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Evaluating the contamination of microcystins in Lake Taihu, China: The application of equivalent total MC-LR concentration

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ABSTRACT

The frequent occurrence and accumulation of microcystins (MCs) in freshwater systems pose serious threats to the drinking water safety and health of human beings. However, determining the overall toxicity and environmental risks from MC exposure is complex because of the variety of MC analogues and their respective toxicities. To address this issue, we conducted a survey of particulate (intracellular) and dissolved (extracellular) MC in Lake Taihu from 14 sampling sites in the northern part of the lake, and 32 stations throughout the entire lake, over a 16-month period. We propose a novel indicator, total MC-LR concentration (TLR), defined as the total concentration of MC-LR after transforming other MC variants (i.e., MC-RR and MC-YR) to an equivalent toxicity of MC-LR. Intracellular concentrations of TLR (iTLR) were usually observed, with the maximum values in July and October 2013 corresponding to periods of peaks in phytoplankton biomass. In contrast, extracellular concentrations of TLR (eTLR) were highest in May and June 2014. These differences in temporal patterns exhibited by LR, TLR, and TMC between intracellular and extracellular MC may be attributed to the influence of environmental variables. In addition, the distribution of iTLR and eTLR in the entire lake showed clear spatial heterogeneity. MC concentrations were greatest in the northern part of the lake during warm months, especially in Meiliang Bay. Based on the strong linear relationships between TLR and the concentration of chlorophyll-a (Chl-a), as well as TLR and the cell density of cyanobacteria, we propose not-to-exceed safety thresholds for Chl-a of 21.28 μg/L in the northern lake and 23.26 μg/L in the whole lake, which are paired with safety thresholds for cyanobacterial cell densities of 2.21×10^8 cells/L and 1.15×10^8 cells/L, respectively. The application of this newly proposed indicator, TLR, may contribute to better evaluation of overall MC toxicity and provide guidance on recommended limits for Chl-a concentration and cyanobacterial cell density in other freshwater ecosystems.

1. Introduction

In the past few decades, cyanobacterial blooms have become a major scientific problem in global freshwater systems. Some large freshwater lakes are increasingly experiencing severe cyanobacterial blooms around the world, such as Lake Erie in USA (Rinta-Kanto et al., 2009), Lake Suwa in Japan (Chan et al., 2007), and Lakes Poyang (Zhang et al., 2015), Chaohu (Yu et al., 2014b), Dianchi (Wu et al., 2014), and Erhai (Yu et al., 2014a) in China. Furthermore, global warming and excessive nutrient inputs are simultaneously contributing to the frequency and intensity of large algal blooms (O'Neil et al., 2012). Harmful cyanobacterial blooms can produce diverse kinds of cyanotoxins, such as microcystins (MCs), which are the most common hepatotoxic compounds. MCs are relatively stable in water and are difficult to remove by traditional water treatment techniques (van Apeldoorn et al., 2007). As a consequence, the occurrence of MCs can pose a serious threat to the drinking water supply, affect aquatic organisms by bioaccumulation (Rezaitabar et al., 2017; Xie et al., 2005; Xie et al., 2007), and even endanger mammals and human beings (Dittmann and Wiegand, 2006; Zhao et al., 2015). Despite numerous reports about the presence and distribution of MCs in lakes and reservoirs throughout the world (Otten et al., 2012; Rinta-Kanto et al., 2009; Sinang et al., 2013; Singh et al., 2015; Zhang et al., 2015), it is still difficult to accurately assess MC contamination in different lake systems. Each lake or reservoir has its own limnological and meteorological characteristics, which may lead to differences in the dominant congener and composition of MCs in the water column (Amé et al., 2010; Gurbuz et al., 2009). At present, there is no universal method to

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compare MC contamination within and among freshwater systems. Given these issues, it is important for resource managers to have an index that can identify the severity of environmental risks caused by MC to gain a better understanding of their overall toxicity in freshwater systems (Xie et al., 2016). Unfortunately, such an indictor has not been proposed to date.

In general, MCs stay in the cell while cyanobacterial cells grow and then are released to the water after cell lysis or death. Therefore, MCs in the water column are usually divided into particulate (intracellular) MC and dissolved (extracellular) MC. Most studies have focused on intracellular MC because about 90% of MC is retained in the cyanobacteria cell. However, Sakai et al. (2013) applied the ratio of extracellular to total MC (i.e. E/T) to evaluate the conditions of a bloom cycle; they found the E/T ratio was a better predictor of clarifying the growth stage of algal blooms than MC concentrations alone (Sakai et al., 2013). This suggests that the most robust evaluation of MC in the water column would include consideration of both intracellular and extracellular MC at the same time.

Over ninety MC variants have been identified to date and each MC analogue has a different degree of toxicity (van Apeldoorn et al., 2007). MC-LR, −RR, and −YR are considered the three most common MC variants, with MC-LR regarded as the most toxic one, followed by MC-RR and MC-YR (Gupta et al., 2003). This difference in toxicity must be taken into account when evaluating the potential toxic influence of MC. For example, an indicator of "relative toxic potential", which takes into consideration both MC diversity and their relative toxicity, and toxicity equivalent factor (TEF) (Wolf and Frank, 2002) was proposed to compare the MC risks among different lakes. However, their study didn't distinguish particulate from dissolved MC. We believe an even more robust approach for risk assessment is to include the equivalent MC-LR values instead of the total MC concentration.

Given the World Health Organization's (WHO) guideline of MC-LR in the drinking water, MC concentrations can be used to evaluate potential safe levels of cyanobacteria biomass (Shang et al., 2015) and identify thresholds of total nitrogen and chlorophyll a (Chl-a) concentrations not to be exceeded (Yuan et al., 2014) based on their strong linear relationships with MC. While many relationships between Chl-a and MC concentrations are positive, they also vary considerably depending on the environmental conditions of the lake (Stumpf et al., 2016). Yuan et al. (2014) recommended a national (USA)-scale threshold of Chl-a concentration of 37 μg/L, paired with a total nitrogen concentration of 570 μg/L, based on frequency of occurrence of MC greater than 1 μg/L. Based on a previous study in Lake Taihu, the possible safety value for Chl-a was suggested as 12.43 μg/L (Otten et al., 2012). The inconsistency in findings relating the association between Chl-a (i.e. cyanobacteria) and MC production among and within different lakes highlights the need for further examination of this relationship before it can be applied to other lakes.

Lake Taihu is the third largest freshwater lake in China and it has suffered from harmful cyanobacterial blooms for many years (Xu et al., 2017). Cyanobacterial blooms occurred only in Meiliang Bay in Lake Taihu prior to the 1990s (Ying et al., 2015). However, due to the influence of economic development and human activities, cyanobacterial blooms are now a frequent occurrence in the northern bays and have spread to the center and southern parts of Lake Taihu over the last two decades (Duan et al., 2009). Regularly occurring blooms, dominated by the MC-producing cyanobacterium, *Microcystis* sp., as well as reports about MC concentrations with different MC congeners, have been well documented in Lake Taihu (Otten et al., 2012; Song et al., 2007; Xu et al., 2008; Ye et al., 2009). The current approach for MC monitoring and risk assessment concentrates mainly on the environmental fate of MC in the water, algal cells, sediment, fish and other aquatic organisms (Gurbuz et al., 2016; Li et al., 2017). These studies are not sufficient to provide an overall evaluation of MC risk in the water column. Thus, it has become increasingly important to have a complete and accurate assessment of MC content, toxicity, and environmental risk, as well as potential safety thresholds for key environmental factors in large, shallow, and eutrophic Lake Taihu. From a lake management perspective, it would be desirable to develop an indicator that could provide an accurate assessment of the overall toxicity of MC. Recommended thresholds for Chl-a concentrations and cyanobacterial abundance are also needed to guarantee the drinking water safety of Lake Taihu.

The objectives of this study were to (1) examine the temporal variation and spatial distribution of cyanobacteria in the entire Lake Taihu, (2) develop a new indictor, TLR, to evaluate the contamination of MC and explore its monthly dynamics and spatial distribution, (3) explore the relationship between environmental factors and intracellular TLR as well as with extracellular TLR in the northern lake and the whole lake, and (4) recommend safety thresholds for the concentration of Chl-a and the cell density of cyanobacteria based on TLR concentration in the water column.

2. Materials and methods

2.1. Study area and sample collection

Lake Taihu (30°56′–31°34′ N, 119°54′–120°36′ E) is located in the delta of the Yangtze River (Fig. 1), with a surface area of 2338 km^2 and an average depth of 1.9 m (Qin et al., 2007). Between July 2013 and December 2014, monthly surface (0–0.5 m) water samples were collected from 14 sampling sites (1, 2, 3, 4, 5, 6, 7, 8, 10, 13, 14, 16, 17, 32) that were located mostly in the northern areas of Lake Taihu; in addition, seasonal water samples were collected from 32 stations (1–32) that were uniformly distributed through the entire lake (Fig. 1). The four seasonal divisions were February (winter), May (spring), August (summer), and November (autumn). All the water samples were collected with a vertical water sampler and transported to the Taihu Laboratory for Lake Ecosystem Research (TLLER) of the Chinese Academy of Sciences, which is located on the shoreline of Meiliang Bay. For each

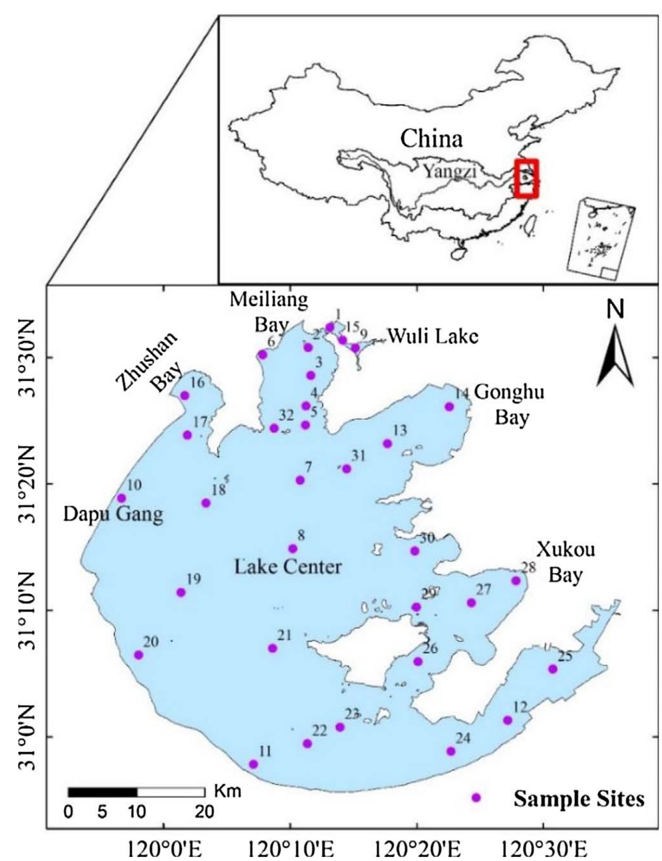

Fig. 1. Map of Lake Taihu and the distribution of sample sites displayed by the dots.