Influence of *Sargassum horneri* Mitigating Odorous Gas Emissions from Swine Manure Storage Facilities

Lavanya Madhavaraj 1, Ho-Dong Lim 1, Kong-Min Kim 1, Dae-Hyuk Kim 2 and Gui Hwan Han 1,∗

1 Center for Industrialization of Agricultural and Livestock Microorganisms (CIALM), 241, Cheomdangwahag-ro, Jeongeup-si 56212, Korea; lavanyamadhavraj@gmail.com (L.M.); eastlake@cialm.or.kr (H.-D.L.); kgm0901@cialm.or.kr (K.-M.K.)

2 Department of Molecular Biology, Institute for Molecular Biology and Genetics, Jeonbuk National University, Jeonju 561–756, Korea; dhkim@jbnu.ac.kr

* Correspondence: ghhan@cialm.or.kr; Tel.: +82-63-536-6713; Fax: +82-63-536-6003

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Abstract: Manures from livestock industries and farmyards should be managed for land application. Currently, a deep pit or barn system is adopted by many swine farms for manure management, therefore releasing harmful gases and rising the total global emissions of GHGs. This research focuses on the effectiveness of the brown seaweed *Sargassum horneri* as a masking agent to mitigate odor-generating gaseous pollutants and reduce the emissions of volatile fatty acids (VFAs) from swine manure storage facilities. Using an optimized procedure, we compared the gaseous emissions from two manure storage barns, one containing swine manure masked with *S. horneri* and the other without masking as a control, over a 30-day period. The results showed that, compared to the control, seaweed masking significantly reduced the sulfide and VFA contents. Furthermore, reductions of 99.48% in H$_2$S, 60 ± 5.21% in NH$_3$ and 74.28 ± 2.14% in gaseous amine emissions were observed within the experimental period. Intriguingly, seaweed masking had beneficial effects, decreasing the total odor content by 97.78 ± 3.15% and increasing the nutrient quality of the manure. *S. horneri* has great potential as a masking agent in swine manure management to control environmental pollution.

Keywords: *S. horneri*; manure additive; mitigation strategy; air quality improvement

1. Introduction

The increase in global animal breeding, such as swine and cattle, is the result of human dietary habits and has led to a significant intensification of livestock farming. In 2016, FAOSTAT stated that 1.5 billion swine and 330 million cattle were raised for meat production [1]. Irritating and noxious gas emissions from swine manure storage facilities are one of the major impacts of livestock industries and have a significant influence on air quality, global climate and surrounding neighborhoods. Unfortunately, storage durations of 3 to 10 months are needed for manure decomposition and land application (a nutrient recycling practice) [2,3]. Consequently, livestock industries pollute the environment by emitting gases such as ammonia (NH$_3$), amines, hydrogen sulfide (H$_2$S) and greenhouse gases (GHGs), which have deleterious effects on the environment and organisms [4]. For instance, NH$_3$ is a harmful pollutant because it plays a vital role in the acidification and eutrophication of ecosystems via its oxidation to nitrous oxide [5]. NH$_3$ is also considered an important neurotoxic substance and has negative effects on the health of animals and humans and the environment. At high concentrations, it can cause ulceration to the eyes and severe irritation to the respiratory tract [6]. Atmospheric H$_2$S concentrations greater than 10 ppm are considered stressors for both humans and swine. For example, H$_2$S gas exposure can lead to neurological diseases, respiratory diseases and eye diseases for animals and humans [7]. Additionally, H$_2$S gas, whose density is higher than air,
can accumulate in the poorly ventilated areas, which intensifies its perilous impact on environment pollution [7, 8]. Therefore, mitigation strategies that reduce gaseous emissions and control noxious odors are necessary. Several methods have been employed to mitigate the gaseous emissions from waste pits and barns; the incorporation of manure additives is one of the most widely practiced methods by swine producers because of its accessibility and cost-effectiveness [9,10]. Various additives that reduce gaseous emissions and odors have been used, such as biochar, sawdust and rice husks. However, biochar is produced via pyrolysis, which takes place at temperatures of 400–800 °C, is relatively expensive and produces a final product with high volatile content and elevated levels of pollutants [11]. Formerly, researchers have utilized biochar for reducing NH₃, for instance, Febrisiantosa et al., [12] showed that coadditive (biochar and FGD gypsum) led to a reduction in NH₃ volatilization by 26–59%. However, it also increased nitrate accumulation by 6.7–7.9 fold, which is supposed to be harmful to the human health and ecosystem [13]. Several researchers represented that biochar has high absorption capacity and can reduce the greenhouse gases and NH₃, although require a high quantity to implement the significant outcome [14]. Some of the strategies used for mitigating odor include organic matter degradation inside manure utilizing biochar, mushroom substrate, rice husks and sawdust as additives and bulking agents during composting process [15–18]. Nevertheless, employing these techniques for odor reduction imposes certain limitations, by the fact that, composting techniques are a slow process, and eventually odor regenerates from the storage manure. Additionally, sawdust and rice husks alone are not effective at reducing gaseous emissions from manure. A summary of the literature describing the performance of these additives in the mitigation of gaseous emissions from livestock industries is shown in Table 1.

Table 1. Summary of the additives utilized to reduce gaseous emissions and compost swine manure in previous studies.

<table>
<thead>
<tr>
<th>Study</th>
<th>Manure Type</th>
<th>Experimental Scale</th>
<th>Additive Utilized</th>
<th>Experimental Duration (days)</th>
<th>Analysis Performed/Gas Emissions Reduction Rate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Present work</td>
<td>Swine manure</td>
<td>Farm</td>
<td>Seaweed</td>
<td>30 days</td>
<td>H₂S: 99.48%, NH₃: 60%, amine: 74.28 and Total odour: 97.78%</td>
</tr>
<tr>
<td>[12]</td>
<td>Swine waste</td>
<td>Small laboratory composters</td>
<td>Biochar and Flue gas desulphurization gypsum</td>
<td>28 days</td>
<td>NH₃ volatilization: 26–59%</td>
</tr>
<tr>
<td>[15]</td>
<td>Mixed manure</td>
<td>Farm</td>
<td>Biochar</td>
<td>60 days</td>
<td>Organic matter degradation *</td>
</tr>
<tr>
<td>[16]</td>
<td>Swine manure</td>
<td>Pilot</td>
<td>Mushroom substrate and rice husks</td>
<td>80 days</td>
<td>Microbial community *</td>
</tr>
<tr>
<td>[19]</td>
<td>Poultry manure</td>
<td>Small laboratory composters</td>
<td>Biochar with wheat straw</td>
<td>42 days</td>
<td>NH₃ reduction: 44%</td>
</tr>
<tr>
<td>[20]</td>
<td>Poultry manure</td>
<td>Small laboratory composters</td>
<td>Bamboo biochar</td>
<td>28 days</td>
<td>NH₃ reduction: 47.1%</td>
</tr>
<tr>
<td>[21]</td>
<td>Cattle slurry and hen manure</td>
<td>Small laboratory composters</td>
<td>Biochar</td>
<td>31 days</td>
<td>CO₂ and CH₄: 11.4–22.5% and 0.004–0.2%, N₂O and NH₃: 0.05–0.1% and 0.8–26.5%</td>
</tr>
<tr>
<td>[22]</td>
<td>Poultry manure</td>
<td>Small laboratory composters</td>
<td>Biochar</td>
<td>42 days</td>
<td>C-CO₂ emission: 6.9% and total C-CO₂ emission: 7.4%</td>
</tr>
<tr>
<td>[17]</td>
<td>Poultry manure</td>
<td>Small laboratory composters</td>
<td>Biochar</td>
<td>210 days</td>
<td>Organic matter degradation: 70%</td>
</tr>
<tr>
<td>[23]</td>
<td>Poultry manure</td>
<td>Small laboratory composters</td>
<td>Bamboo charcoal and bamboo vinegar with sawdust</td>
<td>15 days</td>
<td>NH₃ reduction: 21.5–38.5% and CH₄ reduction: 6.1–22.2%</td>
</tr>
<tr>
<td>[24]</td>
<td>Swine manure</td>
<td>Farm</td>
<td>Sawdust</td>
<td>63 days</td>
<td>Organic matter degradation *</td>
</tr>
</tbody>
</table>

*1, 2, 3 = reduction rate not mentioned.

Therefore, a new strategy that involves utilizing powder of the brown seaweed Sargassum horneri as a masking agent in industrial applications may provide a cost-effective emissions mitigation strategy because such powder is simpler to produce than biochar. S. horneri is one of the main algal species present along the shores of Jeju Island, Republic of Korea and has become a threat to the local coastal biodiversity [24,25]. Besides, this species is considered to have a negative impact on kelp farming, fishery industries and local tourism [25,26]. Moreover, Sargassum-based organic fertilizers have improved soil conditions and growth parameters for field crops [27]. Consequently, the addition
of seaweed to manure as a masking agent has several advantages: (1) an indirect reduction in the amount of seaweed on the coast, thus controlling golden tides [24,25]; (2) the addition of rich nutrients to the manure [28,29]; (3) the odor reduction in swine manure [30]; (4) the development of a sustainable bioadditive for odor mitigation in the livestock industry [30].

In our previous study [30], the effects of *S. horneri* and a microbial consortium (*Bacillus subtilis*, *Saccharomyces cerevisiae* and *Thiobacillus* sp.) alone and the synergistic effects of this brown seaweed with the microbial consortium on the reduction in swine manure odor were compared in laboratory-scale tests. Therein, we determined seaweed swelling on the surface of the manure due to water absorption achieved in thin masking, contributing 98–100% control of odors emissions. Against this background, in the present research, for the first time, we evaluated the mitigation of harmful gas emissions such as H$_2$S, NH$_3$, amine, sulfide emissions; the total odor; and the VFA emissions from storage barns containing swine manure from the swine production industry in the Republic of Korea by *S. horneri* as a masking agent. To the best of our knowledge, this is the first report showing the effects of *S. horneri* as a mitigation strategy for gaseous emissions in field-scale investigations.

2. Materials and Methods

2.1. Materials

Fresh *S. horneri* was collected from Wando, Jeollanam-do, Republic of Korea. After collection, the seaweed was washed, dried, crushed into a fine powder and stored at room temperature. Figure 1 depicts the production of the brown seaweed powder.

![Figure 1. Pictures of the procedure for seaweed powder production: (a) *S. horneri* collection from the sea; (b) seaweed drying in the sun for 6 to 7 days; (c) dried seaweed being crushed; (d) seaweed powder.](image-url)

2.2. Experimental Description

The experimental field site was at Yuhan Farm, a commercial pork production farm located in North Jeolla Province, Jeongeup, Republic of Korea (35°36'00.1'' N and 126°56'09.7'' E). The experiment was performed for 30 days beginning on 29 July 2019. Two similar barns (6.6 m width and 23.7 m length) with a distance of 200 m from each other comprise swine manure (approximately 1-month-old) were used to evaluate the effect of a seaweed masking agent on gaseous emission reduction rates. Throughout the experimental period, manure was maintained at approximately 80% of the total.
volume of each barn. The control barn contained swine manure without seaweed masking (Barn 1), and the test barn contained swine manure with seaweed masking (Barn 2). *S. horneri* (2% w/v) seaweed powder was sprayed with a KZ989 (Kazumi, Osaka, Japan) sprayer from the front and side areas of each barn to form a layer at a thickness of 0.25 cm on the top of the manure [30] (Figure 2). During the experimental period, the average temperature outside and inside each barn (control and test) was recorded every day by a TP-50 thermometer (ThermoPro, Atlanta, Georgia, USA). The gas (NH$_3$, amine and H$_2$S) detections and VFAs content were performed from the front portion of the barns every day at approximately 11 a.m. For a set of 6 consecutive days and the following 3rd day until the end of the experimental period. The total odor and the contents of sulfide and aldehyde gases detections were carried out from the front portion of the barns for the initial and final day of the treatment.

![Pictures of the procedure for seaweed powder production](image)

**Figure 2.** Method and operation performed in the swine farm industry: (a) swine manure storage barn; (b) spraying a solution of seaweed powder as a masking agent on the front of the manure pile; (c) spraying a solution of seaweed powder as a masking agent on the side of the manure pile; (d) manure before seaweed masking; (e) manure after 10 min of seaweed masking; (f) manure after the 30th day of seaweed masking.

### 2.3. Analytical Techniques

Analysis of gaseous ammonia and amine was performed with a Gastec pump (Gastec Corporation, Ayase-city, Kanagawa, Japan) with detector tubes (no. 3L, no. 3M and no. 180, Gastec, Japan). For analysis of the total odor content; the contents of sulfide gases, such as hydrogen sulfide (H$_2$S), methyl mercaptan (CH$_3$S), dimethyl sulfide (C$_2$H$_6$S) and dimethyl disulfide (C$_2$H$_6$S$_2$); the content of gaseous methyl ethyl ketone (C$_4$H$_6$O), and the contents of aldehyde gases, such as formaldehyde (CH$_2$O), acetaldehyde (C$_2$H$_4$O), acrolein (C$_3$H$_4$O), acetone (C$_3$H$_6$O), propionaldehyde (C$_3$H$_6$O), crotonaldehyde (C$_4$H$_6$O), isobutyaldehyde (C$_4$H$_8$O), benzaldehyde (C$_7$H$_6$O), isovaleraldehyde (C$_5$H$_10$O) and valeraldehyde (C$_5$H$_10$O), approximately 2 L gas was collected by a minipump (MP 500NII, SIBATA, Nakane Soka-city, Japan) and stored in polyester aluminum bags (TOP Trading, Bucheon, Korea). Gaseous VFAs were collected using two different types of sorbent tube (ST) gas samplers: (i) a three-bed ST containing Carbopack X, Tenax TA and Carbopack B in a 1:1:1 volume ratio (for aromatics, ketones, esters and alcohols) and (ii) a two-bed ST containing quartz wool and Carbopack C in a 2:8 volume ratio (for VFAs, phenols and indoles). An Agilent J&W 5975C Series GC/MSD (Santa Clara, CA, USA) with an HP-FFAP capillary column (30 m × 0.32 mm × 0.25 mm) was used to separate the VFAs. The injector temperature was maintained at 250 °C. The GC oven...
temperature was set to 60 °C for 2.0 min, ramped at 6 °C min⁻¹ to 145 °C and then at 20 °C min⁻¹ to 240 °C, and held for 3.0 min. Helium was utilized as the carrier gas at a flow rate of 1 mL min⁻¹. For analysis of the total odor content and the contents of sulfide and aldehyde gases, samples were sent to the Siheung Green Environment Center, Siheung, Republic of Korea. The data were analyzed with IBM SPSS Statistics (Version 23.0. Armonk, NY: IBM Corp.) and determined the independent sample t-test and p values. A significant level between control and test was evaluated at a significance level of p < 0.05. The graphs were created and analyzed with SigmaPlot software (SigmaPlot®, version 11).

3. Results

3.1. Effects of Seaweed Masking on Gaseous Ammonia and Amine in Swine Barns

Figure 3 shows the measured gaseous NH₃ and amine concentrations in the control and test barns during the one-month experimental period. The application of the 2% w/v solution of seaweed masking agent resulted in a significant reduction in gaseous NH₃ emissions of 67 ± 2.14% (t = 8.22; p < 0.001) on day 0 (after spraying seaweed) and of 42 ± 0.14–62 ± 2.14% (p = 0.0341) and 60 ± 2.12% over the first 6 days and total month of application, respectively (Figure 3a). On the other hand, the control, with no seaweed masking agent applied, showed no significant reduction in gaseous NH₃ over the entire test period; in fact, gaseous NH₃ emissions increased from 20 ± 5.21 ppm to 25 ± 6.41 ppm. When the temperature increased outside the barn, the gaseous NH₃ concentration increased to approximately 35 ± 5.21 ppm in the control, creating an excessive odor nuisance in and around the storage barns. In contrast, in the test barn, there was no apparent increase in the gaseous NH₃ concentration or odor nuisance when the temperature increased outside the barn (Figure 3a).

Subsequently, Figure 3b presents the gaseous amine concentration in the control and test storage barns. In the control, the amine concentration was 140 ± 2.98 ppm and was constant throughout the experimental period. In the test, the amine concentration was initially similar to that in the control, i.e., 140 ± 1.42 ppm, but after seaweed masking, it decreased to 36 ± 5.71 ppm—a reduction of 74 ± 28.64%—and remained at this concentration until day 3. On day 4 in the test barn, the amine concentration increased to 46 ± 10.02 ppm, where it remained until day 12. This increase was presumably due to the temperature increase inside and outside the barn during that period (Figure 3c). After day 12, the amine concentration varied between 25 ± 12.5 to 36 ± 5.77 ppm, resulting in an overall reduction of approximately 74.28 ± 2.41% (t = 14.95; p = 0.001) compared to that before masking and reflecting good control over the gaseous amine concentration in the test barn.

Finally, Figure 3c demonstrates the variation in temperature outside the barns and in the control and test barns over the entire experimental period. In storage facilities, the biodegradation of manure could fluctuate with the temperature. As presented in Figure 3c, the temperature in the control barn...
was slightly lower than the outside temperature except on days 5 and 12 when high temperatures were recorded in the barn. Notably, large differences in temperature were observed in the test barn compared with the control barn and outside. The highest temperature recorded in the test barn was 32 °C for the entire experimental period, except on day 5, when the outside temperature was very high at approximately 43 °C and the temperature in the test barn was 37 °C, creating a suitable environment for the microbes in the manure.

3.2. Effects of Seaweed Masking on the Contents of Sulfide and Aldehydes in Storage Barns

Gaseous H₂S is emitted from the decomposition and metabolization of undigested proteins and sulfur-containing amino acids such as methionine and cysteine in manure [31]. As shown in the profiles in Figure 4a, the concentration of gaseous H₂S gradually decreased, with a significant overall reduction of 99.49 ± 0.53% (t = 7.43; p = 0.0241) during the experimental period when compared with the control. Furthermore, as illustrated in Figure 4b, the total odor content in the test barn was reduced to 144 ± 6.21 ppb, reflecting a reduction of approximately 97.78 ± 0.49% (t = 8.06; p < 0.001) over the experimental period. Notably, before the seaweed masking agent was applied, the total odor content was 6463 ± 22.12 ppb, which immediately decreased to 4400 ± 20.14 ppb, reflecting a reduction of 32.23%, after the application.

![Figure 4. Effects of seaweed masking on (a) hydrogen sulfide (p = 0.0241) and (b) total odor (p < 0.001) during swine manure storage. The results are the mean of three replicates, and the error bars indicate the standard deviation. BM - before masking.](image)

In Table 2, the contents of CH₄S, C₂H₅S, C₂H₅S₂ and C₄H₈O are shown. No reduction in the initial (day 0) and final (day 30) concentrations of these sulfides were in the control barn. However, in the test barn, a trend level reduction in the sulfide contents of 68.45–99.48% on day 30 was observed, indicating that seaweed masking can decrease the gaseous emissions from swine manure.
In the present study, masking with *S. horneri* had a varied effect on the control over gaseous aldehyde emissions, as few of the aldehyde compounds increased in abundance in the test barn over the experimental period (Table 2). However, C₃H₆O, which can trap hydrogen sulfide, decreased by approximately 97.58 ± 0.54% in the test barn over the test period. Notably, aldehydes such as C₂H₄O and C₃H₆O decreased by 82.63 ± 2.31% and 67.85 ± 1.21%, respectively (trend level difference was observed). In contrast, there was no large difference in the aldehyde content between the initial and final day in the control barn.

### 3.3. Reduction in Volatile Fatty Acid Emissions during the Seaweed Masking Treatment

In Figure 5, acetic acid and propionic acid, which are the major VFAs, composing approximately 60–70% and 10–20% of the VFAs emitted from manure [32], were observed. In the test barn, seaweed masking steadily decreased the acetic acid content by approximately 79.61 ± 2.01% (*t* = 4.39; *p* = 0.0120). In the control, the content of acetic acid (510 ppb) was higher than those of the other acids and did not decrease over the entire experimental period. Similarly, the propionic acid content decreased by 77.56 ± 1.24% (*t* = 10.94; *p* = 0.0380) after seaweed masking, whereas no change in the propionic acid content was observed in the control.

![Figure 5](image-url)

**Figure 5.** Effects of seaweed masking on (a) acetic acid (*p* = 0.0214) and (b) propionic acid (*p* = 0.0380) during swine manure storage. The results are the mean of three replicates, and the error bars indicate the standard deviation. BM—before masking.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Control Initial (Day 0) ppb</th>
<th>Control Final (Day 30) ppb</th>
<th>Seaweed Masking Treatment Initial (Day 0) ppb</th>
<th>Seaweed Masking Treatment Final (Day 30) ppb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulfide Content</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Methyl mercaptan</td>
<td>200 ± 8.22</td>
<td>210 ± 3.45</td>
<td>204 ± 4.11</td>
<td>0.6 ± 0.32</td>
</tr>
<tr>
<td>Dimethyl sulfide</td>
<td>41.5 ± 5.21</td>
<td>48.5 ± 2.14</td>
<td>42.5 ± 2.32</td>
<td>6.71 ± 0.48</td>
</tr>
<tr>
<td>Dimethyl disulfide</td>
<td>161 ± 4.21</td>
<td>170 ± 1.42</td>
<td>166 ± 1.14</td>
<td>11.1 ± 0.21</td>
</tr>
<tr>
<td>Methyl ethyl ketone</td>
<td>1.35 ± 2.12</td>
<td>1.49 ± 1.65</td>
<td>1.49 ± 0.88</td>
<td>0.47 ± 0.42</td>
</tr>
</tbody>
</table>

| Aldehyde Content     |                            |                             |                                               |                                             |
| Acetaldehyde         | 20.21 ± 0.51               | 17.43 ± 0.82                | 18.2 ± 0.22                                  | 3.43 ± 2.53                                 |
| Acrolein             | 0.12 ± 0.05                | 0.11 ± 0.98                 | 0.56 ± 0.11                                  | 0.18 ± 0.57                                 |
| Acetone              | 11.0 ± 0.32                | 12.0 ± 1.24                 | 12.0 ± 0.64                                  | 0.29 ± 0.05                                 |
| Propionaldehyde      | 0.78 ± 0.85                | 0.38 ± 0.62                 | 0.38 ± 0.11                                  | 4.69 ± 0.54                                 |
| Crotonaldehyde       | 0.88 ± 0.62                | 0.97 ± 0.55                 | 0.18 ± 0.32                                  | 0.45 ± 0.12                                 |
| Isobutyraldehyde     | 0.83 ± 0.81                | 0.88 ± 0.72                 | 0.23 ± 0.11                                  | 7.43 ± 0.04                                 |
| Benzaldehyde         | 0.10 ± 0.11                | 0.20 ± 0.33                 | 0.30 ± 0.41                                  | 0.8 ± 0.06                                  |
| Isovaleraldehyde     | 0.42 ± 0.94                | 0.49 ± 0.44                 | 0.39 ± 0.12                                  | 0.36 ± 0.21                                 |
| Valeraldehyde        | 0.11 ± 0.63                | 0.15 ± 0.14                 | 0.38 ± 0.14                                  | 0.3 ± 0.10                                  |
Figure 6a,b show a rapid decrease in butyric and isobutyric acid by $80 \pm 0.42\% \ (t = 10.12; \ p = 0.0851)$ and $88.88 \pm 1.02\% \ (t = 7.13; \ p < 0.001)$, contributing to the suppression of odor emissions from the swine manure in the test barn. On the other hand, Figure 6c,d demonstrate an initial rapid decrease in the contents of valeric acid ($t = 5.82; \ p < 0.001$) and isovaleric acid ($t = 4.40; \ p < 0.001$), followed by a gradual decline and ultimate stabilization at zero, resulting in an overall reduction of 100%.

4. Discussion

Odor emissions from swine storage facilities are due to the fermentation of swine manure—which is a mixture of urine and excreta that contains endogenous end products of digestion—by the microbes in the lower gastrointestinal tract. A variety of simple and complex organic compounds can form, which results in gaseous emissions [33]. The most concerning gases in the swine industry are gaseous ammonia, amines and hydrogen sulfide. Excessive levels of ammonia in the swine industry due to the storage of swine manure for long periods can pose a direct hazard to farmers and the animals themselves [34]. Furthermore, a large amount of gaseous ammonia emissions can release heat and cause thermal injury [35]. In addition, monitoring odor emissions are required in environmental management [36]. It is well known that air quality is an essential consideration for animal caretakers working inside facilities and for the animals themselves due to the respiratory issues that poor air quality can cause [37]. The storage of manure for land application generates more odor than other manure management techniques employed in livestock industries [38]. Smith et al. reported that stored solid manure generates high odor emissions for a long period [39], whereas Moseley et al. reported...
that manure injection can reduce odor emissions by 80–85% compared with emissions from manure land application [40]. In addition, Lau et al. suggested that subsurface deposition (SSD) with the Aerway applicator reduced odor emissions by 8 to 38% compared with land application emissions [41]. However, Flessa and Beese [42] suggested that manure injection has the potential to enhance gaseous nitrous oxide (N2O) emissions when compared with the surface application. Therefore, the present strategy of surface masking with seaweed may be a viable option for controlling the odor from storage barns via an immediate reduction in the total odor and simultaneously controlling NH3, amine and H2S gaseous emissions. The simple reason of utilizing seaweed as a surface masking agent is that they comprise interesting hydrophilic characteristics, which leads to water absorption, therefore contributing to swelling. This swelling on the surface of the manure results in thin masking, hindering the mass transfer from the inside to the outside surface of the manure [30].

Figure 3 shows that application of the 2% w/v seaweed masking reduced NH3 emissions by 60 ± 5.21% and amine emissions by 74 ± 28.64%, respectively. A similar study by Maurer et al. utilizing nonactivated biochar reported a reduction in gaseous NH3 emissions of 13 to 23% and suggested that a thin layer of masking agent can be effective at mitigating gaseous emissions [10]. Intriguingly, research has shown that masking agents are most effective at controlling odors by reducing gaseous emissions from stored manure [43]. Additionally, Smith et al. reported that masking agents can be labile to degradation by microorganisms, hence indicating a potential for rapid odor reduction [39]. However, the selection of a masking agent should be a careful consideration, as Maurer et al. suggested that additives that reduce NH3 emissions are capable of increasing GHG emissions [9]. Reports have shown that the amendment of feed with seaweed, especially for cows, can reduce methane emissions by 99% [44]. It follows that the reduction in and control over the gaseous NH3 and amine concentrations might have been due to the thorough coverage of the swine manure surface by the seaweed, which affected the mass transfer of the gases to the headspace. Thus, the persistent repulsive nuisance odor near the storage barn 1 was suppressed after day 1 until the end of the experimental period. Previous laboratory-scale results from our laboratory demonstrated that S. horneri seaweed masking results in good reductions (90–100%) in gaseous NH3 and amine emissions from swine manure [30]. Consequently, in the current study, the usage of brown seaweed as a masking agent could provide a viable, long-term solution for the reduction in gaseous emissions from storage barns.

Furthermore, Figure 3c represents the variation in temperature inside the test barn due to amendment of seaweed masking and control barn without seaweed masking. Noticeably, an extensive reduction in temperature was observed in the test barn compared with the control barn and outside. Interestingly, reducing the temperature inside barns is one of the proposed strategies to increase the activity of microbes responsible for reducing the gaseous NH3 concentration and nuisance odor [45]. McCrory et al. suggested that higher temperatures inside storage barns favor the dominant pathway of NH3 production [43], and Bleizgys et al. [46] stated that reducing the temperature inside barns could be an excellent way to reduce NH3 emissions. The above strategies align with the results of the current study, as seaweed masking decreased the barn temperature, consequently reducing gaseous NH3 and amine emissions and eliminating the nuisance odor.

Sulfides, especially hydrogen sulfide (H2S), are highly toxic not only for humans but also for swine; approximately 10,000 ppb is the recommended exposure limit for H2S gas; 20,000 ppb or more H2S gas concentration can develop loss of appetite, nervousness and possibly pulmonary edema [47]. Figure 4a represents that the seaweed masking significantly decreased the H2S concentration in the test barn, which indicates the effectiveness of the brown seaweed S. horneri as a masking agent. In general, manure in animal facilities consists of a mixture of urine and feces, containing undigested nutrients including protein, phosphorus, nitrogen and potassium [48]. Gaseous H2S is emitted from the decomposition and metabolization of undigested proteins and sulfur-containing amino acids such as methionine and cysteine in manure [31]. Based on the outcome, seaweed addition could stimulate the activity of sulfate-reducing bacteria (SRB) participating in the degradation of organics to effectively reduce gaseous H2S emissions. This is similar to the findings of Aires et al., who reported that seaweed
enhances the growth of SRB and is involved in sulfate reduction [49]. In this context, Miller et al. suggested that sulfide-utilizing bacteria degrade sulfide compounds present in swine manure, resulting in a reduction in sulfide content in the manure and supporting conditions for the efficient growth and activity of organic matter-digesting bacteria [47].

Table 2 showed the reduction in $\text{CH}_4S$, $\text{C}_2\text{H}_6\text{S}$, $\text{C}_2\text{H}_6\text{S}_2$ and $\text{C}_4\text{H}_8\text{O}$ contents to about 68.45–99.48% in the test barns, whereas no significant differences were observed in the control barn, indicating the effects of seaweed masking. Furthermore, dissimilatory sulfate reduction can occur in seaweed-enriched sediments, as revealed by the detection of proteins encoded by upregulated dsr genes [50]. Therefore, the decrease in the sulfide content might be due to the addition of seaweed to the stored swine manure. Furthermore, the aldehyde contents inside swine waste storage facilities are difficult to accurately determine, as aldehydes can react with hydrogen sulfide as trapping agents [51]. Notwithstanding, there was no large difference in the aldehyde content between the initial and final day in the control barn (Table 2). Sánchez-García et al. mentioned that marine seaweed can produce aldehyde compounds depending on the environmental conditions [52], and Sun et al. reported that aldehyde compounds constitute the odor and flavor of seaweed [53]. Therefore, it is reasonable that the addition of $S. \text{hornerias}$ as a masking agent resulted in significant increases in the aldehyde emission from the stored swine manure. More research is warranted to determine the mechanisms underlying the effect of $S. \text{horneri}$ on aldehyde emissions.

The profiles shown in Figs. 5 and 6 demonstrate the significant reduction in the VFA contents after the addition of the seaweed masking agent, which could have been due to the effects of surface masking and the increased activity of VFA-degrading microorganisms inside the manure. Subsequently, the method of manure storage has a significant effect on the magnitude of volatile fatty acid (VFA) emissions in storage facilities. Inefficient degradation of organic waste creates enormous quantities of VFA [45]. Therefore, it is imperative to increase the activity of organic matter-digesting microorganisms, which enhance the degradation of organic matter usually found in swine manure. In this context, we assessed a strategy to increase the microbiome activities and reduce the emission of VFAs to control the noxious odor emitted from stored swine manure. Figure 5 shows a steady decrease in the acetic acid and propionic acid content compared to the control barn. It should be noted that VFAs are major energy sources for the growth and maintenance of microorganisms: propionic acid is a principal glycolytic precursor, and acetate acid is a primary precursor of fatty acid production [54]. Therefore, the reduction in these acids in manure can ultimately reduce the harmful gas emissions to the environment. In addition, Figure 6 represents the rapid declining of valeric acid and isovaleric acid contents to zero at the end of the experimental period. Furthermore, butyric and isobutyric acid expeditiously decreased, contributing to the removal of noxious odor emissions from the swine manure in the test barn. In conclusion, masking with a 2% w/v seaweed amendment could increase VFA degradation possibly because of the effect of the seaweed on the environmental conditions, such as a reduction in temperature inside the test barn (Figure 3c), creating favorable conditions for VFA-degrading microbes. Sundberg et al. suggested that sustained low temperatures could stimulate VFA-degrading organisms, thereby reducing the associated odor [55]. Nozhevnikova et al. stated that VFA degradation occurred in low-temperature environments via stimulation of hydrogen and acetate-utilizing methanogens [56]. Additionally, in our previous laboratory-scale study, $S. \text{horneri}$ application enhanced the activities of organic matter-degrading bacteria, thereby sustaining a high VFA degradation rate during the treatment of swine manure for odor reduction [26]. Maia et al. reported similar results, in which seaweeds combined with certain substrates decreased VFA production [57]. Overall, $S. \text{horneri}$ could effectively suppress the noxious odor in swine manure storage facilities by increasing the activity of organic matter-degrading microorganisms and consequently decreasing the VFA content—especially under high-temperature environmental conditions—and helping to improve the environment for the animal caretakers and animals. Overall, the application of seaweed as a masking agent could instantaneously control odor and gaseous emissions for improved air quality.
5. Conclusions

A new masking strategy to reduce harmful and nuisance gases in swine manure storage barns was developed. Previous applications of *S. horneri* demonstrated that this species was effective at reducing gaseous emissions from swine manure. However, this is the first assessment of *S. horneri* as a masking agent to reduce the emissions of gaseous H$_2$S, NH$_3$, amines and sulfides and increase the VFA degeneration in a swine manure storage barn, which was tested for one month. Immediate reductions of 23.91% in H$_2$S, 67% in NH$_3$ and 74.28% in gaseous amines were observed 10 min after the addition of the seaweed masking agent, and these low levels were maintained or reduced further over the test period. After 30 days, significant reductions in H$_2$S (99.48%), NH$_3$ (60%) and amine (74.28%) emissions were observed, with a concomitant decrease in VFAs of approximately 77–100%. Interestingly, a reduction (97.78%) in the total odor was observed in and around the test barn.

More work is warranted to compare and evaluate the seaweed masking of different types of manure and analyze the comprehensive control of odors, H$_2$S, NH$_3$ and greenhouse gas (GHG) emissions from the livestock industries. In conclusion, masking manure with *S. horneri* could be an effective approach to rapidly control gaseous emissions and reduce odors from swine manure storage facilities.

**Author Contributions:** G.H.H. conceived the study; L.M. designed and performed the experiments; H.-D.L. and K.-M.K. analyzed the data; D.-H.K. contributed material/analysis tools, and L.M. wrote the paper. All authors have read and agreed to the published version of the manuscript.

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**References**


12. Febrisiantosa, A.; Ravindran, B.; Choi, H.L. The Effect of Co-Additives (Biochar and FGD Gypsum) on Ammonia Volatilization During the Composting of Livestock Waste. *Sustainability* 2018, 10, 795. [CrossRef]


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