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# CONTENTS

#### Aquatic environment

Aquate environment	
Effect of periphyton community structure on heavy metal accumulation in mystery snail (Cipangopaludina chinensis): A case study of the Bai River	
Jingguo Cui, Baoqing Shan, Wenzhong Tang ·····	1723
Enhanced anaerobic digestion and sludge dewaterability by alkaline pretreatment and its mechanism	
Liming Shao, Xiaoyi Wang, Huacheng Xu, Pinjing He	1731
Ammonia pollution characteristics of centralized drinking water sources in China	
Qing Fu, Binghui Zheng, Xingru Zhao, Lijing Wang, Changming Liu	1739
Bulking sludge for PHA production: Energy saving and comparative storage capacity with well-settled sludge	
Qinxue Wen, Zhiqiang Chen, Changyong Wang, Nanqi Ren	1744
Atmospheric environment	
Heterogeneous reaction of NO <sub>2</sub> on the surface of montmorillonite particles	
Zefeng Zhang, Jing Shang, Tong Zhu, Hongjun Li, Defeng Zhao, Yingju Liu, Chunxiang Ye	1753
Heterogeneous uptake of NO2 on soils under variable temperature and relative humidity conditions	
Lei Wang, Weigang Wang, Maofa Ge ·····	1759
Diurnal variation of nitrated polycyclic aromatic hydrocarbons in PM <sub>10</sub> at a roadside site in Xiamen, China	
Shuiping Wu, Bingyu Yang, Xinhong Wang, Huasheng Hong, Chungshin Yuan	1767
Conversion characteristics and mechanism analysis of gaseous dichloromethane degraded by a VUV	
light in different reaction media	
Jianming Yu, Wenji Cai, Jianmeng Chen, Li Feng, Yifeng Jiang, Zhuowei Cheng	1777
Characteristics of odorous carbonyl compounds in the ambient air around a fishery industrial complex of Yeosu, Korea	
Zhongkun Ma, Junmin Jeon, Sangchai Kim, Sangchul Jung, Woobum Lee, Seonggyu Seo	1785
Terrestrial environment	
Identification of rice cultivars with low brown rice mixed cadmium and lead contents and their interactions with the micronutrients iron,	
zinc, nickel and manganese	
Bing Li, Xun Wang, Xiaoli Qi, Lu Huang, Zhihong Ye	1790
In situ stabilization remediation of cadmium contaminated soils of wastewater irrigation region using sepiolite	
Yuebing Sun, Guohong Sun, Yingming Xu, Lin Wang, Dasong Lin, Xuefeng Liang, Xin Shi	1799
Environmental biology	
Kinetic analysis and bacterium metabolization of $\alpha$ -pinene by a novel identified <i>Pseudomonas</i> sp. strain	
Zhuowei Cheng, Pengfei Sun, Yifeng Jiang, Lili Zhang, Jianmeng Chen	1806
Cloning and expression of the first gene for biodegrading microcystin LR by Sphingopyxis sp. USTB-05	
Hai Yan, Huasheng Wang, Junfeng Wang, Chunhua Yin, Song Ma, Xiaolu Liu, Xueyao Yin	1816
Isolation, identification and characterization of an algicidal bacterium from Lake Taihu and preliminary studies on its algicidal compounds	
Chuan Tian, Xianglong Liu, Jing Tan, Shengqin Lin, Daotang Li, Hong Yang	1823
Spatial heterogeneity of cyanobacterial communities and genetic variation of Microcystis populations within large,	
shallow eutrophic lakes (Lake Taihu and Lake Chaohu, China)	
Yuanfeng Cai, Fanxiang Kong, Limei Shi, Yang Yu ·····	1832
Environmental health and toxicology	
Proteomic response of wheat embryos to fosthiazate stress in a protected vegetable soil	
Chunyan Yin, Ying Teng, Yongming Luo, Peter Christie	1843
Pollution level and human health risk assessment of some pesticides and polychlorinated biphenyls in Nantong of Southeast China	
Na Wang, Li Yi, Lili Shi, Deyang Kong, Daoji Cai, Donghua Wang, Zhengjun Shan ·····	1854
Cytotoxicity and genotoxicity evaluation of urban surface waters using freshwater luminescent bacteria	
Vibrio-qinghaiensis spQ67 and Vicia faba root tip	
Xiaoyan Ma, Xiaochang Wang, Yongjun Liu ·····	1861
Environmental catalysis and materials	
Simulated-sunlight-activated photocatalysis of Methylene Blue using cerium-doped SiO <sub>2</sub> /TiO <sub>2</sub> nanostructured fibers	
Yu Liu, Hongbing Yu, Zhenning Lv, Sihui Zhan, Jiangyao Yang, Xinhong Peng, Yixuan Ren, Xiaoyan Wu	1867
TiO <sub>2</sub> /Ag modified penta-bismuth hepta-oxide nitrate and its adsorption performance for azo dye removal	
Eshraq Ahmed Abdullah, Abdul Halim Abdullah, Zulkarnain Zainal, Mohd Zobir Hussein, Tan Kar Ban	1876

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### Ammonia pollution characteristics of centralized drinking water sources in China

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#### Abstract

The characteristics of ammonia in drinking water sources in China were evaluated during 2005–2009. The spatial distribution and seasonal changes of ammonia in different types of drinking water sources of 22 provinces, 5 autonomous regions and 4 municipalities were investigated. The levels of ammonia in drinking water sources follow the order of river > lake/reservoir > groundwater. The levels of ammonia concentration in river sources gradually decreased from 2005 to 2008, while no obvious change was observed in the lakes/reservoirs and groundwater drinking water sources. The proportion of the type of drinking water sources is different in different regions. In river drinking water sources, the ammonia level was varied in different regions and changed seasonally. The highest value and wide range of annual ammonia was found in South East region, while the lowest value was found in Southwest region. In lake/reservoir drinking water sources, the ammonia levels were not varied obviously in different regions. In underground drinking water sources, the ammonia levels were varied obviously in different regions due to the geological permeability and the natural features of regions. In the drinking water sources with higher ammonia levels, there are enterprises and wastewater drainages in the protected areas of the drinking water sources.

Key words: ammonia; centralized drinking water sources; distribution; China DOI: 10.1016/S1001-0742(11)61011-5

#### Introduction

Ammonia-nitrogen, including non-ionized (NH<sub>3</sub>) and ionized  $(NH_4^+)$ , is the predominant pollutant in the main drinking water sources. It originates from human activities in the urban areas, metabolic, agricultural and industrial processes, and from disinfection with chloramine. Ammonia-nitrogen concentration in surface water is affected by hydrogeology and climate change (Delpla et al., 2009; Bates et al., 2008). Ammonia toxicity in waters has been widely investigated: its incomplete nitrification elevates toxic nitrite contents; due to its property to combine chlorine easily, the chlorine necessity during disinfection processes increases, and consequently the content of mutagenic disinfection by-products (DBP) is changing in an ascending mode (Georgieva et al., 2010; Campos et al., 2002; Källqvist and Svenson, 2003; Yu et al., 2007; Puigagut et al., 2005); it also causes the failure of filters for the removal of manganese, and causes taste and odour problems (WHO, 2003).

Ammonia is an important indicator of water quality. It can indicate the possible bacterial, sewage and animal feces pollutions (WHO, 2003). The World Health Organization (WHO, 1993) indicates that the threshold odour concentration of ammonia at alkaline pH is approximately 1.5 mg/L, while the taste threshold is 35 mg/L. The guideline for ammonia concentration is 0.5 mg/L for drinking water in China (GB5749-2006) and European Union (1998). According to the US EPA health risk assessment model, human health risk caused by ammonia content in drinking water is very low (Qin et al., 2011).

Many previous publications investigated the detection method of ammonia (Li, 2007), ammonia removal (Fu et al., 2006), and health risk of ammonia in some drinking water sources (Gu et al., 2000; Ye and Xu, 2000; Qin et al., 2011; Yang et al., 2008; Li et al., 2007; Guo et al., 2007). However, the comprehensive evaluation of ammonia concentration level in the drinking water sources is still lack.

Based on the data from 22 provinces, 5 autonomous regions, and 4 municipalities of China, the present study investigates the spatial distribution and seasonal variations of ammonia concentration in drinking water sources. The ammonia removal during water purification process was also evaluated.

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#### 1 Methods

#### 1.1 Data collection

Water quality data from 3966 centralized drinking water sources including 1214 river sources, 1080 lake/reservoir sources and 1672 groundwater sources in China collected during 2005 to 2009 were used. The studied drinking water sources cover 22 provinces, 5 autonomous regions and 4 municipalities; and serve 37% of China's population (4.8 billion). There is no data for Hong Kong, Macao, and Taiwan. The study area is divided into six geographical regions, namely North region: Liaoning, Jilin and Heilongijang provinces: Northeast region: Beijing and Tianjin cities, Hebei, Jiangsu and Shandong provinces; South East region: Shanghai City, Zhejiang, Fujian, Guangdong and Hainan provinces; Central region: Shanxi, Anhui, Jiangxi, Henan, Hubei and Hunan; Northwest region: Inner Mongolia, Shanxi, Gansu, Qinghai, Ningxia and Xinjiang; Southwest region: Chongqing City, Xizang Autonomous Regions, Guangxi Autonomous Regions, Sichuang, Guizhou, Yunnan provinces

#### 1.2 Data analysis

The analysis for ammonia followed the method of salicylic acid spectrophotometry (GB 7481-87). Statistical analyses were carried out with SPSS for Windows and Monte Carlo method.

#### 2 Results and discussion

#### 2.1 Current ammonia level in drinking water sources

According to the collected data, the annual average ammonia concentration in river water ranged from 0.01 to 4.15 mg/L, with average 0.32 mg/L, median 0.19 mg/L, and standard deviation (SD) 0.903. The probability distribution of ammonia in river drinking water sources is described by logarithm (Fig. 1). The ammonia values with highest frequency are 0.03 and 0.05 mg/L, the 95th percentile is 0.90 mg/L, and the percentage of over 0.5 mg/L is

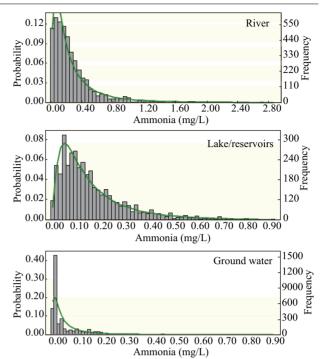


Fig. 1 Probability of ammonia level in river, lake/reservoirs and ground-water drinking water sources in China.

14.12%. The higher values of ammonia are found in river drinking water sources of Shanghai City, Jiaxing of Zhejiang, Guangzhou, Xinyang of Henan, Hengyang of Hunan, Huainan of Anhui and Nanping of Fujian Province.

Table 1 presents the environmental conditions of the drinking water sources with higher ammonia levels, such as protected area, numbers of enterprises, wastewater drainage discharges, and ammonia discharges from industries, urban areas, and non-point sources. In a drinking water source of the upper reaches of Huangpu River in Shanghai, there were 199 enterprises and a wastewater drainage, therefore ammonia is mainly from industrial discharge. The annual average of ammonia levels ranged from 3.87 to 4.15 mg/L between 2005 and 2009, while the monthly mean in 2007 ranged from 1.99 to 6.4 mg/L. Similarly, in a drinking water source of Jiaxing City of

Table 1 Contamination sources of high ammonia levels in drinking water sources

Drinking water	Site	Protected area (km <sup>2</sup> )	Enterprises numbers	Drainage numbers	Wastewater discharge ( $\times 10^4$ tons/yr)			Ammonia discharge (× $10^4$ tons/yr)		
sources					Industry	Urban	Non-point sources	Industry	Urban	Non-point sources
River	Huangpujiang, Shanghai	43.9	199	1	70.4	_	_	4.8	_	-
	Jiaxing City, Zhejiang	0.35	23	5	38	26.3	-	1.2	10.6	-
	Guangzhou City, Guangdong	1.08	-	-	-	16.1	-	-	3.9	-
	Hengyang City, Hunan	0.02	2	2	108	-	-	51	-	-
	Huainan City, Anhui	0.6	8	2	1667	-	-	468	-	-
Lake/reservoirs	Guiyang City, Guizhou	21	1	1	421	-	0.28	-	-	1.01
	Nanchong City, Sichuan	34	-	-	-	0.12	14	-	0.04	1.08
Groundwater	Liaotong City, Inner Mongolia	0.49	4	-	1876	18.2	-	38	11	-
	Chaoyang City, Liaoning	6.62	1	1	17.1	-	-	-	-	-
	Langfang City, Hebei	4.6	-	-	-	1.37	-	-	0.44	-
	Shuangcheng City, Heilongjiang	0.1	-	-	-	0.95	-	-	-	-
	Guiyang City, Guizhou	0.68	_	-	-	-	0.01	-	-	0.02
	Liaoyuan City, Jilin	1.2	-	-	-	-	0.9	-	-	0.1
	Changde City, Hunan	0.01	_	-	_	-	_	-	-	-

"-" No data are available.

No. 10

Zhejiang, there were 23 enterprises and five wastewater drainages, therefore ammonia discharges were mainly from industrial wastewater and urban sewage. The annual average of ammonia levels ranged from 2.42 to 4.34 mg/L between 2005 and 2009, and the monthly mean of ammonia levels ranged from 2.04 to 6.66 mg/L in 2007. In some drinking water sources in Guangzhou, Xinyang of Henan and Nanping of Fujian, there was no enterprise and wastewater drainage outlet in the protected area. In a drinking water source in Guangzhou, the ammonia pollution was mainly from urban sewage with the annual average 1.36 to 2.57 mg/L during 2005 and 2009, while in some drinking water sources of Xinyang of Henan and Nanping of Fujian, the ammonia pollutions were mainly from non-point sources, with annual average 0.77 to 1.29 mg/L during 2005 and 2009. Enterprises and wastewater drainage discharges in the protected area seriously affect the ammonia concentration in drinking water sources.

The annual average ammonia concentration in lake/reservoir drinking water sources ranges from 0.01 to 5.59 mg/L with average 0.2 mg/L, median 0.14 mg/L, and SD 0.26. As shown in Fig. 1b, the ammonia concentration with highest probability is 0.03 mg/L, the 95th percentile is 0.56 mg/L, and the percentage of water sources with over 0.5 mg/L of ammonia is 6.21%. The higher values of ammonia are found in lake/reservoir drinking water sources in Hongfenghu of Guizhou Province, Suzhou City of Jiangsu and Nanchong of Sichuan Province.

In the Hongfenhu lake/reservoir drinking water source, there was an enterprise and a wastewater drainage outlet. Although there is no data from the industry and urban sewage, the annual average of ammonia levels ranged from 1.89 to 5.23 mg/L between 2005 and 2009. In a drinking water source in Nanchong, the ammonia pollution was from urban sewage and non-point sources with annual average ammonia levels ranged from 0.43 to 2.1 mg/L during 2005 and 2009. In general, there are less contaminant sources in lake/reservoir drinking water sources than river drinking water sources in China, therefore the ammonia concentrations in lake/reservoir are lower than that in rivers.

In groundwater drinking water sources, the annual average ammonia ranges from 0.01 to 7.6 mg/L, with average 0.08 mg/L, median 0.03 mg/L and SD 0.29. The ammonia values mainly range from 0.01 to 0.03mg/L, the 95th percentile is 0.22 mg/L, and the percentage water sources with ammonia content exceeded 0.5 mg/L is 1.7% (Fig. 1c). The higher values of ammonia are found in groundwater drinking water sources of Chaoyang City of Liaoning province, Liaoyuan City of Jilin Province, Shuangcheng of Heilongjiang Province and Changde City in Hunan Province.

In a drinking water source in Chaoyang City, there was an enterprise and a wastewater drainage outlet. Although there is no discharge from the industries, the annual average of ammonia concentration was about 5.71 mg/L between 2005 and 2008. In a drinking water source of Liaoyuan, the ammonia pollution was mainly from non-point sources, and the annual average ammonia concentration ranged from 0.90 to 1.16 mg/L between 2005 and 2008. In a drinking water source in Changde, the annual average ammonia ranged from 1.52 to 1.59 mg/L between 2005 and 2008, despite the site where no contaminant source in the protected area. In a drinking water source in Liaotong, the annual average ammonia concentration ranged from 0.18 to 0.24 mg/L between 2005 and 2008; there are 4 enterprises in the protected area, and the ammonia pollution was from industry and urban sewage. Therefore, it can be concluded that human activities can affect the ammonia concentration in underground water.

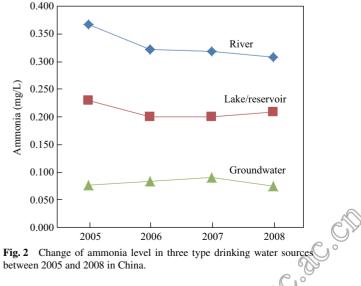
#### 2.2 Trends of ammonia level in drinking water sources of China

The concentrations of ammonia in three types of drinking water sources follow the order of river > lake/reservoir > groundwater (Fig. 2). Although the annual average ammonia of rivers were highest in three types of drinking water sources, it was lower than standard (0.5 mg/L) as indicated in the drinking water guidelines of China. From 2006 to 2008, the ammonia concentration in river drinking water sources was decreased. It may be due to the strengthening management of the wastewater from industries and urban activities in recent year. While there is no obvious change in ammonia concentration in lake/reservoir and underground drinking water sources, it may be due to the long cycle of water exchange and the small environmental changes of the protected area.

#### 2.3 Geographical distribution and seasonal changes of ammonia concentration in China

In terms of geographical distribution, the type of drinking water sources has different proportions in different regions, and the ammonia concentration in surface water quality is affected by hydrogeology and climate change.

In river drinking water sources, ammonia originates from industries, agricultural activities, and domestic wastewater drainage. Between 2005 and 2008, the ammonia levels were varying in different regions (Table 2). The highest value and wide range (0.467-0.541 mg/L) of annual ammonia in river were found in the South East



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Drinking water source	Year	Northeast	South East	North	Central	Northwest	Southwest
River		<i>n</i> = 51	<i>n</i> = 278	<i>n</i> = 71	<i>n</i> = 352	<i>n</i> = 103	<i>n</i> = 316
	2005	0.335	0.520	0.260	0.281	0.274	0.199
	2006	0.357	0.541	0.281	0.264	0.274	0.209
	2007	0.370	0.521	0.250	0.279	0.275	0.195
	2008	0.405	0.467	0.238	0.279	0.253	0.201
Lake/reservoir		<i>n</i> =182	n =303	<i>n</i> =109	<i>n</i> =198	n =75	n =332
	2005	0.183	0.153	0.300	0.199	0.215	0.363
	2006	0.191	0.153	0.248	0.186	0.233	0.239
	2007	0.198	0.164	0.222	0.202	0.199	0.238
	2008	0.158	0.175	0.266	0.180	0.387	0.205
Groundwater		<i>n</i> = 392	<i>n</i> =18	<i>n</i> = 293	n =386	n = 541	n = 105
	2005	0.042	0.020	0.113	0.060	0.081	0.063
	2006	0.044	0.020	0.108	0.071	0.097	0.071
	2007	0.073	0.056	0.100	0.097	0.096	0.068
	2008	0.048	0.020	0.083	0.083	0.081	0.079

n: number of dirnking water sources.

region, while the lowest value (0.195-0.209 mg/L) was found in the Southwest region.

This phenomenon can be explained as follows: in the South East region, it has the natural features of lush vegetation and abundant rainfall, and well-developed social and economic characteristics. The drinking water sources are located at lower reaches of the Changjiang River and the Pearl River. Therefore, there are many possible pollution sources. While in Southwest region, the drinking water sources are located at the upper reaches of the Changjiang River and the Pearl River, where less pollution sources were existent.

In river, the concentrations of ammonia change seasonally (Fig. 3). According to the data collected in 2007, ammonia levels from January to March were obviously higher than the other months. The highest value was found in February, and the lowest value was found in May and November. The changes of ammonia levels were related to the seasonal quantity of water in the river.

In lake/reservoir drinking water sources, ammonia originates from industries, agriculture and domestic wastewater drainage. Between 2005 and 2008, there was no obvious variation in ammonia levels in different regions. It is due to the closed and semi-closed nature of the lake/reservoir drinking water sources, large quantity of water, and the long cycle of water exchange. The highest value (Table 2) of annual ammonia was found in the Southwest, Northwest and North regions, and the lowest value of annual ammonia was found in the South East region. As shown in Fig. 3, the ammonia concentrations varied seasonally, according to the climatic characteristics of the regions. The highest values of ammonia were found in different month in different regions.

In underground drinking water sources, between 2005 and 2008 year, the ammonia levels were varying in different regions. It is due to the geological permeability and the natural features of regions. The highest value (Table 2) of annual ammonia was found in the North and Northwest regions, and the lowest value of annual ammonia was found in the Southwest region.

There was no obvious seasonal change in ammonia concentrations (Fig. 3). Groundwater exists in the pore

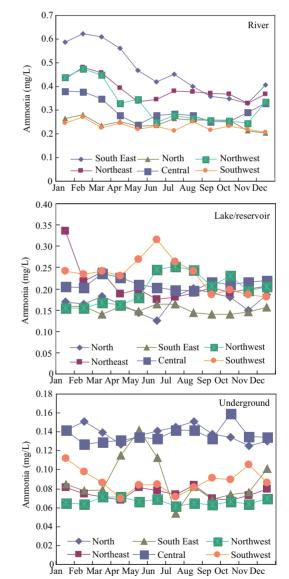


Fig. 3 Seasonal change of ammonia in river, lake/reservoir and underground drinking water sources in different regions of China in 2007.

spaces and fractures in rock and sediment beneath the Earth's surface, thus the effect of climate is limited. As for the ammonia level of groundwater in South East, the higher value found between April and June, it may be due to shallow wells, so the groundwater is related to the surface water.

# 2.4 Water purification process of ammonia in water in China

Ammonia nitrogen is a major pollutant in surface water. It can cause eutrophication and impair self-purification in lakes and rivers. Based on the above discussion, in some drinking water sources in China, the ammonia concentration is much higher than the permissible standard, especially in rivers. It is due to the large quantity of industrial and municipal wastewater discharged into the water resources.

According to the drinking water standard GB5749-2006, the limit value of ammonia is 0.5 mg/L. From 2005 to 2009, the percentages of water sources with more than 0.5 mg/L ammonia were 14.65%, 7.19% and 1.73% in river, lake/reservoir and groundwater drinking water sources, respectively. It is reported that 94.8% of water works adopted the traditional processes of "coagulation-precipitation-filtration-disinfection" (Lu et al., 2010), and the effectiveness of removing ammonia is 15% (Xiao et al., 2001). Therefore, in order to meet the drinking water standard GB5749-2006, about 11.33%, 5.44% and 1.66% of frequency in river, lake/reservoir and groundwater drinking water source respectively needs to adopt intensive water purification process.

#### **3** Conclusions

The levels of ammonia in three types of drinking water sources follow the order of river > lake/reservoir > groundwater, and the levels of ammonia concentration in river sources gradually decreased from 2005 to 2008. The annual average ammonia levels of the three types of drinking water sources were lower than 0.5 mg/L as stipulated by the drinking water guidelines of China.

The ammonia concentrations in surface water quality are affected by hydrogeology, climate change and human activities. They are changing seasonally in river drinking water sources, and varying in different regions due to geological permeability and the natural features of regions. However, there is no obvious seasonal and regional change.

In general, the ammonia content of drinking water sources is mainly from wastewater drainages, and ammonia discharges from industries, urban areas, and nonpoint sources. Wastewater is the predominant contaminant source in most drinking water sources. So, in order to ensure drinking water safety, the management of wastewater should be strengthened, and traditional water treatment systems should be improved.

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