

Monitoring of sulfur dioxide emission resulting from biogas utilization on commercial pig farms in Taiwan

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Abstract The objective of this work tends to promote methane content in biogas and evaluate sulfur dioxide emission from direct biogas combustion without desulfurization. Analytical results of biogas combustion showed that combustion of un-desulfurized biogas exhausted more than 92 % of SO₂ ($P < 0.01$). In the meantime, more than 90 % of hydrogen sulfide was removed during the combustion process using un-desulfurized biogas ($P < 0.01$). Those disappeared hydrogen sulfide may deposit on the surfaces of power generator's engines or burner heads of boilers. Some of them (4.6–9.1 % of H₂S) were converted to SO₂ in exhaust gas. Considering the impacts to human health and living environment, it is better to desulfurize biogas before any applications.

Keywords Livestock biogas · Hydrogen sulfide · Sulfur dioxide · Pig farm · Human health

Introduction

Biogas produced from anaerobic digestion of livestock animal manure may be used for electricity generation or direct combustion on farms. Biogas produced from anaerobic digesters of pig farms mainly contains methane (60.1–77.0 %, $v v^{-1}$) and carbon dioxide (18.2–26.7 %, $v v^{-1}$) (Su et al. 2003). When biogas is released in the atmosphere, it contributes to the greenhouse effect of the livestock sector.

Biogas is an aggressive gas in terms of corrosion, so the used equipment demands special care. This characteristic is a consequence of the presence of 0.1–0.5 % ($v v^{-1}$) of H₂S (Salomon and Silva Lora 2009). The content of H₂S can be higher than 0.5 % ($v v^{-1}$) depending on its biogas source (0.1–0.8 %, $v v^{-1}$) (Huertas et al. 2011). Some studies imposed restrictions to allowable H₂S levels in the biogas for internal combustion engines from 15 to 150 mg m⁻³ (Salomon and Silva Lora 2009; Wellinger and Lindberg 2000). Hydrogen sulfide can be removed from biogas either by non-microbial or microbial processes.

Hydrogen sulfide can be oxidized to form SO₂ ($H_2S + 1.5 O_2 \rightarrow H_2O + SO_2 + 518 \text{ kJ mole}^{-1}$) through combustion of un-desulfurized biogas using power generators, boilers, or steam cookers (Laursen 2007). Sulfur dioxide in the atmosphere can cause acid rain through oxidation ($SO_2 + 0.5 O_2 \rightarrow SO_3 + 99 \text{ kJ mole}^{-1}$) and hydration ($SO_3 + H_2O \rightarrow H_2SO_4 + 101 \text{ kJ mole}^{-1}$), which can pollute our living environment (Ahammad et al. 2008; Laursen 2007). Thus, both H₂S and SO₂ are toxic gases and harmful to human health.

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Latest National Ambient Air Quality Standards for sulfur dioxide was set at $212 \mu\text{g m}^{-3}$ in 1-h (primary standard) on June 22, 2010 (USEPA 2013). The H_2S in the biogas will depend on the feedstock composition and digester's pH (e.g., for a manure sulfur content of 0.2 % and digester pH of 7.2, the raw biogas can contain H_2S in concentrations of nearly 3001 mg m^{-3} . The standard of US Occupational Safety & Health Administration (OSHA) for maximum H_2S permissible exposure level is 30 mg m^{-3} in 10-min maximum duration.) (UNEP 2002). The proposed and final rules for H_2S are 15 mg m^{-3} as an 8-h time weighted average (TWA) and 22.5 mg m^{-3} as a short-term exposure limit (STEL) (URL: <http://www.cdc.gov/niosh/pel88/7783-06.html>). Scrubbing the raw biogas to eliminate its H_2S and NH_3 content will prevent the formation of corrosive sulfur and nitrogen oxides, thus increasing the potential uses of the biogas.

In Taiwan, there are more than 95 % of commercial pig farms (a total of 5.8 million pigs on farms in 2014) equipped with underground, horizontal anaerobic digesters, and wastewater treatment facilities. However, un-desulfurized biogas is applied to heating lamps of piglets in winter and emitting into the atmosphere in summer. Biogas utilization was idled for 20 years in Taiwan until the successful development of biogas bio-filter facility to remove H_2S in livestock biogas (Su et al. 2008, 2013, 2014). However, most pig farmers still do not understand the essence of biogas desulfurization for human health and environmental concerns. The main goal of this study was to evaluate H_2S and SO_2 emission in exhaust gas and human health issue after un-desulfurized biogas combustion.

Materials and methods

Commercial pig farms

Three commercial pig farms equipped with power generators, boilers, or hot water stoves were selected for this study (Table 1). Un-desulfurized biogas was used as the sole fuel for power generators, boilers, and hot water stoves. Pig farm (I) (1000 pigs on farm) has been equipped with a set of biogas bio-filter facility for desulfurizing biogas (Su et al. 2013, 2014). Pig farm (II) (200 pigs on farm) has been equipped with a boiler. Pig farm (III) (300 sows on farm) has been equipped with a hot water stove.

For Pig farm (I), the biogas mixture consisted of desulfurized and un-desulfurized biogas by the ratio of 4:1 resulting from some portion of the un-desulfurized biogas that entered the desulfurized gas pipelines. All exhaust gas samples were detected for H_2S and SO_2 concentrations on site.

Monitoring the exhaust gas from biogas combustion

In addition, another commercial pig farm equipped with power generators was also selected for monitoring H_2S and SO_2 concentrations in the exhaust gas samples by using either desulfurized or un-desulfurized biogas as the sole fuel. A stainless steel sampling tubing was connected to all gas detectors (PortaSens II detector for H_2S ; MX6 iBRID detector for SO_2) for reducing exhaust gas temperature through a Teflon tubing. The sketch of the experiment design was shown in Fig. 1. Biogas produced from anaerobic digesters of the pig farm was introduced to either a bio-filter facility by a biogas vacuum pump or for power generator. Parameters for biogas production and application forms of the three pig farms are shown in Table 1.

Analysis

Determination of H_2S on the farm

Inlet and outlet H_2S were determined using detector tubes (Gastec Co., Japan) (detection range of 0–61,560 mg m^{-3} ; detection limit of 0.14–2.8 mg m^{-3}) and a gas sampling pump (GV-100C; Gastec Co., Japan). A portable electronic H_2S detector was used to complement H_2S measurements for outlet biogas (PortaSens II, Analytical Technology, Inc., USA) (accuracy, $\pm 5 \%$; sensitivity, 1 % of sensor module range). Inlet and outlet temperatures and relative humidity were determined simultaneously by thermal/humidity meters (TES-1364; TES Electrical Co., Taiwan) (accuracy, $\pm 0.5 \text{ }^\circ\text{C}$; $\pm 3 \text{ RH}$ at $25 \text{ }^\circ\text{C}$).

Determination of SO_2 on the farm

Sulfur dioxide in exhaust gas of either the power generator or hot water stoves were determined using detector tubes (Gastec Co., Japan) and a gas sampling pump (GV-100C; Gastec Co., Japan) (detection range of 0.143–10,296 mg m^{-3}). A portable multi-gas leak detector (MX6 iBRID, Industrial Scientific Co., USA)

Table 1 Parameters for biogas production of three commercial pig farms

Pig farm	Biogas source	Daily wastewater volume (m ³ d ⁻¹)	Biogas yield ^a (m ³ d ⁻¹)	Biogas flow rate (L min ⁻¹)	H ₂ S in biogas (mg m ⁻³)	Biogas utilization (specifications)
I	Anaerobic digestion of piggery wastewater	30	100	50 for bio-filter	5575±862	Power generator (30 kW)
II	Anaerobic digestion of piggery wastewater	6	20	NA	2510±1061	Boiler (5.7 kg-biogas h ⁻¹)
III	Anaerobic digestion of piggery wastewater	10	30	NA	4562±1298	Hot water stove (1.7 kg-biogas h ⁻¹)

^a Pig number multiples 0.1 m³ per pig per day (measured parameter on site)

NA not available

(detection range of 0–286 mg m⁻³ SO₂; increments of 0.28 mg m⁻³) was used to complement SO₂ measurements for gas samples.

Statistical analysis

The experimental data of different samples were then analyzed using the ANOVA procedure of data analyzing and graphing software, Origin (OriginLab, Northampton, MA).

Results and discussion

Evaluation of hydrogen sulfide and sulfur dioxide emission from biogas combustion

For Pig farm (I), the concentrations of H₂S and SO₂ were 5575±862 and 19.2±5.6 mg m⁻³ in un-desulfurized biogas, respectively (Table 2). Combustion of un-desulfurized biogas can produce sulfur dioxide (2H₂S+3O₂→2H₂O+2SO₂). When the un-desulfurized biogas was used for the power

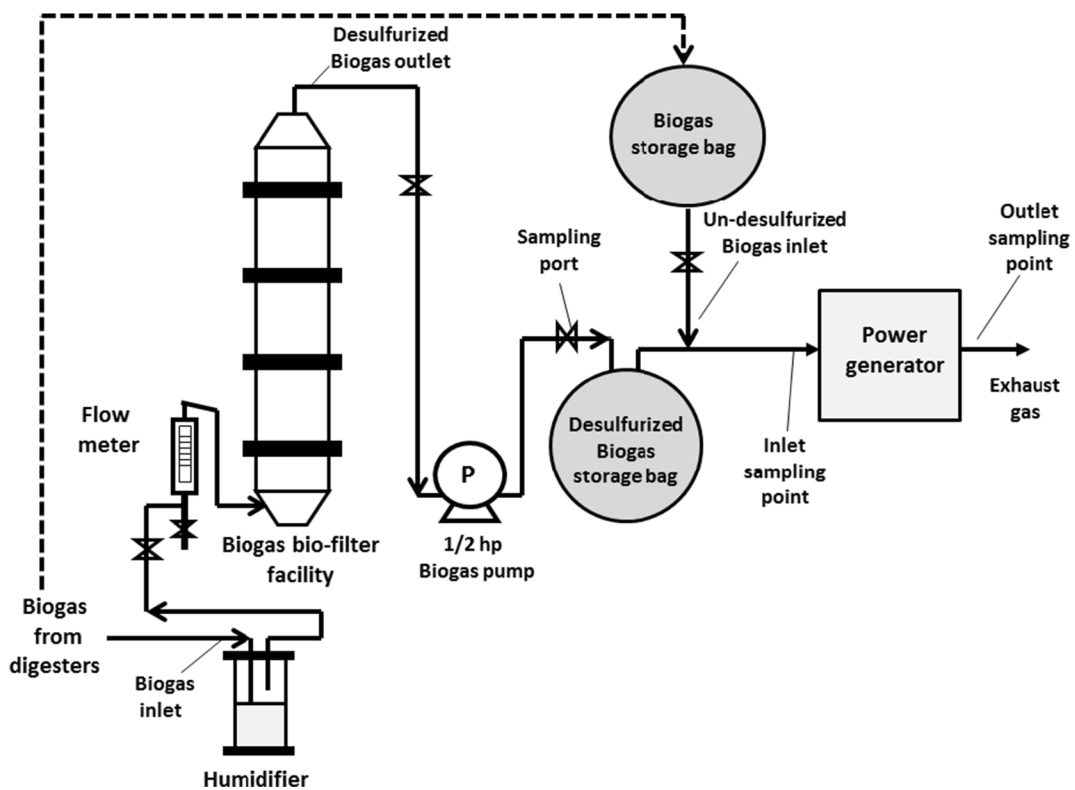


Fig. 1 The sketch of the experimental design for biogas bio-filter facility and power generator

Table 2 Detection of H₂S and SO₂ in gas samples using un-desulfurized biogas for power generation on Pig farm (I)

Concentration (mg m ⁻³)	Biogas	Exhaust gas	Difference (%)	<i>P</i>	H ₂ S conversion degree (%)
Un-desulfurized biogas data (<i>n</i> =18)					
H ₂ S	5575±862	113±43	-98.0	<0.01	95.4
SO ₂	19.2±5.6	256±113	+92.5	<0.01	
Desulfurized biogas mixture data (<i>n</i> =18)					
H ₂ S	3.3±1.8 ^a	19.8±19.1	+83.3	NS	NA
SO ₂	0.3±0.6 ^a	52.7±53.6	+99.4	<0.01	

Data presented as means±SD

NA not applicable, *n* sample size, NS not significant

^a Using desulfurized biogas mixture (desulfurized/un-desulfurized=4:1; v:v) for power generation

generator, the concentrations of H₂S and SO₂ were 113±43 and 256±113 mg m⁻³ in the exhaust gas of the power generator, respectively. Analytical results of biogas combustion showed that combustion of un-desulfurized biogas exhausted about 92.5 % more SO₂ (*P*<0.01). In the meantime, about 98 % of hydrogen sulfide was removed during the combustion process using un-desulfurized biogas (*P*<0.01). Hydrogen sulfide can easily react with metal parts of the power generator and reduce H₂S concentrations in the exhaust gas. Experimental results showed that 95.4 % of H₂S were converted to SO₂, and most SO₂ seemed to deposit on the surface of the internal combustion engine (Table 2). This study was only focused on monitoring H₂S and SO₂ emission from biogas combustion not including H₂SO₄ in liquid phase (i.e., SO₂+1/2O₂→SO₃; SO₃+H₂O→H₂SO₄); thus, SO₃ was not considered.

The concentrations of H₂S and SO₂ were 3.3±1.8 and 0.3±0.6 mg m⁻³ in the desulfurized biogas, respectively (Table 2). The mixed biogas was resulted from biogas recirculation design to avoid over suction pressure of the biogas vacuum pump and prevent vacuum phenomenon inside the biogas bio-filter facility. When the biogas mixture (desulfurized/un-desulfurized=4:1; v:v) was used for the power generator, the concentrations of H₂S and SO₂ were 19.8±19.1 and 52.7±53.6 mg m⁻³ in exhaust gas of the power generator, respectively. Analytical results of biogas combustion showed that the combustion of desulfurized biogas mixture exhausted about 99.4 % more SO₂ (*P*<0.01). In the meantime, about 83.3 % of hydrogen sulfide was removed

during the combustion process using desulfurized biogas mixture. Because exhaust gas from the power generator was directly detected on the farm under an open environment, data variation detected from the exhaust gas was relatively high.

For Pig farm (II), the concentrations of H₂S and SO₂ were 2510±1,061 and 13.3±7.3 mg m⁻³ in un-desulfurized biogas, respectively. When the un-desulfurized biogas was used for a boiler, the concentrations of H₂S and SO₂ were 93±73.4 and 208±82.4 mg m⁻³ in exhaust gas of the boiler, respectively (Table 3). Analytical results of biogas combustion showed that combustion of un-desulfurized biogas exhausted about 93.6 % more SO₂ (*P*<0.01). In the meantime, about 96.6 % of hydrogen sulfide was removed during the combustion process using un-desulfurized biogas (*P*<0.01). Experimental results showed that 91.7 % of H₂S were converted to SO₂ and most SO₂ seemed to deposit inside the boiler (Table 3).

For Pig farm (III), the concentrations of H₂S and SO₂ were 4562±1,298 and 15.8±9.87 mg m⁻³ in un-desulfurized biogas, respectively. When the un-desulfurized biogas was used for a hot water stove, the concentrations of H₂S and SO₂ were 34.8±19.2 and 51.1±40.3 mg m⁻³ in exhaust gas of the hot water stove, respectively (Table 4). Analytical results of biogas combustion showed that combustion of un-desulfurized biogas exhausted about 69.1 % more SO₂ (*P*<0.05). In the meantime, about 99.2 % of hydrogen sulfide was removed during the combustion process using un-desulfurized biogas (*P*<0.01). Experimental results showed that 90.9 % of H₂S were converted to SO₂ and most SO₂ seemed to deposit on the hot water stove (Table 4).

Table 3 Detection of H₂S and SO₂ in gas samples using un-desulfurized biogas for combustion on Pig farm (II)

Concentration (mg m ⁻³)	Biogas (n=11)	Exhaust gas (n=11)	Difference (%)	P	H ₂ S conversion degree (%)
H ₂ S	2510±1061	93±73.4	-96.6	<0.01	91.7
SO ₂	13.3±7.3	208±82.4	+93.6	<0.01	

Data presented as means±SD

n sample size

Human health concern resulting from H₂S to SO₂ emissions

Animal feeding operations (AFOs) can affect air quality through emissions of gases (NH₃ and H₂S), particulate matter (PM), volatile organic compounds (VOC), hazardous air pollutants, microorganisms, and odor. AFOs also produce gases (CO₂ and CH₄) that are associated with climate change. Most of the concern with possible health effects focuses on NH₃, H₂S, and particulate matter, while major ecological effects are associated with NH₃, particulates, CH₄, and oxides of nitrogen (NO_x) (Copeland 2010).

The US Nebraska Department of Environmental Quality found that more than half of the states have standards for H₂S. Standards vary from 1.05 µg m⁻³ for a yearly average (New York) and 7.5 µg m⁻³ averaged over 24 h (Pennsylvania) to Minnesota’s 75 µg m⁻³ not to be exceeded for one half hour twice per year and measured at the AFO property line (Copeland 2010).

Primary particulate materials (PM) from housed livestock contain a much greater proportion of particles of biological origin and/or activity, usually referred to as bio-aerosols, compared with urban or industrial PM (Cambra-López et al. 2010). Secondary PM is rich in SO₄²⁻, NO₃⁻, and NH₄⁺. Gas-to-particle conversion processes can occur to form secondary inorganic particles, in the presence of certain precursor gases such as NH₃, NO_x, SO₂, and volatile organic compounds (VOCs) (Cambra-López et al. 2010).

Hydrogen sulfide is classified as a highly toxic gas with a threshold limit value for proposed exposure of 15 mg m⁻³ (Hamburg 1989). Its distinctive odor can be noticed at concentrations of less than 1.5 mg m⁻³, but the sense of smell sensitivity varies with the individual. At H₂S <15 mg m⁻³, it irritates the eyes and the respiratory tract. Exposure to moderate H₂S concentrations (15–22.5 mg m⁻³) may result in headache, dizziness, clouded vision, nausea, and vomiting. Concentrations of H₂S greater than 1050 mg m⁻³ are fatal in clinical experience (Hamburg 1989).

The concentrations of H₂S in the un-desulfurized biogas were 5575±862, 2510±1061, and 4562±1298 mg m⁻³ for Pig farms (I) to (III), respectively. The H₂S in un-desulfurized biogas has reached fatal concentration (H₂S >1050 mg m⁻³). Based on the experimental results, the concentrations of H₂S in the exhaust gas after biogas combustion were from 34.8±19.2 to 113±43 mg m⁻³ and that can cause symptoms of headache, dizziness, clouded vision, nausea, and vomiting. Also, frequent H₂S emission can cause corrosion for all metal parts of pig houses and facilities.

There are limits only for SO₂ (primary standard—365 µg m⁻³ and secondary standard—100 µg m⁻³ within 24 h), also considered as biogas combustion products and harmful to our health (Salomon and Silva Lora 2009). Primary standards (0.212 mg m⁻³ of SO₂ for 1 h; 99th percentile of 1-h daily maximum concentrations, averaged over 3 years) by the US EPA provide public health protection, including protecting the health of sensitive populations such as asthmatics, children, and the elderly. Secondary standards (1.41 mg m⁻³ of

Table 4 Detection of H₂S and SO₂ in gas samples using un-desulfurized biogas for combustion on Pig farm (III)

Concentration (mg m ⁻³)	Biogas (n=10)	Exhaust gas (n=10)	Difference (%)	P	H ₂ S conversion degree (%)
H ₂ S	4562±1298	34.8±19.2	-99.2	<0.01	90.9
SO ₂	15.8±9.87	51.1±40.3	+69.1	<0.05	

Data presented as means±SD

n sample size

SO₂ for 3 h; not to be exceeded more than once per year) by the USEPA provide public welfare protection, including protection against decreased visibility and damage to animals, crops, vegetation, and buildings (URL: <http://www.epa.gov/air/criteria.html>).

Sulfur dioxide is classified as an irritant gas, and it is generally present in the atmosphere of industrial and urban centers at 2.6–2620 µg m⁻³. Symptoms of upper respiratory irritation have been reported at levels of 5.24–13.1 mg m⁻³, and concentrations above 52.4 mg m⁻³ have noticeable irritant, choking, and sneeze-inducing effects (Hamburg 1989). Based on the experimental results, the concentrations of SO₂ in the exhaust gas after biogas combustion were from 51.1±40.3 to 256±113 mg m⁻³ and that can cause symptoms of noticeable irritant, choking, and sneeze-inducing effects. Also, all SO₂ emission concentrations of this study were much higher than all USEPA's SO₂ standards for allowance emission.

Control of H₂S and SO₂ emissions from livestock biogas combustion

Hydrogen sulfide is oxidized into sulfur dioxide which dissolves as sulfuric acid; extremely acidic electrolytes dissolve metals rapidly and accelerate the corrosion process (Martin 2008). Combustion of 1 L of methane (CH₄+O₂→CO₂+2H₂O) can produce 1.4 mL of liquid water and 0.9 L of carbon dioxide under standard conditions. The water produced is an electrolyte, and the oxygen is the oxidizing agent needed for corrosion. The carbon dioxide speeds up the corrosion by making the electrolytic solution more acid which speeds up the dissolution of the metal into ions (Martin 2008).

Thus, hydrogen sulfide can be oxidized to SO₂ and deposited inside the engines of power generators or on the surface of burner heads of boilers or stoves. Experimental results indicated that biogas must be desulfurized before any applications for either power generation or direct combustion to avoid sulfur dioxide emission and acid rain formation. A novel biogas bio-filter facility was developed and extended to pig farmers by our research team in Taiwan from 2009. More than 93 % of H₂S can be removed and methane content remains above 60 % (v v⁻¹) in desulfurized biogas by biogas bio-filter facility under ambient temperature and limited oxygen conditions (Su et al. 2008, 2013, 2014).

Since the biogas vacuum pump was not ensured to be completely airtight, some air could be introduced to the biogas stream during the experimental periods. Thus, airtight pumps will be recommended in the future. The initial design of the humidifier was trying to keep biogas humidified prior to bio-filter facility; however, the humidifier is now used to collect condensed water from raw biogas (i.e., remove humidity from raw biogas) (Fig. 1). Biogas desulfurization is essential to prevent corrosion and reduce mechanical maintenance cost of power generators or boilers.

Hydrogen sulfide can be removed from biogas either by non-microbial or microbial processes. Non-microbial processes include either dry oxidation (e.g., iron oxide pellets) or liquid phase oxidation (e.g., liquid scrubber) processes (Kapdi et al. 2005; Petersson and Wellinger 2009), while microbial processes use certain bacteria immobilized in bioreactors to remove H₂S from gas streams (Potivichayanon et al. 2005; Syed et al. 2006; Kantachote et al. 2008; Song et al. 2013). A novel bio-filter is a three-phase bioreactor (gas, liquid, and bio-carriers) made with a highly porous filter bed with buffer capability, nutrient availability, and moisture retention capability to promote the growth of sulfur oxidizing bacteria, which is successfully applied to commercial pig farms (Su et al. 2008, 2013, 2014).

There are five groups of technical SO₂ emission control options as follows (Cofala and Syri 1998): (1) use of low sulfur fuels (e.g., fuel desulfurization); (2) in-furnace control of SO₂ emissions (e.g., through limestone injection or with fluidized bed combustion) (with emission reduction between 40 and 80 % at relatively low costs); (3) conventional wet flue gas desulfurization (WFGD) processes (with emission reduction between 85 and 99 % at moderate costs); (4) advanced, high efficiency methods for capturing sulfur from flue gas (with emission reduction up to 99 % at relatively high costs); and (5) measures to control process emissions.

No hydrogen sulfide and sulfur dioxide regulatory emission limits are set for livestock farming in Taiwan up to date. Biogas produced from pig farms is released to ambient atmospheres in the summer. Some un-desulfurized biogas is used for heating lamps for piglets in the winter. Combustion of un-desulfurized biogas can produce sulfur dioxide and can cause air pollution problems. More than 96 % of hydrogen sulfide in biogas was reduced from three commercial pig farms for power generation or direct combustion using boilers and hot

water stoves (Tables 2 to 4). Thus, the best way to control hydrogen sulfide and sulfur dioxide emission is to lower the hydrogen sulfide amount in livestock biogas before any forms of biogas combustion.

Conclusions

Sulfur dioxide emission resulting from H₂S oxidation was detected from exhaust gas samples of power generators or boilers on pig farms when un-desulfurized livestock biogas was applied. Experimental results of this study implied that some SO₂ produced from biogas combustion could deposit on the surfaces of power generator engines or metal parts of boilers. However, some other SO₂ was emitted to the atmosphere and pollute the air of our living areas. Thus, regulations related to H₂S and SO₂ allowance emission must be established in Taiwan in order to manage air quality resulting from livestock biogas combustion. Livestock biogas is the most efficient renewable energy produced from livestock wastes. It is essential to set up H₂S and SO₂ emission standards according to human health while using livestock biogas as a new energy in the near future.

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