKN (g N/kg fresh mass)	ATNL (g)
Treatment 1: manure treated with whey	Replicate 1	F1	3.50	5.41	63
	Replicate 2	F2	3.46	5.37	61
	Replicate 3	F3	3.48	5.40	60
Treatment 2: manure treated with waste of citrus juice industry	Replicate 1	F4	2.73	4.81	51
	Replicate 2	F5	2.75	4.77	51
	Replicate 3	F6	2.71	4.79	49
Treatment 3: manure treated with waste of orange juice industry	Replicate 1	F7	2.81	4.99	54
	Replicate 2	F8	2.85	4.93	53
	Replicate 3	F9	2.88	4.94	50
Treatment 4: control, a mixture of dairy manure and water	-	F10	2.51	3.79	148

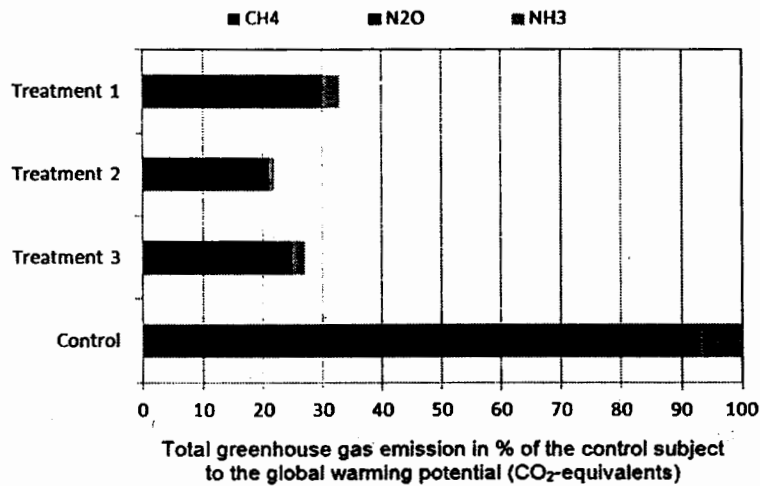


Figure (3): Total greenhouse gas emission of the different treatments subject to the global warming potential (CO₂-equivalents).

Manure treatment with acidic liquid biowastes enabled high reduction rates of gaseous emissions from manure. However, it was noticed that the pH-values slightly increased by about 0.4 with the progress of the investigations (through the 180 days of investigations), and consequently the gaseous emissions slightly increased with the progress of the investigations. This can be explained by the statement of *Raghav et al. (2012)* that the organic acids degrade with time. Nevertheless, manure treatment with acidic liquid biowastes is still effective in lowering manure pH and reducing greenhouse gases and ammonia emissions.

The results of this study agreed with the results of several studies (*Stevens et al., 1989; Oenema and Velthof, 1993; Hendriks and Vrieling, 1997; Kroodsmas and Ogink, 1997; Martinez et al., 1997; Beck and Burton, 1998; Pedersen, 2003*) which concluded that manipulating the balance between ammonia and ammonium by lowering the pH-value of slurry is a measure to reduce emissions. The results of this study confirmed the statements of several studies (*Al-Kanani et al., 1992; Berg et al., 2006a,b; Husted et al., 1991; Li et al., 2006; Pain et al., 1990; Stevens et al., 1989*) that a slurry pH around 5.5 can reduce ammonia emission by 80 to 90%. Additionally, the results of this study agreed with those of *Lay et al. (1997)* and *Berg et al. (2006a,b)* that higher methane production occurs, when the pH-value is between 6 and 7 and a slurry pH below 6 is necessary to reduce methane emission and below 5.0 causes a noticeable reduction of methane emissions, where the pH-value influences the activities of microorganisms. On the other hand, the results of this study agreed with the statements of *Berg and Hoernig (1997), Berg and Pazsiczki (2003) and Berg (2003)* who stated that using organic acids is a promising possibility to reduce methane, nitrous oxide and ammonia emissions. However, the organic acids are expensive. Therefore, in this study, acidic liquid biowastes -containing organic acids- were implemented to manipulate the manure pH, where the investigated biowastes are cheap and the costs are limited to transportation.

The implemented acidic liquid biowastes in this study have several advantages, which are: low-pH biomaterials, cheap where the costs are limited to transportation, and available from food industries. An additional advantage is developing an important usage for some of the biowastes of

the food industries. This manure treatment is expected to produce a high value biofertilizer. Manure treated with biomaterials and then distributed as biofertilizer for the plants leads to avoiding the disadvantages of the mineral and inorganic additives which contaminate the soils and increase their salt contents. This treatment is expected to provide cost effective and safe biofertilizers (manure treated with different acidic liquid biowastes) free of soil contaminants with lower greenhouse gases and ammonia emissions. One key issue is that the effect of these biofertilizers on soils' structures, nutrients content, chemical and physical properties after being incorporated into soils, should be investigated thoroughly. Although the effect of treated manure with acidic liquid biowastes on soils' properties still unknown, these biowastes are simply biomaterials which are degradable in soils. Additionally, the used biowastes do neither contain heavy metals nor pollutants. Therefore, these biowastes are not expected to be soil contaminants. Anyway, further investigations should be carried out to discover their effects on soils.

Eventually, this process is an integrated waste management of both manure and acidic liquid biowastes of food industries. Important outcomes are complying with Kyoto Protocol by mitigating the emissions of greenhouse gases, and conforming to Gothenburg Protocol by abating the ammonia emissions which allow revising and updating the present emissions inventories when this emissions abatement technique is applied.

4. Future developments

Further future investigations are planned and will be carried out to determine the effectiveness of this technique to mitigate gas emissions from beef cattle, buffalo, sheep, goat, horse, pig and poultry manure. Other additives are planned to be investigated, such as the acidic biowastes of the of food/juice industries for the following materials: mandarin, grape fruit and peach juices. Furthermore, additional materials are under consideration such as: lignite, perlite and humate products. On the other hand, the results of this study will be further tested through on-farm experiments. The same procedures carried out through the laboratory experiments are planned to be conducted on-farm with full-scale experiments. This requires a small manure tank in a dairy farm to be treated with the biowastes recommended by the results of this study. Subsequently, the

manure tank will be covered with a membrane cover similar to that used in biogas plants but under aerobic conditions, this will allow the accumulation of the gases under the cover where a tube will suck a sample to the multi-gas monitor to quantify the concentration of the different gases.

CONCLUSIONS

Lowering manure pH can reduce methane, nitrous oxide and ammonia emissions. Manure treatment with acidic liquid biowastes (waste of citrus, orange and milk industries) which contain organic acids, reduces the manure pH which inhibits the bacterial activities and, consequently, mitigates the gaseous emissions from manure. Manure treatment with acidic liquid biowastes enabled high reduction rates of gaseous emissions from manure. However, the pH-values slightly increased with the progress of the investigations (through the 180 days of investigations). This can be explained by the fact that the organic acids degrade with time. Nevertheless, manure treatment with acidic liquid biowastes is still effective in lowering manure pH and reducing greenhouse gases and ammonia emissions.

Manure treatment with waste of citrus juice industry was the most effective manure treatment among the investigated treatments. Additionally, manure treatment with waste of orange juice industry was effective in reducing methane, nitrous oxide, and ammonia emissions. Although whey was effective in reducing the gaseous emissions, it was the less effective manure treatment. Eventually, the results of this study recommended manure treatment with acidic liquid biowastes as an effective emissions abatement technique.

Considering the different global warming potentials and the measured emission rates, methane is the predominant greenhouse gas emitted from liquid manure storage facilities. The total greenhouse gas emission in percentage of the control sample calculated on the basis of the global warming potential (CO₂-equivalents) showed that manure treatment with waste of citrus juice industry delivered the lowest CO₂-equivalents, whereas manure treatment with waste of orange juice industry delivered slightly higher CO₂-equivalents. However, manure treatment with whey delivered the highest CO₂-equivalents among the three additives.

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المخلص العربي

معاملة الفضلات السائلة بمخلفات مصانع الاغذية للحد من انبعاثات الميثان، وأكسيد النيتروز والأمونيا

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روث الماشية هو المصدر الرئيسي لانبعاثات غاز الأمونيا (NH_3) و يعتبر مصدرا هاما من مصادر غازات الاحتباس الحراري (غازات الدفيئة) وخاصة غاز الميثان (CH_4) وأكسيد النيتروز (N_2O). هذا و تساهم انبعاثات الأمونيا في زيادة المغذيات بكلا من المياه والتربة والنظم الإيكولوجية ورفع درجة الحموضة بها. كما تساهم الغازات الدفيئة في ظاهرة الاحتباس الحراري. ويمكن تخفيض هذه الانبعاثات الغازية عن طريق رفع درجة حموضة روث الماشية مما يحد من انبعاثات كلا من غاز الميثان، وأكسيد النيتروز والأمونيا. على الرغم من أن الأحماض غير العضوية مهمة الا أن لها عيوب عديدة منها تلوث التربة. يعتبر استخدام الأحماض العضوية وسيلة فعالة لكنها مكلفة؛ وبالتالي فإن استخدام مخلفات التصنيع الغذائي التي تحتوي على الأحماض العضوية هي مجدية للغاية. الهدف من هذا البحث هو دراسة تأثيرات استخدام مخلفات مصانع الاغذية كإضافات لروث الماشية على انبعاثات كلا من غاز الميثان، وأكسيد النيتروز والأمونيا. تم معالجة الروث الناتج من الأبقار الحلوب بمخلفات التصنيع الغذائي (شرش اللبن و مخلفات صناعة عسائر البرتقال و الحمضيات) ثم قياس معدل تدفق كلا من غاز الميثان، وأكسيد النيتروز والأمونيا باستخدام نظام مصمم خصيصا لهذا الغرض والذي يتكون من عدة قوارير و عداد لتسجيل انبعاثات الغازات المختلفة. أظهرت النتائج أن تدفقات الغاز المنبعث من روث الماشية المعامل مع مخلفات صناعة عسائر الحمضيات كانت ١٣, ٠,٢١٩ و ٢,٥٢٣ جرام/م^٢يوم لكلا من الميثان، وأكسيد النيتروز والأمونيا على التوالي. و تدفقات الغاز المنبعث من روث الماشية المعامل بمخلفات صناعة عسائر البرتقال كانت ١٣,٥٨ و ٠,٢٢٣ و ٢,٥٨١ جرام/م^٢يوم لكلا من الميثان، وأكسيد النيتروز والأمونيا على التوالي. بينما كانت تدفقات الغاز المنبعث من روث الماشية المعامل بشرش اللبن ١٧,٤٥، ٠,٢٧٩ و ٣,٠٦٣ جرام/م^٢يوم لكلا من الميثان، وأكسيد النيتروز والأمونيا على التوالي. في حين أن تدفقات الغاز المنبعث من العينة الطبيعيه (خليط من روث الماشية والمياه) كانت ٥٨,٢١، ٠,٣٤٧ و ١٨,٩ جرام/م^٢يوم لكلا من الميثان، وأكسيد النيتروز والأمونيا على التوالي. وبناء على ذلك، فإن معالجة روث الماشية بمخلفات مصانع الاغذية قادره على الحد من الانبعاثات الغازية من روث الماشية.

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