

Uses of Agri By-Product in Inhibiting Ammonia and Methane from Cattle Slurry During Storage

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Received: 16 February 2023

Accepted: 14 June 2023

Abstract

Liquid manures storages produce significant amounts of ammonia (NH₃) and methane (CH₄) gas from biological anaerobic fermentation. Small-scale observation studies using 3 kg of slurry for each treatment were carried out to investigate a simple approach to mitigate NH₃ and CH₄ emissions during slurry storage. There were five treatments using agriculture by-products. The agriculture by-products from rice husk, rice straw, cocopeat and wood chips were placed on top of cattle slurry filled inside a small container as physical cover to reduce NH₃ volatilization and CH₄ emission. During the forty-five days of the undisturbed storage period, an immediate reduction of CH₄ and NH₃ gas fluxes were observed after the application (day 1). The cumulative NH₃ releases were lowered between 28.6% and 66.7% as compared to untreated (Ctrl) after 30 days of observations, and CH₄ was inhibited by 4 to 33% after 45 days of storage. Ricestraw was found to be the best as a physical cover in inhibiting both gases (NH₃ and CH₄) with cumulative inhibition percentages of 31 and 33%, respectively. These preliminary findings provide insight into future manure management systems applicable as one of the greenhouse gas mitigation strategies.

Keywords: methane emission, ammonia volatilization, slurry storage, greenhouse gas.

Introduction

The handling and use of manure on livestock farms contribute to emissions of the greenhouse gases methane (CH₄) and nitrous oxide (N₂O), especially on slurry animal waste typically swine, dairy and beef cattle farming. Proper manure management is important as manure storage contributes to the release of greenhouse gas (GHG) emissions up to 18% of the total agriculture emission (Pattey *et al.*, 2005; Chadwick *et al.*, 2011; Sommer *et al.*, 2013). Although Malaysia's manure GHG emission is stagnant at an average 5.4%, much lower than global reports,

this emission is reflected to the current population at 612,294 beef cattle and 45,113 dairy cattle (at year 2019). These number are only representing the current SSL of around 21%.

The release of GHGs, particularly methane (CH₄), during the storage of slurry manure is significant, however measures are being developed and applied to limit these emissions. To date, Malaysian livestock activities have contributed to 1202.0 Gg CO₂ equivalent from emission from manure management and indirect nitrous oxide (N₂O) emission from manure in the year 2019

(NRECC, 2022). Producing biogas from a bio-digester in which the captured CH_4 is converted to CO_2 by flaring or utilized to generate renewable energy is one option to limit this emission. However, this method is inapplicable to small farmers because it requires a significant investment, high maintenance costs, and a large number of animals to be economically feasible and productive. Therefore, alternative measures are required to reduce CH_4 and other greenhouse gas emissions from slurry storage facilities, particularly for small- to medium-scale livestock farmers in Malaysia.

Slurry storage acts as a point source of GHG gas emissions during manure storage, and permeable surface coverings (natural crusts and artificial covers) are increasingly recognised for their capacity to lower different gaseous emissions, including ammonia (NH_3) (Berg *et al.*, 2006). Surface cover using agricultural waste not only can act as a physical barrier but also to retain gaseous emission (CH_4 and NH_3) (Guarino *et al.*, 2006), as a medium for microbes to oxidize methane and as an excellent medium for nitrification and denitrification process (Portejoie *et al.*, 2003; Guarino *et al.*, 2006; Petersen & Ambus, 2006; Hansen *et al.*, 2009).

The slurry crust is defined as a fibrous layer on slurry surface, which developed during the slurry storage (Pain & Menzi, 2011). Slurry crusts form due to solids in the slurry being carried to the slurry surface by gas bubbles which are produced by microbes during microbial degradation of the organic matter (Misselbrook *et al.*, 2005). Crusts that formed on cattle slurry by natural processes can be promoted by adding chopped straws or fibrous materials (Sommer and Husted, 1995; Pain & Menzi, 2011; Aguerre *et al.*, 2012). The existence of a crust on the surface of the slurry may provide a suitable environment for the growth methane oxidizing microbes

(Duan, 2012; Duan *et al.*, 2013). This oxidation by slurry crusts offers a potentially important sink for the CH_4 generated by methanogen in the liquid slurry beneath it, but may increase the N_2O emission due to nitrification of NH_4^+ and subsequent denitrification of NO_3^- in the crust microenvironment (Hansen *et al.*, 2009).

Methane gas is a potent gas produced by methanogen in strictly anaerobic environments with a global warming potentials on a 100-year time horizon that are 34 times greater than carbon dioxide (Myhre *et al.*, 2013). In Malaysia, there was minimal awareness of CH_4 emissions from slurry since they may have no short-term effects on the nearby environment, animals, or farmers, and have no immediate ramifications for the farm's income. Meanwhile, NH_3 volatilisation is the transfer of nitrogen from animal dung to the atmosphere. They indicate a loss of valuable fertiliser N content from manure. Furthermore, anthropogenic NH_3 emissions to the troposphere produce indirect environmental damage such as soil acidification and eutrophication of watercourses (Portejoie *et al.*, 2003; Petersen *et al.*, 2012). Earlier studies showed a significant reductions of up to 80% in NH_3 emissions from stored slurry with natural crusts or floating covers (Portejoie *et al.*, 2003; Misselbrook *et al.*, 2005).

The addition of substrate as slurry cover is considered as an additional cost. Thus, recycling agriculture by-products is a convenient way of reducing farmer's cost. The objective of this study was to provide an early investigation on the uses of agriculture waste and by-product to mitigate emissions from stored slurry. It was hypothesized that physical barrier by the uses of agri-waste and by-product promotes crust formation which help to reduce CH_4 emission and NH_3 volatilization from slurry surface.

Materials and Methods

Slurry agriculture biomass as physical cover

Fresh cattle slurry (FS) from mixtures of cattle breed was obtained from a reception pit and slurry handling pond of cattle farm, Pedas, Negeri Sembilan. The cattle were at a range of 1-4 years old, weighed around 50-300 kg. These animals were fed with total mixed ration (TMR) at 3% dry matter basis of bodyweight, which comprises of 60% concentrates and 40% fresh grass / pastures (% dry matter basis). The concentrates contents are mainly palm kernel expeller (PKE), grinded corn, soya bean meal, soya bean hull, grinded rice hull, crude palm oil (CPO), molasses, and limestone with addition of less than 0.002% minerals and trace elements for the animal growth requirement. The slurry obtained kept in a 130 L drum container and stored under cover for 48-92 hr prior to use. The slurry physicochemical composition (pH; oxidation redox potential, ORP; dry matter, DM; volatile solid, VS; carbon and nitrogen was analysed before the experimental design was carried out. Initial slurry characteristics were $9.0 \pm 0.18\%$ dry matter Kg^{-1} slurry (DM), $70.2 \pm 0.4\%$ volatile solid Kg^{-1} DM (VS), total carbon (C) 321.5 ± 62.70 mg Kg^{-1} slurry, total nitrogen (N) 51.2 ± 25.14 mg Kg^{-1} slurry, C:N ratio 6.28:1, and pH 7.18 ± 0.01 .

Cattle slurry was transferred into 5 L high-density polyethylene (HDPE) plastic pail [18.3 and 21 cm (bottom and top) diameter x 22.0 cm height], such that each pail received 3 Kg cattle slurry and with and without (3 cm thickness) agriculture by products as biological cover. The experiment was designed based on 4 different types of agriculture by products, namely, i) slurry + woodchipped (WC); ii) slurry + rice husk (Sekam); iii) slurry + rice straw at 3- 5cm length (Ricestraw); iv) slurry + cocopeat (Cocopeat) and a control treatment with untreated slurry without any substrate (Ctrl).

The lab scale experiment was carried out on December 2021 for a period of 45 days.

Moisture loss, temperature, pH and oxidation redox potential

The slurry moisture loss was weighted using digital weighing scale FX5000i (AND Company Limited, Japan). While, slurry temperature, pH and ORP were measured using a Hanna pH electrode probe (model HI 991003; Hanna Instrument, USA). Those measurement was conducted only at Ctrl treatment slurry to avoid disturbances of the slurry surface. Meanwhile, the ambient temperature and humidity was recorded using EasyLog EL-USB-2-LCD (Lascar Electronic, United Kingdom).

Slurry dry matter (DM) and volatile solids (VS) content

Slurry dry matter (DM) and volatile solids (VS) were determined by drying 10 g slurry samples at 80° - 105°C to constant weight (16 hr) and as loss on ignition at 450°C for 16 hr in muffle furnace Carbolite CWF 1200 (Carbolite Ltd, UK).

Total carbon (C) and nitrogen (N)

The total C and N of fresh slurry were measured by Elementar Analyzer (CHNOS) model Vario Macro Cube (Elementar, Germany).

Greenhouse gases measurement

Methane emission

Greenhouse gas fluxes were sampled from the ca. 2 litres vessel headspace above the pail through a butyl rubber septum during closed system. Headspace gas samples were taken immediately (T0) securing the lid in place, after 30 (T30) and 60 minutes (T60). Gas samples were placed in 20 mL pre-evacuated gas vials and analysed using Agilent 7890B gas chromatogram (GC). The

GC was equipped with J &W Scientific CS-Gaspro 45 m X 0.320 μm capillary columns, and equipped with a flame ionized detector (FID). Gas fluxes were calculated based on the linear increase in gas concentration between the T0 and T60 samples over the one-hour period, headspace volume and slurry weight. Cumulative gas emissions for the storage period were calculated by interpolating the measurements between adjacent sampling points using the trapezoidal rule (Cardenas *et al.*, 2010).

Relative ammonia volatilization

Measurement was carried during closed system concurrent with gas sampling period. Relative NH_3 volatilisation was determined using a 0.02 M orthophosphoric acid (H_3PO_4) passive trap placed in a non-ventilated (sealed) environment (Misselbrook & Powell, 2005). The acid trap was carefully suspended inside this headspace, 3 cm from the lip of the vessel. Following the one-hour 'incubation' when the lid remained in place, the traps were removed and the ammonium-N (NH_4^+N) concentration in the H_3PO_4 acid determined as per described by (Mulvaney, 1996). Prior to incubation at 30°C, 15 μL of 6% Na_2EDTA , 60 μL of Na-Salicylate-nitroprusside and 30 μL of hypochlorite solution were added. Na-Salicylate-nitroprusside solution consisted of 7.8% (w/v) Na-Salicylate and 0.125% (w/v) Na-nitroprusside, while hypochlorite solution (pH 13) contained 2.96% (w/v) NaOH, 9.96% K_2HPO_4 (w/v) and 10% (v/v) Na-hypochlorite. Absorbance readings were measured after 30 min incubation using a SpectraMax ABS microplate reader at a wavelength of 667nm and analysed by SoftMax Pro 7 (Molecular Devices, USA).

Results and discussion

Slurry agriculture biomass as physical cover

Recycling agricultural by-products to induce crust formation on the slurry surface is

one method of reducing CH_4 emission. The crust generated in this study was thin and not as solid as normal crust found in anaerobic lagoons in European countries. As a result, there was a chance that fewer or no prospective methane oxidising bacteria (MOB) and ammonia oxidising bacteria (AOB) would dominate the crust that used CH_4 and NH_3 emission (Petersen & Ambus, 2006; Hansen *et al.*, 2009; Duan, 2012; Nielsen *et al.*, 2013). These bacteria are responsible for lowering the amount of gas discharged into the atmosphere in a biological or natural way. The Ctrl slurries indicated that the pH of the slurry stayed constant (pH 7.1-7.5) during initial 30 days (Figure 1). However, the last pH recorded at day 45 showed increased to pH 8.1, and it's not clear causes of this. Probably due to high moisture loss which at 45.6% (Figure 1; Day 45) and were the highest compared to others. During this incubation period, the ORP reading indicated that the slurries remain constant strictly anaerobic condition. Slurries characteristics (pH, DM, and VS %) and the moisture loss during observation study were not significantly altered, albeit the DM content was determined to be lower at the end of the trial (Table 1). This was due to microbial digestion and organic matter deposition at the bottom of the pails.

Ammonia volatilization and methane emission

The uses an agriculture by-product had showed significant ($P < 0.05$) reduction in NH_3 volatilization on day 0 except WC, compared to Ctrl (Figure 2; i and ii). Agriculture by-product act as physical barrier and such volatilization is retarded immediately. However, as the NH_3 accumulated and its volatilization passing the thin layer of the agriculture by-products as its not hardened as typical crust. The NH_3 volatilization loss fluctuated and decreased on the day 30 as the biomass used is stabilized and crusted became

as physical barrier. The formed barrier alike a crust significantly inhibiting the NH_3 volatilization from the slurry surface. Therefore, the cumulative emission indicated the agriculture by-products which are WC, Rice husk, Ricestraw and Cocopeat showed a reduction on NH_3 volatilization at 66.7, 47.2, 33.0 and 28.6% respectively. Lower NH_3 emission on WC may associated with lower

crust pH (not measured) that retain solubilized form NH_4^+ from volatilize similar to Berg *et al.*, 2006 and Guarino *et al.*, 2006. Crust formed and lower pH value than neutral is a significant approach to inhibit greenhouse gas emission including NH_3 (Berg *et al.*, 2006; Guarino *et al.*, 2006; Smith *et al.*, 2007; Hansen *et al.*, 2009).

Table 1. Slurry characteristic during storage observation

Treatment	Start			
	Dry Matter % DM (\pm SEM)	Volatile solid % VS (\pm SEM)	Ammonium content $\text{NH}_4^+\text{-N}$ (g kg^{-1} Slurry)	C: N Ratio
Rice husk (Sekam)	8.1 \pm 0.11	42.20 \pm 2.53	0.12 \pm 0.004	6.66
Cocopeat	8.6 \pm 0.08	40.55 \pm 2.75	0.12 \pm 0.009	6.85
Ctrl	8.0 \pm 0.06	39.51 \pm 2.59	0.11 \pm 0.006	6.28
Woodchipped (WC)	8.1 \pm 0.19	40.77 \pm 0.91	0.12 \pm 0.003	9.24
Ricestraw	8.1 \pm 0.08	41.82 \pm 0.57	0.11 \pm 0.005	8.38
End				
Rice husk (Sekam)	12.4 \pm 0.65	83.52 \pm 1.76	43.0 \pm 5.14	8.80
Cocopeat	11.8 \pm 1.54	81.91 \pm 1.94	39.2 \pm 4.53	10.61
Ctrl	10.7 \pm 0.46	78.32 \pm 1.48	31.5 \pm 6.46	9.35
Woodchipped (WC)	11.1 \pm 0.24	82.07 \pm 1.10	35.6 \pm 7.67	10.93
Ricestraw	11.6 \pm 0.49	82.46 \pm 1.26	37.5 \pm 4.95	9.52

The uses of agriculture by-product act as physical barrier to inhibit anthropogenic GHG emission mainly CH₄. All treatments showed immediate fluxes inhibition at day 1 (Rice husk 35.3%, Ricestraw 50.8%, WC 16.6% and Cocopeat 40.8%) however, only Ricestraw shows significant ($P<0.05$) inhibition compared to Ctrl (Figure 3; i). This inhibition was replaced with stronger emission fluxes in following sampling point except for Ricestraw (Figure 3). These is probably the addition of biomass organic matter as precursor for microbial fermentation activities. The by-products physical shape, characteristic and surface area dipped to slurry surface where influence the level of microbial activities may contributed to those emissions. The cumulative CH₄ during 45 days storage period showed the Ctrl and Rice husk emitted 45.9 g Kg⁻¹ VS and 46.3 g Kg⁻¹ VS CH₄, respectively. Lowest emission was from Ricestraw recorded CH₄ emission inhibited 31%, CW and Cocopeat emitted 4 and 17% CH₄ lower than Ctrl (Figure 3; ii). The uses of Ricestraw in inhibiting CH₄ this study contradicted to previous result (Berg *et al.*, 2006). Reduction of CH₄ emission in this study were slightly lower than a study by Matulaitis *et al.* (2015) using pig slurry and at lower environment temperature. It emits more CH₄ when applied at 25°C and similar to Berg *et al.* (2006). While, Misselbrook *et al.* (2016) reported that clay granules as covers did not give any impact on CH₄, but reduced NH₃ by 77%.

Ammonia volatilization and CH₄ emission during slurry storage is unavoidable, unless full covered lagoon is applied, but it can be reduced by managing a few factors such as in this study.

According to the Intergovernmental Panel on Climate Change, the global warming potential of CH₄ is 28 times larger than that of carbon dioxide, and the global warming potential of nitrous oxide (N₂O) is 265 times more than that of carbon dioxide (CO₂) (IPCC, 2021). Adding biological cover to the surface of slurry may not be favoured by farmers because to the expense and profit margin implications. In addition, biological cover must remain on the surface of the slurry until the next maintenance processes, and may affecting the quantity and cost of the biological cover in addition to the number and size of waste ponds used.

However, if compared to physical and chemical approach, this biological cover proved to be more viable and cost-effective. The uses of agriculture by-product as biomass cover has similar benefit to bio-cover uses in bad odours absorption (English & Fleming, 2006). Biological cover did enhanced the aerobic condition in lagoon surface and biologically retarded the odorants (Zhang *et al.*, 2013). Furthermore, comparing to physical and chemical methods, biological methods were more feasible and economical. This is important as Malaysia is dominated by small-to-medium-sized farm (Mohd Saufi, 2022).

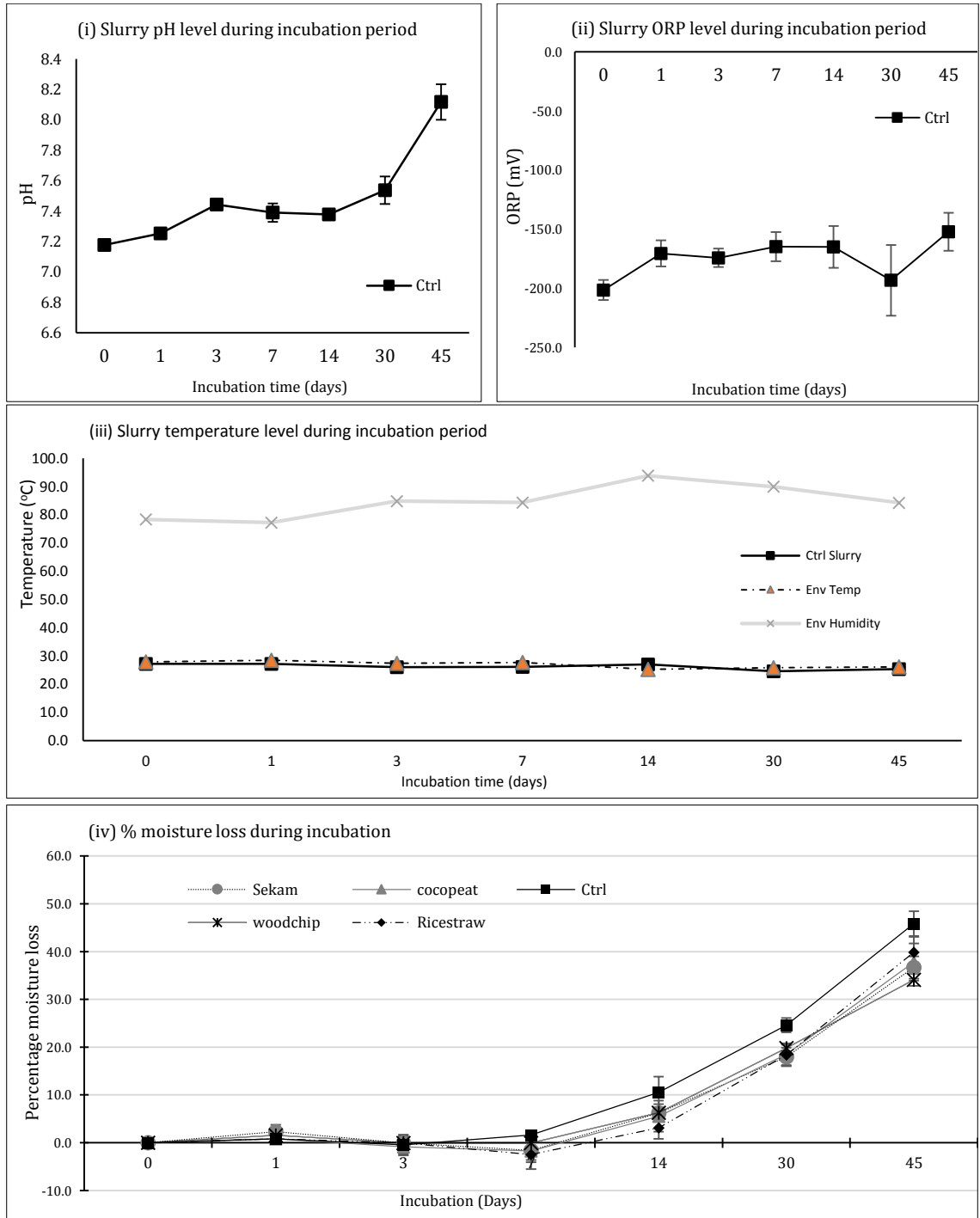


Figure 1. Observation on slurry pH, ORP temperature and moisture loss

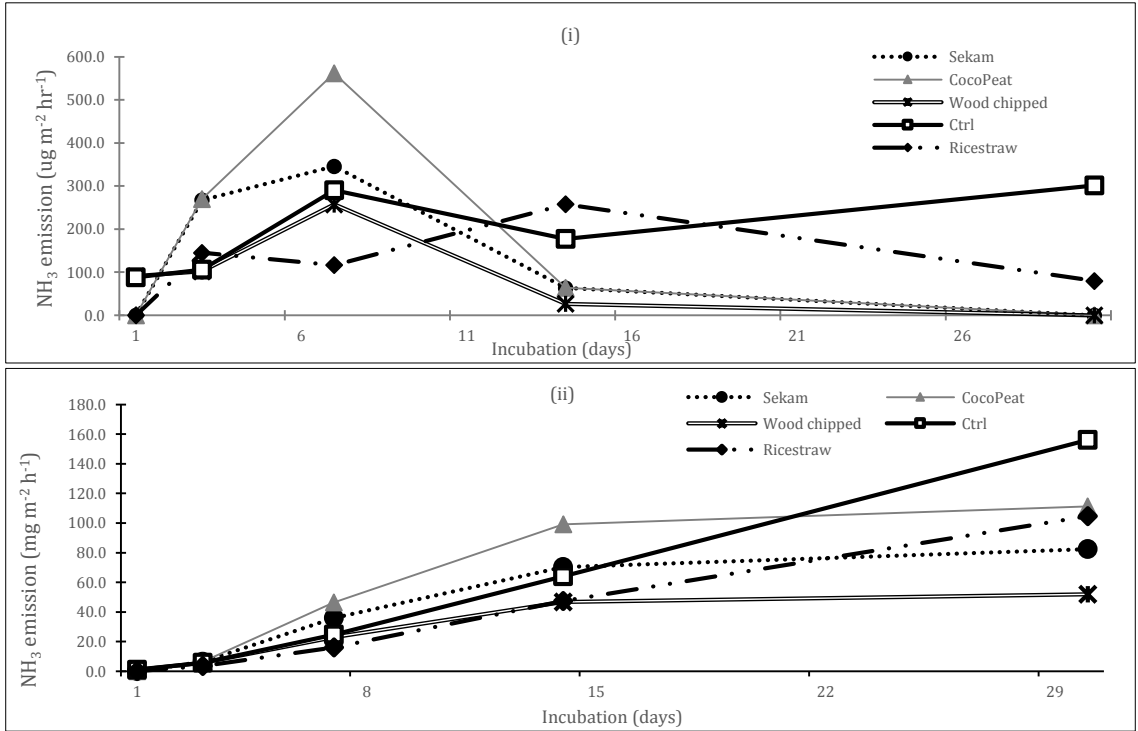


Figure 2. Ammonia fluxes (i) and cumulative emission (ii) during slurry storage with an agriculture by-product.

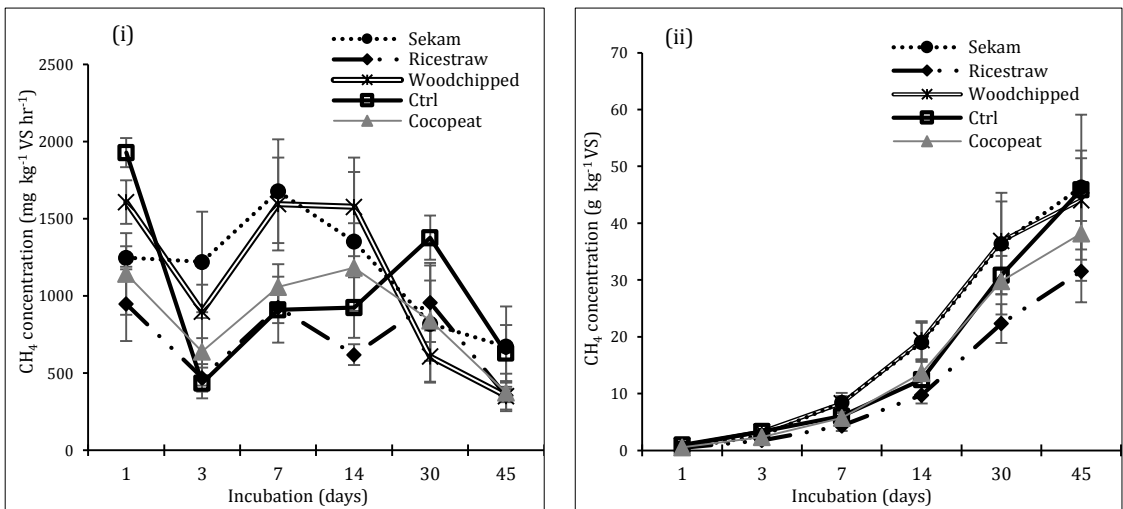


Figure 3. Methane fluxes (i) and cumulative emission (ii) during slurry storage with an agriculture by-product.

Conclusion

The covering method using agriculture by-products were found to effectively reduce the emission of potent gases from slurry. Depends on the target gases, NH_3 volatilization is best inhibited by the use of WC as covering, while for CH_4 emission, highest reduction was found with the use of Ricestraw. This study concludes that Ricestraw is the best agri-waste that can be utilised as physical cover to inhibit both NH_3 and CH_4 emission from stored slurry with inhibition at 33% and 31%.

In this study, covering ruminant liquid waste was determined to be a promising manure management strategy for combating climate change and global warming. Larger scale experiment may be useful in evaluating the uses of agriwaste as biological cover in inhibiting potent gas emission. As the world population expands, so does the need for animal protein, necessitating quick mitigation on all fronts in order to keep livestock production sustainable without adding more anthropogenic gases to the troposphere.

Acknowledgements

This research was conducted at Agrobiodiversity & The Environment Research Centre, MARDI and funded by the 12th Malaysia Plan and the Malaysian Agricultural Research and Development Institute, Malaysia through project code P21400010170505.

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