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Toxic cyanobacteria in water supply systems: data analysis to map global challenges and demonstrate the benefits of multi-barrier treatment approaches

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Abstract

The occurrence of toxic cyanobacteria in surface waters and their impact on drinking water treatment plants (WTPs) is a growing, global concern. The main objective of this paper was to assess the presence of cyanobacteria in surface water sources and associated cell removal efficiency in full-scale WTPs across the world. Previously unpublished data was collected from WTPs experiencing cyanobacterial blooms in either their managed surface waters or recreational waters. In total, data were collected from 31 surface water sources and 21 WTPs in North and South America, Europe, Asia, and Australia. The most commonly detected species were identified in both the surface waters, including *Microcystis*, *Anabaena*, *Nostoc*, *Oscillatoria*, and *Planktolyngbya*, and water treatment plant intakes, including *Microcystis*, *Cylindrospermopsis*, *Anabaena*, *Pseudanabaena*, and *Aphanizomenon*. In the intakes, cyanotoxins and taste and odor (T&O) compounds frequently co-occurred (80%) as did multiple cyanotoxins (39%). Conventional treatment saw a wide range of removal depending on the density of cells, species, and metabolites. Although more than 28% of sampling events displayed negligible or even negative removals of metabolites or cells due to accumulation within the clarifier, filtration, or water recycling, the presence of multiple treatment barriers, particularly advanced treatments like granular activated carbon and nanofiltration, allowed for the cells and their metabolites of concern to be removed to below guideline values. During treatment, total microcystins were often removed without releasing their intracellular fraction, whereas cylindrospermopsin, geosmin, and 2-MIB were commonly detected as entirely extracellular at the plant's intake. The maximum tolerable cell (MTC) counts for cyanotoxin- or T&O-producing cells were calculated using guideline values, average removal efficiency, and the average cell quota derived from data. The 21 WTPs in this work were found to be able to tolerate approximately 74,000 cells/mL for microcystins, 8,000 cells/mL for cylindrospermopsin, and 1,200 cells/mL for geosmin and 2-MIB before exceeding guideline values. These levels provide guidance for water treatment plant operators to assess the potential risk associated with cells capable of producing cyanotoxins or T&O compounds.

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Key words: cell quota, cyanobacteria, cyanotoxins, drinking water treatment, maximum tolerable cell count, taste and odor compounds

Highlights

- Distinguish between the abundance and presence of cyanobacteria.
- Assess the performance of a full-scale treatment process for the successful removal of cyanobacteria and their associated metabolites.
- Determine the range of cyanotoxin or T&O compound concentration per cell.

Graphical Abstract

INTRODUCTION

In recent years, potentially toxic cyanobacteria have been increasingly detected in water supply systems across the planet (Svrcek & Smith 2004; AFSSA 2006; Hobson *et al.* 2010; Newcombe *et al.* 2010; Bishop *et al.* 2018; Zamyadi *et al.* 2019). Their occurrence in drinking water sources and the consequent impact on water treatment plants (WTPs) is an increasing global concern (Chen *et al.* 2019; Zamyadi *et al.* 2019). Climate change effects, e.g., rising water temperature, human activities including increased agricultural nutrient loads, and population growth, may intensify toxic cyanobacterial blooms, even in more temperate climates (Wiedner *et al.* 2007; Elliott 2012; Paerl & Paul 2012; Sinha *et al.* 2012; Zamyadi 2014). However, factors affecting the composition of species and the presence of cyanotoxins are complex (AFSSA 2006; Zamyadi *et al.* 2019).

The most undesirable effects of cyanobacterial blooms in water supply systems are related to the human health risks, esthetic water quality, and the operation of WTPs (Chorus & Bartram 1999; Carmichael *et al.* 2001; Kommineni *et al.* 2009; Newcombe *et al.* 2010; Zamyadi *et al.* 2013a, 2013b; Chen *et al.* 2019). However, the production and breakthrough of potent cyanobacteria and their cyanotoxins (e.g., microcystins, cylindrospermopsin, saxitoxin, and anatoxins) is the most important public health risk associated with these blooms (Chorus & Bartram 1999; Newcombe *et al.* 2010; Zamyadi *et al.* 2012; Chen *et al.* 2019). In addition to gastrointestinal, cytotoxic, and hepatotoxic effects, cyanotoxins have been hypothesized to be a possible cause of neurodegenerative illness (Holtcamp 2012; Bradley *et al.* 2013). While releases of taste and odor (T&O) compounds, e.g., geosmin and 2-methylisoborneol (2-MIB), during these blooms do not affect human health, they negatively affect the esthetic water quality and result in customer complaints (Chen *et al.* 2019). Anatoxins and T&O compounds are difficult to remove via conventional treatment, including coagulation/clarification, filtration, and oxidation, because they are not effectively degraded by the most commonly employed oxidant, chlorine (Ho *et al.* 2007; Kommineni *et al.* 2009; Faruqi *et al.* 2018; Chen *et al.* 2019). Furthermore, sudden changes in raw water characteristics (e.g., pH) due to cyanobacterial cells result in diverse impacts on the treatment processes including poor flocculation performance, increases in the consumption of coagulant, and the generation of disinfection byproducts from the organic matter released by the algae (Coral *et al.* 2013; Zamyadi *et al.* 2015a, 2019; Chen *et al.* 2019). Furthermore, the treatment upsets could lead to cell accumulation during treatment processes within WTPs; there are documented instances of cyanobacteria accumulation occurring in sludge bed of clarifiers, in surface water inside sedimentation tank, in filter media, in surface water

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inside filtration basin, and in sludge thickener or lagoons (Zamyadi 2014; Pestana *et al.* 2016; Chen *et al.* 2019; Zamyadi *et al.* 2019).

Results of several years of monitoring in different surface water sources across the globe demonstrate that water authorities and water treatment utilities need to have a proper knowledge of the presence of potentially toxic and T&O-producing cyanobacteria and their proliferation in (a) source waters and (b) within treatment plants (AFSSA 2006; Zamyadi *et al.* 2012, 2013b, 2019; Chen *et al.* 2019). Understanding the treatment barriers that are commonly employed for drinking water production and assessment of their limitations is essential for proper evaluation of the breakthrough risk associated with cyanobacterial cells and their metabolites. Thus, this work presents a compilation and analysis of data from across the globe, relating to cyanobacteria and their metabolites in surface waters and WTPs. The data were extracted from the peer-reviewed literature, case studies and obtained through personal correspondence. The aim was to compare data in order to (i) distinguish between the abundance and presence of genera, (ii) assess the performance of a full-scale treatment process for the successful removal of cyanobacteria and their associated metabolites, and (iii) determine the range of cyanotoxin or T&O compound concentration per cell.

DATA COLLECTION AND ANALYSIS

Data on the occurrence of cyanobacteria in surface waters were collected from the published literature. The data included 31 different locations with 2,140 sampling events that occurred during cyanobacterial blooms of varying intensities (range from <1,000 up to >200,000 cells/mL) in North and South America, Europe, Africa, Asia, and Australia, from 1994 to 2019 (Heresztyn & Nicholson 1997; Bumke-Vogt *et al.* 1999; Fastner *et al.* 1999; Tarczyska *et al.* 2001; Maatouk *et al.* 2002; Pietsch *et al.* 2002; Schmidt *et al.* 2002; Zurawell 2002; Ballot *et al.* 2003; Höger 2003; Nasri *et al.* 2004; Albay *et al.* 2005; Hotto *et al.* 2005; Jurczak *et al.* 2005; Mankiewicz *et al.* 2005; Costa *et al.* 2006; Messineo *et al.* 2006; Ndebele & Magadza 2006; Haddix *et al.* 2007; Izaguirre & Taylor 2007; Törökné *et al.* 2007; Yen *et al.* 2007; Dai *et al.* 2008; Kagalou *et al.* 2008; Al-Tebrineh *et al.* 2012; Lürling & Faassen 2012; Zamyadi *et al.* 2012, 2013a, 2013b, 2019; Somdee *et al.* 2013; Amrani *et al.* 2014; Duong *et al.* 2014; Bowling *et al.* 2016; Chiu *et al.* 2016a, 2016b). A map of the locations where data were collected is shown in Supplementary Material, Figure S1. The collected data included taxonomic enumeration and speciation analysis of cyanobacteria, and their associated cyanotoxins, including cylindrospermopsin (CYN), microcystin (MC), saxitoxin (SAX), and T&O compounds, including geosmin and 2-MIB. For analysis, samples were divided into those collected during very intense blooms (>200,000 cells/mL) and the blooms below that threshold. The maximum cyanotoxin or T&O compound concentration per cell, also called the cell quota (fg/cell) or C_{quota} value, was calculated for five metabolites (Table 1).

Table 1 | Average, minimum, and maximum cell quota (concentration of metabolite in fg per cell) for five cyanobacterial metabolites

Metabolite	Average (fg/cell)	Minimum (fg/cell)	Maximum (fg/cell)
Microcystin	142	23	443
Cylindrospermopsin	610	0.4	13,500
Saxitoxin	13	5	20
Geosmin	21	1	53
2-MIB	7	3	12

Generated from surface water and intake WTP samples.

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An additional 62 sampling events were obtained from 21 WTPs (Supplementary Material, Figure S1) through the peer-reviewed literature, case studies and personal correspondence. Similar to the source water sites, data were collected from WTPs experiencing cyanobacterial blooms or detectable levels of cyanobacteria metabolites. The following regions were represented in the data: Queensland, Australia; Quebec, Canada; Sulejow, Poland, Finland, and Wisconsin, USA; Lengg, Switzerland; Saint-Caprais, France; and Tai Hu, Taiwan. The data from full-scale plants were collected between 1998 and 2019 (Karner *et al.* 2001; Tarczyska *et al.* 2001; Maatouk *et al.* 2002; Schmidt *et al.* 2002; Hoeger *et al.* 2005; Rapala *et al.* 2006; Zamyadi *et al.* 2012, 2013a, 2013b, 2016, 2019; Chiu *et al.* 2016a, 2016b). These sampling events included the following treatment processes: pre-treatment (ozonation, potassium permanganate, or copper sulfate), powdered activated carbon (PAC), coagulation/flocculation/sedimentation, coagulation/flocculation/dissolved air flotation (DAF), coagulation/flocculation/direct filtration, filtration (including rapid, slow, ultra, and nano), ultraviolet (UV) disinfection, granular activated carbon (GAC), and oxidation (chlorination and ozonation). The specific processes employed at the different WTPs along with the number of sampling events for each plant are shown in Supplementary Material, Table S1. The type of data collected was not consistent between sampling events due to bloom variability; for example, total 2-MIB was reported for some events, whereas total cells, cyanobacteria genera, and intracellular and extracellular information for all five metabolites were reported for other events.

Cumulative removal was calculated after each treatment process to compare the total cells, cyanotoxins, and T&O compounds to the levels in the raw water. To compare aggregate data, Student's *t*-tests with unequal variance were applied. The maximum, minimum, first and third quartile, median, and outliers were calculated to present the data collected from different treatment processes within boxplot graphs. Using the minimum, average, and maximum removal efficiency (η) of each treatment process, the maximum tolerable cell count (MTC) in cells/mL is calculated using the following equation:

$$\mathrm{MTC} = \frac{\mathrm{GV}}{C_{\mathrm{quota}} \times (1 - \eta)} \tag{1}$$

where GV is the guideline value for the cyanotoxin or T&O compound in treated water and C_{quota} is the average metabolite concentration per cell (Table 1). Due to the international nature of the reviewed case studies, the World Health Organization (WHO) recommended GVs of 1 μg/L for MC-LR, 0.7 μg/L for CYN (chronic value), and 3 μg/L for SAX (acute value) were adopted in this work (Chorus & Bartram 1999; Newcombe *et al.* 2010). For geosmin and 2-MIB, the GV was 10 ng/L or the level at which humans can detect T&O compounds (Hobson *et al.* 2010).

RESULTS AND DISCUSSION

Presence of cyanobacteria in surface waters

Of the more than 2,000 samples collected from 31 sites around the globe, 23 genera were detected with individual counts ranging over ten orders of magnitude (Figure 1). *Microcystis* sp. (*M. flos-aquae* and *M. aeruginosa*) were present in 84.4% of the source water samples compiled, followed by *Anabaena* sp. (Figure 1(a)). Apart from *Microcystis* sp., the next four commonly detected genera (*Anabaena*, *Nostoc*, *Oscillatoria*, and *Planktolyngbya*) represent the primary cyanobacterial genera of interest for surface waters that may be used for recreational purposes (Figure 1(a)).

Multiple potentially toxic (i.e., microcystin-, cylindrospermopsin-, and saxitoxin-producing) cyanobacterial species (between 2 and 11 species) were found to co-occur in the surface water samples,

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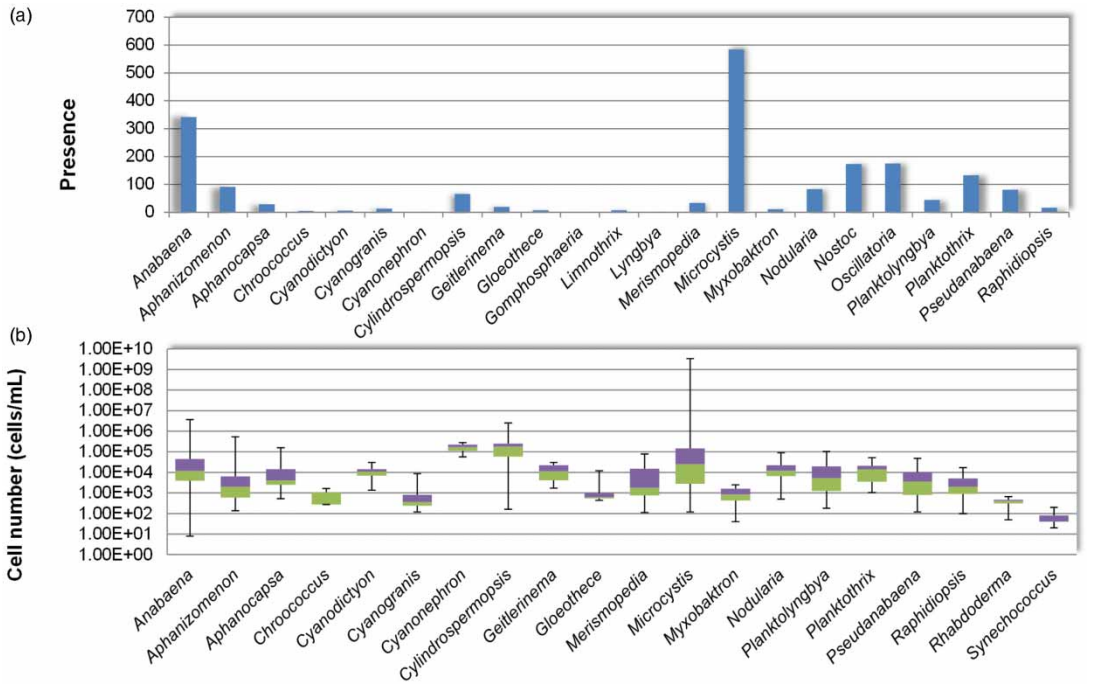

Figure 1 | (a) Cyanobacteria genera identification (presence) and (b) genera abundance (cell count) in 31 surface waters.

showing clearly that blooms frequently contain more than one potentially toxic species (Figure 1). On the basis of cell counts, the average ratio of potentially toxic species to the total phytoplankton community in all collected samples ranged from 34 to 61%. However, when the very intense bloom samples (i.e., cell numbers over 200,000 cells/mL) were separated from the remaining water samples, up to 99% of the phytoplankton community comprised potentially toxic cyanobacterial species. Notably, the quantitative extent will depend on a number of factors including sampling effort. This trend is indicative of the ability of potentially toxic cyanobacteria to outcompete other phytoplankton communities (Elliott 2012; Paerl & Paul 2012). An evaluation of inland lakes of the United States saw similar results with cyanobacteria dominating 76% of samples and 95% of samples containing species capable of producing microcystins (Loftin *et al.* 2016). In this work (Loftin *et al.* 2016), the detection of toxin-producing cyanobacteria translated to high levels of microcystin in the surface waters. The very intense blooms had a maximum of 9,750 μg/L total microcystins or an order of magnitude higher than that found in the samples containing <200,000 cells/mL at 980 μg/L. Both these levels significantly exceed the guideline values recommended for issuing swimming advisories for recreational waters at 20 or 8 μg/L from Health Canada and the U.S. Environmental Protection Agency (U.S. EPA), respectively (Health Canada 2012; U.S. EPA 2017); however, not all of the surface waters in this dataset are used for recreational purposes.

Removal of cyanobacteria during water treatment

In the surface waters that act as drinking water intakes, cyanobacterial blooms are of concern due to their ability to negatively impact treatment, e.g., disruption of clarification and filtration, and the potential to release toxic metabolites into finished drinking water (Kommineni *et al.* 2009; Zamyadi *et al.* 2019). In the current study, the cyanobacterial cell data from 21 WTPs were analyzed to determine the cumulative removal after each stage of treatment (as outlined in Supplementary Material, Table S1). A range of concentrations and species were detected in the raw water of the WTPs with total cell counts ranging from <500 cells/mL up to a maximum of 2.1×10^6 cells/mL. When the raw water cell concentrations were binned according to the Water Quality Research Australia

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(WQRA) alert levels, samples with total cell counts >65,000 cells/mL made up 56% of the data, 34% had between >6,500 and 65,000 cells/mL, and 10% were <6,500 cells/mL (Newcombe *et al.* 2010). The dominant species observed in the raw water were similar to those found in the surface waters and included *Microcystis* sp., *Cylindrospermopsis*, *Anabaena* sp., *Pseudanabaena* sp., and *Aphanizomenon*.

Cumulative removal for total cyanobacteria cells following the last stage of treatment where data were collected (e.g., post-GAC, post-chlorination, or post-sand filtration) averaged 98 ± 9.3% with the bulk of the removal occurring during the first or second stage of treatment, e.g., coagulation/flocculation/sedimentation, coagulation/flocculation/DAF, or sand filtration (Figure 2). Although the multi-barrier treatment trains were able to mitigate the total cell counts, 8% of samples ($n = 50$) saw higher levels of cell density relative to the raw water post-sedimentation or DAF. Following filtration (sand, UF, and NF), 28% of sites had their total cell count rise relative to the influent; however, the post-filtration cell counts never exceeded the levels in raw water (Figure 2). The scale of these increases depended on the mechanism behind it with increases post-sedimentation or DAF in the range of 10^4 cells/mL compared with 10^1–10^3 cells/mL following NF, UF, and sand filtration. For sedimentation, the removal failure was attributed to the accumulation of cells within the clarifier (e.g., as a scum on the surface or in the sludge bed) due to poor coagulation. For DAF, recycling of the supernatant from a lagoon treating the filter backwash water and DAF sludge float was the cause of this increase (Pestana *et al.* 2016). The breakthrough of cells post-filtration was attributed to the accumulation of cells on the filtration media (Pestana *et al.* 2016; Zamyadi *et al.* 2019).

The mean cumulative removal post-sedimentation (65 ± 96%, $n = 23$) was not statistically different from that observed for post-DAF (58 ± 90%, $n = 24$), due to the high variability in data. To reduce this variability, the outlier (negative) values were removed, but still no significant difference was observed. This did not match with the anticipated outcomes, in which the hypothesis was that separation via the DAF process would be more successful for cyanobacteria due to their buoyancy (AWWA 2010; Newcombe *et al.* 2010). However, the results presented in the current review were likely driven by the WTP raw water cyanobacteria levels: the DAF plants had an average of 680,000 ± 5% cells/mL in the raw water which was far higher than the average cell concentration entering the sedimentation plants of 99,000 cells/mL. Notably, separation for these processes is only as successful as the upstream coagulation–flocculation process. Post-filtration the WTPs with DAF upstream (99 ± 0.75%) were

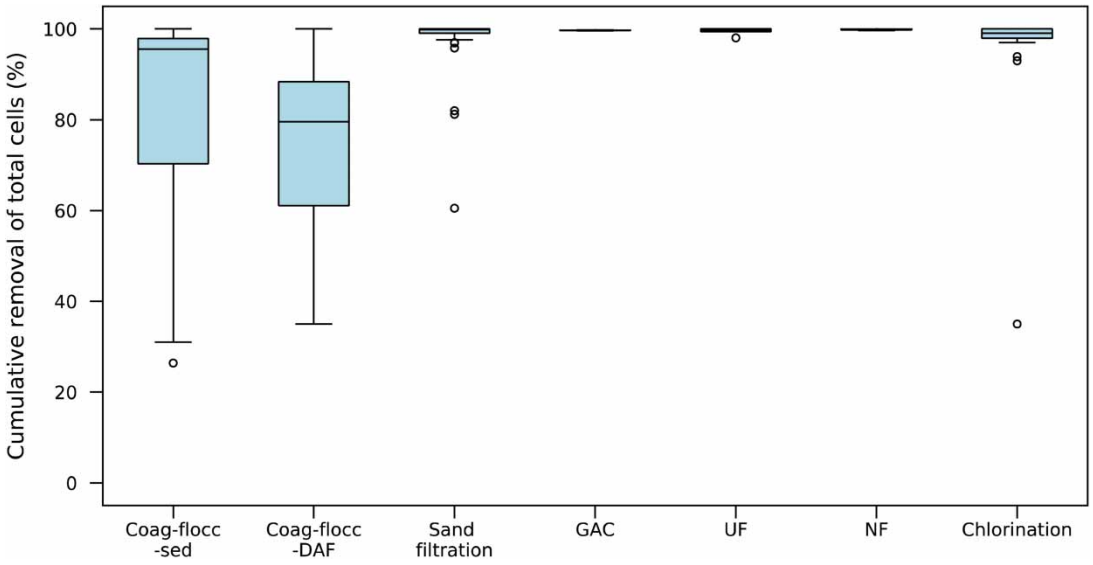

Figure 2 | Cumulative removal of total cyanobacterial cells during treatment ($n = 50$). Boxplots show the minimum, maximum, median, first quartile, and third quartile, and the circles reflect statistical outliers.

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slightly more effective than those employing sedimentation (96 ± 9.3%), though not significantly. Subsequent treatment processes (e.g., GAC, UF, NF) appeared to provide more consistent removal (cumulative removal was always >98%) but experienced lower cell density at their influent (Figure 2).

In addition to monitoring the total cyanobacterial cell removal, the removal of select species (*Microcystis* sp., *Cylindrospermopsis*, *Anabaena* sp., *Aphanizomenon*, and *Pseudanabaena* sp.) were also evaluated (Supplementary Material, Figure S2). Following coagulation–flocculation–sedimentation, *Microcystis* sp. (98 ± 5.3%, $n = 22$) and *Anabaena* sp. (95 ± 12%, $n = 23$) cells were removed more efficiently on average than the *Aphanizomenon* (43 ± 81%, $n = 21$) and *Pseudanabaena* sp. (86 ± 29%, $n = 20$), due to the incidences of clarifier acclimation for *Aphanizomenon* and *Pseudanabaena* sp. This effect carried over to sand filtration, likely due to a combination of higher cells at the head of the filtration system and additional acclimation in the filters. As a result, *Microcystis* sp. saw significantly better mitigation. *Microcystis* sp. (94 ± 8%, $n = 11$) also had significantly higher removal post-DAF as compared with *Cylindrospermopsis* (83 ± 19%, $n = 21$).

These data highlight the need to monitor cyanobacterial cells following coagulation processes, water recycling, and every type of filtration (e.g., conventional sand and advanced treatment with UF/NF), rather than relying only on the raw water values. Although the increases following treatment were primarily observed at warm-weather locations, e.g., Taiwan and Australia, the potential for cell acclimation during filtration was also observed during the summer months in Canada. The impact of these increases has negative consequences for downstream processes and can lead to treatment plants relying on cells to be removed by processes not intended for that purpose, e.g., GAC and chlorination (Supplementary Material, Figure S2).

Removal of cyanotoxins and T&O compounds during water treatment

The removal of total cyanotoxins and T&O compounds during treatment is critical to the safety and customer acceptance of the finished water. To effectively remove the total metabolite, treatment must address both the intracellular and the extracellular fractions. In the 21 WTPs, cumulative removal was monitored for three cyanotoxins: microcystins, cylindrospermopsin, and saxitoxin and two T&O compounds: 2-MIB and geosmin (Figures 3 and 4). For the 25 sampling events where T&O compounds and the three cyanotoxins were monitored, 80% saw the co-occurrence of at least one cyanotoxin,

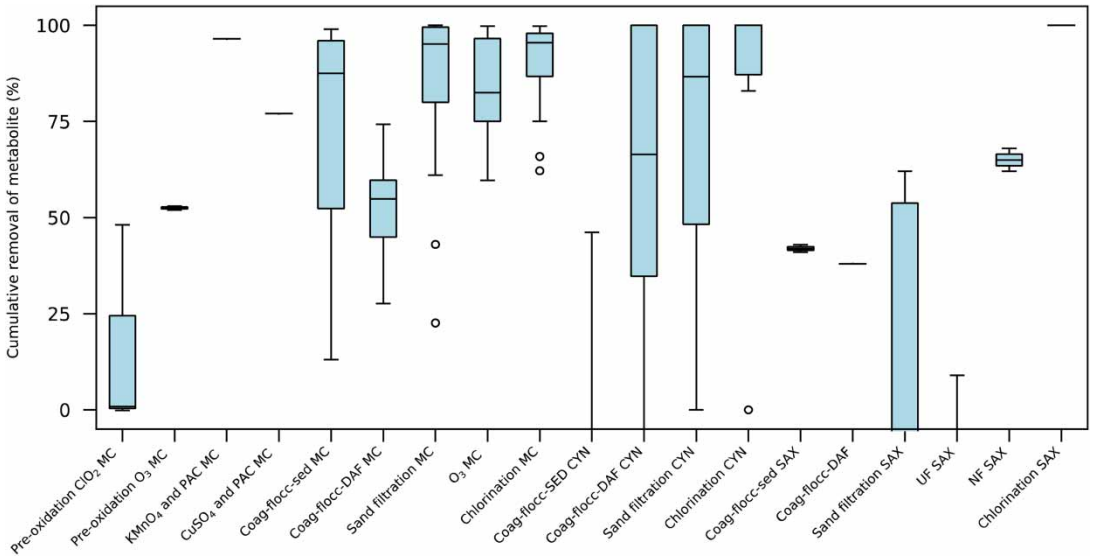

Figure 3 | Cumulative removal of total MC (microcystins, $n = 38$), CYN (cylindrospermosin, $n = 19$), and SAX (saxitoxin, $n = 4$). Boxplots show the minimum, maximum, median, first quartile, and third quartile, and the circles reflect statistical outliers.

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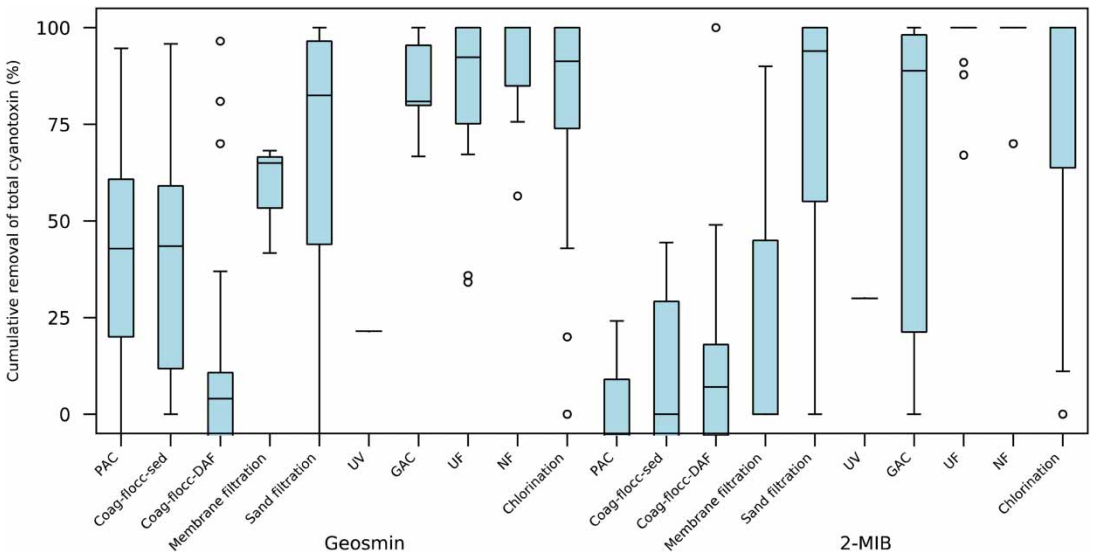

Figure 4 | Cumulative removal of total geosmin ($n = 41$) and 2-MIB ($n = 36$). Boxplots show the minimum, maximum, median, first quartile, and third quartile, and the circles reflect statistical outliers.

typically cylindrospermopsin, alongside both T&O compounds during treatment. The co-occurrence of multiple cyanotoxins was observed in 39% of the 28 sampling events. Similarly, an evaluation of inland U.S. lakes observed that cyanotoxins and T&O compounds co-occurred in 91% of samples and multiple cyanotoxins were found in 48% of samples (Graham *et al.* 2010).

For the cyanotoxins, pre-treatment varied depending on the plant with some plants employing PAC, ozone, chlorine dioxide, or a combination of potassium permanganate and PAC or copper sulfate and PAC (Figure 3). For total microcystins, ozonation was found to be the most effective pre-oxidation technique, followed by a combination of copper sulfate (8-day exposure) and PAC, potassium permanganate and PAC, and finally chlorine dioxide. Ozonation and potassium permanganate are effective against both the intracellular and extracellular fraction of microcystins at appropriate doses, but copper sulfate and chlorine dioxide cannot remove extracellular microcystins (Coral *et al.* 2013; Fan *et al.* 2013; Zamyadi *et al.* 2015b). Pre-oxidation is often applied to meet treatment goals other than cyanobacteria, e.g., control of biofilm, enhanced coagulation, or reduction of disinfection byproducts. As a result, the dose may be insufficient to both lyse the cyanobacterial cells and oxidize the released extracellular fraction. PAC dosed after the initial oxidant exposure can be effective to sorb the released metabolite but requires sufficient exposure time and doses (Ho *et al.* 2011). The timing of PAC addition to post-oxidation or -algicide application is also important. Recent work has shown that low pre-oxidation doses that initially (after 30-minutes) show no impact on cells can result in delayed release of extracellular cyanotoxins (Zamyadi *et al.* 2020). The full-impact (in terms of extracellular cyanotoxin release) of an oxidant or algicide may not be realized until up to 7 days after the initial exposure, even when oxidant is no longer present (Zamyadi *et al.* 2020). These releases can be mitigated by downstream processes, e.g., chlorination, GAC, or NF, but multi-barrier treatment is necessary to eliminate extracellular toxins after the delayed release.

Following pre-oxidation, coagulation–flocculation–sedimentation and DAF were evaluated for both total microcystins and cylindrospermopsin (Figure 3). Although both processes have the potential to eliminate cells (i.e., intracellular cyanotoxins), they do not mitigate the extracellular fraction (Ho *et al.* 2013). The average total microcystin removal post-sedimentation ($62 \pm 30\%$) was slightly higher than that of post-DAF ($52 \pm 14\%$). No negative removal was observed after DAF and only one instance was reported post-sedimentation. This indicates that the microcystin-producing cells did not accumulate during clarification nor were they returned to the head of the plant via the water recycling system. The subsequent treatment process, e.g., filtration, ozonation and oxidation via chlorine or ozone were

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effective for microcystin treatment. Sand filtration has the potential to provide both physical separation of cells in addition to biodegradation, provided that the columns are acclimated to the presence of cyanotoxins (Ho *et al.* 2012; Maghsoudi *et al.* 2015). Secondary chlorination and ozonation have the potential to effectively remove total microcystins through cell lysis and subsequent oxidation, but their efficacy depends on the dose, species, state of bloom, and background matrix, e.g., organic matter and pH (Fan *et al.* 2013). Poor removal only occurred when total cyanotoxins were intracellular at this stage of treatment.

For cylindrospermopsin, inconsistent removal was observed following DAF (range of −48 to 99%) due to a combination of the incomplete cell removal, the limited impact on extracellular cyanotoxins, and the negative removals from the recycled water entering the head of the plant. Sand filtration also saw a broad range of removals (0–99%), due to the lower biodegradation potential of cylindrospermopsin compared with microcystins and the release of cells post-filtration (Ho *et al.* 2012). A limited dataset was available for saxitoxin, but the median removal of post-sedimentation and post-DAF was similar to that observed for microcystins and cylindrospermopsin. Relative to other cyanotoxins, sand filtration of saxitoxin had the lowest median value, likely owing to the resistance of saxitoxin to biodegradation (Ho *et al.* 2012). Intracellular and extracellular saxitoxins were removed poorly during ultrafiltration, but nanofiltration was able to effectively mitigate both intracellular and extracellular saxitoxin (Coral *et al.* 2011; Fan *et al.* 2014). Chlorination was effective for the oxidation of saxitoxins as well as cylindrospermopsin.

The other two metabolites evaluated as a part of the current review were geosmin and 2-MIB (Figure 4). Standalone PAC removal was ineffective (<25% on average) for 2-MIB and highly variable for geosmin. In this instance, the low removal was attributed to a combination of the inclusion of data from WTPs experiencing an influx of cyanobacteria during water recycling and limited removal of intracellular metabolites. For geosmin and 2-MIB, coagulation–flocculation–sedimentation or DAF had lower average removals (<20%) relative to the three cyanotoxins. Furthermore, DAF was less effective than sedimentation for geosmin and no difference was observed for 2-MIB. This is in contrast to previous work showing that DAF has the potential to enhance removal for extracellular T&O compounds via volatilization (Chen *et al.* 2019).

The filtration processes (membrane or sand) were both more effective in mitigating total T&O compounds than the initial coagulation–flocculation and sedimentation/DAF; however, membrane filtration (average of 58% for geosmin and 30% for 2-MIB) was not as effective as sand filtration (average of 70% for geosmin and 54% for 2-MIB), likely due to the added removal process of biodegradation for extracellular geosmin and 2-MIB (Ho *et al.* 2007; Faruqi *et al.* 2018). Although UV has the potential to inactivate cells, its ability to degrade the metabolites requires significantly higher doses than those applied for disinfection, which led to poor removals for both T&O compounds (<30%). GAC was an effective mechanism for the removal of intracellular and extracellular geosmin, but variable for 2-MIB. Information on the age of the GAC was not available, but the removal of metabolites can occur via sorption or biodegradation (if the adsorber was biological) (Ho *et al.* 2007). The application of UF and NF (post-DAF) were both effective for the total T&O compounds, though NF would likely only be appropriate for the T&O compounds. Although chlorination was likely ineffective in degrading extracellular geosmin and 2-MIB, it did contribute to the release of intracellular metabolites.

To better understand at what point in treatment extracellular metabolites were present or released, the fraction of extracellular to total metabolite after each stage of treatment was calculated (Supplementary Material, Figure S3). Geosmin and 2-MIB saw progressive increases in this fraction until after the final stage of treatment where the total T&O compound was on average ~80% released. In contrast, the two cyanotoxins ended at an average extracellular fraction of <10%. To better understand these trends, the fraction of data collected where the extracellular metabolite was equal to the total metabolite was determined (Figure 5). A category was reserved for cells that were removed without the detection of extracellular metabolite and one in which the cells were neither lysed nor

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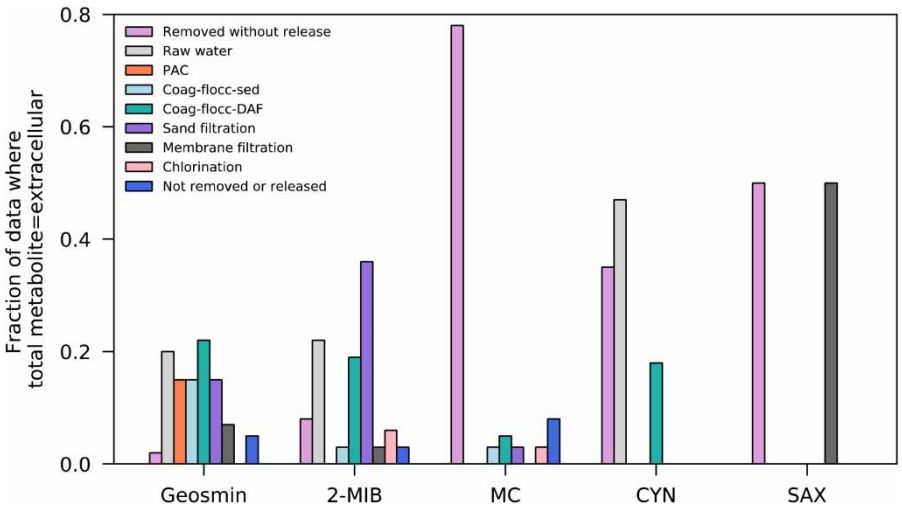

Figure 5 | Stage of treatment where the concentration of extracellular metabolite was equal to the total metabolite concentration as a fraction of the total sampling events for geosmin, 2-MIB, MC (microcystin), CYN (cylindrospermopsin), and SAX (saxitoxin).

removed during treatment (Figure 5). For geosmin and 2-MIB, a very low fraction of sampling events saw removal without release, which was reflected in the poor total removal than observed for these compounds and in the fraction of extracellular relative to total (Supplementary Material, Figure S3). In contrast, the three cyanotoxins were frequently removed without release, post-sedimentation/-DAF or sand filtration, which led to the behavior shown in Supplementary Material, Figure S3.

In approximately 20% of the geosmin and 2-MIB sampling events, the metabolites were entirely extracellular when they entered the plant. For the T&O compounds, this result coincides with what has been observed in surface waters where the extracellular fraction often dominates and has been found to be up to 80% of the total metabolite (Jüttner & Watson 2007). Cylindrospermopsin has been shown to follow the same trend with the majority (>50%) of metabolite detected as extracellular (Rücker *et al.* 2007). This was reflected in the data where 47% of the raw water sampling events had the extracellular cylindrospermopsin equivalent to the total. In contrast, microcystins and saxitoxins are primarily found as intracellular in surface waters. As a result, there was no sampling event in which the raw water contained only extracellular cyanotoxin (Figure 5).

For geosmin, each subsequent treatment step contributed to the release of intracellular toxins with an almost equal distribution in post-PAC, post-sedimentation, post-DAF, and post-sand filtration. Although none of these processes is designed to produce release, death of the cells over time or the stress-induced during treatment, e.g., DAF, likely contributed to release (Zamyadi *et al.* 2018). The other T&O compound, 2-MIB, was split between sand filtration and DAF. Microcystins and cylindrospermopsin saw similar trends with DAF contributing to the release of intracellular toxins in more instances than sedimentation or sand filtration. Because saxitoxin was evaluated in a limited number of sampling events, the frequency of removal without release or post-membrane filtration is likely not representative of the behavior long-term. Given that blooms have the potential to release their intracellular metabolites throughout treatment (regardless of whether the treatment is intended for that purpose or not), operations must be wary of the release of cyanotoxins and T&O compounds during the later stages of treatment.

MTC numbers in source waters

The wide range of treatment removals observed in the previous section illustrates the difficulty in providing specific guidance values for the cell count numbers that represent a potential risk for WTPs.

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Furthermore, the mechanisms behind the production of cyanotoxins are complex and difficult to predict as the levels can be influenced by the cell growth cycle, abiotic factors, e.g., light, temperature, and nutrients, as well as ecological factors, e.g., strain composition, grazing, abundance, and growth of competitors, and transfer within the food web (El-Shehawy *et al.* 2012). During the collection of surface water data in this work, the cell quota was calculated whenever possible to provide a wide range of potential values (Table 1). For microcystin, this was 23–443 fg/cell and an average of 142 fg/cell. This range is similar to previous estimates of 200 fg/cell (Chorus & Bartram 1999) and 330–420 fg/cell from the Great Lakes region in Canada (Almuhtaram *et al.* 2018) but is below an average value of 630 pg/cell generated from *Microcystis* sp. collected from a lake in New Zealand (Ministry for the Environment 2009). The cylindrospermopsin cell quota ranged from 0.4 up to 13,500 fg/cell with an average value of 610 fg/cell, which is a dramatically wider range than that reported by the WHO of 4–190 fg/cell for a range of lab-cultured cylindrospermopsin-producing cyanobacteria (WHO 2019a). Saxitoxin contained 5–20 fg/cell with an average of 13 fg/cell or below the range of 40–1,300 fg/cell from *Scytonema* sp., *Aphanizomenon* sp., and *Dolichospermum circinale* reported by the WHO (WHO 2019b). Similar to cyanotoxins, the concentration of T&O compounds per cell can vary depending on the stage of the growth cycle in addition to the environmental conditions. Geosmin and 2-MIB ranged from a concentration of 1–53 and 3–12 fg/cell, respectively, with averages of 21 fg/cell for geosmin and 7 fg/cell for 2-MIB. Previously reported values have varied from 15 to 838 fg/cell for *Dolichospermum circinale* in reservoirs in South Australia and Victoria (Hobson *et al.* 2010; Tsao *et al.* 2014; Pestana *et al.* 2016). The 2-MIB cell quotas were similar to previously reported values from lab-cultured *Pseudanabaena* cells at 5–51 fg/cell (Chiu *et al.* 2016a, 2016b; Zamyadi *et al.* 2016).

A range of MTC counts was established by using the minimum, maximum, and average removal along with the average cell quota (Figure 6). When the effluent values for a given process resulted in a concentration of metabolite below detection and no detection limits were reported, a maximum removal was set at 99%. The calculated MTC values were compared against the alert levels from guidance agencies, WHO and WQRA, that recommend sampling and treatment actions that should be taken by WTPs. WHO alert levels are set at 1, 2,000, and >100,000 cells/mL and WQRA levels are ≥500 to <2,000 cells/mL, ≥2,000 to <6,500 cells/mL, and ≥6,500 and ≥65,000 cells/mL (Chorus &

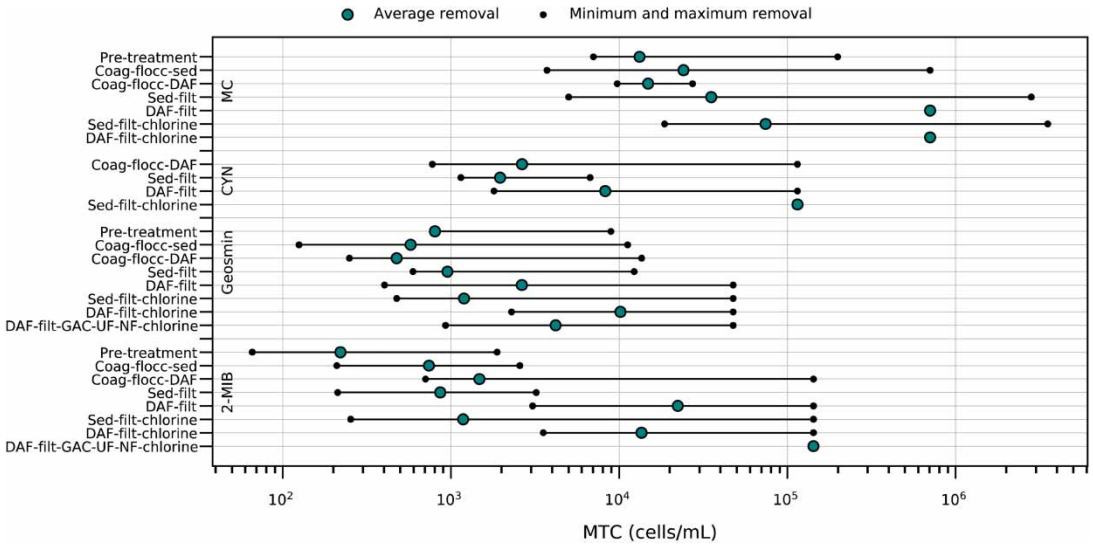

Figure 6 | MTC for MC (microcystin), CYN (cylindrospermopsin), geosmin, and 2-MIB following the minimum, average, and maximum percent removal during treatment employing the average cell quota. Pre-treatment includes PAC, pre-oxidation with ClO_2, $KMnO_4$, or O_3, and $CuSO_4$ and PAC. In cases where the metabolite was below its detection limit in the effluent, a removal of 99% was applied. SAX did not have enough reported removal values to be included in this analysis.

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Bartram 1999; Newcombe *et al.* 2010). The alerts are based around *Microcystis aeruginosa* blooms or those where a cyanotoxin producer is dominant relative to the total biovolume.

The four metabolites had variable MTC values depending on the type of treatment employed (Figure 6). Plants were separated after coagulation–flocculation into those employing DAF and those with sedimentation. Advanced treatment, e.g., GAC adsorption, UF, and NF, was only evaluated for metabolites at the plants that employed DAF. A plant with DAF as their post-coagulation–flocculation step along with sand filtration and chlorination could effectively treat 700,000 cells/mL microcystin-producing cells and not exceed the drinking water microcystin GV of 1 μg/L. Comparing these results to the collected data verifies this result. For the 12% of the sampling events with cell counts on the order of 10^6 cells/mL, no GVs were exceeded for any metabolites, though T&O compounds were often present at detectable levels in the finished water. Plants based around sedimentation were less effective with an MTC of 74,000 cells/mL. A bloom dominated by cylindrospermopsin-producing cells could be removed on average at influent levels of 8,000 and 115,000 cells/mL for DAF–sand filtration–chlorination and sedimentation–sand filtration–chlorination, respectively. The T&O compounds had similar MTC values, despite using the 10 ng/L GV. The geosmin and 2-MIB MTC levels were both at approximately 1,200 cells/mL for sedimentation and 12,000 cells/mL for DAF. The addition of advanced treatment, e.g., GAC, UF/NF, to the DAF plant did not enhance the MTC for geosmin but increased it by an order of magnitude for 2-MIB.

The levels observed here agree with those recommended by guidance agencies and treatment plants would be appropriately placed into high-risk categories when they were at risk for exceeding the cyanotoxin GVs. However, the calculation of MTC values, in this instance, relied on average removal and average cell quota, which means that there is potential for lower cell tolerance if the maximum cell quota were employed. This would impact all metabolites, but particularly cylindrospermopsin, which has a maximum cell quota that is approximately 20 times higher than its average. If possible, WTPs should evaluate the cell quotas for their individual blooms to accurately assess the potential tolerance for their plant.

CONCLUSIONS

The removal of toxic cyanobacterial cells, cyanotoxins, and T&O compounds was examined from a global dataset of 31 surface waters and 21 WTPs in 20 countries. In the surface waters examined, multiple toxic species (i.e., cells capable of producing microcystins, cylindrospermopsin, or saxitoxin) were found to co-occur. This translates to an increased risk for WTPs where effective removal of multiple cyanotoxins or T&O compounds must occur simultaneously. The efficiency of conventional and advanced water treatment barriers varied depending on the concentration and species of cells present in the raw water, the metabolites, and the operational parameters, e.g., a dose of PAC or pre-oxidation. The cumulative removal of post-chlorination for microcystins, cylindrospermopsin, and saxitoxin was 89 ± 19, 92 ± 27, and 99%, respectively. The averages for geosmin and 2-MIB were 80 ± 27 and 62 ± 27%, respectively, which was lower than that observed for the three cyanotoxins monitored. Negative cell removals were observed in approximately 28% of the sampling events ($n = 50$) due to the accumulation of cells in the clarifier, during water recycling processes (e.g., sludge management), or within filtration. This trend translated into negative removals for the total (e.g., intracellular and extracellular) cyanotoxins and T&O compounds, though GVs were never exceeded in the finished effluent. Overall, these data highlight the need for WTPs to have real-time monitoring and treatment plans to prevent breakthrough into finished water.

MTC values were determined using the average cell quota and average anticipated removal. The MTCs for plants employing coagulation–flocculation–DAF were generally higher than those with coagulation–flocculation–sedimentation indicating the efficacy of this treatment in removing

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intracellular metabolites. A WTP employing sedimentation would be able to tolerate less than 1,200 cells/mL of geosmin or 2-MIB-producing cells before exceeding the 10 ng/L human detection threshold whereas a plant with DAF could have a bloom with approximately 12,000 cells/mL before exposure. For microcystins, these levels were significantly higher at approximately 74,000 cells/mL for sedimentation and 700,000 cells/mL for DAF before passing the 1 μg/L GV. WTP tolerance was lower for cylindrospermosin-producing cells with an average removal of 8,000 cells/mL before exceedance. These MTC values provide water treatment operators with useful data on the potential risk associated with blooms dominated by a given cyanotoxin- or T&O compound-producing cyanobacteria.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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