

Desorption behavior of ammonia nitrogen from sediment in cellar rainfall collection in a typical village in Northwest China

Guo-zhen Zhang, Jia-hui Sun, Ya-ting Mu, Fu-ping Wu, Ke Ma and Hao Yang

ABSTRACT

The desorption behavior of ammonia nitrogen (NH₃-N) in cellar sediment was studied to illustrate the influence of sediment on the quality of water in cellar rain collection. The impacts of three factors on the ability of cellar sediment to desorb NH₃-N were analyzed, which include the concentration of the cellar sediment, temperature and the degree of disturbance. In addition, the isothermal balance and kinetics fitting were studied. The results show that sediment concentration, temperature, and the disturbance degree greatly affect the NH₃-N desorption process. The pseudo-first-order and pseudo-second-order kinetics equations are suitable for describing the desorption process ($R^2 > 0.86$) under the different conditions of cellar sediment concentration. The Langmuir isothermal model is more suitable for describing the equilibrium desorption of the different sediment concentrations than the Freundlich isothermal model. Ultimately, the NH₃-N desorption process has a significant influence on cellar water quality. Such results could be a theoretical basis for collection, treatment, and maintenance of cellar water.

Key words | ammonia nitrogen, desorption, sediment, water cellar

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INTRODUCTION

In recent years, a shortage of drinking water in China's northwest region has been partly mitigated by the '121 Rainwater Harvesting Project' and 'Water-saving Irrigation Project'. However, a problem remains regarding the safety of the drinking water derived from the rainwater harvest. Many factors contribute to the pollution of collected rainwater, such as regional differences, overlying surfaces, rainfall volume and cellar materials (Wu *et al.* 2014; Tao *et al.* 2015). When a rainstorm occurs, rainwater, along with soil, sand and a large number of pollutants on the ground can jointly flow into water cellars. The materials in the flow will then permeate into cellar sediment. The pollutants can be absorbed by cellar sediment and undergo a series of biochemical reactions with cellar sediment, thus

affecting the water quality in the cellar. Therefore, cellar sediment plays an important role in cellar water quality. Obviously, it is highly desirable to investigate the impact of cellar sediment. This forms the basis of the current study.

From the literature (Li 2014; Zhang *et al.* 2014), cellar water studies are mainly focused on four issues: the standard of water quality; characteristics of water quality and pre-treatment technologies; advanced treatment technologies; and the management and maintenance technology/strategy of cellar water. To the best of our knowledge, there is no existing literature regarding the role of cellar sediment on water quality (Liu *et al.* 2016).

In this study, sediment from a household cellar rainwater collecting system in Fangli Town, Qingcheng

County, Qingyang City, Gansu Province, was sampled to investigate how the desorption of ammonia nitrogen from the cellar sediment influences the cellar water quality. A kinetics simulation was conducted with a goal to provide an approach for maintaining the water quality.

MATERIALS AND METHODS

Materials

Qingyang City, Gansu Province, is a typical arid and dry area in northwest China (Figure 1), where cellar water is an important source of drinking water. In Fangli Village, as in other villages in this area, a cellar was built in every household and the rainwater was collected from the roofed courtyard. The characteristics of a typical cellar and the water are shown in Table 1.

The sediment used in this study was collected under the surface of the water in the center of the cellar using a bucket and rope (Figure 2). The thickness of accumulated sediment in the selected cellars was approximately 40 cm. After collection, the sediment samples were naturally air-dried, ground and sieved using a 120-mesh to prevent large impurities before use.



Figure 1 | Location of Qingyang City.

Table 1 | Profile of water cellar

Cellar material	Structure	Volume (m ³)	Type of collecting surface	Size (m ²)	Water quality	Main purpose of water	Years used
Red mud	Canned	30	Roofed garden	300	Medium	Domestic water	30

The distribution of particle sizes of the cellar sediment is shown in Table 2. The clay ($\leq 2 \mu\text{m}$), powder ($2\text{--}50 \mu\text{m}$) and sand ($50\text{--}1,000 \mu\text{m}$) accounted for 9.00%, 70.77% and 20.24% of the sample, respectively. The median diameter (D_m) of the cellar sediment was $22.75 \mu\text{m}$. Physical and chemical characteristics of the cellar sediment are shown in Table 3.

Experimental design

Experimental water samples were prepared by the addition of different quantities of cellar sediments into 200 mL of deionized water in a series of 250 mL conical flasks. In the absence of previous research related to water-cellar sediment, the varying concentrations of sediment were chosen based on the varying sediment of rivers, lakes and reservoirs. The concentrations of the sediments were set as 0.5, 1, 2, 5 and 10 g/L. Flasks were put on an oscillation facility while samples were collected at different times of oscillation under different conditions of sediment concentration, temperature and oscillation speed, shown in Table 4. Samples of the supernatant were filtered through a $0.45 \mu\text{m}$ micro-porous membrane while $\text{NH}_3\text{-N}$ was measured to determine the desorbed amount, which was used for kinetics simulation and isothermal equilibrium analysis of desorption. The main analysis methods used for the test are shown in Table 5.

Desorption model

Kinetics equation of desorption

Kinetics were used to study the influence of various factors on the rate of pollutant desorption reactions. The process that causes the sediment to desorb pollutants is generally described by the pseudo-first-order and/or pseudo-second-order kinetics equations, Weber and Morris intra-particle diffusion model or Bangham pore diffusion model. In this study, the pseudo-first-order and pseudo-second-order



Figure 2 | Appearance of cellar and sediment collection.

Table 2 | Mechanical composition and elements of cellar sediment

Median size (μm)	Clay ($\leq 2 \mu\text{m}$)	Powders (2–50 μm)	Sand (50–1,000 μm)	Fe_2O_3 (%)	SiO_2 (%)	Al_2O_3 (%)	MgO (%)	CaO (%)
22.752	8.997%	70.765%	20.238%	5.05	53.24	12.72	2.39	9.52

Table 3 | Main physical and chemical indicators of cellar sediment collected

Parameter	Moisture (%)	Ignition loss (%)	Organic matter (%)	Total phosphorus (mg/kg)	Total nitrogen (mg/kg)	Ammonia nitrogen (mg/kg)	Nitrate nitrogen (mg/kg)	Nitrite nitrogen (mg/kg)
Content	32.3	9.321	0.756	480	505	273	10.061	0.042

kinetics equations were used to analyze the desorption kinetics based on the assumption that chemisorption is a control mechanism, which makes an electron public or

Table 4 | Control parameters

Temperature ($^{\circ}\text{C}$)	V (mL)	Concentration of cellar sediments (g/L)	Disorder degree (oscillation speed) (r/min)	Sampling time (h)
20 ± 0.5	200	0.5	20 ± 0.5	48 h
		1.0		
		2.0		
		5.0		
		10.0		
20	200	5 g/L	190 ± 5	
25				
27				
30				
20 ± 0.5	200	5 g/L	140	
			165	
			190	
			215	
			240	

causes electron transfer between the adsorbent and adsorbate. The Weber–Morris model was used to analyze the reaction control step and the obtained adsorbent particle diffusion rate constant. The Bangham pore diffusion model was used to describe the mechanism of diffusion channels (Wang 2014).

The pseudo-first-order kinetics equation is expressed as:

$$\frac{dq}{dt} = k_1(q_e - q) \quad (1)$$

Table 5 | Experimental methods

Test name	Test method
Ammonia nitrogen	Nessler's reagent spectrophotometry
Organic matter	Potassium dichromate volumetric method
Particle size	Fully automatic laser granulometer, Mastersizer 2000, produced by British Malvern Instrument Co. Ltd
Chemical composition	X-ray fluorescence spectrometer, MagixPW2403, produced by Dutch Philips Co. Ltd

The pseudo-second-order kinetics equation is expressed as:

$$\frac{dq}{dt} = k_2(q_e - q)^2 \quad (2)$$

where q is the amount of ammonia nitrogen desorbed by sediments at t , (mg/kg); q_e is the balanced amount of ammonia nitrogen desorbed by sediments, (mg/kg); k_1 and k_2 are the rate constant, (h^{-1}); and t is the duration of the desorption process, (h).

Isothermal equilibrium desorption model

Isothermal equilibrium desorption models are often used to describe the mechanism of desorption. The Langmuir and Freundlich isothermal desorption models are widely recognized.

The Langmuir (L) isothermal desorption equation is expressed as:

$$Q_e = \frac{S_m K_1 C_e}{1 + K_1 C_e} \quad (3)$$

The Freundlich (F) isothermal desorption equation is expressed as:

$$Q_e = K_f C_e^n \quad (4)$$

where S_m is the maximum amount of ammonia nitrogen desorbed by sediments, (mg/kg); Q_e is the balanced amount of ammonia nitrogen desorbed, (mg/kg); K_1 is a desorption coefficient; C_e is the equilibrium concentration of pollutants, (mg/kg); K_f represents the equilibrium desorption coefficient; and n is the desorption index, which is a constant.

RESULTS

Effect of the cellar sediment concentration on the desorption of ammonia nitrogen

When the concentration of cellar sediment is 0.5, 1, 2, 5 and 10 g/L, respectively, the amount of desorbed $\text{NH}_3\text{-N}$ is as presented in Figure 3. The results were derived from using an oscillation rate of 190 ± 5 r/min and temperature of $20 \pm 0.5^\circ\text{C}$. The $\text{NH}_3\text{-N}$ released from cellar sediments

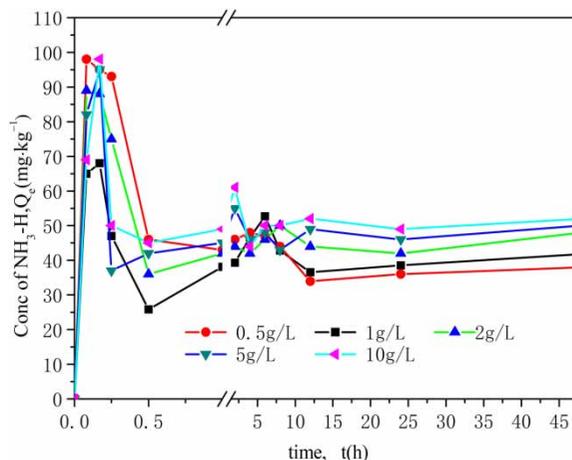


Figure 3 | Temporal variation of desorbed $\text{NH}_3\text{-N}$ at different cellar sediment concentrations.

shows a significant increase in the first 30 min, while the maximum value was reached at 30 min. The concentration rapidly declined in the subsequent 30 to 60 min, and then it slowly rose again and gradually approached the equilibrium value. At the beginning of oscillation, the water and sediment particles loosely bonded together, resulting in the rapid desorption of ammonia nitrogen and thus the rapid increase in the concentration of desorbed $\text{NH}_3\text{-N}$. Subsequently, particles inside the sediment were gradually exposed as the mixing of water and sediment progressed, which allowed more opportunity for $\text{NH}_3\text{-N}$ to release from the solid surfaces, therefore increasing the concentration of adsorption. The ensuing decrease of $\text{NH}_3\text{-N}$ release might be explained by the fact that it is difficult to transfer and desorb $\text{NH}_3\text{-N}$ atoms which tightly bond with particles inside the sediment after the loosely joined $\text{NH}_3\text{-N}$ atoms are released. At the end of the first increasing and then decreasing process (at approximately 4 hours), the distribution of $\text{NH}_3\text{-N}$ in the water and sediment became balanced and the concentration of desorption also became relatively stable, although there were still some fluctuations.

As shown in Figure 3, the equilibrium amount of desorbed $\text{NH}_3\text{-N}$ increased with the increasing concentration of cellar sediments, but the increase in desorbed $\text{NH}_3\text{-N}$ was minor. A higher concentration of water-cellar sediment means more desorption activators (namely, water-cellar sediment) and desorption points, depending on the condition of the water. Thus, the desorption rate is increased, leading to a greater amount of desorbed $\text{NH}_3\text{-N}$. However,

the adsorption of $\text{NH}_3\text{-N}$ is also increased with an increase in sediment concentration in the cellar. That is the reason why the equilibrium amount of desorbed $\text{NH}_3\text{-N}$ slightly increases (Li *et al.* 2014; Wang *et al.* 2014; Li *et al.* 2016).

Effect of temperature on desorption of ammonia nitrogen

When the temperature of the water was 20°C, 25°C, 27°C and 30°C, respectively, the correlative amounts of desorbed $\text{NH}_3\text{-N}$ under an oscillation rate of 190 ± 5 r/min and a sediment concentration of 5 g/L were as shown in Figure 4. It shows that the maximum $\text{NH}_3\text{-N}$ concentration was obtained at 10 min. Subsequently, the $\text{NH}_3\text{-N}$ concentration rapidly decreased. Thereafter, the concentration slowly rose and reached a second maximum at 2 hours. After that, a similar decrease and increase in concentration occurred again. An equilibrium was reached at 4 hours. The initial increase in $\text{NH}_3\text{-N}$ concentration occurred because temperature has an effect on the surface adsorption capability of sediment particles and colloids. As temperature increases, a series of physical and chemical reactions occur in the sediment, which may cause more $\text{NH}_3\text{-N}$ release. The cause of the subsequent rapid decrease is similar to the mechanism mentioned above. There are two reasons that the second maximum occurs and an equilibrium is gradually reached. Firstly, Brownian motion is intensified as temperature goes up. Secondly, as the temperature increases, the surface

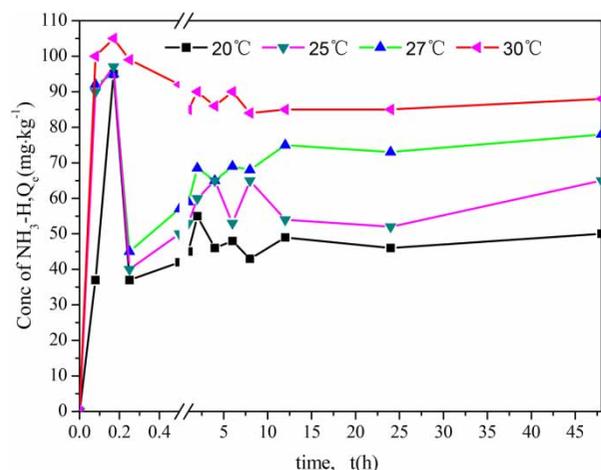


Figure 4 | Temporal variation of desorbed $\text{NH}_3\text{-N}$ at different temperatures.

tension of the liquid generally weakens, molecular spacing expands and desorption strengthens.

As indicated in Figure 4, the higher the temperature is, the higher the equilibrium concentration of desorbed $\text{NH}_3\text{-N}$. This suggests that the desorption reaction is an endothermic reaction. From the perspective of physical desorption, desorption is only a physical effect when there is no electron transfer, formation and destruction of chemical bonds or atomic rearrangement (Lei *et al.* 2016). The necessary mechanisms are as follows: (a) the need for a small amount of heat; (b) low stability of adsorption; (c) a high rate of adsorption and desorption; (d) no need for activation energy; and (e) no correlation between the rate of adsorption and desorption and temperature. In general, chemical desorption is fairly intensive. It occurs when the chemical bond is forced between desorbed substances and causes desorption of materials. The main mechanisms are as follows: (a) a need for relatively high adsorption heat; (b) selective adsorption; (c) a relatively stable reaction; (d) activation energy; and (e) an increasing rate of adsorption and desorption as temperature goes up. Therefore, it can be concluded that both physical and chemical desorption rates are intensified as temperature goes up.

Effect of disorder degree on desorption of ammonia nitrogen

Figure 5 presents the amount of the desorbed $\text{NH}_3\text{-N}$ when the degree of disorder (i.e., oscillation speed) was 140, 165, 190, 215 and 240 r/min, respectively. The experiments were done at a temperature of $20 \pm 0.5^\circ\text{C}$ and a water-cellar sediment concentration of 5 g/L. The concentration of $\text{NH}_3\text{-N}$ rapidly increased in the early stage, then rapidly declined and finally reached equilibrium. At the beginning of the experiment, the concentration of $\text{NH}_3\text{-N}$ in the overlying water was low. There was a relatively high gradient in $\text{NH}_3\text{-N}$ concentration between the water and underlying sediments. This caused the physically adsorbed $\text{NH}_3\text{-N}$ to be quickly released, breaking the original balance. Subsequently, particles inside the sediment were gradually exposed. New adsorption sites were generated and $\text{NH}_3\text{-N}$ in the overlying water was absorbed, which reduced the amount of $\text{NH}_3\text{-N}$ in the overlying water. Then, as oscillation continued, the reaction between the sediment and

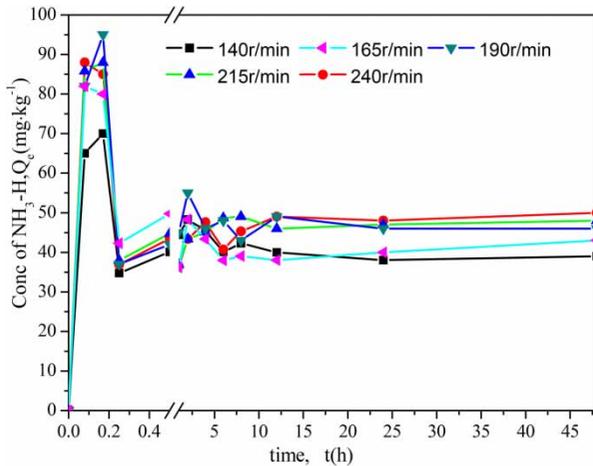


Figure 5 | Temporal variation of desorbed $\text{NH}_3\text{-N}$ under different disorder degrees.

overlying water continued and gradually reached a new equilibrium.

It can be seen from Figure 5 that the degree of disorder had a small influence on the equilibrium concentration of desorbed $\text{NH}_3\text{-N}$. The equilibrium desorption concentration of $\text{NH}_3\text{-N}$ was 40 mg/kg when the disorder degree (oscillation speed) was 140r/min, whereas it was 50 mg/kg when the disorder degree was 240r/min.

Kinetics simulation of the desorption of ammonia nitrogen

The concentration of desorbed $\text{NH}_3\text{-N}$ was recorded at an oscillation rate of 190 ± 5 r/min, water temperature of $20 \pm 0.5^\circ\text{C}$ and different sediment concentrations of 0.5, 1, 2, 5 and 10 g/L, respectively. The resultant data were fitted using pseudo-first-order and pseudo-second-order kinetics equations. The isotherms are shown in Figures 6 and 7 and the kinetic parameters are shown in Table 6.

As shown in Figures 6 and 7, it seems not perfect to use both the pseudo-first-order and pseudo-second-order kinetics equations to reflect the entire $\text{NH}_3\text{-N}$ desorption process ($R^2 = 0.2\text{--}0.6$). The reason for this is that the data did not fit the early stage when the $\text{NH}_3\text{-N}$ concentration rapidly increased due to the release of $\text{NH}_3\text{-N}$ in the sediment, followed by a decrease. Therefore, data for the first 30 min were removed and the remaining data were fitted using the pseudo-first-order and pseudo-second-order kinetics equations.

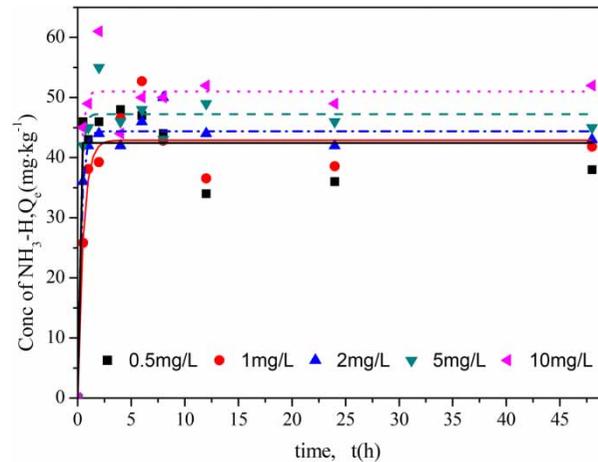


Figure 6 | Pseudo-first-order kinetics for desorption of $\text{NH}_3\text{-N}$ under the conditions of different sediment concentrations.

As shown in Table 6, good correlations ($R^2 = 0.86\text{--}0.97$) were then obtained using the pseudo-first-order and pseudo-second-order kinetics equations after the removal of the first 30 min data. It demonstrated that the long-term desorption process of $\text{NH}_3\text{-N}$ in water-cellar sediment can be relatively well fitted by the pseudo-first-order and pseudo-second-order kinetics equations, although they are not suitable for describing the initial dynamic process (Xia et al. 2013; Wu 2014). The Q_e (the equilibrium amount of ammonia nitrogen that a unit of sediment desorbs) increases as the concentration of sediments increases. This indicates that there is a positive correlation between the equilibrium amount of $\text{NH}_3\text{-N}$ that a unit of sediment desorbs and the concentration of sediment

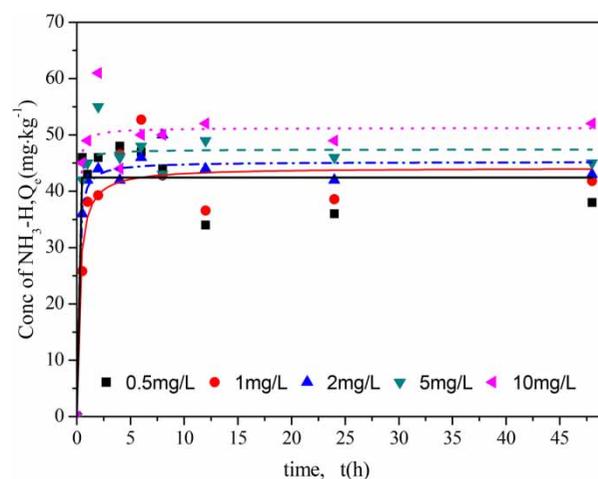


Figure 7 | Pseudo-second-order kinetics for desorption of $\text{NH}_3\text{-N}$ under the conditions of different sediment concentrations.

Table 6 | Parameters of kinetics equations under the conditions of different sediment concentrations

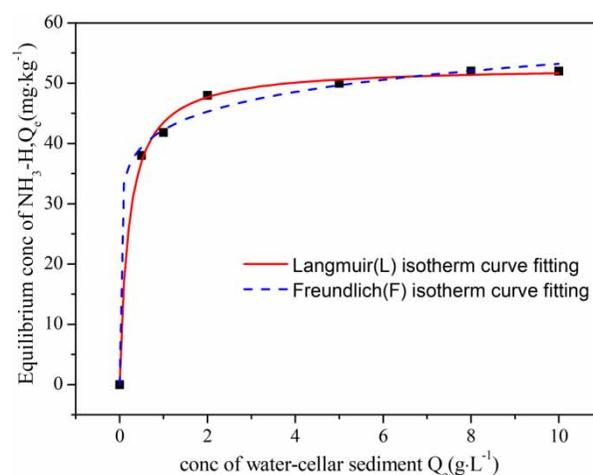
Sediment concentration (g/L)	Pseudo-first-order kinetics equation			Pseudo-second-order kinetics equation		
	Q_e (mg/L)	k_1 (h^{-1})	R^2	Q_e (mg/L)	k_2 (h^{-1})	R^2
0.5	42.4	8.265E6	0.8698	42.4	1.117E26	0.8698
1	42.9	1.916	0.8930	44.2	92.91	0.8645
2	44.4	3.270	0.9697	45.2	205.0	0.9662
5	47.2	47.2	0.9496	47.4	506.6	0.9423
10	51.0	4.205	0.9274	51.3	427.7	0.9201

at a certain temperature and oscillation speed. Q_e obtained from the pseudo-first-order kinetics equation is slightly larger than that from the pseudo-second-order kinetics equation. In addition, compared with an increased gradient of the sediment concentration, the increase in the gradient of Q_e is small. Also seen from Table 6, kinetics coefficients (k_1 and k_2) gradually increase as sediment concentration increases, except when the concentration of sediment is 0.5 g/L. When the concentration of sediment is 0.5 g/L, k_1 and k_2 reach their maximum values (8.265E6 and 1.117E26, respectively). This indicates that there is a small mutual influence among sediment particles on the release of NH_3 -N in low concentrations (0.5 g/L) and that an equilibrium can quickly be reached. When the concentration of sediment increases from 1 g/L to 10 g/L, k_1 increases from 1.916 to 4.205, while k_2 increases from 92.91 to 506.6. Obviously, k_1 is the lowest under the condition of 1 g/L because of both the mutual influence among sediment particles on NH_3 -N release and the strengthened absorption that occurs with the increase in the concentration of sediment.

Isothermal equation of desorption of ammonia nitrogen

When sediment concentrations were 0.5, 1, 2, 5 and 10 g/L, respectively, the equilibrium amount of NH_3 -N was recorded under the conditions of an oscillation rate of 190 ± 5 r/min and a water temperature of $20 \pm 0.5^\circ C$. The results were fitted using the Langmuir (L) isothermal equation and Freundlich (F) isothermal equation. The isotherms are shown in Figure 8, while the kinetic parameters are shown in Table 7.

As shown in Figure 8, the equilibrium amount of desorbed NH_3 -N gradually increased until stability was

**Figure 8** | Isothermal equations.

obtained with the increase in the concentration of cellular sediment. It shows that the concentration of cellular sediment has significant impact on the equilibrium concentration of ammonia nitrogen (Zhao *et al.* 2015).

In Table 7, K_1 (the desorption equilibrium constant of the Langmuir isothermal equation) is equal to the ratio of adsorption and desorption. It is related to the surface combined energy of the sediment particles and reflects the spontaneous degree of the absorption and desorption reaction. The higher its value, the higher the spontaneous degree of the absorption and desorption reactions and the stronger the desorption ability of the desorption material.

Table 7 | Parameters of isothermal equations

Langmuir fitting parameters			Freundlich fitting parameters		
S_m (mg/kg)	k_1 (h^{-1})	R^2	K_f (h^{-1})	n	R^2
0.0528	4.6894	0.9972	0.0423	0.1002	0.9935

S_m (the maximum amount of contaminant desorbed by the sediment) represents the ability of the sediment to desorb the contaminant. The value of S_m divided by C (the concentration of $\text{NH}_3\text{-N}$ in the sediment of the raw water) is 19.34%, indicating that the storm or artificial disturbance causes $\text{NH}_3\text{-N}$ in the sediment to be released again into the water and thus causes secondary pollution of the water. Overall, it is evidenced from R^2 (0.9972 and 0.9935) that both the Langmuir isothermal and Freundlich isothermal equations are suitable for fitting the process of sediment desorbing $\text{NH}_3\text{-N}$.

CONCLUSIONS

Rainwater collected in cellars is used as drinking water in villages in northwest China. An anoxic environment is easily formed in water cellars, which increases microbial reproduction and impairs water quality. The desorption behavior of $\text{NH}_3\text{-N}$ from cellar sediment was studied to provide useful information on cellar water safety. The experimental results demonstrate that the concentration of sediment, the temperature and the degree of disorder have positive correlations with the equilibrium amount of desorbed $\text{NH}_3\text{-N}$. Among them, the sediment concentration is the most significant factor to influence $\text{NH}_3\text{-N}$ desorption, thus influencing the cellar water quality. This provides insight into the improvement of the cellar water quality by controlling the pollutants in the sediment. Both pseudo-first-order and pseudo-second-order kinetics equations are suitable for fitting the $\text{NH}_3\text{-N}$ desorption process. The Langmuir isothermal equation is more suitable for fitting the relationship between initial and equilibrium concentrations of $\text{NH}_3\text{-N}$ than the Freundlich isothermal equation.

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