

Metals and Metalloids in the Water–Bloom-Forming Cyanobacteria and Ambient Water from Nanquan Coast of Taihu Lake, China

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Abstract Concentrations of 12 metal(oid)s were investigated in the bloom-forming cyanobacteria and ambient water samples collected monthly between March 2009 and February 2010 at the Nanquan coast of Taihu Lake, China. The metal(oid) concentrations in ambient water decreased in the order $\text{Fe} > \text{Zn} > \text{Ni} \approx \text{As} \approx \text{Cu} > \text{Mn} > \text{Ag} > \text{Cr} > \text{Se} > \text{Cd} > \text{Co} > \text{Tl}$, while those in cyanobacteria followed a sequence $\text{Fe} > \text{Mn} > \text{Zn} > \text{Cu} \approx \text{Ni} > \text{Co} > \text{Ag} > \text{Cr} \approx \text{As} > \text{Cd} > \text{Tl} > \text{Se}$. The metal(loid) burdens removed by cyanobacteria were estimated as 164 t Fe, 12.4 t Mn, 3.6 t Zn, 2.0 t Ni, 2.0 t Cu, 0.5 t As, 0.5 t Cr, 0.4 t Cd, 0.9 t Ag, 1.1 t Co, 0.2 t Tl, and 0.09 t Se during the 2008–2010 bloom seasons.

Keywords Taihu Lake · Metal · Metalloid · Water–bloom-forming cyanobacteria

Lake Taihu, located in the delta of the Yangtze River in eastern China, is the third largest freshwater lake in China (2,428 km²) and plays a vital role as a drinking water supply, for navigation, and fishery. The lake has been experienced accelerated eutrophication and the water quality has been adversely affected by domestic and industrial effluent. Heavy cyanobacterial blooms have broken out frequently in the lake every year since the late 1990's, and caused a severe drinking water crisis in Wuxi city in May, 2007. In the bloom forming process uptake of metals by cyanobacteria may critically influence the

cyanobacterial growth, bloom mechanisms, and toxin production, in addition to phosphorus and nitrogen stimulated growth (Baptista and Vasconcelos 2006; Gadd 2010). Furthermore, the sequestration, reclamation, immobilization or detoxification of metal(loids) by cyanobacteria may represent an opportunity for bioremediation of metal pollutants, using the cyanobacteria as scavengers of metals from water bodies (El-Enany and Issa 2000; Mejáre and Bülow 2001; Gadd 2010). We therefore investigated the uptake and bioconcentration of metals and metalloids in bloom-forming cyanobacteria in Taihu Lake, to clarify the interactions of cyanobacterial blooms with metals and metalloids, and to identify possible cyanobacterial metal (loid) sinks and their fate through the food web in Taihu Lake ecosystem. In the present study, concentrations of twelve metal(loid)s were, for the first time, analyzed during cyanobacterial blooms in Taihu Lake. The aims of the present research were to investigate: (1) the characteristics and capacity of metal(loid) biouptake, bioconcentration, and bioenrichment by the lake's cyanobacteria, (2) the possible physicochemical functions of different metal(loid) species on cyanobacterial bloom mechanisms, and (3) the feasibility of using cyanobacteria as contamination/nutrient bioremediation, control or bio-mining agents in Taihu Lake.

Materials and Methods

The water–bloom-forming cyanobacteria (the blooming season from May to November) and ambient water (a year round) samples were collected monthly and immediately transported to the laboratory, between March 2009 to February 2010, from Nanquan coast in western Taihu Lake, where heavy cyanobacterial blooms occur every year. Lake

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water was first filtered by 0.22 µm syringe Millex filters (Millipore Co. Ltd., USA) to obtain water samples (30 mL/sample). An additional 1.5 mL of purified HNO₃ (MOS grade; Sinopharm Chemical Reagent Co., Ltd., Shanghai, China) was added to provide same acid basis as other solutions.

The cyanobacteria were concentrated through 75 µm sieves, rinsed six times with Milli-Q water (Millipore Co. Ltd., USA; resistivity, 18.2 MΩ/cm), dried to a constant weight at 80°C for 24 h, and measured for moisture content (Average ± SD: 95.61 ± 1.5 %). Dried samples were homogenized by grinding in a porcelain mortar, then packed with weighing paper, and then stored in a desiccator until required. A 0.1 ± 0.005 g amount of dried cyanobacteria per sample was weighed in a Teflon PTFE tube (Milestone Inc., Italy) to which 10 mL of purified nitric acid (MOS grade; Sinopharm Chemical Reagent Co., Ltd., Shanghai, China) was added. After decomposing with an ETHOS A T260 microwave digestion system (Milestone Inc., Italy), the solution was diluted to a final volume of 200 mL with Milli-Q water and transferred into acid-washed polypropylene bottles.

Concentrations of Fe, Mn, Zn, Cu, Ni, As, Cr, Cd, Ag, Co, Se and Tl were determined by an Agilent 7500ce ICP-MS (Agilent Technologies, USA) using Li, Sc, Ge, Y, In and Bi as internal standards. Quality assurance and quality control was checked by spike recoveries (94.2 %–110 % for water samples; 83.1 %–131 % for cyanobacteria samples). All concentration data are presented as µg/L for water, and µg/g dry weight for cyanobacteria, except for those specially marked.

All statistical analyses in this study were performed with SPSS version 16 (SPSS Inc., Chicago, IL, USA). Bioconcentration factor (BCF) as: $BCF = C_{algae}/C_{water}$, where C_{algae} is the metal(loid) concentration on a wet weight basis in washed algal cells, and C_{water} is that of the same metal(loid) in the growth media (e.g., ambient water).

Results and Discussion

The highest concentrations of metals were Fe and Zn in water samples of the Taihu Lake. The waterborne metal and metalloid concentrations generally decreased in the order Fe > Zn > Ni ≈ As ≈ Cu > Mn > Ag > Cr > Se > Cd > Co > Tl (Table 1). The maximum level of Fe was higher than that (100 µg/L) in a shallow and highly eutrophic Lake Kasumigaura in Japan (Inaba et al. 1997). Iron concentrations in water samples of this study varied seasonally, being higher in summer and lower in winter. Especially, Fe increased significantly in July ($p < 0.05$) and reached the highest value at October. Photosynthetic

Table 1 Annual concentrations of metals and metalloids in water samples collected from Nanquan coast of Taihu Lake, China (µg/L)

	March (n = 6)	April (n = 6)	May (n = 6)	June (n = 6)	July (n = 6)	August (n = 6)	September (n = 6)	October (n = 6)	November (n = 6)	December (n = 6)	January (n = 4)	February (n = 6)
As	1.3 ± 0.088 ^{fg}	0.20 ± 0.14 ^h	1.6 ± 0.13 ^e	1.7 ± 0.05 ^e	1.7 ± 0.042 ^e	3.3 ± 0.21 ^e	3.7 ± 0.08 ^b	4.3 ± 0.11 ^a	2.1 ± 0.07 ^d	1.4 ± 0.04 ^f	1.2 ± 0.07 ^g	1.3 ± 0.05 ^g
Cr	0.063 ± 0.154 ^d	–	0.96 ± 0.27 ^b	0.071 ± 0.174 ^d	–	1.8 ± 0.57 ^a	–	–	–	0.20 ± 0.49 ^{cd}	0.40 ± 0.11 ^e	0.81 ± 0.29 ^b
Cd	0.023 ± 0.026 ^d	2.2 ± 0.01 ^a	0.042 ± 0.008 ^d	0.036 ± 0.007 ^d	0.029 ± 0.011 ^d	0.45 ± 0.01 ^b	0.036 ± 0.014 ^d	–	0.087 ± 0.042 ^c	0.022 ± 0.032 ^d	0.055 ± 0.045 ^{cd}	–
Ag	0.015 ± 0.022 ^e	1.5 ± 0.01 ^b	0.008 ± 0.013 ^{ef}	–	0.003 ± 0.007 ^f	0.69 ± 0.04 ^c	0.002 ± 0.002 ^{ef}	4.0 ± 0.01 ^a	0.093 ± 0.105 ^d	0.013 ± 0.016 ^{ef}	0.001 ± 0.001 ^f	–
Mn	1.0 ± 0.25 ^{ef}	–	0.66 ± 0.09 ^{gh}	0.40 ± 0.14 ⁱ	0.58 ± 0.09 ^h	1.6 ± 0.19 ^c	8.0 ± 0.25 ^a	3.8 ± 0.03 ^b	1.4 ± 0.13 ^{cd}	1.3 ± 0.11 ^{de}	1.1 ± 0.03 ^f	0.73 ± 0.10 ^g
Fe	24 ± 5.6 ^f	–	32 ± 4.1 ^e	16 ± 1.4 ^g	86 ± 4.8 ^b	165 ± 4.4 ^a	21 ± 7.0 ^g	171 ± 14 ^a	52 ± 4.2 ^c	38 ± 3.2 ^d	12 ± 2.3 ^h	29 ± 3.8 ^{ef}
Ni	2.2 ± 0.13 ^{cde}	2.3 ± 0.22 ^{cd}	2.4 ± 0.10 ^c	2.1 ± 0.06 ^{de}	2.0 ± 0.06 ^{ef}	2.0 ± 0.07 ^f	2.2 ± 0.04 ^d	1.8 ± 0.05 ^g	2.2 ± 0.11 ^{de}	2.1 ± 0.05 ^{ef}	3.3 ± 0.06 ^a	2.6 ± 0.04 ^b
Cu	2.5 ± 0.41 ^d	3.5 ± 0.03 ^b	1.9 ± 0.09 ^e	1.5 ± 0.03 ^g	1.7 ± 0.06 ^f	1.7 ± 0.29 ^{ef}	1.0 ± 0.02 ^h	11 ± 0.03 ^a	0.31 ± 0.76 ⁱ	3.3 ± 0.15 ^c	2.7 ± 0.16 ^d	3.2 ± 0.08 ^c
Zn	15 ± 24 ^{deg}	6.8 ± 0.88 ^{gh}	15 ± 1.4 ^{ef}	5.0 ± 2.4 ^h	3.9 ± 7.5 th	30 ± 21 ^c	61 ± 20 ^{ab}	30 ± 19 ^{cdeg}	30 ± 12 ^{cd}	77 ± 26 ^a	41 ± 9.1 ^{bed}	68 ± 24 ^a
Se	1.7 ± 0.70 ^{bc}	–	0.70 ± 0.20 ^{deg}	0.93 ± 0.74 ^{cdef}	1.4 ± 0.81 ^{bed}	2.3 ± 0.47 ^b	0.59 ± 0.30 ^{ef}	1.8 ± 1.0 ^{bcd}	3.8 ± 0.68 ^a	0.29 ± 0.19 ^f	0.58 ± 0.20 ^{ef}	0.26 ± 0.44 ^{fg}
Co	0.06 ± 0.02 ^f	–	0.13 ± 0.006 ^g	0.11 ± 0.017 ^{gh}	0.10 ± 0.02 ^h	0.89 ± 0.01 ^b	0.20 ± 0.01 ^e	3.9 ± 0.01 ^a	0.45 ± 0.09 ^c	0.22 ± 0.03 ^{de}	0.28 ± 0.006 ^d	0.16 ± 0.02 ^f
Tl	0.02 ± 0.03 ^{ef}	1.4 ± 0.02 ^c	0.041 ± 0.014 ^e	0.021 ± 0.003 ^f	0.021 ± 0.011 ^f	4.4 ± 0.01 ^b	0.019 ± 0.012 ^f	4.5 ± 0.01 ^a	0.12 ± 0.13 ^d	0.044 ± 0.034 ^{ef}	0.011 ± 0.014 ^f	–

* The data with different superscripts in the same row are significantly different ($p < 0.05$)

– Not detected

Table 2 Temporal concentrations of metals and metalloids in water–bloom-forming cyanobacteria collected from Nanquan coast of Taihu Lake, China ($\mu\text{g/g}$ dry weight)

	May (n = 6)	June (n = 5)	July (n = 5)	August (n = 6)	September (n = 6)	October (n = 6)	November (n = 3)
As	1.1 \pm 0.29 ^{e*}	7.0 \pm 0.45 ^c	15 \pm 0.50 ^a	5.1 \pm 0.58 ^d	8.2 \pm 0.42 ^b	–	–
Cr	19 \pm 0.57 ^a	2.5 \pm 1.1 ^c	16 \pm 1.2 ^b	0.70 \pm 0.87 ^d	0.95 \pm 0.49 ^d	–	–
Cd	26 \pm 0.28 ^a	1.4 \pm 0.07 ^c	2.1 \pm 0.03 ^b	1.5 \pm 0.20 ^c	1.2 \pm 0.05 ^d	–	–
Ag	–	–	–	34 \pm 0.62 ^a	35 \pm 0.85 ^a	–	–
Mn	41 \pm 0.95 ^d	43 \pm 1.2 ^c	661 \pm 11 ^a	40 \pm 1.1 ^d	131 \pm 2.2 ^b	9.6 \pm 0.37 ^c	42 \pm 1.3 ^{cd}
Fe	328 \pm 24 ^d	1,104 \pm 31 ^b	10,332 \pm 258 ^a	228 \pm 50 ^c	747 \pm 39 ^c	0	4.2 \pm 7.3 ^f
Ni	36 \pm 0.78 ^b	1.9 \pm 0.72 ^c	40 \pm 0.71 ^a	13 \pm 1.5 ^e	15 \pm 0.62 ^d	13 \pm 0.22 ^c	18 \pm 0.27 ^c
Cu	24 \pm 0.31 ^c	5.1 \pm 0.13 ^e	38 \pm 0.86 ^a	35 \pm 1.2 ^b	38 \pm 2.2 ^a	0	14 \pm 0.27 ^d
Zn	27 \pm 2.1 ^b	20 \pm 1.2 ^c	10.6 \pm 3.2 ^a	14 \pm 3.5 ^d	27 \pm 6.5 ^b	5.1 \pm 1.2 ^e	82 \pm 18 ^a
Se	0	0	1.2 \pm 0.21 ^b	1.0 \pm 0.55 ^b	1.4 \pm 0.71 ^b	0	3.2 \pm 0.24 ^a
Co	18 \pm 0.19 ^b	0.52 \pm 0.03 ^d	4.8 \pm 0.11 ^c	31 \pm 0.58 ^a	31 \pm 0.76 ^a	0	0
Tl	0.15 \pm 0.01 ^c	0	0.002 \pm 0.005 ^d	3.0 \pm 0.06 ^b	3.02 \pm 0.07 ^b	3.6 \pm 0.23 ^a	3.6 \pm 0.11 ^a

* The data with different superscripts in the same row are significantly different ($p < 0.05$)

– Not detected

activity drastically decreased under restricted Fe conditions in freshwater lakes (Imai et al. 1999). Highest Fe levels were generally confined to July–October, coinciding with dramatic increases in cyanobacterial biomass during the same season (Su et al. 2011). This suggests that Fe might play an important role during the occurrence and development of cyanobacterial blooms in Taihu Lake. Zinc levels ranged from 3.9 to 77 $\mu\text{g/L}$ and showed an appreciable increase in August ($p < 0.05$), and reached the highest level in December. Although levels of As and Cu varied monthly, their mean concentrations usually ranged between 1 and several $\mu\text{g/L}$. Copper has been found to be poisonous to green algae *Chlorella pyrenoidosa* when dissolved Cu was $>31.8 \mu\text{g/L}$ (Yan et al. 2001), which is much higher than values from the present study. Low levels of Mn were usually maintained until July, but those elevated in August to highest level in September. Dissolved Mn increases the growth and reproduction of *Prorocentrum* at 0–20 $\mu\text{mol/mL}$ (Zhang et al. 2002).

Nickel, As, Cr, Cd and Ag are major toxic metal(loid)s. Algae growth could be enhanced by waterborne Ni at $<0.1 \text{ mg/L}$, and inhibited at 0.4 mg/L , respectively (Jiang and Lin 1995). The levels of Ni of this study ranged from 1.8 to 3.3 $\mu\text{g/L}$, suggesting that dissolved Ni might enhance cyanobacterial growth in Taihu Lake if other nutrients were present in excess. The average concentrations of waterborne As were significantly higher in August–October ($p < 0.05$) than in any other months. Elevated level of the form of arsenate (As^{3+}) is highly toxic to algae cell structure, and also reduces algae community diversity (Gao et al. 1997). Surprisingly, As^{3+} favors fast growth of *Microcystis* and stimulates microcystin production (Gong et al. 2009). In Taihu Lake, the

dominant species of the water–bloom-forming cyanobacteria are *Microcystis* (See hereafter). Higher dissolved As may probably promote the growth of *Microcystis* and inhibit that of other cyanobacterial species during water–bloom season in the lake. Highest average concentration of dissolved Cr was found in August. Likewise, the fate of Cr in the aquatic environment will vary depending on its chemical form. Trivalent is advantageous for plant growth, whereas Cr^{6+} is carcinogenic and toxic to plants (Shanke et al. 2005). In comparison with the peak level in April, dissolved Cd was always present at significantly lower levels in any other month ($p < 0.05$). The highest level of Ag in water of this study was in October, being significantly higher than in any other month ($p < 0.05$). Green algae *Chlamydomonas* and *Pseudokirchneriella subcapitata* could be inhibited by exposure to dissolved Ag (Hiriart-Baer et al. 2006), and there were variable Ag tolerances among diverse phytoplankton taxa (Zhang et al. 2006). The maximum average concentrations of waterborne As, Cd, Cr of the present study were 4.3, 2.2, and $1.8 \mu\text{g/L}$, respectively (Table 2), even though, which are below those for acceptable drinking water criteria of WHO (i.e., As $10 \mu\text{g/L}$, Cd $3 \mu\text{g/L}$, Cr $50 \mu\text{g/L}$) and US EPA (i.e., As $10 \mu\text{g/L}$, Cd $5 \mu\text{g/L}$, Cr $100 \mu\text{g/L}$) (Frisbie et al. 2002). These data suggested that “background” concentrations of these metals/metalloids might be very low in the water of Taihu Lake.

In Nanquan site, cyanobacterial blooms were only observed in May–November 2010 during our study period, and *Microcystis* was the dominant genus (91.4 %–99.5 %) in the cyanobacterial community during the aforementioned blooming season (Su et al. 2011). Bloom-forming cyanobacteria turned out to be the strongest

Table 3 Temporal variations in bioconcentration factors of water–bloom-forming cyanobacteria collected from Nanquan coast of Taihu Lake, China

	May	June	July	August	September	October	November
As	28.4	268	544	59.4	97.6	0	0
Cr	821	2,277	–	14.8	–	0	0
Cd	25,377	2,543	4,532	131	1,478	0	0
Ag	0	0	0	1,955	760,980	0	0
Mn	2,542	6,940	70,381	959	724	79.1	762
Fe	429	4,586	7,401	53.8	1,532	0	2.12
Ni	635	573.7	1,211	258	305	214	221
Cu	524	223.3	1,357	812	1,630	0	1,188
Zn	749	264.5	1,670	18.0	19.2	5.22	70.9
Co	5,622	292.7	3,000	1,356	6,827	0	0
Se	0	0	52.9	16.8	106	0	22.3
Tl	147	0	5.90	26.5	6,987	25.1	790

– Not detected in water, 0 not detected in cyanobacteria

accumulator for Fe and Mn in Taihu Lake. The highest concentrations of metals/metalloids were Fe and Mn, generally following a sequence $\text{Fe} > \text{Mn} > \text{Zn} > \text{Cu} \approx \text{Ni} > \text{Co} > \text{Ag} > \text{Cr} \approx \text{As} > \text{Cd} > \text{Tl} > \text{Se}$ (Table 2). Uptake of metals/metalloids by cyanobacteria in Taihu Lake seemed to correlate with the stages of cyanobacterial blooms. Cyanobacterial Fe concentration increased significantly in June, and reached the highest value in July, then decreased sharply from August ($p < 0.05$). Moreover, concentrations of cyanobacterial Mn, Zn, Cu, and Ni were all highest in July, and were much higher than those in any other month ($p < 0.05$). Heavy cyanobacterial blooms usually occurred around June and July in Nanquan site of Taihu Lake. The plausible reason may be that Fe, Mn, Zn, Cu, and Ni preferentially favored algae growth and promoted the occurrence of water bloom or the demand of these elements is simply high when growth and biomass were maximal. In contrast, concentrations of cyanobacterial Se, Co and Tl in cyanobacteria were higher at the late stage of cyanobacterial blooms (September–November).

The toxic metals As, Cd, Cr, and Ag showed visible temporal fluctuations throughout cyanobacterial bloom season in Nanquan site of Taihu Lake (Table 2). The highest concentrations of cyanobacterial As were in July, significantly higher than those in any other month ($p < 0.05$), whereas those of Cr and Cd were in May. Cyanobacterial Ag was only detected in August and September. Cyanobacteria, as effective metal sorbents, are an important sink for metals in aquatic environment and may play an important role in determining metal speciation and bioavailability (Baptista and Vasconcelos 2006). Therefore, the fate of cyanobacterial metals/metalloids need to be primary concern, if they are transferred and accumulated through the food web of Taihu Lake due to the dominant role of cyanobacteria as primary producers.

The highest BCFs of As, Mn, Fe, Ni and Zn in water–bloom-forming cyanobacteria were in July. Meanwhile, that of Cd was in May, Cr was June, and Ag, Cu, Co, Se and Tl were in September, respectively. Generally, the maximum BCFs of the metals/metalloids ranked in decreasing order: $\text{Ag} > \text{Cd} > \text{Mn} > \text{Fe} > \text{Tl} > \text{Co} > \text{Cr} > \text{Zn} > \text{Cu} > \text{Ni} > \text{As} > \text{Se}$ (Table 3). The potential of bloom-forming cyanobacteria biomass for metal binding must lead to the focus on the ecological and commercial significance of removal and recovery of metal/metalloid in Taihu Lake, because bound metal(loid) loads will be released from cyanobacteria when they die and decay after the bloom season, possibly playing a role in future eutrophication. According to the data of Chinese government, a total of 2,050,000 tons/wet weight (ca. 90,000 ton/dry weight) of cyanobacteria biomass had been removed from Taihu Lake in the bloom seasons of 2008–2010 (NDRC 2010). Therefore, the burdens of each metal(loid) in the cyanobacteria biomass (dry weight, calculated using the average moisture content of cyanobacteria of this study) removed during the aforementioned bloom seasons, by calculated combinedly the average concentration data in Table 2, can be estimated as 164 t of Fe, 12.4 t of Mn, 3.6 t of Zn, 2.0 t of Ni, 2.0 t of Cu, 0.5 t of As, 0.5 t of Cr, 0.4 t of Cd, 0.9 t of Ag, 1.1 t of Co, 0.2 t of Tl and 0.09 t of Se. Namely, all of these burdens were possibly removed from Taihu Lake in the same seasons. Therefore, cyanobacteria biomass removal may possibly serve as an eco-friendly and economic alternative approach (Alluri et al. 2007) to water remediation for removing waterborne metal/metalloid ions from the water bodies like Taihu Lake. The results of the present study strongly suggest that bloom-forming cyanobacteria can be possible used as an important “bio-mining” resource, due to their huge amount of biomass and high bioconcentration of the metals/metalloids. In addition, as the highest BCFs of most metals/metalloids were in July, this month seems to be the most important time for cyanobacteria biomass removal in Taihu Lake.

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