Gases emissions estimation and analysis by using carbon dioxide balance method in natural-ventilated dairy cow barns

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Abstract: Contaminated gases emissions from livestock industry are becoming one of the most significant contributors to the increasingly serious environmental pollution. To find a way to reduce gases emissions, it is essential to reveal the factors that can affect the gases emissions. In this study, the concentrations of typical gases (including ammonia (NH₃), carbon dioxide (CO₂), hydrogen sulfide (H₂S), and sulfur dioxide (SO₂)) generated from naturally-ventilated dairy cow barns were detected through the sample-data method in Tianjin, northern China. Indoor environmental conditions, such as temperature (T) and relative humidity (RH), were measured simultaneously. After applying the carbon dioxide mass balance method, ammonia, hydrogen sulfide and sulfur dioxide emissions were determined. The correlation analysis and regression analysis between the climate condition and gas emissions were conducted to assess the data collected in dairy cow barns during the whole study period. There was a significant relationship between environmental conditions and gas emissions. NH₃, H₂S and SO₂ emissions from the building are in the range of 0.98-2.36 g/LU h, 0-0.034 g/LU h, and 0-0.069 g/LU h, respectively. The numerical analysis shows that the NH₃ emission is highly correlated with the temperature and relative humidity. The ventilation rate shows a positive correlation with all the three gases.

Keywords: gas emissions, environmental conditions, correlation analysis, regression analysis, carbon dioxide, H_2S emission, NH_3 emission, SO_2 emission

DOI: 10.25165/j.ijabe.20201302.4802

Citation: Zou B, Shi Z X, Du S H. Gases emissions estimation and analysis by using carbon dioxide balance method in natural-ventilated dairy cow barns. Int J Agric & Biol Eng, 2020; 13(2): 41–47.

1 Introduction

The thermal environment and gases emissions from the dairy cow houses not only directly affect the production performance of dairy cows, but also have an impact on the global ecological environment. The main factors that affect the performance of dairy cows are genetics, physiology, nutrition, and environment^[1]. When the genetic factors and feeding conditions are fixed, the production performance depends on the environment. Generally, the greatest influence from ambient environment on dairy cows is the temperature, relative humidity, and the concentration of ammonia (NH₃), carbon dioxide (CO₂), hydrogen sulfide (H₂S) in the cow house. Dairy cows are homoeothermic animals and need to maintain a constant body temperature^[2]. They are sensitive to factors, which influence their thermal exchange with the environment. These factors include air temperature, radiant temperature, air velocity, and relative humidity^[3].

When environmental conditions exceed a threshold that increases the core-body temperature, heat stress occurs^[4]. Heat stress has a negative effect on the production performance of dairy cows. It can decrease the milk production and reproductive success^[5-10]. In addition, it can reduce the comfort level of dairy

cows and has a negative stimulus, such as hunger, thirst, and frustration^[6, 11-13]. The gases from the dairy barns are harmful to the cows and human health and consequently have a negative impact on the milk production. Usually, most gases in cattle houses include NH₃, CO₂, H₂S, etc. Ammonia in cattle houses is mainly derived from cow's feces and urine, spoilage of forage and so on. Ammonia is a kind of poisonous gas, which not only reduces the growth and productivity of dairy cow^[14], but also increases the mortality^[15]. Livestock NH₃ emissions are the most important source of global NH3 emissions, accounting for 39% of NH₃ emissions worldwide^[16], and NH₃ is considered to be the most harmful gas to affect the air quality^[17]. CO₂ generated from a cowshed is primarily derived from enteric fermentation and respiration^[18]. CO₂ is a kind of greenhouse gas^[19] and the emissions of CO2 can affect to the undergoing significant warming of global climate $^{\left[20\right] }.$ $H_{2}S$ from the dairy barns is produced by anaerobic bacterial decomposition of protein or other sulfur-containing organic matter^[21]. It is toxic and associated with numerous animal and human mortalities in livestock facilities^[22]. SO₂ is an oxidation product of sulfur containing gases^[23], and can be evaporated into the atmosphere where it combines with vapor to form sulphur acid and nitric acid^[24].

Since the performance of dairy cows is associated with the environment factors, and appropriate actions can protect animals from heat stress^[25], researches have been conducted to test the relationship. In 2007, air emission data was collected continuously from five dairy farms over a period of 2 years, including concentrations of particulate matter (PM), ammonia (NH₃), hydrogen sulfide (H₂S), and non-methane volatile organic compounds (NMVOC) from five dairy barns is detected by Heber^[26-28]. The concentrations of CH₄, N₂O, NH₃, and CO₂ from

Received date: 2018-11-25 Accepted date: 2020-01-15

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a naturally-ventilated dairy barn were measured with a multi-location method by Ngwabie^[29]. Saha et al.^[30] carried out an identical experimental set-up about the seasonal and diel variations of ammonia (NH₃) and methane (CH₄) emissions to cover three seasons of winter, spring, and summer, and two autumn seasons in 2010, 2011, and 2012. An 18 daily measurement was conducted in six livestock buildings for dairy cows located in Wielkopolska Voivodship to determine the values of greenhouse gases (CH₄ and N₂O) and NH₃ emission factors^[31]. Wang^[32] measured barn NH₃ emission factors per animal unit (20.6-21.2 g/d AU) based on two passive samplers. In summary, the previous studies are mostly in USA and Europe. This research focuses on the measurements of temperature, relative humidity, and the concentrations of NH₃, CO₂, H₂S and SO₂ generated from the naturally ventilated dairy barns in North China. By applying the carbon dioxide balance method, the gases emissions are calculated and the relationship between gases emissions and the temperature and humidity are revealed.

2 Materials and methods

2.1 Experimental site

The experimental sites are selected in a farm with 3132 dairy cows, which is located in Tianjin in North China. Gas concentration monitoring is conducted in two of them. The barn is naturally ventilated. The doors on the two sides of the barns are open in the experimental period to increase the ventilation rate. The experimental barn is 132 m long and 30 m wide and housed 343 cows as shown in Figure 1. The width of center channel is 5 m. The width of feeding channel is 4 m. The width of cow's bed is 5 m. The width of dung manholes near the wall is 3.5 meters.

All the cows in the barns are adult cows, including lactating cows, pregnant cows, and postpartum cows. The calves are separately raised in calf islands. The milking parlor is separate with the barns. It is divided into four sections: A-D, as shown in Figure 1. Section A is the obstetric bed. Section B is used to hold the prenatal dairy cows. Section C is used to hold the postpartum prenatal dairy cows. The data sensors are installed in Section B and Section C. Section D is the veterinarian studio where the experimental data collection control center is set up.

The manure in the barns is scraped using scraping manure board by the cleaning truck three times a day, which occurs at 8:00 am, 1:30 pm, and 6 p.m. During the measurement period, there are 190-210 dairy cows with an estimated average body weight of 650-750 kg in the barn. Daily activities in the barn include milking and feeding for three times per day. The first cycle starts with milking at 6:50 am followed by feeding. A second cycle started at 2:20 pm with milking and feeding. The third milking is at 10:20 pm.

Dung manholes	Dung manholes		
A Cow's bed	B Cow's bed		
Feeding channel	Feeding channel		
Ce	nter channel		
	Feeding channel		
D veterinarian studio	C Cow's bed		
	Dung manholes		

Figure 1 Layout of the dairy cattle building

2.2 Dairy cows feeding

The feeding of dairy cows includes concentrated feed, rough feed, and excipients feed. The concentrated feed includes cottonseeds, milk powder, soybean, rapeseed, corn protein flour, etc. Rough feed includes silage corn, fresh yarrow, wheat straw, field hay, etc. Excipients feed includes beer grains, pomace, beetroot, etc.

2.3 Instrumental setup

The concentrations of NH₃, SO₂, and H₂S in the air of the cattle building are measured using electrochemical sensors MOT300 series sensors, which are produced by Korno Electronic Technology Co., Ltd, Shenzhen, China. The measurement range of the sensors is 0-196 mg/m³. The measuring accuracy for the sensors according to data sheets from the manufacturer is $\pm 3\%$. The output signals are 4-20 mA. The concentrations of CO₂ in the air of the cattle building are measured using an infrared detection gas sensor MOT300-CO₂-IR. The measurement range of the sensors is 0-3928 mg/m³. The detection threshold of the gas is 1.96 mg/m³. The measuring accuracy for the sensors according to data sheets from the manufacturer is $\pm 3\%$. The output signals are 4-20 mA.

The temperature and relative humidity of the air of the cattle building are measured using QFA3171. The QFA3171 is used in ventilation and air conditioning buildings where high accuracy and short response times for measuring relative humidity are required. The measuring range covers the entire humidity range of 0-100% and the temperature range of $-40 \,\text{C}$ to $+70 \,\text{C}$. The instruments are placed on a 2.5 m high crossbeam in the center of the buildings. All the gas detectors gather gas concentrations with the natural diffusion detection method, which can decrease the impacts of tube material to gases concentrations compared with the pump suction detection method.

2.4 Data acquisition

Measurements are carried out during the hot weather period from July 2017 to September 2017 in the dairy cow buildings. Gas concentrations, temperature, and relative humidity of the air are measured at 1 min intervals throughout the sampling period for 24 h a day continuously. Data is transferred between sensors and host machine via Modbus TCP protocol with an Ethernet I/O module. This module is a web-based Ethernet I/O module Block. There are eight input analog signals and four output digital signals in the module which allow three gases detection and temperature and humidity detection simultaneously. Then the data is logged in the MySQL database.

2.5 Ventilation and emission rate numerical aspects

Since sensors have detected the concentrations of gases, the emissions of gases can be figured out as long as the value of ventilation rate (VR) is calculated. The ventilation rate of a building can be determined by calculating the mass balance of CO_2 flow. The ventilation rate in dairy cow barns is calculated by the CO_2 mass balance method^[33], as presented in Equation (1).

$$V_{\rm CO_2} = \frac{C_{pro} \cdot A_{\rm CO_2}}{(C_{cin} - C_{cout}) \times 10^{-6}}$$
(1)

where, V_{CO_2} is the ventilation flow in m³/h from CO₂ balance; C_{pro} is the production of CO₂ in m³/h on a 24 h basis; A_{CO_2} is a coefficient for the diurnal adjustment of the CO₂ production due to animal activity; and C_{cin} and C_{out} are the CO₂ concentrations in the indoor and outdoor air in ppm. In this study, a value of 0.185 m³/h hpu (where 1 hpu = 1000 W of total heat produced by the animals at 20 °C) for the total CO₂ production in the barn is

selected for the CO₂ balance^[34]. The emission rates of NH₃, CO₂, H₂S, and SO₂ are calculated using V_{CO_2} in Equation (1) and the enhanced concentrations in the dairy barn as shown in Equation (2):

$$E = V_{\rm CO_2} \left(C_{in} - C_{out} \right) \tag{2}$$

where, *E* is the emission rate, mg/h; V_{CO_2} is the ventilation rate in m^{3/}h on a 24 h basis; C_{in} and C_{out} are the gases concentrations inside and outside the dairy barn, respectively, mg/m³.

3 Results and discussion

As the dairy barn used in the experiment is a naturally ventilated cowshed, the gases concentrations and temperature and humidity values inside the cowshed are closely related to the environment in the surrounding atmosphere. We collected the average temperature and humidity values inside and outside the dairy barns, as shown in Table 1. The wind speed data is obtained from the China meteorological data network.

 Table 1
 Daily average environmental conditions for representative days

Date	Wind/m s ⁻¹ –	Tempe	rature/°C	Relative h	Relative humidity/%	
		Indoor	Outdoor	Indoor	Outdoor	
7.1	2.68	28.6	24.67	56.98	73.67	
7.2	1.79	32.5	27.45	42.23	60.67	
7.3	2.23	33.1	26.13	52.57	76	
7.4	2.23	30.8	25.39	59.92	74.25	
7.5	1.79	28.5	25	63.48	70.25	
7.6	1.79	32.6	26.86	47.53	66.75	
7.7	2.23	31	27.28	65.05	72.21	
7.8	2.68	29.6	27.54	67.07	78.46	

3.1 Gases concentrations

Daily variations were observed for all gases with higher concentrations of NH_3 , CO_2 , and H_2S inside the dairy barn than in the outdoor air (Table 2). The indoor concentrations were typically 2-3 times as high as the outdoor concentrations (Table 2). The indoor concentrations of NH_3 and CO_2 show higher concentrations than the outdoor concentrations, but there is no obvious linear relationship. An exception was SO_2 where outdoor concentrations of the results of research, the most representative days were chosen for the needs of statistical analysis. These days are characteristics for their concentration of gases of the year.

 Table 2
 Gases concentrations average values inside and outside of the barns

Date -	NH ₃ /ppm		CO ₂ /ppm		SO ₂ /ppm		H ₂ S/ppm	
	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
7.1	4.58	1.52	1360.92	423.67	0.007	0.005	0.081	0.029
7.2	5.16	1.98	1395.99	430.41	0.009	0.006	0.096	0.016
7.3	5.24	2.01	1374	476.32	0.004	0.005	0.074	0.015
7.4	4.91	1.77	1390.21	459.31	0.003	0.006	0.072	0.008
7.5	4.87	1.75	1410.03	507	0.006	0.007	0.068	0.012
7.6	5.65	2.37	1390.19	466.79	0.011	0.01	0.067	0.01
7.7	4.63	1.74	1368.28	436.5	0.008	0.006	0.082	0.026
7.8	4.19	1.31	1401.53	456.34	0.007	0.004	0.048	0.008

In the period of measuring, the one-day indoor NH_3 concentration in the dairy cow barns was in the range of 0.87-7.5 ppm. In order to analyze the factors related to ammonia concentration in a day, graphs of concentration changing with time are shown in Figure 2. The concentration profile of NH_3 with significantly visible peaks is shown in Figure 2. The peaks occur

in the morning at 9:50, 14:00 and in the evening at 18:00, which corresponds to the manure cleaning time. The barn uses the scraping manure board to clean the manure. The NH₃ concentrations could increase by 70% to 75%^[35]. In addition, there are smaller peaks, which are shown at about 7:00 am, 2:30 pm, and 22:10 pm, which correspond to the feeding and milking time. During the feeding and milking time, there are increased dairy cow activities including walking around, urination, defecation and mixing of the manure on the floor, which increase the NH₃ concentration. During the nighttime, from midnight to 6:00 in the morning, the ammonia concentrations show the continuous downward trend. Most of the dairy cows sleep occurs at night when the barn is least disruptive, which means less activities, such as feeding, urination, defecation, so the ammonia concentration is less during that time period.



Figure 2 Variation of NH₃ concentration inside the barn during a representative day

The main source of CO_2 emission from dairy barns is the animal respiration, with smaller contributions from microbial respiration in manure. Higher air temperature and surface air velocity can significantly enhance the CO_2 emissions from the manure^[36]. There is a gentle rising peak of CO_2 concentration around 9:00 am, as shown in Figure 3, which is related to the feeding time. It means more metabolic activities and more CO_2 emission. At the other time of the day, the concentration curve shows a smooth trend which is related to the ventilation rate in the building.



Figure 3 Variation of CO₂ concentration inside the barn during a representative day

 H_2S is the major sulfur compound emitted from livestock production^[37]. In dairy farms, sulfur excretion mainly comes from animal feces and urine. H_2S is formed by intermediate sulfur containing compounds originating from organic compounds containing sulfur break down^[38]. As manure decomposes anaerobically, some part of sulfur is used as an electron acceptor to form sulfide compounds, such as sulfur dioxide. In general, the concentrations of hydrogen sulfide and sulfur dioxide are related with the manure amount and decompositions in the barn. During the research period, the highest levels for hydrogen sulfide and sulfur dioxide are measured during the morning when there is little air movement inside the dairy barn, the minimum and maximum indoor hydrogen sulfide and sulfur dioxide concentrations

measured for the entire sampling period is as follows: 0-0.465 ppm H_2S , 0-0.219 ppm SO₂.

3.2 Temperature and relative humidity measurement

Dairy cows are sensitive to high temperature and humidity in summer. Thermal weather directly affects feed intake thereby, reduces growth rate, milk yield, reproductive performance, and even death in extreme cases^[39]. Temperature Humidity Index (THI) is a measure to o estimate impact of thermal weather conditions of dairy cows^[40]. The temperature and humidity index (THI) can be calculated by the following formula^[41]:

 $THI = 1.8T + 32 - (0.55 - 5.5 \times 10^{-3}RH) \times (1.8T - 26)$ (3) where, *T* is dry bulb temperature, °C; *RH* is relative humidity, %.

Average hourly temperature, relative humidity and THI values in a day are shown in Figure 4. Throughout the whole measurement period, the indoor temperature varies from $21.18 \,^{\circ}$ C to $35.63 \,^{\circ}$ C while relative humidity ranges from 50% to 87.36%. Despite there is a large variation in relative humidity, the indoor temperature remains little interval. THI shows the same trends as the temperature data is observed.



Figure 4 Diurnal variation in indoor environmental conditions during study period

3.3 Relationship of gases concentration and environmental conditions

The production of polluting gases is affected by airflow, temperature, and humidity inside the barn which strongly depend on the continuously changing weather conditions^[42]. To find the relationship among the temperature and relative humidity and gases concentration, the data is normalized and displayed in Figure 5. Complementary to the former insight that high temperature increases emissions, we find that ammonia concentration increases a rise in temperature, while the opposite situation occurs under relative humidity. The influence of temperature on ammonia concentration is mainly through the influence of urease activity in excrement and urine^[43]. It is found that urease activity increases with the increase of temperature. Therefore, the increase of temperature in dairy barns will lead to the increase of urease activity, urea decomposition and ammonia emission. In general, higher temperature leads to a higher amount of gaseous NH₃^[44], which is manifested by the increase of ammonia concentration. When the relative humidity of the air is high which implies there is more water in the air, the amount of gaseous ammonia decreases due to the fact that ammonia is easily dissolved in water^[45]. As shown in Figure 5, there is no significant relationship between temperature and carbon dioxide concentration. However, the relative humidity (RH) has a positive effect on carbon dioxide Typically, high relative humidity means concentration. insufficient ventilation, which can explain the high carbon dioxide Ammonia concentration and carbon dioxide concentration. concentration show the same trend during some certain time intervals is observed. Low levels of hydrogen sulfide and sulfur dioxide is detected during the study suggested that measurement methods and instructions with higher accuracy may give better estimates of H_2S and SO_2 concentration.



Figure 5 Diurnal variation in indoor environmental conditions during study period

3.4 Measurements of ventilation and gases emissions rates

In the present study, according to Equation (1), the calculated ventilation rate (VR) is in the range of 343-590 m³/LU h and the mean VR is 503 m³/LU h with measuring periods of one minute. According to Equation (2), the NH₃, SO₂, and H₂S emissions are calculated in Table III. The emission profiles of NH₃, SO₂ and H₂S emission with significantly visible emission are shown in Figure 6. Due to lower amounts of manure in the building and less animal activities, the lower NH₃ emission during the housing period occurs usually at night. Variations of the SO₂ emission are smaller as compared with the H₂S emission.



Figure 6 Diurnal variations in NH₃, SO₂, H₂S emission from the dairy cattle building during a period

 Table 3
 Gases emission values during the measuring period

$(\mathbf{g} \mathbf{L} \mathbf{U}^{-1} \mathbf{h}^{-1})$					
	ỹ	SD	Max.	Min	
NH ₃ emission	1.52	2658.26	2.36	0.98	
H ₂ S emission	0.001	22.4765	0.069	0	
SO ₂ emission	0.002	10.975	0.034	0	

Note: \tilde{y} , Mean; SD, standard deviation; Max, maximum; Min, minimum; 1 LU =500 kg animal weight.

3.5 Correlation analyzes

Aiming to examine the impact of the indoor climate conditions on NH₃, SO₂, H₂S emissions, correlation analysis about emissions of gases related to temperature, relative humidity values, and THI and ventilation rate is carried out in the research. In addition, regression analyzes are applied on NH₃, SO₂, H₂S emissions and indoor climatic conditions.

The correlation coefficient and p-value between gases emissions and climatic conditions are presented in Table 5. According to correlation analysis, the correlation coefficient shows a positive relationship between temperature and NH₃, SO₂, H₂S emissions (Table 5), while this relationship to SO₂ is quite weak (r=0.084). Relative humidity has a week positive effect on SO₂ and H₂S emissions, however showed a negative impact on NH₃ emission (r=-0.478).

There is a negative correlation between NH₃, SO₂, H₂S



emissions and ventilation rate, and the correlation between SO_2 emissions and ventilation rate shows the most significant correlation (r = 0542). Because of condition with insufficient ventilation, gases are accumulated inside the dairy barn, which leads to higher gases concentrations. Temperature Humidity Index (THI) typically accounts for the combined effects of environmental temperature and relative humidity. There are no any researches about the impact of THI on the gases emissions. Based on our research findings, it shows some correlation statistics between THI and gases emissions (Table 4).

 Table 4 Correlation coefficient r and p-value between gases emissions and climatic conditions

		T/°C	<i>RH</i> /%	THI	VR
NH ₃ emission	r	0.331172	-0.4777	0.208444	-0.368749
	<i>p</i> -value	0.0003	< 0.0001	0.0017	< 0.0001
H ₂ S emission	r	0.363324	0.057217	0.362277	-0.388716
	p-value	< 0.0001	0.439171	< 0.0001	< 0.0001
SO ₂ emission	r	0.084	0.018141	0.121881	-0.541697
	p-value	0.282	0.817655	0.120018	< 0.0001

3.6 Regression analyzes

According to the results of correlation analyzes, the significant factors are analyzed with the linear regression analysis, as shown in Figure 7.



Figure 7 Regression analysis between gases emissions and temperature, relative humidity and ventilation rate

According to the regression analysis, the temperature dependency of the NH_3 , SO_2 , H_2S emissions rates of both gases is significantly different. The NH_3 emission increases with temperature with the linear regression coefficient of 0.583. The impact of temperature presents a trigonometric correlation on the H_2S emission. In contrast, no correlation is found between temperature and SO_2 emission. Relative humidity correlates negatively with NH_3 emission with the regression coefficient 0.536. However, no significant correlation is observed between relative humidity and the emissions of SO_2 and H_2S . Ventilation rate shows a positive correlation with all the three gases, but all the linear correlation coefficients are weak.

4 Conclusions

In this study, the concentrations of NH₃, SO₂, H₂S, CO₂ and environmental conditions were measured in a dairy cow barn with frequent removal of manure. The ventilation rate was calculated based on CO₂ mass balance method and the emissions of NH₃, SO₂, and H2S are subsequently determined. The following conclusions could be drawn: the concentrations of NH₃, SO₂, H₂S, and CO₂ varied considerably in time inside the naturally ventilated barn. Low concentrations of SO₂ and H₂S were measured, suggesting that dairy barns with frequent manure removal did not constitute a major source of sulfur compounds. NH₃, SO₂, and H₂S emissions from the building were respectively in the range of 0.98-2.36 g/LU h, 0-0.034 g/LU h, 0-0.069 g/LU h. NH3 emission is highly correlated with temperature and relative humidity. Ventilation rate shows a positive weak correlation with all the three gases. Not all the observed correlation coefficients are high during the linear correlation and regression analysis, a linear mixed model or an ARIMA model might be a better choice to improve the accuracy of the model.

Acknowledgements

This work was supported in part by China Agriculture Research System under Grant CARS-36.

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