Bull Environ Contam Toxicol (2010) 85:279–286
DOI 10.1007/s00128-010-0085-y

Fluxes and Influencing Factors of Ammonia Emission from Monosodium Glutamate Production in Shenyang, China

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Received: 6 January 2009 / Accepted: 9 July 2010 / Published online: 27 July 2010

Abstract In order to solve environmental problems resulting from ammonia released into the atmosphere, the emission of ammonia contamination from monosodium glutamate (MSG) production was quantitatively observed, and the relationships with relevant influencing factors (ammonium-N, nitrate/nitrite-N and pH) were statistically analyzed. The results indicated that the release of gaseous ammonia from MSG production including the treatment and discharge of wastewater was highly dependent on the technical processes utilised. The flux of ammonia released from the fermentation workshop was highest, up to 8.98×10^5 mg m^{-3} min^{-1}, and the flux from the sugar-refining workshop was lowest, only 85.1 mg m^{-3} min^{-1}. The release of gaseous ammonia during the whole MSG production was significantly proportional to the concentration of ammonium-N in the discharged solution, and exponentially proportional to the concentrations of nitrate–nitrogen and nitrite-nitrogen in the discharged solution. Although there was no linear relationship between the flux of ammonia released during the whole MSG production and pH values in the discharged solution, pH was significantly related to the flux of ammonia released during the treatment and discharge of wastewater.

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Keywords Ammonia emission · Air pollution · Nitrogen cycle · Monosodium glutamate production

In the natural environment, normal release of ammonia (NH_3) is an important component in biogeochemical cycles of nitrogen (Zhou and Huang 2001). However, there is an increasing emission of ammonia with the development of industrial and agricultural production in recent years (Prasertsak et al. 2001; Perrino and Catrambone 2004). Due to increasing release of gaseous ammonia, some original biogeochemical modes and natural ecological balances related to nitrogen cycling are being disrupted (Hyde et al. 2003; Jung et al. 2004; Thompson and Meisinger 2005). Particularly, an increase in ammonia in the atmosphere is potentially promoting induction of environmental degradation and ecological damage (Prasertsak et al. 2001). Land deterioration, water eutrophication and formation of toxic minute particulates and aerosols are indirectly and/or directly involved in elevating ammonia levels in the environment (Zhou 2001; Shostell and Bukaveckas 2004).

It cannot be denied that production of monosodium glutamate (MSG) is becoming an important contributor of ammonia contamination in the atmosphere. As early as the beginning of the twentieth century, MSG was introduced as a condiment in human foods. Nowadays the production of MSG and demand for MSG is still being expanded with the improvement in people's levels of living (Fuke and Shimizu 1993; Zhou et al. 2004). China has become a major producer and consumer of MSG. The annual output of MSG in this country is up to half of the entire world's production. With the rapid development in the MSG industry, more and more wastewater and ammonia is being discharged into the environment. Due to the high chemical oxygen demand (COD), organic pollutants and

ammonium-N, as well as low pH, wastewater from the MSG industry is usually difficult to be treated effectively. In the past decade, more and more attention has been paid to water pollution problems resulting from MSG wastewater, and development and improvement of its treatment technology (Hellinga et al. 1998; Schmidt and Sliekers 2003). However, the contribution of ammonia contamination from the MSG industry as an important environmental problem has been neglected. Thus, to quantify the flux of ammonia released from the MSG industry and to discuss influencing factors related to ammonia release from the MSG production are of practical significance (Theobald et al. 2005). The purpose of this study was to provide a scientific basis for the control of ammonia contamination in the atmosphere and the recovery of the environment (Zhou et al. 2004; Zhou and Hua 2004).

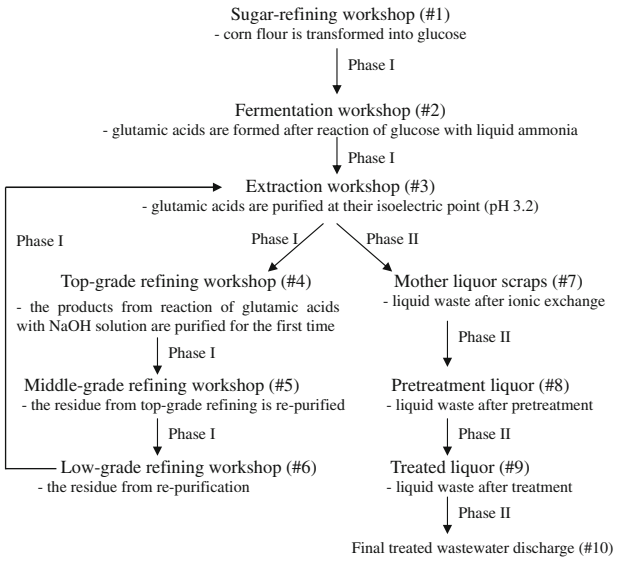

Fig. 1 Working procedures of MSG production and sampling sites in the study

Materials and Methods

A big MSG factory in the west suburb of Shenyang as the most important old industrial base in China was chosen as the observed locality for this study. The geographical location of this factory built in 1939 is 41°82′N, 123°38′E. The average annual output of MSG from the factory has increased from about 2.0×10^6 kg when MSG production began up to 5.0×10^7 kg today.

The whole MSG production process related to the release of gaseous ammonia includes the synthesis of MSG (Phase I) and the treatment and discharge of wastewater (Phase II). Each technical/working process of MSG production was briefly described in Fig. 1. As shown in Fig. 1, sugar-refining, fermentation, and extraction, as well as top-grade, middle-grade and low-grade MSG refining processes are involved in Phase I and mother liquor scraps, pretreatment liquor, treated liquor and final treated wastewater discharge are involved in Phase II. Phase I is associated with the release of mother liquor scraps, pretreatment liquor and treated liquor in Phase II. In accordance with the MSG production processes, 10 sampling sites (Fig. 1) were selected.

Gaseous samples for the determination of ammonia levels were collected by an air-sampling apparatus (mode of TMP-1500 model, made in Jiangsu Province, China). In Fig. 1, each of the 10 sections is independently force-air ventilated. There is only one outlet for air from each unit used for sampling. After the sugar-refining process, formed glucose could be transferred to the fermentation jars when its pH value was adjusted to about 7.0 by NaOH solution, and special bacterium was added, and then anhydrous liquid ammonia was added in order to keep pH at nearly 7.0. After thirty-hour reaction under conditions of 37°C and 0.1 Mpa, glutamic acid was formed. According to the national standard method (GB/T 18204.25 2000) of the People's Republic of China for the determination of ammonia in air, the air-sampling apparatus was sequentially installed above 1.2 m of the distance from liquid outflow from the working procedures and all the wastewater discharge orifices. For example, in the extraction workshop, the apparatus was emplaced above 1.2 m of the distance from the extract solution. Because each workshop or production unit is a relatively close system, the speed and rate of air movement at the each exhaust port sampled is approximatively zero. In order to avoid the heterogeneity of gaseous samples at each sampling site, the sampling time lasted for 45 min in each site and each site was thrice sampled. The velocity of air flow through the apparatus was about 1.0 L min^{-1}. The gaseous samples collected by the air-sampling apparatus were bubbled through 25 ml of dilute sulfuric acid. At the end of a sampling period, sulphuric acid solution used for absorption of ammonia was washed with 40 ml of distilled water and collected for ammonia analysis.

Based on 1 week sampling interval, wastewater samples were sequentially collected at the 10 sites mentioned above (Fig. 1) on May 3, 10, 17 and 24, 2004 and November 6, 13, 20 and 27, 2005. After 5 h precipitation of ferment extractants from the extraction workshop, the supernatant liquid in the jar was collected for analysis. The other sampling sites were set at the outfall of wastewater discharge from the other workshops. The volume of each collected water sample was 5 L. After collection, all the samples were immediately transferred to analytical laboratories and stored at 4°C.

The determination of ammonia was carried out in the Shenyang Station of Environmental Monitoring in 24 h

after the samples were collected. All the sulphuric acid solution samples absorbing ammonia were analyzed for ammonia using the improved Nessler spectrophotometric method (EPA-China 2004, 1989). Using the national standard method (GB/T 18204.25 2000) of the People's Republic of China, the lowest detection limit of ammonia in air is 0.4 mg m^{-3}.

Levels of ammonium-N, nitrate–N and nitrite-N in wastewater were measured using the Nessler spectrophotometric method and the sulfanilamide spectrophotometric method (EPA-China 1989) in 24 h after the water samples were collected. The measurement of pH in wastewater was carried out using the 3-B pH meter in 1 week after the water samples were collected.

The average NH_3 flux was calculated using the following expression:

$$V = M/T \quad (1)$$

Where M is the NH_3 concentration (mg m^{-3}) collected at the time T, T is the sampling time (min), and V is the average NH_3 flux (mg m^{-3} min^{-1}).

All the data from the above analytical experiments were statistically analyzed using Microsoft Excel and Origin 7.0 on a computer. The statistical analyses included calculation of average values, and regression of ammonia release and ammonium-N, nitrate–N + nitrite-N, and pH, respectively. When the significance level (p value) of a regression equation is lower than 0.01, there is a significant correlation relationship; when $p < 0.005$, the regression relationship is very significant (He 2004).

Results and Discussion

All the 8 observations indicated that ammonia was continuously released into the air in every working procedure of the whole MSG process including MSG synthesis, wastewater discharge and treatment. However, there was a major difference in the quantities of released ammonia between various working procedures. In particular, the release of ammonia in the fermentation workshop (sampling site #2) was at the highest, up to 8.98×10^5 mg m^{-3} min^{-1} (Fig. 2a) and as high as 83.7% of the total amount released during the whole MSG process. The high release is responsible for insufficient reaction between aqueous ammonia and glucose, which results in the escape of gaseous ammonia, when superfluous aqueous ammonia is added to fermentation jars after the sugar-refining process in order to form glutamic acids during the synthesis of MSG.

Besides the fermentation workshop (sampling site #2), the quantity of ammonia released from the extraction workshop (sampling site #3) was also high, up to 1.29×10^5 mg m^{-3} min^{-1}, because the high temperature

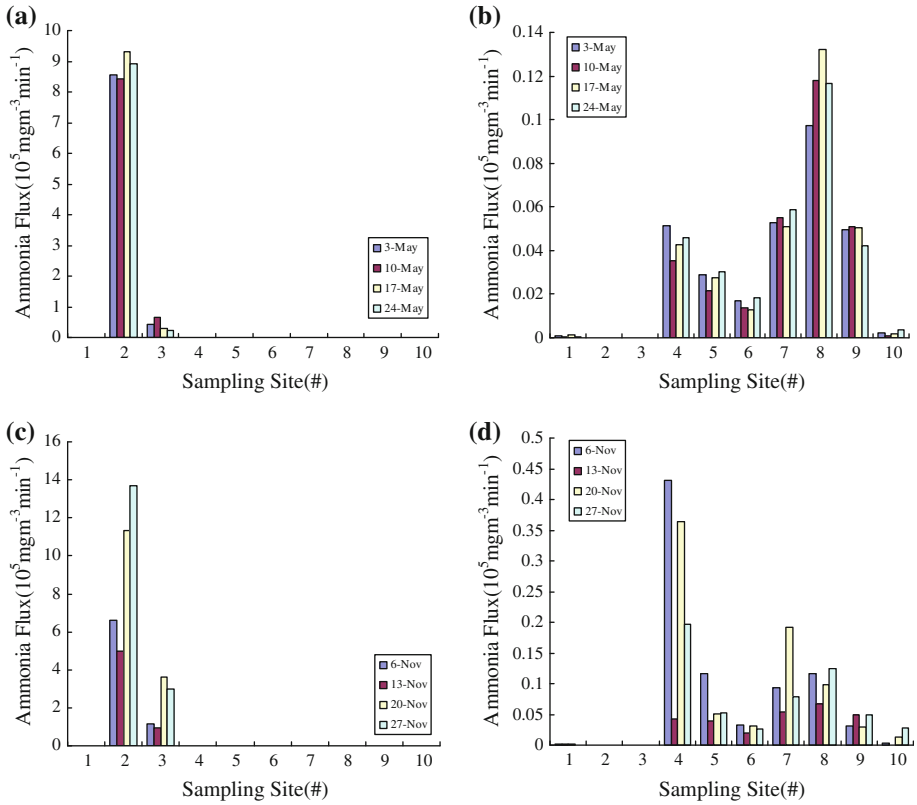

Fig. 2 Average fluxes of ammonia emission in the process of producing MSG: **a** fermentation and extraction in May 2004; **b** other processes except fermentation and extraction in May 2004; **c** fermentation and extraction in November 2005, and **d** other processes except fermentation and extraction in November 2005

(about 38°C) of extractants during the extraction process of MSG can perhaps promote volatilization of gaseous ammonia (Hellinga et al. 1998). However, the release of ammonia from the extraction workshop (sampling site #3) was only 1/7 of that from the fermentation workshop (sampling site #2). The emission of ammonia became less during the other working procedures. The average quantity of ammonia from the top-grade refining workshop was 1.50×10^4 mg m^{-3} min^{-1}, only 1/9 of that from the extraction workshop (sampling site #3). In particular, the average quantity of ammonia from the sugar-refining workshop (sampling site #1) was at the lowest, only 85.1 mg m^{-3} min^{-1} (Fig. 2b), but it was greatly higher than the background level of ammonia in the atmosphere (1.1 mg m^{-3}).

The flux of ammonia released from the treatment and discharge of MSG wastewater (Phase II) was less than that from the synthesis of MSG (Phase I). The release of ammonia from the pretreatment liquor (sampling site #8) was at the highest during Phase II. There were 3 main reasons including (1) reaction of sodium polyacrylate added for wastewater treatment with mother liquor scraps; (2) volatilization of ammonia promoted by aeration wastewater treatment; (3) escaping of ammonia from solution by increasing temperature (up to 40°C) during the pretreatment of wastewater. The average quantity of ammonia released from the treated liquor (sampling site #9) was up to 4.41×10^3 mg m^{-3} min^{-1}, because calcium carbonate added during the wastewater treatment can react with various acids in wastewater, thus promoting the formation of gaseous ammonia. The release of ammonia from the final wastewater discharge outlet was low, only 675.2 mg m^{-3} min^{-1}.

It was indicated in Fig. 3 that the quantity of ammonia released into the atmosphere changed with the sampling time (4 intervals) at the same sampling site. The most obvious difference in the changes of ammonia release with time took place in the fermentation workshop (sampling site #2). The range of ammonia release was 4.99×10^5–1.37×10^6 mg m^{-3} min^{-1}. This is probably due to unstable technical processes of the working procedure. There was also a wide range of ammonia release in the extraction workshop (sampling site #3). As for Phase II, the greatest variation in the release of ammonia happened at the working procedure of pretreatment liquor (sampling site #8), and the quantity of ammonia released from sampling site #8 was 9.72×10^3 mg m^{-3} min^{-1} on May 3, and changed to 1.32×10^4 mg m^{-3} min^{-1} on May 17, 2004. This variation may result from changing aeration and different amount of sodium polyacrylate added during the wastewater pretreatment.

According to all the 8 observations, it can be calculated that the annual quantity of ammonia released from the whole MSG process was up to 5.64×10^5 kg m^{-3}. The

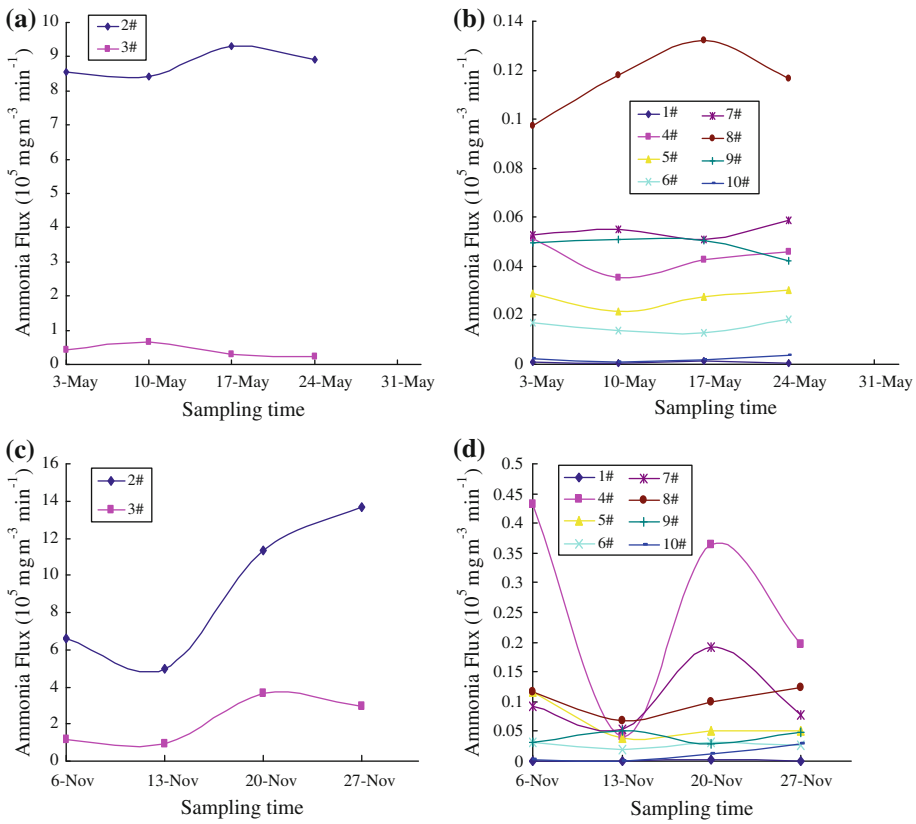

Fig. 3 Dynamical changes in the average flux of ammonia released with time during the MSG production: **a** fermentation and extraction in May 2004; **b** other processes except fermentation and extraction in May 2004; **c** fermentation and extraction in November 2005, and **d** other processes except fermentation and extraction in November 2005

quantity from Phase I was 5.51 × 10^5 kg m^{-3}, accounting for 97.7% of the total ammonia-releasing flux due to the MSG production. The quantity of ammonia released from Phase II was 1.26 × 10^4 kg m^{-3}, only 2.3% of the total.

In theory, the release of ammonia should be proportional to the concentration of ammonium-N in outflow and wastewater from the MSG process because ammonium-N in solution can be easily transformed to gaseous ammonia under certain conditions. By examining the data from the 8 observations, it was indicated that there was a definite linear relationship between the quantity of ammonia released from the MSG process and the concentration of ammonium-N in outflow and wastewater from the MSG production (Fig. 4a). The significant relationship can be expressed by following regression equation:

$$\Phi(NH_3) = 0.0003\left[NH^{4+}\right] - 0.87 \quad (r = 0.702, n = 80, p < 0.005) \tag{2}$$

Where $\Phi(NH_3)$ is the flux (mg m^{-3} min^{-1}) of ammonia emission. According to Eq. 2, the flux of ammonia released from the MSG process can be directly restricted by the concentration of ammonium-N in solution from the MSG production. In particular, the quantity of ammonia released from Phase I had a closer relationship with the concentration of ammonium-N in the outflow from the MSG production because the significant level of the regression equation increased as follows (Fig. 4b):

$$\Phi(NH_3) = 0.0005\left[NH^{4+}\right] - 1.18 \quad (r = 0.902, n = 48, p < 0.005) \tag{3}$$

The relationship can testify that ammonia released from Phase I is mostly attributed to transformation of ammonium-N in the outflow. When the concentration of ammonium-N in the outflow increased, it can easily change into gaseous ammonia under certain conditions. Similarly, there was also a significantly linear relationship between the quantity of ammonia released during Phase II and the concentration of ammonium-N in wastewater (Fig. 4c), which can be described by following regression equation:

$$\Phi(NH_3) = 6E-06\left[NH^{4+}\right] + 0.027 \quad (r = 0.671, n = 32, p < 0.005) \tag{4}$$

The relationship also showed that transformation of ammonium-N into gaseous ammonia is a very important biochemical process (Kuai and Verstraete 1998; Zhou and Huang 2001; Schmidt and Sliekers 2003) during the discharge and treatment of MSG wastewater.

Even regression of the individual observations showed that there were independent linear relationships between the levels of ammonia released from the whole MSG process and the concentration of ammonium-N in outflow and wastewater from the MSG production (Table 1). However, the significant level ($p < 0.05$–0.025) of the individual observations was obviously less than the total level of all observations. Similarly, the level of ammonia released during Phase I was related to the concentration of ammonium-N in outflow from Phase I, and the significant level based on the regression of the individual observations was also lower than the total level of all observations. However, it was indicated that the quantity of ammonia released during Phase II was not significantly ($p > 0.05$) related to the concentration of ammonium-N in wastewater from Phase II based on regression of the individual observations.

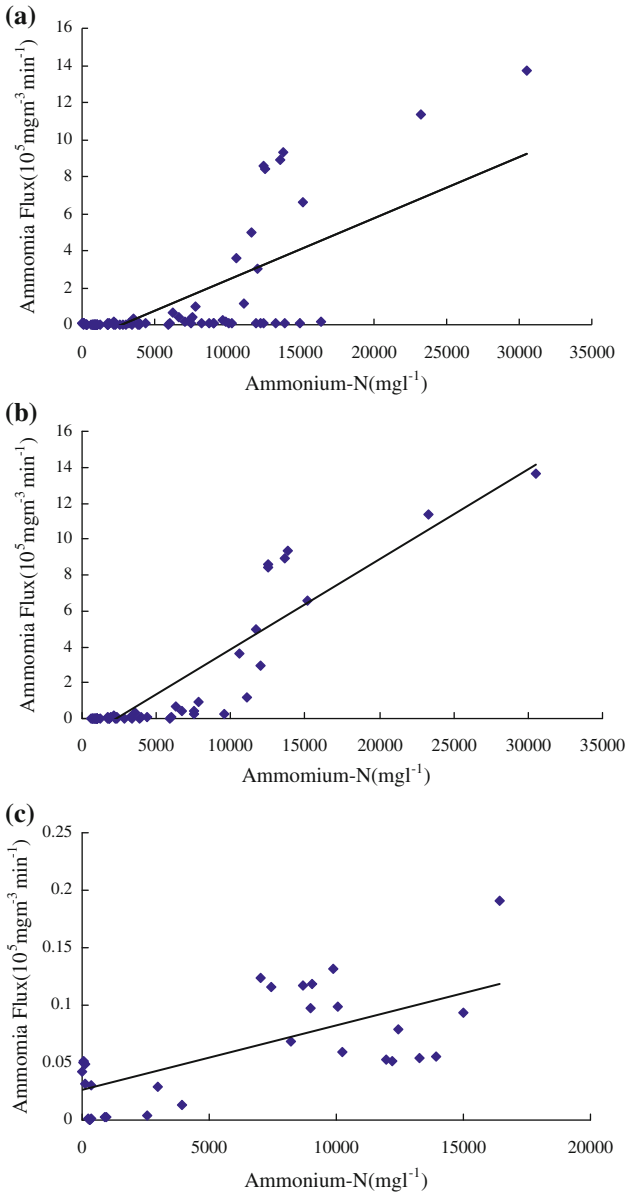

Fig. 4 Relationships between the average flux of ammonia emission and ammonium-N in the discharged solution from the whole MSG process (**a**), Phase I—the MSG synthesis (**b**), and Phase II—the wastewater treatment and discharge (**c**)

Table 1 Changes in the relationships between NH_3 emission and ammonium-N with time

Sampling time	Phase	Regression equation	n	r	p
May 3	Whole process	$\Phi(NH_3) = 0.0312[NH_4^+] - 58.78$	10	0.567	<0.05
	Phase I	$\Phi(NH_3) = 0.0654[NH_4^+] - 140.03$	6	0.876	<0.005
	Phase II	$\Phi(NH_3) = 0.0004[NH_4^+] + 2.91$	4	0.595	>0.1
May 10	Whole process	$\Phi(NH_3) = 0.0256[NH_4^+] - 22.12$	10	0.532	<0.05
	Phase I	$\Phi(NH_3) = 0.0642[NH_4^+] - 82.75$	6	0.924	<0.005
	Phase II	$\Phi(NH_3) = 0.0004[NH_4^+] + 3.43$	4	0.536	>0.1
May 17	Whole process	$\Phi(NH_3) = 0.0322[NH_4^+] - 57.56$	10	0.595	<0.005
	Phase I	$\Phi(NH_3) = 0.0635[NH_4^+] - 116.94$	6	0.891	<0.005
	Phase II	$\Phi(NH_3) = 0.0005[NH_4^+] + 3.05$	4	0.591	>0.1
May 24	Whole process	$\Phi(NH_3) = 0.0402[NH_4^+] - 139.26$	10	0.634	<0.025
	Phase I	$\Phi(NH_3) = 0.0658[NH_4^+] - 247.94$	6	0.825	<0.025
	Phase II	$\Phi(NH_3) = 0.0006[NH_4^+] + 2.25$	4	0.637	>0.1
Nov. 6	Whole process	$\Phi(NH_3) = 0.022[NH_4^+] - 66.69$	10	0.587	<0.05
	Phase I	$\Phi(NH_3) = 0.044[NH_4^+] - 200.33$	6	0.837	<0.01
	Phase II	$\Phi(NH_3) = 0.0006[NH_4^+] + 2.64$	4	0.767	>0.05
Nov. 13	Whole process	$\Phi(NH_3) = 0.0166[NH_4^+] - 21.16$	10	0.522	<0.05
	Phase I	$\Phi(NH_3) = 0.0415[NH_4^+] - 93.83$	6	0.888	<0.005
	Phase II	$\Phi(NH_3) = 0.0003[NH_4^+] + 2.83$	4	0.585	>0.05
Nov. 20	Whole process	$\Phi(NH_3) = 0.0379[NH_4^+] - 130.46$	10	0.781	<0.005
	Phase I	$\Phi(NH_3) = 0.0541[NH_4^+] - 150.41$	6	0.996	<0.005
	Phase II	$\Phi(NH_3) = 0.0011[NH_4^+] - 0.0586$	4	0.951	<0.01
Nov. 27	Whole process	$\Phi(NH_3) = 0.042[NH_4^+] - 126.75$	10	0.917	<0.005
	Phase I	$\Phi(NH_3) = 0.0457[NH_4^+] - 87.79$	6	0.988	<0.005
	Phase II	$\Phi(NH_3) = 0.0004[NH_4^+] + 4.61$	4	0.542	>0.05

Nitrate–N and nitrite-N are two important species which are present in the biogeochemical cycling of nitrogen (Zhou and Huang 2001). Some researchers pointed out that nitrate–N, nitrite-N, ammonium-N and gaseous ammonia can mutually transform under certain conditions (Hellinga et al. 1998; Prasertsak et al. 2001; van Dongen et al. 2001). Thus, this is an interesting problem whether gaseous ammonia released into the atmosphere is significantly related to nitrate–N and nitrite-N in outflow and wastewater from the MSG production due to the mutual transformation mechanism mentioned above. By examining all the data from the 8 observations, it was indicated that there were no significantly ($p > 0.1$) linear relationships between the quantity of ammonia released from the whole MSG process and the concentration summation of nitrate–N and nitrite-N in outflow and wastewater from the MSG production. In other words, there is no directly mutual relationship between the two. Similarly, the flux of ammonia released during Phase I or II had no significantly ($p > 0.1$ and $p \gg 0.1$) linear relationships with the concentration summation of nitrate–N and nitrite-N in outflow from Phase I or wastewater from Phase II. Thus, we surmise that there were no direct relationships between the quantity of ammonia released from the MSG process and the concentration summation of nitrate–N and nitrite-N in outflow and wastewater from the MSG production.

In fact, gaseous ammonia released into the atmosphere should have a certain relationship with nitrate–N and/or nitrite-N in solution (Zhou and Huang 2001). By further examining all the data of the 8 observations, it was found that there was a significantly exponential relationship between the flux of ammonia released from the whole MSG process and the concentration summation of nitrate–N and nitrite-N in outflow and wastewater from the MSG production. The relationship can be expressed by the following regression equation:

$$\Phi(NH_3) = 44.49e^{0.0015[NO_3^- + NO_2^-]} \quad (r = 0.719,\ n = 80,\ p < 0.001) \tag{5}$$

Thus, the flux of ammonia released from the MSG production is indirectly affected by the concentration of nitrate–N and nitrite-N in outflow and wastewater. In other words, nitrate–N and nitrite-N in outflow and wastewater can restrict emission of ammonia from the MSG production to some extent.

In theory, acidic or alkaline properties of a solution may play a key role in the transformation of nitrate–N, nitrite-N,

and ammonium-N into gaseous ammonia (Zhou and Huang 2001). In particular, the solution (outflow) form the MSG production is adjusted using NaOH solution during Phase I, and pH in wastewater is raised by adding calcium carbonate during Phase II. Thus, it can be deduced that the flux of ammonia released from the whole MSG process should directly relate to pH in outflow and wastewater from the MSG production.

By examining all the data from the 8 observations, it was indicated that the quantity of ammonia released from the whole MSG process was not linearly related to pH in outflow and wastewater from the MSG production, because the correlation coefficient of the corresponding regression equation was only 0.173 (significance level $p > 0.5$). Thus, it can be concluded that the quantity of ammonia released from the MSG process may not be directly restricted by pH in outflow and wastewater from the MSG production. Besides pH, other important factors to be further explored may have greater significance in the release of gaseous ammonia from solution (Strous et al. 1997; Zhou and Huang 2001). Similarly, there was no linear relationship between the quantity of ammonia released during Phase I and pH in outflow from Phase I because the correlation coefficient was only 0.201 (significance level $p > 0.25$). However, the quantity of ammonia released during Phase II had a significantly linear relationship with pH in wastewater from Phase II, and the corresponding regression equation is listed as follows:

$$\Phi(NH_3) = 1.37pH + 8.66 \quad (r = 0.895, n = 32, p < 0.005) \tag{6}$$

The promotion of releasing gaseous ammonia from wastewater may be probably attributed to an increase of pH in wastewater with the addition of $CaCO_3$ during the wastewater treatment. Of course, ammonium-N, as well as nitrate–N and nitrite-N, is usually unstable in solution. Various nitrogen species in solution can change into gaseous ammonia with the change in pH from an acidic to an alkaline environment (Zhou and Huang 2001).

All the working procedures of the whole MSG process including the synthesis of MSG (Phase I) and the treatment and discharge of wastewater (Phase II) can release gaseous ammonia, but there was a big difference in the quantity of ammonia released among these working procedures. The flux of ammonia released from the fermentation workshop was at the highest, up to 8.98×10^5 mg m^{-3} min^{-1} at the average value. The sugar-refining workshop had the lowest ammonia-releasing flux, only 85.1 mg m^{-3} min^{-1}. The release of gaseous ammonia from each working procedure was in the sequence 2# (the fermentation workshop) >3# (the extraction workshop) >4# (the top-grade MSG refining workshop) >8# (the pretreatment liquor) >7# (the mother liquor scraps) >5# (the middle-grade MSG refining workshop) >9# (the treated liquor) >6# (the low-grade MSG refining workshop) >10# (the final discharge outfall) >1# (the sugar refining workshop). The average annual quantity of ammonia released from the whole MSG process was up to 5.64×10^5 kg m^{-3}, where the synthesis of MSG (Phase I) accounted for 97.7%, and the discharge and treatment of wastewater (Phase II) accounted for 2.3%.

The quantity of ammonia released from the whole MSG process, particularly the synthesis of MSG (Phase I), not only had a linear relationship with the concentration of ammonium-N in solution, but also significantly had an exponential relationship with the concentration summation of nitrate–N and nitrite-N in solution. Thus, ammonium-N in solution may be a determinant factor for the release of gaseous ammonia into the atmosphere. However, there was no significantly linear or exponential relationship between the quantity of ammonia released from the whole MSG process including Phase I and pH in solution. On the contrary, that from the treatment and discharge of wastewater (Phase II) had a significantly linear relationship with pH in wastewater. During Phase II, pH in solution may be an important factor affecting the release of gaseous ammonia into the atmosphere.

Acknowledgments This work was financially supported by the Ministry of Education, People's Republic of China as a grand fostering project (No. 707011) and the National Natural Science Foundation of China for a Young Scholars Project (No. 30900204).

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