

Article

Sustainable Management of Buffalo Manure Digestate: Environmental and Economic Assessment of Biochar-Based Cover for Ammonia Emission Mitigation

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Abstract

Storage of livestock effluents represents a major source of ammonia (NH₃) emissions into the atmosphere. Reducing ammonia volatilisation is essential to improve the efficiency of nutrient use, mitigate air pollution, and enhance the overall sustainability of livestock production systems aligned with the principles of the circular economy. Therefore, identifying efficient and sustainable mitigation strategies is crucial. Conventional floating covers are commonly used to reduce emissions; however, they present limitations in terms of management, durability, and cost. This study proposes a novel approach by comparing traditional floating cover materials like straw and light expanded clay with biochar as an innovative and sustainable mitigation strategy to reduce ammonia volatilisation from the liquid fraction of buffalo digestate obtained from an anaerobic digestion plant in southern Italy. All cover materials were applied at a uniform thickness of 2 cm under laboratory conditions using a dynamic chamber technique. Additionally, a cost analysis was performed considering the material purchase cost for an average storage tank of 700 m² and two hypothetical reduction efficiencies (50% and 70%). Results indicated that biochar was the most effective cover, achieving a 67% reduction in ammonia emissions compared with the uncovered control. Light expanded clay exhibited the lowest efficiency, likely due to its insufficient sealing capacity at the applied thickness. From an economic perspective, biochar becomes increasingly competitive when emission reduction efficiency is accounted for, owing to its favourable physical–chemical properties. The results highlight the potential of using biochar as a sustainable, circular strategy for mitigating atmospheric emissions and improving nitrogen conservation, while also creating opportunities for its subsequent reuse in agriculture as a soil amendment.

Keywords: ammonia emission reduction; digestate storage; techno-economic prospect; dairy buffalo



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1. Introduction

Approximately 81% of the total worldwide emissions of ammonia (NH₃) are caused by activities in the agricultural sector [1]. Specifically, livestock farming contributes to the production of 60–90% of the NH₃ emissions produced from agriculture [2,3]. Once volatilised, NH₃ adversely affects soil, air, and water quality [4]. Moreover, ammonia

volatilisation is also responsible for the formation of secondary particulate matter (PM 2.5) responsible for human illnesses, such as cardiovascular disease [5–7]. Livestock activities generate large quantities of manure, which must be stored before being used as fertiliser to comply with the new RENURE legislation (REcovered Nitrogen from manURE), developed by EU Member States, which set strict guidelines for the management of organic and inorganic nutrients, restricting storage, rate, and timing of land application to reduce diffuse nutrient losses from agricultural sources. Considerable research has focused on mitigating ammonia emissions from livestock production. Public agencies, regulators, and farmers are recognizing the environmental impact of ammonia emissions and the need to reduce them. Within the framework of sustainable agriculture and circular bioeconomy strategies, improving digestate management is increasingly recognised as a priority for reducing environmental impact while preserving nutrient value [8,9]. Technologies that can mitigate gaseous emissions and enhance nitrogen retention simultaneously can contribute to more resource-efficient livestock systems and support the transition towards low-emission agriculture [10,11]. One potential solution is applying covers to slurry during storage to reduce NH_3 emissions [9,12]. The exposed surface of manure can be covered with different materials, natural or synthetic. Cover application can reduce ammonia emissions by limiting air movement and decreasing nitrogen transfer from manure to the atmosphere [13–15]. Permeable and organic covers are susceptible to weather conditions and can sink or drift, blocking pumping drainage systems [16,17]. Natural covers, unlike synthetic ones, do not require removal of meteoric water. They are typically low-cost and effective in emission reduction, with organic covers achieving up to 87% reduction [15,16]. Several natural materials are suitable for this purpose. Straw, for instance, is a crop residue that, if already present on-farm as leftover, can be considered a low-cost material. Research studies have demonstrated that the application of straw, whether untreated or pre-treated, as a material on stored manure, has the capacity to significantly reduce ammonia emissions [18,19]. Additionally, when reed straw was added to manure during the composting process, a reduction of circa 12% was registered. This outcome is probably consequent to the fact that straw is an N-poor material and can immobilize ammonia [11]. It should be noted that straw is a material with a low density, susceptible to damage from wind and rain. Numerous studies have evaluated straw's capacity to maintain its position in relation to its effectiveness in reducing ammonia emissions [15,16]. In order to prevent straw from sinking into the stored slurry, the total solids of the slurry should be at least 4% [15]. Although different straw types such as reed straw, wheat straw, and rice straw have been assessed in the literature, a comparison between the different aptitudes and efficacy in reducing emissions among these different materials has been poorly assessed [18,19]. Clay is a low-cost, porous, water-impermeable mineral that is commonly used as an insulating material. It can float on liquid surfaces, such as slurry [13]. Light expanded clay aggregates (LECA[®]) are an inexpensive option for covering slurry [14]. However, their effectiveness in reducing ammonia emissions is inconsistent. LECA[®] granules only reduce ammonia emissions when applied at a thickness of 14 cm. Thinner layers may be less effective and can increase NH_3 volatilisation [8]. Additionally, the surface of LECA[®] promotes microbial growth, which can degrade organic matter and increase NH_4^+ release and NH_3 emissions. This microbial activity can lead to bio-plugging of the LECA[®], further reducing its performance [16,20].

Biochar, a material produced by the pyrolysis of various biomasses, has gained attention as a method of reducing ammonia emissions when applied to stored livestock manure. In addition to its use in agriculture, biochar is employed in nutrient recovery, livestock farming, pharmaceuticals, and water treatment [21,22]. Biochar is obtained from the pyrolysis of different biomasses [21]. The pyrolyzed biomass material, as well as the

pyrolysis conditions and processes, can influence the final characteristics of the biochar [22]. The benefit that biochar brings to the soil when applied as a soil conditioner has made biochar widely studied in recent years [23]. In addition, its effectiveness in reducing the emission of NH_3 when used as a cover on stored livestock manure is due to the fact that it acts as a physical barrier and has an adsorption effect [24,25]. However, previous work on the same biochar and digestate showed that nitrogen adsorption alone does not fully explain the observed reduction in ammonia emissions, suggesting that additional physical and mass-transfer mechanisms are involved [26]. The characteristics of biochar, including its chemical functional groups, porosity, surface area, and hydrophobicity, affect its ability to absorb NH_3 and ammonium ions (NH_4^+). Its hydrophobic nature enables it to float on manure and reduce liquid absorption [27–29]. Studies show that biochar acts as a physical barrier and an adsorbent, effectively reducing ammonia emissions. When applied to swine manure, a biochar cover reduced ammonia emissions by 33%. Other studies have reported reductions ranging between 4–78%, depending on the biochar's properties and production methods. However, weather conditions and manure characteristics can influence biochar's performance [29,30]. Researchers are continuing to study its potential for large-scale applications and how different pyrolysis processes impact its effectiveness [29].

Biochar production is generally related to a local niche industry, and large-scale producers are not widely common [20,30]. Economic feasibility remains a key factor in the adoption of mitigation technologies at the farm level. Therefore, it is essential to evaluate both environmental performance and implementation costs in order to identify sustainable solutions for livestock producers. Although biochar fits well in the circular economy concept, economic viability and market competitiveness are necessary to facilitate broader-scale biochar production and agricultural sector adoption [31]. Since biochar is a highly heterogeneous material, its physicochemical properties can vary substantially depending on the feedstock and production conditions. Consequently, its purchase and selling prices can differ widely based on these properties. The low market demand for biochar presents a significant challenge to the generation of profit from its production. Furthermore, the market value of biochar has not yet been clearly established [32,33]. To date, only a few studies have examined the effectiveness of natural cover materials in reducing ammonia emissions from stored buffalo manure [28,34]. This study aims to compare the effectiveness of various natural cover materials, including biochar, in reducing ammonia emissions from buffalo digestate storage. Beyond emission mitigation performance, the study integrates environmental, economic, and resource-management perspectives to assess the practical applicability of these strategies in livestock systems.

2. Materials and Methods

2.1. Experimental Setup

The efficiency in reducing ammonia emissions of three natural floating covers from the stored liquid fraction of buffalo digestate was tested for 34 days under laboratory conditions. Each experimental unit, used to simulate the average storage situation of farm tanks, consisted of a 5 L capacity glass pot with a height of 25 cm and diameter of 16 cm. Each pot was filled with 3 L of liquid fraction of digestate. Corn straw, biochar, and clay LECA[®] (Laterlite S.p.A., Rubbiano di Solignano, Italy) were tested in triplicate. Before weighing and application, all cover materials were dried at 105 °C for 24 h to standardise initial moisture content and reduce variability associated with storage and transport conditions. This laboratory procedure was applied to ensure comparability among materials and does not reflect field application practices, where such pre-treatment would not be feasible. The different covers were brought to room temperature before the application. All cover layers were standardized at a thickness of 2 cm, ensuring a consistent

basis for comparison with the biochar layer [35]. The choice was based on a previous study on biochar, where a 2 cm layer showed effective ammonia emission reduction. The aim of this study was to compare the different cover materials with biochar [34]. To measure the thickness of each cover (2 cm), an empty container of the same type that was used to store the liquid digestate was used. The thickness was measured using a tape measure, and the material was weighed before application. The straw cover was manually pressed to the bottom of the jar to create a stable, uniform layer, enabling a direct comparison with other cover materials. After the glass pots were filled with digestate (3 L), the covers were manually applied to the digestate surface, providing a layer of 2 cm of 12.8 g of straw (S), 84 g of biochar (B), and 180 g of clay LECA® (L), respectively. Furthermore, a control experimental unit (NC) consisting of only digestate without any cover was set up, for a total of 12 jars. The experimental units were stored during the whole test period in a walk-in closet with a constant temperature set at 18 °C. The storage and gas measurements were carried out in a controlled environment. The experimental setup is showed in Figure 1.

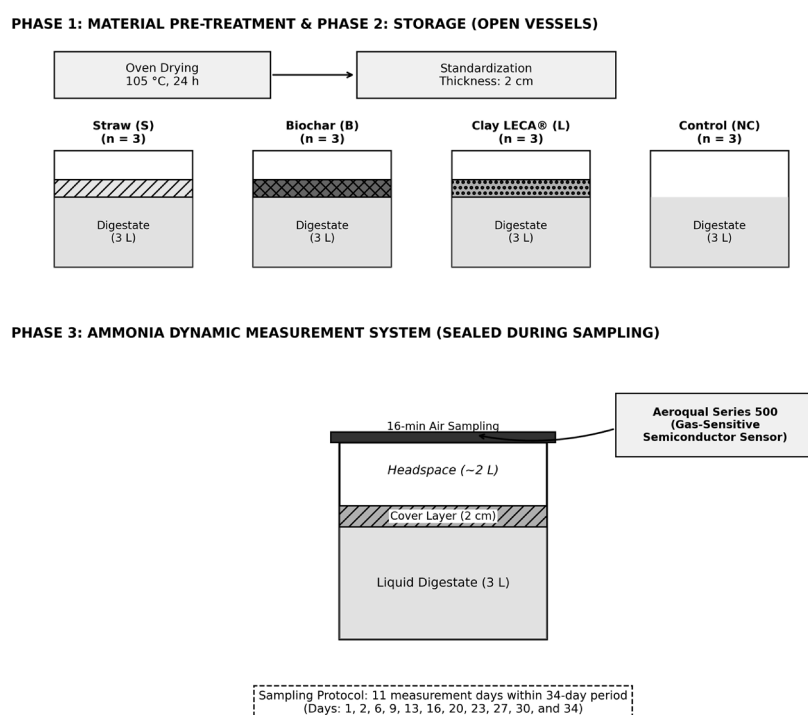


Figure 1. Methodological workflow.

2.2. Buffalo Digestate Characteristics and Analysis

The plant, located in the province of Caserta, processes buffalo slurry originating from nearby dairy buffalo farms. The liquid fraction of the digestate was collected immediately before entering the storage tank and transported to the laboratory for determination of its physicochemical properties, including pH, dry matter (DM), total Kjeldahl nitrogen (TKN), and total ammoniacal nitrogen (TAN), according to standard methods [27]. Samples were also taken at the end of the storage period. All analyses were carried out in duplicate. Digestate DM was determined by drying the digestate in an oven at 105 °C for 24 h. The digestate TKN content was assessed by applying the Kjeldahl method. The TAN content was defined after distillation of the digestate sampled using a titrimetric method.

2.3. Cover Material Characteristics

The biochar was purchased from Nera Biochar Srl (Turin, Italy) and used in a study carried out by Scotto di Perta et al., 2020 [26]. The pyrolyzed biomass was made from

wood chips from Piedmont: elm tree, ash tree, chestnut, and conifers, as reported by the manufacturer. A single commercial batch of biochar was used for all experimental treatments, and no different biochar types or mixtures were applied. The production process consisted of a 30 min pyrolysis at a temperature of 550 °C. Table 1 summarizes biochar characteristics [34].

Table 1. Biochar characteristics: Ash, C (carbon), H (hydrogen), N (nitrogen), and O (oxygen) are expressed as wt% db (dry basis); H/Corg: hydrogen-to-organic carbon atomic ratio; pH: acidity/alkalinity; pHPZC: pH at the point of zero charge; OH groups: surface hydroxyl groups (mmol g^{−1}); BET surface area: specific surface area determined by the Brunauer–Emmett–Teller method (m² g^{−1}).

Ash wt% db	C wt% db	H wt% db	N wt% db	O wt% db	H/Corg	pH	pHPZC	OH Groups mmol g ^{−1}	BET m ² g ^{−1}
3.4	74.6	2.0	0.7	19.3	0.7	10	8.38	0.0697	350

The particle size distribution of the biochar was as follows: 6.5% of particles passed through a 0.5 mm sieve, 74.6% through a 2 mm sieve, and 100% through a 5 mm sieve.

LECA[®] (Light Expanded Clay Aggregate) was used as received, with a particle size of 10–20 mm as provided by the manufacturer. LECA[®] is reported as a material resistant to high temperatures, moisture, and chemical agent attacks. Moreover, the iron oxides present in the clay mineral structure can act as a catalyst for the chemisorption of hydrogen sulphide (H₂S), ammonia, methane, and several other organic compounds.

The corn straw utilized in this experiment was obtained from a dairy cattle farm in southern Italy, where it was primarily employed as bedding material for the weaning of calves and in select other areas of the farm. Subsequent to collection, the straw underwent manual trimming to a length of approximately 2 centimetres.

2.4. Ammonia Emission Measurement

To define the effect of different treatments on ammonia emissions during manure storage under controlled experimental conditions, the ammonia emissions were measured following the dynamic chamber method described by [15,36]. Specifically, the jars were only sealed during the measurement process to allow ventilation of the headspace. Each experimental unit consisted of a 5 L glass vessel containing 3 L of digestate, resulting in an approximate headspace volume of about 2 L. To seal the containers, a screw cap was used that was designed to facilitate airflow and connect the container to an expansion chamber. The flow rate was regulated using a precision flowmeter (Series RM Rate-Master[®] Polycarbonate Flowmeter, 5'' scale, equipped with a needle valve, accuracy ±3% full scale) connected to the system to ensure controlled and repeatable airflow conditions. Each experimental unit consisted of a 5 L glass vessel containing 3 L of digestate, resulting in an approximate headspace volume of about 2 L. The digestate was stored accordingly in open vessels with no lid in a temperature-controlled room. Then, air was sampled for 16 min and analysed in real time using a gas-sensitive semiconductor sensor (Aeroqual Series 500), which was used to quantify NH₃ concentration. The instrument produced by Aeroqual Limited, Auckland, New Zealand has a detection range of 0–100 ppm, with a detection limit of 0.2 ppm and an accuracy of ±0.5 ppm + 10%. The measurement occurred on days 1, 2, 6, 9, 13, 16, 20, 23, 27, 30, and 34, for a total of 11 measurements in 34 days. To maintain consistency and reduce variability, measurements were conducted in a fixed sequence, preserving a constant interval between successive samples. The pH and the manure temperature were monitored before the gas detection started. To avoid disturbance of the sample surface due to the pH probe immersion in the liquid, a plastic tube was applied to the jar wall, before being filled with manure, to create easy

access. The tube was therefore closed with a plastic cap and opened only during the pH measurement. The pH was measured with a portable pH meter (MT51302523, Mettler Toledo, Greifensee, Switzerland).

2.5. Flux Calculations

The gaseous emission fluxes were evaluated as follows:

$$F = Q \frac{C_{out} - C_{in}}{A} \quad (1)$$

C_{in} is the gas concentration of air inlet into the chamber in mg m^{-3} ; C_{out} is the gas concentration of air outlet from the chamber in mg m^{-3} ; Q is the airflow rate through the chamber in $\text{m}^3 \text{h}^{-1}$; and A is the circular area of the emitting surface in m^2 . Cumulative emissions from each manure type over the storage period were evaluated by averaging net flux rates between two sampling points and by multiplying by the time interval between sampling points [15].

The emission factor (EF) was defined as the percentage of N applied as digestate that is emitted as NH_3 , as described by [29].

The reduction efficiency (RE) was calculated as the percentage decrease in cumulative NH_3 emissions relative to the untreated control [35]. Positive values indicate emission reductions, whereas negative values indicate an increase in emissions compared with the control.

$$RE\% = \left[\frac{E_{control} - E_{treatment}}{E_{control}} \right] \times 100 \quad (2)$$

$E_{treatment}$ and $E_{control}$ refer to cumulative emissions of treated (covered) and control samples, respectively.

2.6. Statistical Analysis

The data collected during the observation periods were then organized into datasets, with cumulative NH_3 emissions calculated for each treatment. Statistical analysis was performed using one-way analysis of variance (ANOVA) followed by a post hoc Tukey test to identify significant differences between treatments. A p -value threshold of <0.05 was employed for hypothesis rejection, indicating a significant difference in ammonia emission reduction across treatments. The cumulative emissions were analysed at 24 h, on the 9th day, and on the 34th day of the measurement period. All statistical analyses were conducted using IBM SPSS Statistics for Windows (Version 28.0, IBM Corp., Armonk, NY, USA).

Additionally, to evaluate the statistical robustness and sensitivity of the experimental design given the sample size ($n = 3$ per treatment, $N = 12$ in total), a post hoc statistical power analysis was performed. The effect size was quantified using η^2 and Cohen's f . The achieved power ($1 - \beta$) was calculated based on the observed effect size and a significance level of $\alpha = 0.05$ using R statistical software (v4.5.0; pwr package).

2.7. Nitrogen Mass Balance

A nitrogen mass balance was performed to quantify the distribution of nitrogen among emitted, retained, and remaining fractions. Initial nitrogen stock was calculated from digestate volume (3 L) and TKN concentration (g N kg^{-1}). Nitrogen losses were estimated from cumulative $\text{NH}_3\text{-N}$ emissions. Remaining nitrogen was calculated as the difference between initial and emitted nitrogen. The closure of the mass balance was evaluated for each treatment.

2.8. Comparative Analysis

A scaling-up calculation was conducted considering a full-sized manure storage tank, followed by a purchasing prices comparison across the tested materials. The comparison of biochar, straw, and clay was performed considering the same reduction efficiency (RE). Two hypotheses were considered: namely, RE of 50% (RE50) and 70% (RE70) were chosen. This comparative approach was adopted to evaluate the effectiveness of the tested materials within sustainable livestock waste management systems and their potential applicability. Bibliographic research was conducted to build a correlation between thickness of covers and ammonia RE. In the selected data, thickness and RE were explicitly stated. In Table 2, used data are reported with their corresponding reference.

Table 2. Data used to build the relationship between thicknesses (cm) of different cover materials and relative reduction efficiency (RE). Negative values indicate an enhancement of ammonia emissions.

Materials	Slurry Type	Manure Dry Matter	Storage Days	Layer Thickness cm	RE %	References
Biochar	Pig Slurry	3.3	30	0.635	16	[37]
	Pig Slurry	3.3	30	0.635	25	[37]
	Pig Slurry	3.3	7	0.635	38	[37]
	Pig Slurry	3.3	7	0.635	30	[37]
	Buffalo Digestate	6.6	34	2	67	This study
	Cattle Slurry	9.3	134	2	40	[38]
	Cattle Slurry	5.39	56	5	72	[30]
	Cattle Slurry	0.64	56	5	82	[30]
LECA [®]	Buffalo Digestate	6.6	34	2	−13	This work
	Cattle Slurry	6.14	30	7	17	[18]
	Cattle Slurry	7.87	113	20.8	2	[35]
	Pig Slurry	6.15	70	7	77	[39]
	Pig Slurry	2.33	30	14	75.1	[18]
	Pig Slurry	7	162	82	6	[13]
	Cattle Slurry	9.3	134	83	10	[38]
Straw	Pig Slurry	7.5	189.5	75	8	[20]
	Buffalo Digestate	6.6	34	2	36	This work
	Pig Slurry	2.33	30	7	34	[18]
	Pig Slurry	2.33	30	14	86	[18]
	Buffalo Slurry	1.96	20	1	7.3	[40]
	Cattle Slurry	5.39	56	53	5	[30]
	Cattle Slurry	0.64	56	70	5	[30]

To account for the heterogeneity of the literature data used in the comparative assessment, an additional statistical analysis was performed. A linear mixed-effects model was fitted using reduction efficiency (RE) as the response variable, while cover material, slurry type, dry matter content (DM), storage duration (days), and cover thickness (cm) were included as fixed effects. Study identity was included as a random intercept to account for between-study variability. Furthermore, separate regression analyses were performed for

each cover material (biochar, straw, and LECA®) to investigate the relationship between cover thickness and reduction efficiency. All analyses were conducted using R statistical software (version 4.5.0).

Data were interpolated to retrieve the cover thickness necessary to reach a specific RE for all the materials. For this purpose, both logarithmic and exponential trend lines were used when appropriate, depending on the R^2 value. These calculations were undertaken not only to determine the optimal thickness and associated costs of covers for the storage tanks but also to define the storage volume of the materials and transportation considerations. The calculation was carried out considering a manure storage tank with an emitting surface of 700 m², corresponding to the average dimensions of a storage facility inside a buffalo farm with about 300 lactating animals, which is the average size of a buffalo farm in the region where samples were taken.

A sensitivity analysis was performed on the biochar market price to assess the robustness of the techno-economic comparison. Three price scenarios were considered: a baseline price of €500 m³, corresponding to the purchase price of the biochar used in this study; and two alternative scenarios representing $\pm 20\%$ variation (€400 and €600 m³) [41]. For each scenario, the total cost required to cover a 700 m² manure storage tank was recalculated for two target ammonia reduction efficiencies (RE50 and RE70), with all other parameters kept constant. These costs were then compared with those estimated for LECA® and straw to evaluate how sensitive the economic ranking of the cover materials is to fluctuations in the biochar market price.

3. Results

3.1. Manure Composition

Digestate characteristics before and after the trial are reported in Table 3.

Table 3. Digestate characteristics before and after the trial. Cattle slurry properties at the beginning (Before Exp) and at the end of the experiment (After Exp). NC, control; B, biochar; S, straw; L, LECA®.

		DM	TKN	TAN	pH
		g/kg			
Before Exp	Digestate	66.66	3.5	1.66	8.2
After Exp	NC	81.3	2.9	1.1	8.4
	B	76.2	2.9	1.3	8.1
	S	75.0	2.9	1.2	8.3
	L	73.4	3.1	1.4	7.7

As reported in Table 3, at the end of the experiment, DM values increased in all the tested samples. As a matter of fact, during the storage period, water evaporation occurred and a consequent decrease in weight and digestate level was registered in all the samples, affecting the final DM content. In line with this, the cover applications proved to influence the evaporation of the stored digestate. The highest DM increase of 22% was registered in NC, and the lowest 10% in L. As shown in Figure 2, at the beginning of the experiment, the pH of the untreated manure was 8.3. The lowest value of pH was detected on the 16th day of storage; the decrease occurred in all the samples, but clay showed the lowest value.

The same day was also registered as a decrease in digestate temperature. On the 30th day, the highest peak of pH occurred in all the monitored samples, shifting from a mean of 8.5 to a mean of 8.92. Generally, manure covered with clay had the lowest pH values.

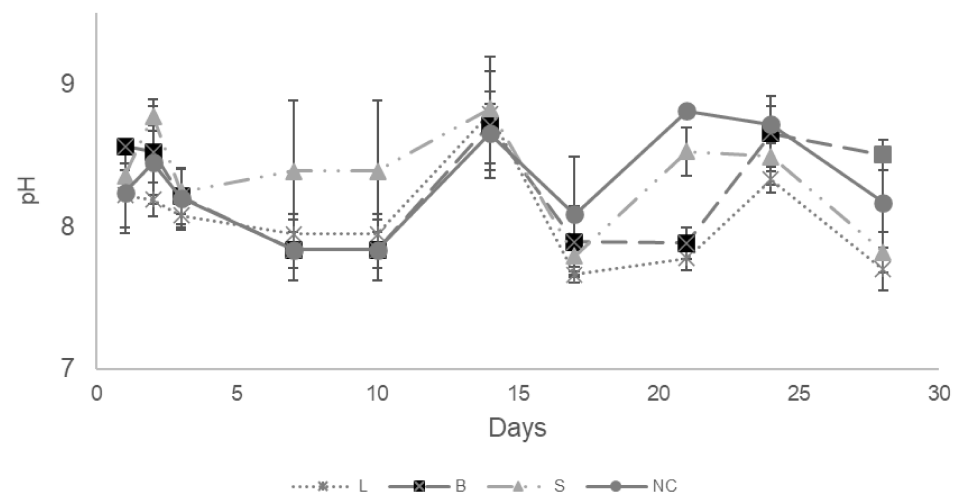


Figure 2. pH trends of digestate covered with clay (L), straw (S), biochar (B), and uncovered control (NC), during the monitored period.

With regard to the nitrogen content (TKN), a decline was observed in all the samples at the conclusion of the experiment. A less marked decrease was observed in L, with a decline of 11.4% being recorded. The reduction register for all other treatment, inclusive of NC, accounted for 17.1%. As with TKN, TAN content exhibited a similar trend across the various treatments. The reduction in TAN content was found to be lowest in L and highest in NC, at 15.7% and 33.7%, respectively.

To better evaluate nitrogen dynamics during the storage period and to address nitrogen partitioning within the system, a simplified nitrogen mass balance was performed considering initial nitrogen content, final TKN and TAN concentrations, and cumulative NH_3 emissions. The results are reported in Table 4. Overall, the observed nitrogen losses through NH_3 volatilisation were consistent with the reduction in TAN content, indicating that the mitigation effect of the tested cover materials was associated with improved nitrogen retention in the liquid fraction. The variation in ΔN was generally consistent with the measured NH_3 emissions, although direct quantitative agreement is not expected because NH_3 emissions were monitored discontinuously, and short-term emission peaks may not have been captured.

Table 4. Nitrogen mass balance across treatments. Initial N refers to nitrogen content at the start of the experiment, final N represents the remaining nitrogen after 34 days. NH_3 emissions refer N losses as cumulative emissions for each treatment. ΔN was calculated as initial N – final N.

	Initial N	Final N	NH_3 Emission	ΔN
	G			
NC	10.50	8.50	0.253	2.00
B	10.50	8.90	0.082	1.60
S	10.50	8.70	0.163	1.80
L	10.50	8.40	0.287	2.10

3.2. Ammonia Emissions

The cumulative NH_3 emission at the end of the trial (34 days) accounted for 4.1, 8.1, 14.3, 12.6 $\text{gN-NH}_3 \text{ m}^{-2}$ for B, S, L and NC, respectively. One-way ANOVA revealed significant differences among treatments at all evaluated time points (24 h, 9 d, and 34 d), as shown in Table 5.

Table 5. One-way ANOVA results for cumulative NH₃ emissions at 24 h, 9 days, and 34 days. F-values, degrees of freedom, and exact *p*-values are reported for each time point. Significant treatment effects were detected at all sampling times (*p* < 0.001).

Time	F	df	<i>p</i> -Value
24 h	59.90	3, 8	7.98×10^{-6}
9 d	45.23	3, 8	2.31×10^{-5}
34 d	26.36	3, 8	1.69×10^{-4}

Table 6 shows the cumulative ammonia emission during the trial; all treatments had the same emission pattern, showing their effectiveness in mitigating ammonia emissions from the first hours of measurement.

Table 6. Mean cumulative NH₃ emissions $\pm$ standard deviation during the experimental period (24 h, 9 days, and 34 days). Different superscript letters (a,b,c) within each column indicate statistically significant differences among treatments (*p* < 0.05, Tukey post hoc test); identical letters indicate no significant differences.

Time Steps	24 h	9 Days	34 Days
g NH ₃ -N m ⁻²			
Treatment			
NC	0.7 ^a $\pm$ 0.1	4.0 ^a $\pm$ 0.3	12.6 ^a $\pm$ 0.4
B	0.0 ^b $\pm$ 0.0	0.3 ^b $\pm$ 0.1	4.1 ^b $\pm$ 0.3
S	0.4 ^c $\pm$ 0.0	2.1 ^c $\pm$ 0.3	8.1 ^b $\pm$ 1.3
L	0.5 ^c $\pm$ 0.1	3.6 ^a $\pm$ 0.2	14.3 ^a $\pm$ 1.2

B was the most effective cover material, reducing NH₃ emissions by 67% compared with NC after 34 days (*p* < 0.05) and achieving the highest reduction efficiency (RE), reported in Table 7. During the trial, the biochar cover remained visibly intact in all replicates, with no cracks or breaks forming on the surface. Although the biochar gradually settled and became unevenly distributed over time, particularly after two weeks, the surface layer remained continuous and free of visible cracks throughout the experimental period. The data obtained are broadly consistent with the major trends showing that biochar cover can be effective in mitigating NH₃ emissions [27]. In good accordance with our findings, [38] reported a significant reduction (*p* < 0.05) in NH₃ emissions following the application of a 1.5 cm biochar cover to swine manure. The same mitigation effect was not registered when thinner layers of 1 cm and 0.75 cm of this material were tested [13,22]. Other authors assessed that superficial biochar application reduced the cumulative ammonia emissions by 48% compared with the control [19]. In other studies, the possibility of making a second application was investigated due to the temporary effect of the biochar [37]. A reapplication of biochar, thereby creating a thicker cover, resulted in a much higher statistically significant percentage reduction. This finding suggests that biochar cover could be a good method to reduce ammonia emission when it is able to create a layer thick enough over the manure surface to influence mass transfer to the headspace.

In the present work, straw cover showed a good mitigation effect as well as a good floating aptitude. S, which was compacted before application, gradually lost density during storage and showed a less uniform surface distribution after the first week. However, only the layer directly in contact with the manure became wet, and the cover remained afloat until day 23. This result, however, is in contrast to a previous study [40], where the application of straw on stored manure did not significantly reduce NH₃ emissions after 6 days. Indeed, straw cover started to sink after the beginning of the trial. This difference in cover behaviour may be attributed to differences in manure characteristics, particularly dry

matter (DM) content, or, more likely, to the thermal pre-treatment applied to the cover prior to testing. Overall, S reduced NH_3 emissions by 36% compared with NC during the first 24 h ($p < 0.05$). In accordance with our findings, ref. [18] reported an 86% reduction in NH_3 emissions ($p < 0.05$) when a 14 cm straw layer was applied, whereas a 7 cm layer resulted in a non-significant reduction of 32% ($p > 0.05$). On the contrary, other tests assessing a layer of 8 cm of treated and untreated straw showed good emission reduction compared to the control ($p < 0.05$) [13]. Furthermore, straw was shown to be effective in mitigating ammonia emission in combination with zeolites not only during manure storage but also during the composting process [42]. When a layer of 30 cm of straw was applied on the manure surface during a pilot scale experiment, after 122 days of storage, the cumulative emissions were reduced by 90% compared to the control [43], showing how a thicker cover layer can enhance its capacity to mitigate ammonia emissions. All the cited authors demonstrated the feasibility of using straw cover to reduce ammonia emissions. Different layer thicknesses and pre-treatment are all factors influencing the reduction efficiency.

Table 7. Emission factors (EFs) referred to TKN and TAN calculated after 34 days of storage. NH_3 emissions reduction efficiency (RE %) of the tested cover compared to the control. Values refer to 24 h, 9 days, 34 days of measurement. Negative values indicate an enhancement of ammonia emissions compared with control. Values reported as 100% at 24 h for biochar reflect emissions below or close to the analytical detection limit in the majority of replicates, rather than absolute zero emissions.

	EF (%)		RE (%)		
	TKN	TAN	24 h	9 Days	34 Days
NC	2.41	5.08	-	-	-
B	0.78	1.65	100 ± 0.0	93 ± 0.02	67 ± 0.06
S	1.55	3.27	52 ± 0.07	49 ± 0.07	36 ± 0.17
L	2.73	5.76	33 ± 0.02	10 ± 0.01	−13 ± 0.22

After 24 h of storage, the LECA[®] cover (L) reduced NH_3 emissions by 29% compared with the control ($p < 0.05$); however, it showed the lowest reduction efficiency among all treatments.

From day 2 to day 13 the cover's effectiveness continued to decrease (Figure 3). At the end of the trial, L showed the lowest RE (Table 7). For the whole trial duration, L was the only cover to remain physically stable. This stability was likely due to the buoyancy of the LECA[®] granules, which remained partially submerged while maintaining exposure above the manure surface. The applied granules completely covered the manure surface, forming a single layer of LECA[®] spheres. Throughout the trial, the granules were incorporated into the manure surface, creating a solid cover less mobile than the others tested. The higher cumulative ammonia emissions observed for L are consistent with the findings of a previous study, which also reported increased emissions from covered manure compared with the control when a 5 cm clay layer was applied to fresh dairy manure in a field trial [12]. Only with a thicker LECA[®] cover corresponding to a 14 cm layer was a significant reduction of ammonia volatilisation observed [13]. For a thinner cover of 7 cm, a reduction of 16.81% compared to the control was assessed, but was not defined as significant [18]. A LECA[®] surface layer depth of 2 cm reduced NH_3 emissions by 21% when applied to stored liquid manure [35]. According to Nartey et al. [12], even when the LECA[®] cover forms a firm surface crust, the gaps between the granules allow NH_3 volatilisation, making the superficial cover application ineffective.

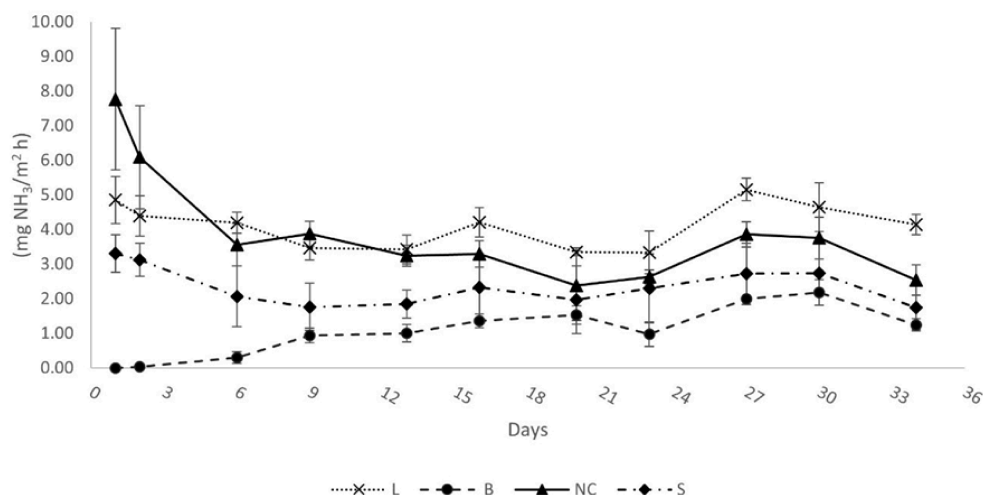


Figure 3. Daily ammonia emission measured during the trial. Error bars indicate SEM.

Lastly, NC showed emissions ranging from 7.8 to 2.4 g m² h^{−1}. On the second day, the maximum emissions started to decrease, not maintaining a stable trend. From day 13 onwards, a floating surface crust developed in all replicates. Although the crust was heterogeneous and prone to cracking, it remained afloat throughout the trial. The minimum temperature between all the tested groups was registered in NC and measured 17.1 °C. Ammonia reduction efficiency and the emission pattern appeared to be consistent throughout the trial. Emission factors (EF), expressed as the percentage of total nitrogen emitted as NH₃ relative to the initial nitrogen content, ranged from 0.78 to 5.76% (Table 7). The lowest EF was observed for B and the highest for L, further confirming the effectiveness of biochar in mitigating ammonia emissions.

Ammonia reduction efficiency (RE) during the trial and among the treatments is shown in Table 7. The measured RE highlight how B had emissions close to 0 for the entire first duration of the trial, while L during the same period had almost lost the mitigation effect. Indeed, L at the end of the trial showed a negative RE.

3.3. Statistical Meta-Modelling of Emission Factors

The robust mixed-effects model (LMM) revealed that cover thickness emerged as the strongest fixed predictor of mitigation efficiency (estimate = 8.26 ± 1.28, *t* = 6.47), indicating that thicker cover layers substantially improved emission reduction regardless of the baseline matrix. Storage duration also positively influenced mitigation performance (estimate = 0.42 ± 0.15, *t* = 2.82), suggesting that cover effectiveness increased over time under controlled conditions.

The marginal effects predicted by the robust mixed-effects model are presented in Figure 4. Cover thickness exhibited the strongest positive relationship with emission reduction efficiency (Figure 4E), followed by storage duration (Figure 4D). In contrast, dry matter content showed only a weak negative trend (Figure 4C). Among the cover materials, biochar achieved the highest predicted mitigation efficiency, whereas LECA[®] displayed the lowest performance (Figure 4A). Differences among slurry types were comparatively small and characterized by broad confidence intervals (Figure 4B).

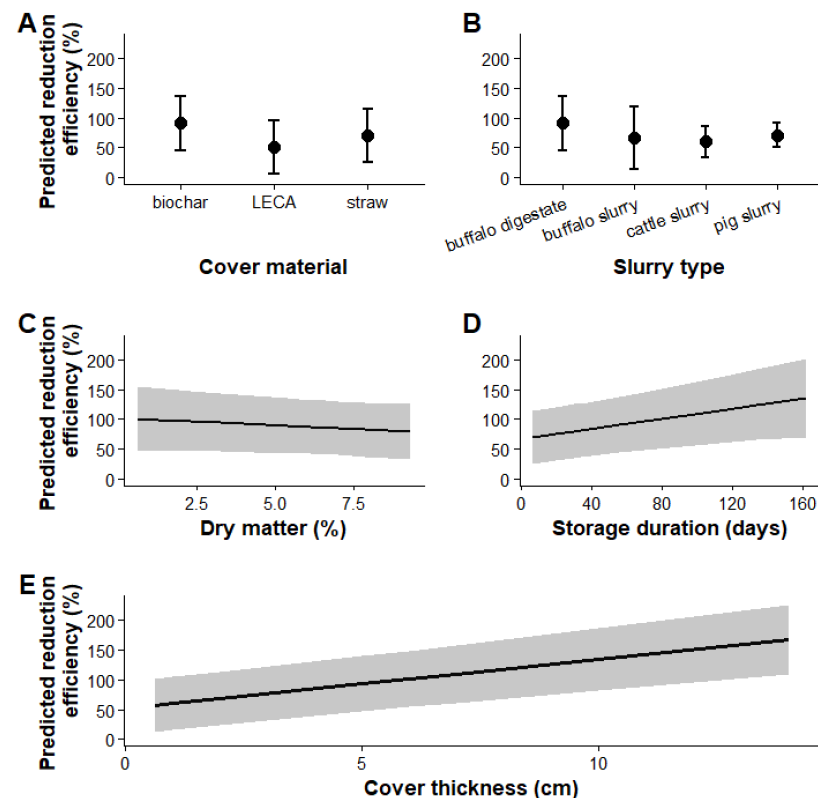


Figure 4. Predicted marginal effects of cover material (A), slurry type (B), dry matter content (C), storage duration (D), and cover thickness (E) on emission reduction efficiency, estimated using a robust mixed-effects model. Shaded areas and error bars indicate 95% confidence intervals. Study identity was included as a random effect to account for between-study variability.

The nearly linear increase in reduction efficiency with increasing cover thickness suggests that thicker floating layers provide a more effective physical barrier to gaseous diffusion. This observation is consistent with previous studies reporting enhanced emission mitigation as the resistance to gas transfer increases. The positive effect of storage duration may indicate progressive stabilization of the cover layer and the establishment of conditions that further restrict gas exchange at the slurry surface. In contrast, slurry type and dry matter content exerted comparatively minor effects, suggesting that the physical characteristics of the cover are more influential than the intrinsic properties of the stored slurry under the conditions represented in this meta-analysis. A limitation of the present analysis is the relatively small number of observations available for some cover materials and slurry categories in the literature. Consequently, the estimated effects should be interpreted as indicative trends rather than definitive quantitative relationships. Additional studies covering a broader range of cover configurations and storage conditions would improve the robustness of future meta-analytical assessments.

Nonetheless, given the dominant effect of cover thickness observed in the mixed-effects model, an exploratory analysis was conducted to evaluate the relationship between cover thickness and reduction efficiency within each cover material category. Separate regression models were fitted using cover thickness as the sole predictor. Linear, logarithmic, and exponential models were evaluated, and model performance was assessed using the coefficient of determination (R^2).

For all cover materials, the logarithmic model provided the best fit to the data. The strongest relationship was observed for biochar ($R^2 = 0.836$), indicating that cover thickness explained approximately 84% of the observed variability in emission reduction efficiency.

Straw also showed a strong logarithmic relationship ($R^2 = 0.729$), whereas the relationship was weaker, although still substantial, for LECA ($R^2 = 0.597$).

The consistency of the logarithmic response across all cover materials indicates that emission reduction efficiency increases rapidly at low cover thicknesses and progressively approaches a plateau as thickness increases.

Figure 5 Material-specific logarithmic relationships between cover thickness and emission reduction efficiency for biochar (A), LECA (B), and straw (C). Logarithmic functions provided the best fit for all cover materials, explaining 83.6%, 59.7%, and 72.9% of the observed variability, respectively. The results indicate diminishing returns in mitigation efficiency with increasing cover thickness, with the greatest gains occurring at lower thickness values. The strong logarithmic relationships observed for biochar and straw suggest that cover thickness is a primary driver of ammonia mitigation efficiency, largely independent of cover material type. These findings support the interpretation that increasing cover thickness enhances resistance to gas transfer, although the magnitude of the benefit progressively decreases as cover thickness increases. The material-specific regression models were subsequently used to estimate the cover thicknesses required to achieve predefined mitigation targets under practical storage conditions.

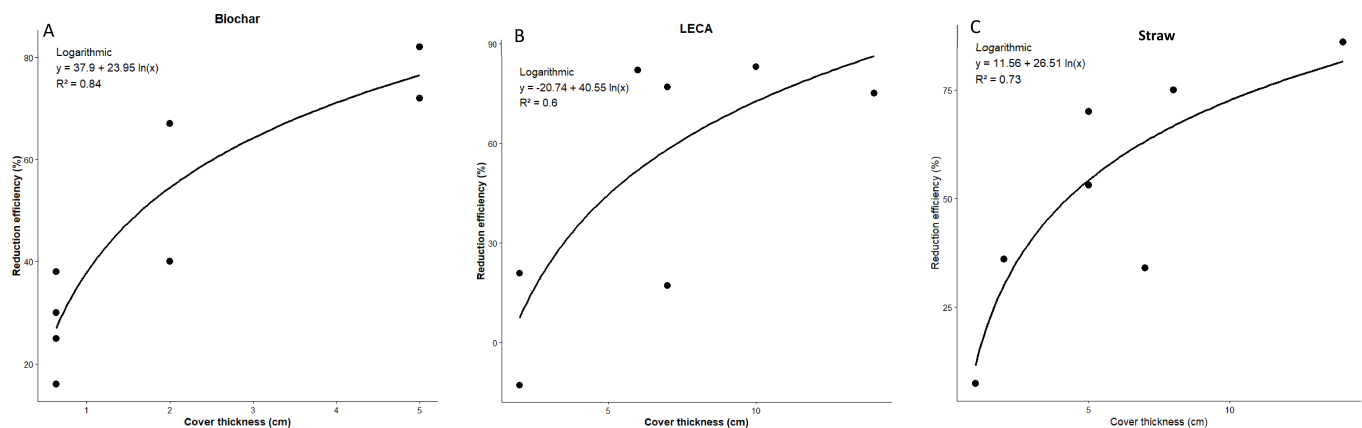


Figure 5. Material-specific logarithmic relationships between cover thickness and emission reduction efficiency for biochar (A), LECA[®] (B), and straw (C). Logarithmic models provided the best fit for all cover materials ($R^2 = 0.836$, 0.597 , and 0.729 for biochar, LECA[®], and straw, respectively), indicating a progressive decrease in mitigation gains with increasing cover thickness.

Given the trend equation shown in Figure 5, the cover thicknesses determined, considering a storage tank with an emitting surface of 700 m^2 and the targeted RE, in RE50 were 1.66 cm, 5.72 cm, 4.26 cm, for biochar, LECA[®], and straw, respectively. The relative volumes required to cover the storage tank with the calculated cover layer were 12.4 m^3 , 92.2 m^3 , 68.3 m^3 for biochar, LECA[®], and straw, respectively. The cover thicknesses for RE70 were 3.82 cm, 9.38 cm, 9.06 cm with corresponding required volumes of 22.7 m^3 , 107.9 m^3 , 84.8 m^3 for biochar, LECA[®], and straw, respectively. In order to assess the economic impact, the unit price of biochar, including transport costs, was set at 500 €/m^3 , as indicated by the seller from whom the biochar was purchased. According to available online sources, the price range for LECA[®] is between $\text{€}8$ and $\text{€}10$ for 50 litres, while straw costs between $\text{€}150$ and $\text{€}170$ per tonne. The total cost of covering a 700 m^2 storage tank with the selected materials is $\text{€}6191$, $\text{€}16,602$, and $\text{€}6146$ in RE50 and $\text{€}11,366$, $\text{€}19,420$, and $\text{€}7634$ in RE70 for biochar, LECA[®], and straw, respectively. These results are reported in Table 8.

Table 8. Cover layer thickness, required volume, and total cost for different materials at two target ammonia reduction efficiencies (RE 50% and RE 70%) for a 700 m² storage tank.

Material	RE Target (%)	Cover Thickness (cm)	Required Volume (m ³)	Total Cost (€)
Biochar	50	1.66	12.4	6191
	70	3.82	22.7	11,366
LECA [®]	50	5.72	92.2	16,602
	70	9.38	107.9	19,420
Straw	50	4.26	68.3	6146
	70	9.06	84.8	7634

A sensitivity analysis was conducted to evaluate the impact of variability in the market price of biochar on the techno-economic results [44]. Three price scenarios were considered, with a baseline value of €500 m^{−3} and variations of −20% (€400 m^{−3}) and +20% (€600 m^{−3}) [41]. Based on the biochar volumes required to achieve the target ammonia reduction efficiencies reported in Table 8, the total cost of covering a 700 m² storage tank ranged from €4953 to €7429 for the RE50 target and from €9093 to €13,639 for the RE70 target. As expected, total costs varied proportionally with the assumed biochar price. Across all scenarios, biochar remained substantially less expensive than LECA[®]. At RE50, the economic ranking between biochar and straw was sensitive to biochar price fluctuations, with biochar being slightly less expensive than straw at the −20% scenario and slightly more expensive at the +20% scenario. In contrast, at RE70, straw remained the least expensive option across all scenarios. Overall, the results indicate that moderate variations in biochar market price do not substantially alter the techno-economic comparison among the evaluated cover materials.

3.4. Limitations of the Study and Future Work

Despite the encouraging outcomes observed in controlled laboratory settings, it is important to acknowledge several limitations when interpreting the findings of this study.

One limitation concerns the number of replicates ($n = 3$ per treatment), which was selected according to the time required for ammonia measurements to ensure that all experimental units could be analysed within the same working day under consistent conditions. A formal a priori power analysis was not conducted prior to the experiment. A post hoc power analysis based on the observed treatment effects for cumulative NH₃ emissions at 34 days indicated a very large effect size (Cohen's $f = 3.14$) and a statistical power of 0.999 ($\alpha = 0.05$). These values indicate a high probability of detecting the observed treatment differences under the experimental conditions. The limited replication ($n = 3$ per treatment) may have reduced sensitivity to smaller effects among treatments. The experimental duration of 35 days is comparatively brief when set against typical on-farm storage times, which may extend over several months. Consequently, the long-term stability and persistence of the mitigation effect remain uncertain. Furthermore, the experimental setup failed to replicate certain environmental dynamics that occur under real farm conditions. Factors such as wind, rainfall, temperature fluctuations, and the periodic addition of fresh slurry to the storage tank can significantly influence the cover integrity and ammonia volatilisation rates. These variables have the potential to modify the physical composition of the cover materials, particularly lightweight ones such as straw and biochar, thereby impacting their efficiency over time. Moreover, no post-experiment characterization of the cover materials was performed. Previous studies on the same biochar and digestate indicated that adsorption alone does not explain the observed reduction in ammonia emissions, suggesting that physical mechanisms likely contribute significantly.

Furthermore, it is important to note that the test conditions may not fully represent the variability in digestate composition that occurs in practice. This is due to the fact that differences in feedstock, anaerobic digestion process, and storage management can influence ammonia emissions. The uniform layer thickness of 2 cm, which was utilised for all materials in this study, was selected for the purpose of comparative analysis. However, it is acknowledged that this thickness may not align with the optimal operational thickness under field conditions. It is recommended that future research efforts concentrate on the execution of full-scale trials. The objective of these trials should be to evaluate the performance, durability, and management requirements of biochar and other floating covers under variable climatic and operational conditions. Such investigations will provide valuable insights into the practicality and sustainability of mitigation strategies in commercial livestock operations.

4. Conclusions

In this study, three natural cover materials (biochar, straw, and LECA[®]) were compared under controlled conditions. Under the tested laboratory conditions, biochar achieved the highest NH₃ emission reduction (67%), followed by straw (36%), while LECA[®] was ineffective and resulted in higher cumulative emissions than the control. This approach contributes to reducing environmental impacts associated with livestock waste management. The results indicate that a 2 cm LECA[®] layer is insufficient to mitigate NH₃ emissions effectively. The comparative assessment showed that biochar achieved equivalent NH₃ reduction targets with substantially lower cover thickness requirements than straw and LECA[®], resulting in costs comparable to straw and considerably lower than LECA[®]. The behaviour of biochar must be studied to better understand how to properly manage a cover when applied under farm scale conditions in order to assess the effect of wind and rain on biochar's mitigation effect. Further studies are also needed to investigate the main physical properties of biochar in relation to the pyrolysis process and how the production techniques can influence gaseous emission mitigations and production costs. However, it should be noted that this study was conducted under controlled laboratory conditions, which do not fully represent on-farm storage systems. Therefore, while the results allow a comparative assessment of the different cover materials, further validation under full-scale field conditions is required before practical implementation.

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