The Impact of Lake Water Quality on the Performance of Mature Artificial Recharge Ponds

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Abstract: Artificial groundwater recharge is commonly used for drinking water supply. The resulting water quality is highly dependent on the raw water quality. In many cases, pre-treatment is required. Pre-treatment improves the drinking water quality, although how and to what extent it affects the subsequent pond water quality and infiltration process, is still unknown. We evaluated two treatment systems by applying different pre-treatment methods for raw water from a eutrophic and temperate lake. An artificial recharge pond was divided into two parts, where one received raw water, only filtered through a micro-screen with 500 µm pores (control treatment), while the other part received pre-treated lake water using chemical flocculation with polyaluminium chloride (PACl) combined with sand filtration, i.e. continuous contact filtration (contact filter treatment). Water quality such as cyanobacterial biomass, microcystin-LR as well as organic matter and nutrients were measured in both treatment processes. We found cyanobacterial biomass and microcystin-LR level after the contact filter treatment was significantly different from the control treatment and also significantly different in the pond water. In addition, with contact filter treatment, total phosphorus (TP) and organic matter removal were significantly improved in the end water, TP was reduced by 96 % (< 20 µg/L) and the total organic carbon (TOC) was reduced by 66 % instead of 55 % (TOC content around 2.1 mg/L instead of 3.0 mg/L). This full-scale onsite experiment demonstrated effective pre-treatment would benefit a more stable water quality system, with less variance and lower cyanotoxin risk. In a broader drinking water management perspective, the presented method is promising to reduce cyanotoxin risk, as well as TP and TOC, which are all predicted to increase with global warming and extreme weather.

Keyword: Groundwater, Pre-treatment, Contact filtration, Infiltration ponds, Nutrients removal, TP, Cyanobacteria, Cyanotoxin, Microcystin-LR, Eutrophic lakes, TOC

1. Introduction

Artificial groundwater recharge plays an essential role in the sustainable management of groundwater resources as the demand for high quality drinking water continuously increases with growing populations combined with over exploiting of groundwater resources [1]. Using artificial recharge of surface water for drinking water supply is commonly used in Sweden and elsewhere, such as in the Netherlands, Australia, China and US [2,3,4,5,6]. About 100 treatment plants in Sweden, representing about 25 % of the Swedish municipal drinking water supply in volume, apply artificial recharge [7]. A standard way of artificial groundwater recharge is to divert raw water, such as lake water, directly into recharge ponds where the water slowly infiltrates and becomes groundwater. The water treatment plant then withdraws groundwater through wells and distributes it to consumers following disinfection and post-treatment, such as filtration or adjustments in hardness and pH. The water quality after the artificial infiltration is important for drinking water safety and is highly dependent on the raw water quality. Hägg et. al. showed that the final drinking water quality in terms of organic content might follow the same pattern as the raw water quality in conditions without pre-treatment, according to his study on 16 artificial groundwater treatment

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plants in Sweden [8]. Moreover, experience from Finnish waterworks by examining the occurrence of microcystin in raw water sources and treated drinking water showed that efforts should be made to stabilize the operational conditions of the artificial recharge to meet the high and rapid variations in the raw water quality [9]. Therefore, pre-treatment is often required before the water enters the artificial recharge ponds. As if the incoming water is stabilized, the final drinking water quality in general improves. While, to what extent pre-treatment can reduce cyanobacteria and cyanotoxin risk is rarely studied.

Pre-treatment is highly valuable for drinking water from a hypereutrophic lake, such as Lake Vomb (south, Sweden), particularly during algal bloom season [10]. On one hand, there might be accumulation of nutrients and cells in the artificial recharge ponds as well as cyanobacteria regrowth due to favourable condition such as easier light access and warmer water for a small water body. On the other hand, infiltration ponds for removing cyanotoxins might not be enough if the infiltration rate is high or the water temperature is low, which might lower the biodegradation activities [11]. Therefore, a complementary approach such as pre-treatment is needed especially in the autumn, when cyanobacteria cells might decay and release toxins [10]. Groundwater contamination by microcystin from toxic cyanobacteria blooms posed a significant health risk for residents in Lake Chaohu, China, when it is used for drinking water [12]. Moreover, rivers and lakes in boreal regions or those surrounded by large agricultural areas tend to trigger algal blooms [13,14]. Cyanobacterial blooms in freshwaters have become a worldwide problem for both ecosystem function and human health [11].

Hence, to protect and safe-guard surface water supplies, such as lakes and rivers, has become challenging for drinking water supply. Multi-barrier approach is needed for managing toxic cyanobacteria in drinking water, including prevention, source control, optimizing treatment operations for toxin and cell removal, and implementing a detection and monitoring system to facilitate effective treatment and provide an early warning to the operators and the public in case of elevated levels of toxins in treated water [16]. Introducing pre-treatment is a way to optimizing treatment operations and preventing cyanobacteria regrowth and reducing cyanotoxin risk.

To examine the nutrient condition in the pond water is of great importance, especially total phosphorus, which if above 20 μg/L may provide re-growing condition for cyanobacteria [17]. Eutrophic conditions tend to enhance the growth and bloom formation of algae, specifically cyanobacteria [18]. The removal of natural organic matter (quantified as TOC) from lake water is also of considerable importance, as it may affect the drinking water quality in various ways such as causing formation of disinfection by-products and supporting biofilm in the distribution systems [15]. Besides, recent studies have also demonstrated that synergies between climate warming and increasing levels of natural organic matter are able to trigger an increase in cyanobacteria biomass [19], as well as a reduction in the biodiversity of phytoplankton [20,21].

The best opportunity for cyanotoxin removal is to remove intact cyanobacterial cells physically [22]. This can be effectively achieved by standard drinking water treatment processes (coagulation, flocculation, sedimentation and filtration) or highly effectively achieved by microfiltration and ultrafiltration [23,24]. Others have also shown that chemical flocculation and filtration treatments are effective in removing cyanobacteria cells and other harmful compounds [23,24]. While for extracellular toxins, they are more complex to remove. Activated carbon, membrane filtration and chemical inactivation (Ultraviolet (UV), disinfectants and oxidants) are common treatment techniques for the removal of extracellular toxins [25]. Furthermore, Svrcek and Smith (2004) thoroughly reviewed the efficiency of different treatment processes for the removal of cyanotoxins from potable water [22] and the American Environmental Protection Agency (EPA) summarized treatment processes and relative effectives for both intact cells and dissolved cyanotoxin removal [25]. Swedish National Food Agency also published treatment methods' efficiency for removing different type of cyanotoxins [26]. Like artificial recharge infiltration, bank filtration (sand filtration) is commonly used for removing cyanobacteria and cyanotoxins. Romero and his colleagues

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summarized a few studies and did a pilot study at the Lake Lagoa do Peri in Brazil showed that bank filtration is efficient in removing phytoplankton and Cylindrospermospsis raciborskii (105–106 cells/mL) and removing up to 7.26 µg STX Eq./L in raw water before reaching a sampling well at 12-meter-deep and located 20 m from the lakeshore. The biomass was mainly removed in the upper 2 cm of the sand and no cyanotoxins were found in the monitored well water [27].

One of the biggest challenges in cyanotoxin detection is due to its complexity with various variants and many types are not able to detect even with advanced analytical devices. No single analytical application can detect all cyanotoxin variants in an environmental sample. Most of the research about cyanotoxins were done mainly focused on microcystins, normally on the most toxic variant: microcystin-LR (MC-LR)[28] or microcystin-LR equivalents (converting other congeners to MC-LR). It has been associated with most of the incidents of toxicity involving microcystins in most countries [29]. There are over 100 microcystin variants but the most common one is microcystin-LR. This method is not particular for any specific variant. The best approach for monitoring of microcystin-LR is to use a combination of screening and more sophisticated quantification methods such as HPLC or LC-MS/MS [30]. Commercially available Enzyme-Linked Immunosorbent Assay (ELISA) test kits are one of the more commonly utilized cyanotoxin testing methods due to low cost and less training required. It measures all variants with results expressed as microcystin-LR equivalence. It is also recommended by Swedish National Food Agency as a screening tool for microcystins monitoring in drinking water resources [26]. While, there are much more microsystin variants existing in Swedish source water, most commonly MC-LR, MC-RR, MC-YR, MC-D-Asp3-LR and D-Asp3-RR [31], as well as other types of cyantoxins such as beyond anatoxins, cylindrospermopsins. There is huge gap between what can be measured and what exist.

In our study site, one of the largest drinking water treatment plants in south Sweden, mature artificial recharge ponds have been applied for drinking water supply for more than 60 years. Past observation of trace amounts of cyanotoxins in groundwater samples (around 0.3 µg/l and highest 0.88 µg/l) (mainly using a screening test and confirmed by analytical analysis at Swedish Food Agency) had prompted operators to improve the treatment process by introducing pre-treatment to complement the pond infiltration performance [10]. The purpose of this project was to test how much nutrients, cyanobacteria biomass and cyanotoxins were reduced through pre-treatment process, and how it would affect the pond water quality and final infiltrated water. Higher concentrations of cyanotoxin in the recharged water may occur more often due to hypereutrophic condition of raw water and likely warmer water temperature due to climate change [10,32].

Our study aims at supporting decision-making at waterworks to identify proper pre-treatment methods/barriers for safe drinking water supply to meet future challenges accompanying e.g. climate warming. Our study also contributes to the develop management strategies and decision support for monitoring and effective management of cyanobacterial issues in practice such as preventing cyanobacteria re-growth in the artificial recharge ponds, eliminating cyanotoxin in drinking water.

2. Material and Methods

2.1 Case study description

The study was performed at the Vomb Waterworks, an artificial groundwater treatment plant in south Sweden, which has been applying artificial recharge for more than 60 years. Vomb Waterworks is run by the largest drinking water supply company in south Sweden (Scania), Sydvatten, representing about 900 000 inhabitants from 17 municipalities. Raw water with a flow rate around 1000 L/s is taken from the Lake Vomb which is situated about 3 km northeast of the treatment plant and water is pumped to a sieve station with a micro-screen of 500 µm pore size, removing macro-particles and reeds. The water is then distributed into infiltration ponds around the waterworks. There is in total 54 ponds (covering a surface of 430 000 square meter) but only half of them are in use at the same time. The water seeps slowly, with an average velocity of 0.4 m/d through

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the fine sand (grain size is about 0.5-0.8 mm), and it takes about 2 to 3 months for the water to reach the groundwater wells. The water is then pumped up through 120 wells and led to the treatment plant. In the drinking water treatment plant, the main treatment process is to remove iron, reduce the hardness, adjust pH and disinfect the water before distribution to consumers [2].

The catchment area of the lake is 450 km^2. The treatment plant has been used since 1948 for drinking water production due to the unique natural features. The area is rich in natural groundwater and suitable for artificial groundwater recharge as the area south of the lake is mostly composed of sand, pebbles and gravel [33]. With an average precipitation of 750 mm/year, it is estimated that about 5m^3/s water on average infiltrated into the groundwater aquifer [33].

In this study, one of the fifty artificial recharge ponds at Vomb Waterworks was used. The pond was selected for this experimental test based on both practical and technical considerations such the access to the pre-treatment facility, heterogeneity of the natural layers of the whole area and the location of the area where residence time was longer compared to the average residence time of all ponds. That was to avoid negative effect on groundwater quality if anything went wrong. The pond was divided into two parts, where one received untreated lake water, while the other received pre-treated water through chemical flocculation combined with continuous contact filtration.

2.2 Continuous Contact filtration

The continuous contact filtration technique in this study used an Upflow Solids Contact Filter which eliminates the need for settling tanks, since liquid-solid separation, filtration, and sludge removal are done in a single unit process. After addition of coagulant, flocs which are formed are removed continuously in a moving sand bed. This purifies water and at the same time washes the sand without interruption and additional maintenance (for a detailed description of the technology, see [34]). The filters are commercially available. In our study, Dynasand filters (Nordic Water, Gothenburg, Sweden) were applied. The process involves chemical precipitants (Polyaluminium Chloride) which are added to a mixer in the pipe system before it reaches the filters. Hydroxide flocks are formed and then filtered and separated directly inside the filter bed. The sand grain size is around 1.2 to 2.0 mm, and the filter covers an area of 3 m^2 with a filter bed height about 2 meters. This process is suitable for treatment of surface water and is commonly used in small scale water treatment plants (< 20,000 m^3/day) in Sweden, examples can be seen in, Kalmar, Karlskoga and Karlskrona [8].

The coagulant dosage was tested at the beginning of the project as it is an important factor for the removal of fine suspended solids, dissolved organic matter and phosphate. At the end, 195 µl/l PAX 15 (polyaluminium chloride (PACl), KEMIRA, Sweden) was used for this pilot test. To guarantee good efficiency, the following condition were chosen: the maximum flow was 10.5 L/s; sand movement average speed was 9.79 mm/min; pH level was around 6.4. The consumption of PAX 15 was 177 L/ day. A week consumption of Pax15 was around 1240 L.

2.3 Study design

The project design is illustrated in Fig.1. The artificial recharge pond was divided into two separate parts of similar size and shape, one receiving lake water only treated through a micro-screen (500 µm pores) (control treatment) and one after coagulation, flocculation and continuous contact filtration (contact filter treatment). Both sides of the pond were maintained in the same way and received the same water flow. To be able to follow the treatment process during the study, 4 observation wells were used for groundwater sampling. They were located 2 meters from the sides of each pond (Fig. 1). The ground level was used as reference. The pond was 2 meters deep. The 4 observation wells were on average 13 meters deep and the groundwater table was around 5-7 meters under the ground. The pond was divided by a bank of fine material in the middle, approximately 0.5 meters under the surface of the sand. Water flows entered the pond from the furthest sides, the groundwater collected in the closest wells from both sides was assumed to be unaffected by water from the opposite pond.The experiment set-up started in the second week of April 2014. It took two months (in June) to have a stable system before taking samples from both inlets of the pond and it took four months (in August) to have enough

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standing water in the both half ponds to take pond water samples.

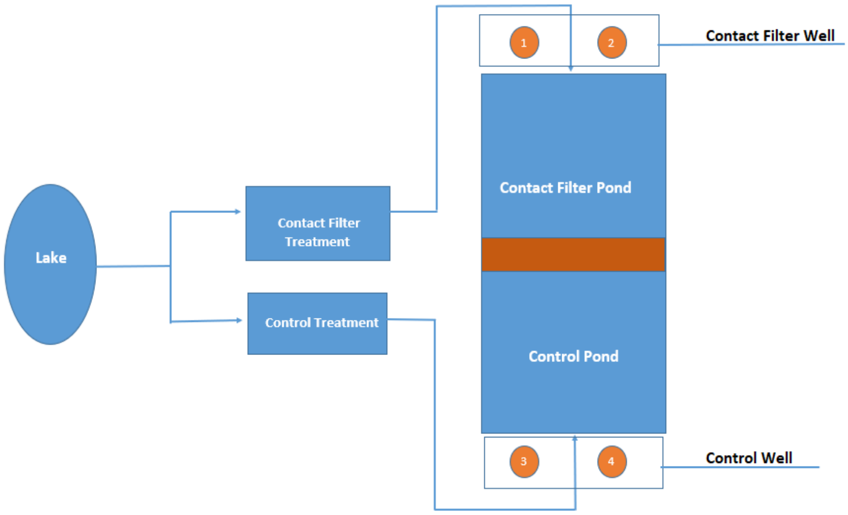

Figure 1. Study site overview. The Lake is Lake Vomb from which water is led either through contact filter treatment or control treatment to their corresponding pond, i.e. contact filter pond and control pond.

2.4 Samplings

Samples were taken from 10th of June to 20th of October 2014 (every the other week) at several sites along the treatment process, including from the incoming lake water, pre-treated water after contact filter treatment and control treatment, water from the contact filter pond and control pond (from the end of August) and infiltrated water from all four wells (Fig. 1). The number of samples for each parameter are summarized in Supplement Table 1.

Water quality parameters that were sampled were: turbidity (FAU), UVA_{254nm}, color (436nm), Chemical Oxygen Demand (COD mg /L), total phosphorus (mg/L), pH, Total Organic Carbon (TOC mg /L), nitrate (mg/L), orthophosphate (mg/L), cyanobacteria and microcystin-LR.

A Hach Ratio XR Turbidimeter (Hach Lange GmbH, Germany) was used for turbidity measurements and a WTW 197-S pH meter (Xylem Analytics, Weilheim, Germany) for pH measurements. An DR6000™ UV VIS Spectrophotometer (Hach Lange GmbH, Germany) was used to measure: UV(SAC)254nm and VIS_{436nm}-absorbance (water color). Colourimetric titration with potassium permanganate was used for COD measurement. TOC samples were sent to specialized laboratory (Eurofins, Sweden). Orthophosphate and total phosphorus were measured by HACH LANGE test kits at the above spectrophotometer. Algae samples were fixed by lugol and kept overnight to sink on the bottom of Utermöhl chamber before analysis. A reverse microscope with maximal enlargement of 400x was then used to count cells. The cell density was transformed to a biomass scale so that the number represents the percentage of algae area covered on the chamber area (1= 10%, 2= 20% and so on). This was used to give a simple estimation which gives an overview of the abundance of cyanobacteria. The following taxonomic groups were distinguished by their morphological characteristics:

-1: *Woronichinia/ Snowella / Mycrocystis / Radiocystis*
-2: *Chroococcus*
-3: *Beggiatoa*
-4: *Achroonema / Limnothrix / Planktolyngbya*
-5: *Anabaena / Nostoc.*

Beacon Microcystin Tube Kit was used for microcystin analysis(Beacon Analytical Systems Inc., 2019, Saco, Maine, USA). It is a polyclonal antibody based, semi-quantitative method. The test process refers to [35]. Several microcystin congeners can be detected due to cross reactivity. According to the instructional brochure, the percentage cross reactivity (% CR) of other microcystin congeners relative to

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microcystin-LR are microcystin-LR (100%), microcystin-RR(73%), microcystin-YR (58%), microcystin-LA(2%), microcystin-LF (3%), microcystin-LW(4%) and nodularin(126%). This may pose microcystin test kit is suitable as a screening method.

The evaluation of this method as a quick analysis for field tests has been demonstrated in previous work, where it showed 20 % false ''positive'' compared to analytical method HPLC for microcystin-LR measurements [17]. One reason might be due to the cross-reactivity with other microcystin congeners shown above, which might also be its advantage as a screening method for microcystin and its congeners in water. HPLC has higher accuracy while it only detects the types applied as standard. The official detection level for the microcystin test is 0.3 µg/L [35]. According to the test kit supplier, through spike and recovery experiment with standard solution, values below 0.3 µg/L might also be valid. In this study, it was assumed that all the data measured could be used for analysis.

The guideline value of microcystin-LR monitoring in drinking water suggested by the World Health Organization (WHO) is 1 µg/L [36]. This value was newly listed in the revised Drinking Water Directive proposal (2018) which is the main piece of EU legislation in regards to regulate the water quality intended for human consumption [37]. A group of expertise from EU project CYANOCOST proposed that the parametric value 1.0 µg/L should include all microcystin variants, not only microcystin-LR [38]. As most tested microcystin variants exhibit strong toxicity, the inadequacy of current microcystin guidelines based only on microcystin-LR might pose health risk for drinking water supply [38]. More cautious microcystin-LR values are used in some regions, such as 0.16 µg/L in Vermont [39] and 0.1 µg/L in Minnesota in U.S.A [40].

2.5 Bootstrapping resampling

Data resampling refers to methods for economically using a collected dataset to improve the estimate of the population parameter and help to quantify the uncertainty of the estimate. Bootstrapping is one of the resampling methods, which relies on random sampling with replacement. It allows assigning measures of accuracy, defined in terms of confidence intervals in this study for example, to the estimated resamples. This technique allows estimation of the sampling distribution of almost any statistic using random sampling methods [41].

3. Results

Site observation showed continuous contact filter treated water developed slower resistance in the pond and higher infiltration rates. Until the end of August, i.e. after running project for 4 months, the whole bottom in the control pond was covered by water, while only 2/3 of the contact filter pond was covered by water. Water quality with mean value and Min.-Max. values at different stages along the treatment process are presented in Supplementary Table 1. Below sections focus on the removal of organic matter, nutrients, cyanobacteria and microcystin-LR.

3.1 Organic matter and particles removal

One major task for drinking water producers is to remove particulate materials, i.e. inorganic and organic particles. Particulate materials from the raw water and flocs formed during chemical flocculation are physically removed by filtration processes. This can be seen in Fig.2, where contact filter treatment removes about 50 % of particles from the raw water, measured as turbidity (FAU) and it remains low level in the following pond water. On the control treatment side, turbidity has a slight decrease due to the micro-screen which was designed to remove particles larger than 500 µm. The increase of turbidity with big variance in the pond water might partly be due to algae re-growth in the control pond (Fig.5).

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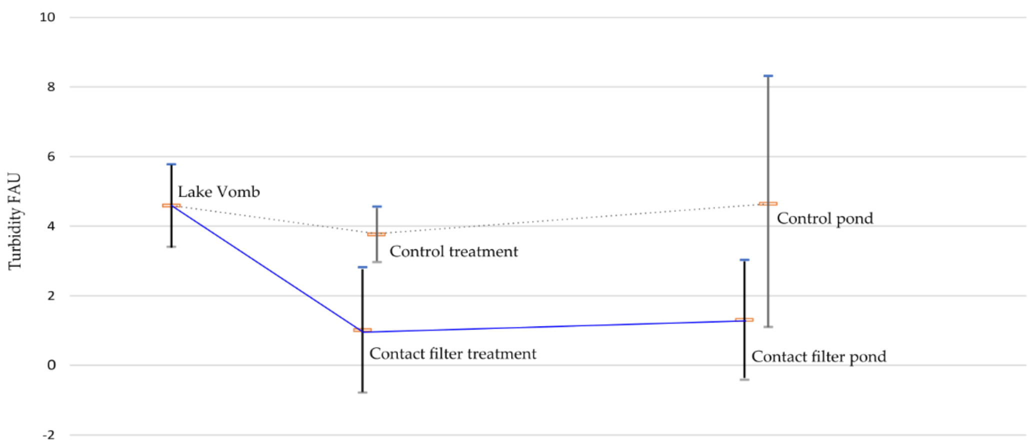

Figure 2. Turbidity measurements at different stages along the two treatment processes (FAU) (The middle bar is the mean value and the top and lower bars show the standard deviation. The dotted line is the contact filter treatment process and the solid line is the control treatment process.)

The organic content removal in the form of TOC-content shown in Fig. 3 is much more efficient by contact filter treatment process than the control treatment. TOC-content was already reduced by 50%, after contact filter. This is a similar result to the previous studies on flocculation for TOC-content removal by using the same source water [42] and within the observed range of other WTP's in Sweden using contact filtration [8,42,43]. The final TOC-content in the well water on the contact filter side was on average 2.1 mg/ L, while in the control wells, the TOC-content is on average 3.0 mg/ L.

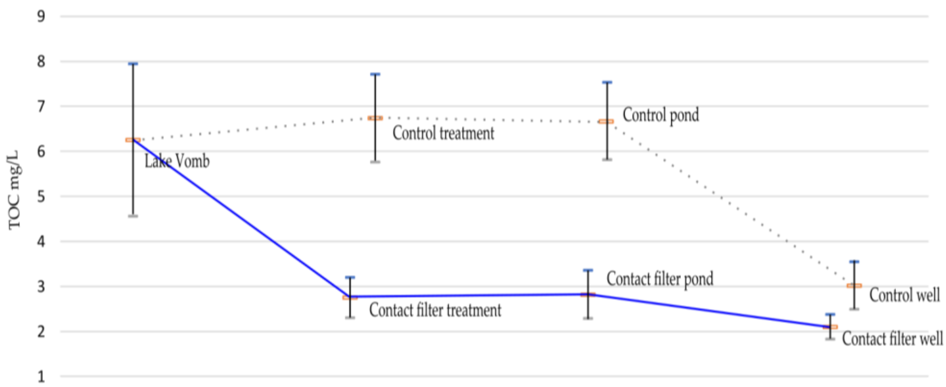

Figure 3. TOC measurements at different stages along the two treatment processes (mg/L) (The middle bar is the mean value and the top and lower bars show the standard deviation. The dotted line is the contact filter treatment process and the solid line is the control treatment process.)

Similar trends were observed for other indicators of natural organic matter content (NOM), i.e. chemical oxygen demand (COD) and UV-absorbance (UVA), where these parameters were significantly reduced after contact filter process before infiltration process (S. Fig. 1 and 2).

3.2 Nutrients removal

Phosphorus removal using chemical coagulation and rapid sand filtration has been proved to be very effective in previous studies [45]. This is also verified by this study where up to 95% of the total phosphorus (TP) was removed after contact filter treatment (Fig. 4) and the TP concentration was reduced below 20 μg/L. To keep a low concentration of phosphorus (< 20 μg/L) is crucial for cyanobacterial bloom control [17]. This can be seen in Fig.5 that intensive cyanobacterial blooms presented in the control pond where the phosphorus remained the same as in the raw water while very clear water presented in the contact filter pond where TP content was low.

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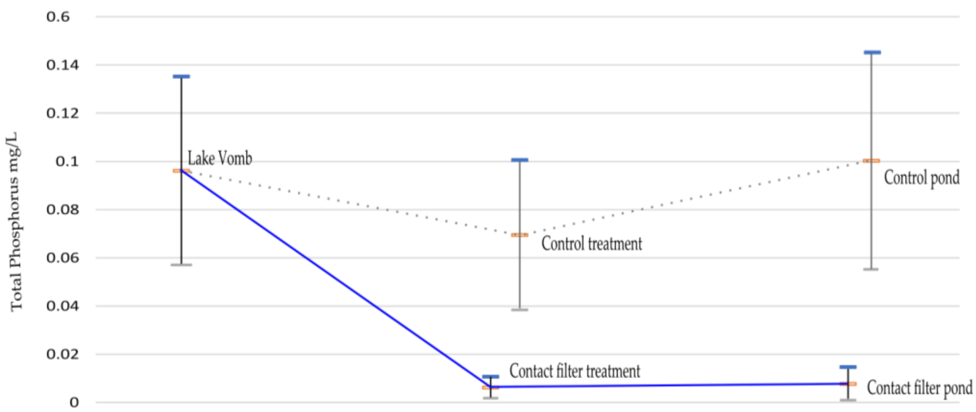

Figure 4. Total phosphorus removal at different stages along the two treatment processes (The middle bar is the mean value and the top and lower bars show the standard deviation. The dotted line is the contact filter treatment process and the solid line is the control treatment process.)

Figure 5. Onsite observation of two sides of pond, left: control pond, right: contact filter pond, August 2014 (Photo: taken by Marie Baehr and Petra Larsson)

3.3 Cyanobacteria removal

On site observation in Fig.5 has already showed clear evidence of water quality improvement by introducing contact filter treatment. The cyanobacteria biomass in the control pond was about 8 times more than in the contact filter pond. The result of on-site visual inspection is shown in Supplementary Table 2 and a subjective evaluation result about the comparative amount of cyanobacteria in different groups is shown in S. Fig.5.

The dominating species of cyanobacteria in the control pond are group 1 (*Woronichinia*/ *Snowella* / *Mycrocystis* / *Radiocystis*) followed by group 5 (*Anabaena* / *Nostoc*.) and group 2 (*Chroococcus*) (S. Fig. 5). Only group 1 (*Woronichinia*/ *Snowella* / *Mycrocystis* / *Radiocystis*) and 4 (*Achroonema* / *Limnothrix* / *Planktolyngbya*) are present in the contact filter pond (S. Fig. 5). The difference of those groups are mainly their morphology and size. Some cyanobacteria species with smaller cell size passed the contact filter, such as group 1 (2-3 µm) and group 4 (1-3 µm). The group with larger cell size and filaments might be filtered out or limited to grow due to low phosphorus supply in the pond. There were no traces of cyanobacteria in the well samples, suggesting that the infiltration process in the pond was able to remove cyanobacteria even when there were intensive blooms in the control pond.

Microcystis and *Anabaena* (*Dolichospermum*) are listed as the most toxic cyanobacterial genera and often succeed each other during harmful algal blooms [46]. *Woronichinia naegeliana* appears frequently in freshwater and one study showed that blooms of *W. naegeliana* was toxic towards invertebrate zooplankton [47].

One way to control cyanobacteria biomass is to reduce the phosphorus concentration, as discussed

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above. The ratio of nitrogen and phosphorus should also be monitored. Nitrogen limiting condition favors nitrogen fixing cyanobacterial blooms. In this case, *Woronichinia naegeliana* [48] presented in the control pond where nitrogen and phosphorus ratio was below 10. While in the contact filter pond, where nitrogen and phosphorus ratio was above 40, the nitrogen fixing specie *Anabaena* did not show in the pond water although it was found after the contact filtration process (S. Fig. 5). In a previous study, it was also discussed how low N:P ratios may trigger higher cyanobacterial blooms [49].

3.4 Cyanotoxins analysis result

The microcystins are hepatotoxic products of freshwater cyanobacterial blooms of *Microcystis, Anabena* and *Nostoc* with *M. aeruginosa* being the most common [41,36]. *Anabaena* might also produce anatoxin and saxitoxins [36]. Different variants of microcystins have been detected from blooms where W. naegeliana was the dominant species among secondary metabolites[51,52]. The biological effects on planktonic crustaceans were investigated with relation to a fraction containing microginin-FR3 [52].

A clear seasonal pattern of microcystin-LR content in water was observed, that higher concentration appeared during late summer and early autumn (July to October) and remained very low (less than 0.1 μg/L) during the winter and spring time. Boxplot results of microcystin-LR levels at different stages (S. Fig. 6) show clearly that contact filter treatment system has much less variance than the control system, although all median values are similar (thick line in the middle).

During the experiment period, the microcystin content both after pretreatment and in the pond water follow the same trend in raw water, with much lower values and less variance in the contact filtration process (left in Fig.6). A steady development of microysin-LR in the control pond (green diamond in the figure on the right, Fig.6) is observed from early September to the middle of October 2014. It was likely partly produced by the cyanobacteria regrowth in the pond, specifically on 13th of Oct. 2014. This situation was under control on the left figure, the microcystin-LR increases along with the content increase in the raw water at beginning, while it does not follow at certain level. The higher microcystin-R concentration on 29th of Sep. and 13th of Oct. in raw water were prevented to enter the artificial recharge system.

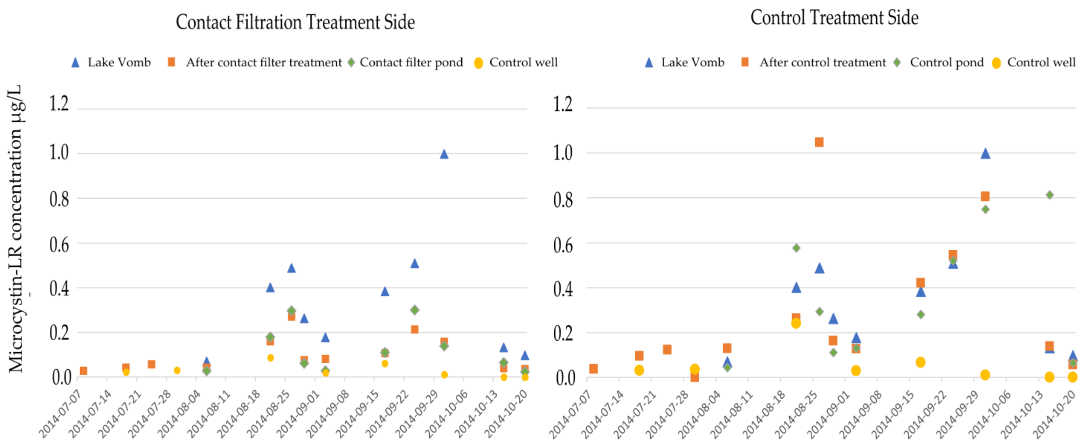

Figure 6. Microcystin development along the sampling period (left was the microcystin condition in the contact filtration treatment process and right is the control situation)

Bootstrapping method was used to test the significance of the difference of microcytin-LR after two treatment processes and also in the pond water during the bloom season (June to October 2014). The advantage to use this method is because, it does not require any test for its distribution type and can do resampling and generating a close to population distribution based on limited sampling points such as in this case. Comparison of the mean value, variance and standard deviation were done and 1000 resampling iterations were generated. In every iteration (random sampling from the original set of data), for example, the mean difference was calculated. After 1000 literation the distribution of the mean difference thereafter was plotted as seen in Fig.7. It demonstrates all possible difference between these two sets of data. The sample difference 0.2 (the blue line) which is close to the median of the distribution, suggesting high possibility to happen. The mean difference of microcystin-LR concentrations after two pretreatment steps is significant based on 95% confidence (between the red lines). This applies to the mean difference 0.24 in the pond water. Their variance and standard deviation difference are also significant. There is no significant difference of the microcystin-LR concentration in the well water.

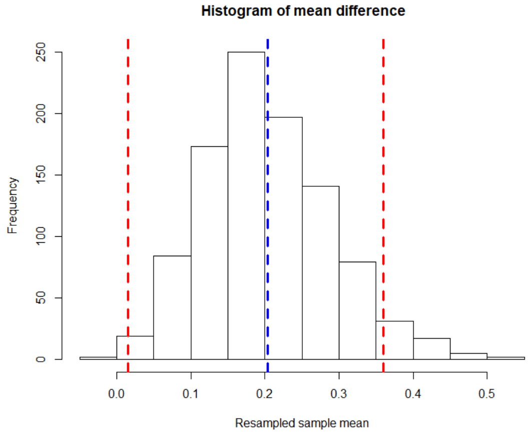

Figure 7. The distribution of mean difference of toxin level after reference treatment and new treatment during the bloom months (June to October 2014)

4. Discussion

This on-situ, full-scale experiment demonstrated the efficiency of pre-treatment on the removal of nutrients and organic contaminants and its impact on pond water quality, reducing cyanotoxin risk in drinking water. Such is shown in S. T. 1 that it resulted in more stable water quality with less variance, reduced total organic matter, prevented cyanobacteria regrowth and eliminated cyanotoxin in the pond water. While there are still a lot of issues needed to be discussed as below.

4.1 Regarding to organic matter removal

Pre-treating the raw water reduced high variability of organic matter entering the pond, as well as during the following pond infiltration process. The variance in turbidity, TOC and COD measurements in the control pond was higher than the contact filter pond. It is also consistent in the TOC and COD measurements in the well water.

Efficient reduction of particles and natural organic matter, on one hand, will result in less clogging and maintenance of artificial recharge ponds and reducing organic pollutants in drinking water, on the other hand, certain amount of organic matter is also expected contributing certain resistance in the infiltration process for longer absorption and degradation time and is also used as food for microbial organisms. Unfortunately, we were not able to observe the starting phrase. Because it took two months to get the system stabilized before taking samples. For a seeping rate of 0.4 m/day and average depth of the groundwater table to the bottom of the pond around 3 to 5 meters, it was estimated that it might just take around 10 days to reach the closest observation wells.

The pre-treatment complemented the TOC-content removal during the pond infiltration process where microbial degradation occurred. This resulted an overall TOC-reduction from 52% to 66%. This was achieved partly by the two different technique´s abilities to remove different fractions of organic matter. Organic matter in the form of particles and cells are removed by physical separation. The different factions in the dissolved form (DOM: dissolved organic matter) which can be removed selectively by different treatment methods. An examination of DOM composition in drinking water production showed that coagulation was highly selective towards to remove fraction resembled

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terrestrial endmembers and DOM with microbial endmember characteristics, was especially reactive during slow sand filtration [53].

4.2 Regarding to the nutrient removal

Continuous contact filtration has been known for its high removal of phosphorus and it was also observed in our case study. This brought positive impact on preventing the cyanobacteria regrowth in the pond water. The general nitrogen concentration was low and did not change along the treatment processes (S. Fig. 3 and 4). To monitor nitrogen level is important for final drinking water quality and controlling cyanobacteria favourable growth condition. As in a nitrogen limiting condition with enough phosphorus, nitrogen-fixing cyanobacteria might get a chance to grow by fixing the nitrogen in the air. The nitrate level in the final production from the control treatment even showed increased values as it might be contributed by the decay of algae, which might fix nitrogen from the air, followed by nitrification process converting ammonia to nitrate (S. Fig. 3 and 4). This might also show that this infiltration layer is under anaerobic condition, which might be due to high TOC-load in water and from cell growth in the pond.

4.3 Regarding the removal of cyanobacteria and cyanotoxin

Our experiment allowed us to see the fate of different groups of cyanobacteria influenced by pre-treatment. It was highly dependent on their morphology and size as well as the changed growing condition. Reduced total phosphorus and the reduced nitrogen and phosphorus ratio prohibited the cyanobacteria regrowth in the artificial recharge ponds. This is important confirmation on how to influence nutrients condition in water for preventing cyanobacteria growth, reducing the risk of cyanotoxin in drinking water.

The micro-screen had big pore size, it could only remove small number of cells, and neither intracellular nor extracellular toxins were removed. In addition to the cyanobacteria regrowth in the control pond water, this resulted significant difference of the cyanobacteria cells and microcystin-LR between the two sides both after pre-treatment and in the pond water.

In the current system, infiltration ponds are the only barriers for cyanobacteria and cyanotoxin removal. The historical trace amount of microcystin in the groundwater was a warning, posing the need for complementary treatment methods. Artificial recharge infiltration ponds were recorded to effectively remove cyanotoxins with help of microbial degradation [54], which was also observed during in this study. Coagulation might also remove certain extracellular cyanotoxins that are hydrophobic such as microcystin variants RR, YR, LR and LA [23]. The majority of the extracellular are removed in the infiltration pond, likely by microbial degradation[55]. While biodegradation on cyanotoxins was dependent on the pH and temperature [56], it might require other barrier to complement microbial activities especially when it comes to the autumn, when it is likely that cyanobacteria start to decay and release toxins in water [46]. We did not find this in our study site during the study time. While it might not be the situation for all the other ponds and other time. Furthermore, the mechanism of cyanotoxin removal by microorganisms needs to be further studied as microorganisms mainly consist of organic carbon, nitrogen and phosphorus. Therefore, the C:P:N ratio in the surrounding environment might also affect their full potential for water treatment [57].

4.4 Discussion around microcystin detection, and monitoring

We have already discussed the complexity of detecting microcystin due to its various variants and that only using microcystin-LR to indicate toxicity situation is not ideal. There might present other types of toxins present, such as anatoxin and saxitoxins, posing potential non-detectable risk. Pre-treatment would help to reduce the hazard risk in general. Accumulation of cyanobacteria and toxin in drinking water process [58] might happen even the toxin level is not high in the raw water, such as high toxin content in the sludge. This presents more challenges for drinking water operators for both monitoring and treating cyanotoxins.

Immunological based microcystin test kit which can detect other microcystin variants due to its cross-reactivity might be a good start as a screening method. While this also requires analytical methods to confirm the concentration level as well as to examine the risk level of other type of cyanotoxins such as anatoxin and saxitoxins that are likely to be produced by the species presented in the sources water. In practice, the confirmation was occasionally done by Swedish Food Agency for suspicious samples with high microcystin-LR concentrations[10,17].

The current guideline values applied in Sweden, is microcystin-LR <1 µg/L which might be legislated as EU law as suggested in the revised draft for EU Drinking water directive. As we discussed before that experts in the research field also suggest this parametric value to involve all type of microcystin variants. Examples of Minnesota and Vermont showed that a stricter level due to health concern or public initiative might push water supplier to initiate targeted monitoring [39]. Drinking water operators need more practical support for cyanotoxin detection and monitoring.

4.5 Regarding to the impact of final water quality

This project demonstrated the positive aspect of introducing effective pre-treatment to reduce the risk of cyanotoxin presence in the drinking water. The effect was limited, however, since the toxin was analyzed in well water taken after a pond which was selected to have better performance than the average level of all the ponds as for its original safety concern. On one hand, we fulfilled the project purpose that is to examine the continuous contact filtration's effect on cyanobacteria cell, nutrients and cyanotoxin removal and impact on the pond water quality, and on the other hand we could also get an indication that during the experiment period, under the current toxin level represented by microcystins' variants, the selected pond was capable to deal with cyanotoxin. However, this does not mean that it would be the same situation for other ponds with less retention time and in the future, when global warming continues, cyanotoxin levels are likely to increase.

As this is a small-scale study with a 10 L/s flow rate and with the consideration of mixing with natural ground water and a final production of 1000 L/s from the waterworks, the effect of the experiment on the total drinking water production is almost neglected. Furthermore, the setup took space to build. It might not be suitable for a large water treatment plant like Vomb Waterworks. Future pre-treatment investigation might be focused on filter system that with smaller pore size like 10 or 30 µm, for organic matter removal such as algae [59].

5. Conclusion

Cyanobacteria in drinking water is a complex issue and multi-barriers approach should be applied. On a small-scale in situ experiment, we have demonstrated how and to what extent pre-treatment of raw water can reduce the cyanobacteria and cyanotoxin risk in the artificial recharge ponds before infiltration and how the treatment process can benefit from introducing effective pre-treatment. It might not be suitable for a large drinking water treatment plant as Vomb Waterworks. While it might be highly likely applicable for a small-scale water treatment plant, such as ca. 20 000 m^3/d.

In general, pre-treating raw water contributes to more stable water quality in the following treatment processes, reduced organic load and reduced phosphorus in the infiltration ponds, and higher water quality in the infiltrated water. Water quality supplied to the infiltration pond can be improved considerably, especially regarding TOC-reduction (on average by 66%), COD-reduction (on average by 75%), UVA-reduction (on average 74%) and TP-reduction (on average by 95%). Compared to the control treatment, this led to a reduction of the total TOC-concentration in the final product from around 3.0 mg/L to 2.1 mg/L, total phosphorus below 20 µg/L and significance decrease of cyanobacteria and microcystin-LR level. This results in preventing intensive cyanobacteria blooms in the artificial recharge ponds and reducing the risk of additional formation of cyanotoxin.

The detection of cyanotoxin in drinking water is challenged in practice. Detection methods that can cover most of the toxic variants are needed. Water operators are encouraged to expand to other toxin types based on the local situation such as anatoxin and saxitoxins in this case. In the future, there might

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be continuous discussion around the parametric value of microcystin for drinking water and more types of cyanotoxin should also be included in the legislation.

Data Availability Statement

The datasets generated during and/or analyzed during the current study are not publicly available due to request from Sydvatten, but are available from the corresponding author on reasonable request.

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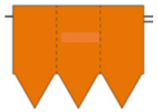
Supplementary Table 1. Water quality at different stages along the treatment processes make changes of the toxin datathe

Treatment train	Raw			Pretreatment					Artificial recharge Pond					Well water				
Water quality				After contact filter treatment 		After control treatment 			Contact filter pond 		Control treatment pond 			Contact Filter Well		Control Filter Well		
	Avergae	Min-Max	No.	Avergae	Min-Max	Avergae	Min-Max	No.	Avergae	Min-Max	Avergae	Min-Max	No.	Avergae	Min-Max	Avergae	Min-Max	No.
TOC	6.25	4.3-9.1	n=6	2.75	2-3.3	6.7375	5.5-8.5	n=8	2.82	2.1-3.2	6.67	5.1-7.4	n=6	2.1	1.9-2.5	3.01	2.1-3.5	n=7
COD	4.955	4.13-5.64	n=10	1.70	1.42-2.36	4.76	4.03-5.51	n=14	1.71	1.27-2.42	5.333	4.27-6.37	n=8	1.26	1-1.55	1.55	0.67-12.28	n=8
Turbidity	4.59	(2.4-6.5)	n=9	1.01	0.3-5.82	3.76	2.7-4.5	n=9	1.3	0.25-5.75	4.64	(3-14.2)	n=9					
UV (A254 nm)	17.19	(15.9-17.7)	n=9	5.51	4.53-7.15	16.44	15.1-17.1	n= 13	5.18	4.7-5.75	16.33	(15.1-16.4)	n=8	4.42	1.14-6.93	5.16	1.22-7.24	n=9
Colour	2.09	1.56-2.18	n=9	0.33	0.24-0.63	1.46	1.01-1.94	n= 13	0.51	0.211-0.593	1.56	1.14-1.72	n=8	0.87	0.24-1.32	0.38	0.04-1.41	n=8
TP	0.096	0.041-0.194	n=11	<0.02	<0.02	0.0694	0.02-0.106	n= 15	<0.02	0-0.018	0.100	0.06-0.216	n=9	<0.02	<0.02	<0.02	<0.02	n=9
Ammonium	0.069	0.014-0.234	n=9	0.057	0.024-0.1896	0.052	0.018-0.164	n= 13	0.044	0.011-0.096	0.049	0.03-0.099	n=6	0.0082	0-0.021	0.0075	0-0.02	n=5
Nitrate	0.3687	0.203-0.989	n=10	0.516	0.035-1.6	0.559	0.207-1.66	n=15	0.392	0.206-1.08	0.318	0.143-1.02	n=6	0.55125	0.149-1.63	0.833	0.277-1.56	n=8
Orthphosphate	0.1685	0.049-1.02	n=10	<0.02	<0.02	0.0694	0.02-0.106	n=15	<0.02	0-0.019	0.100	0.06-0.216	n=9	0.0488	<0.02	<0.02	<0.02	n=8
Cyanobacteria	Present			NO		present			NO		present a lot							
Microcystin	0.138	0-1	n=30	0.053 n=33	0.01-0.27	0.132 n=34	0.01-1.05		0.0548	0.01-0.30	0.138	0.01-0.75	n=30	0.026	0-0.09	0.029	0-0.24	n=19

Supplementary Table 2. Global amount of cyanobacteria in different steps of the pre-treatment process

	Raw	After control treatment	After contact filter treatment	Control treatment pond	Contact filter pond	Contact filter well	Control filter well
24th of July	x				x		
21st of Aug.	x					x	x
26th of Aug.							
29th of Aug.						x	x
3rd of Sep.	x						
15th of Sep.						x	x
17th of Sep.							
1st of Oct.				x	x	x	x
6th of Oct.	x					x	x
15th of Oct						x	x
20thof Oct						x	x

	Extensive amount to blooms
	Large amount
	Medium
	few to medium
	Zero to few
x	No samples

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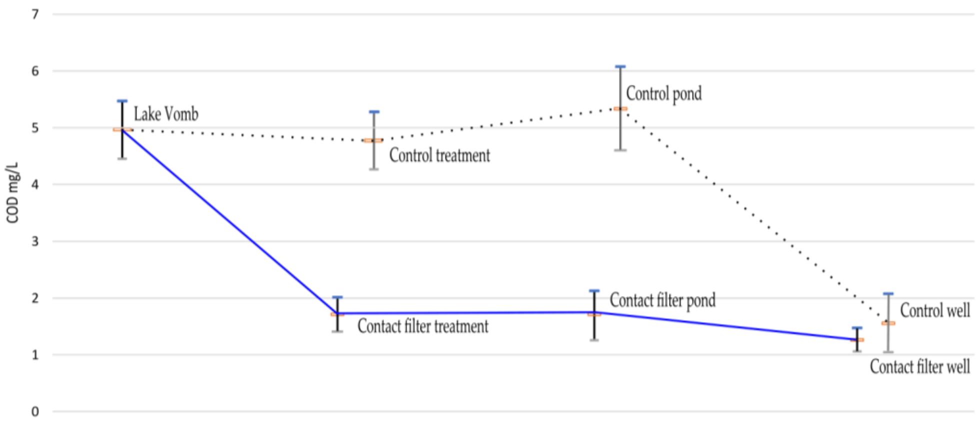
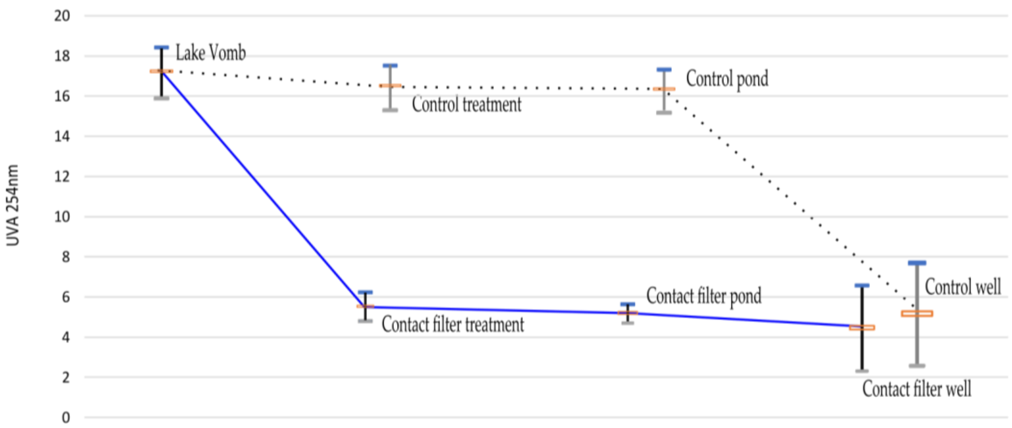
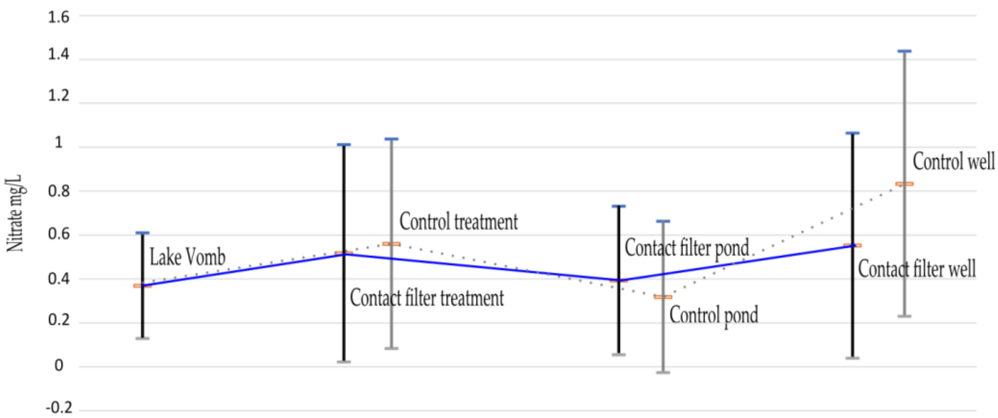


Supplementary Figure 1. COD measurements at different stages along the two treatment processes (mg/l) (The middle bar is the mean value and the top and lower bars show the standard deviation. The dotted line is the contact filter treatment process and the solid line is the control treatment process.)



Supplementary Figure 2. UVA 254 nm at different stages along the two treatment processes (The middle bar is the mean value and the top and lower bars show the standard deviation. The dotted line is the contact filter treatment process and the solid line is the control treatment process.)



Supplementary Figure 3. Nitrate content at different stages along the two treatment processes (mg/l) (The

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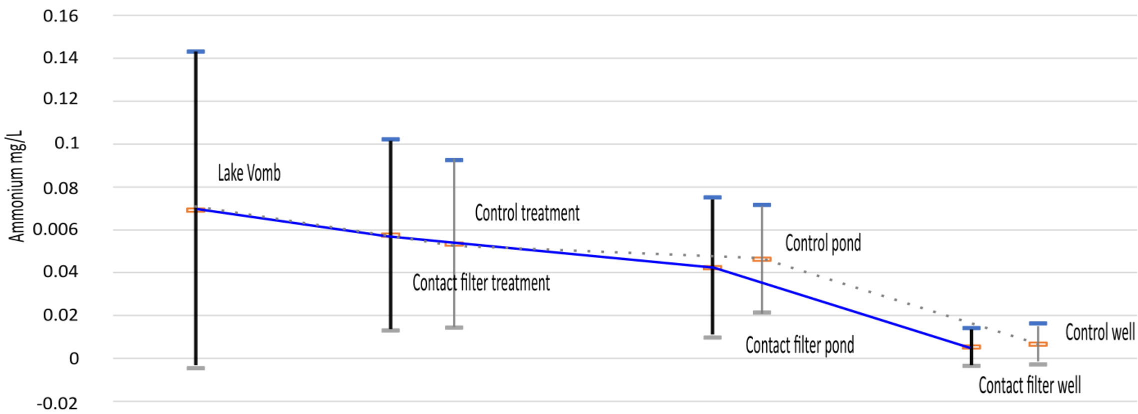
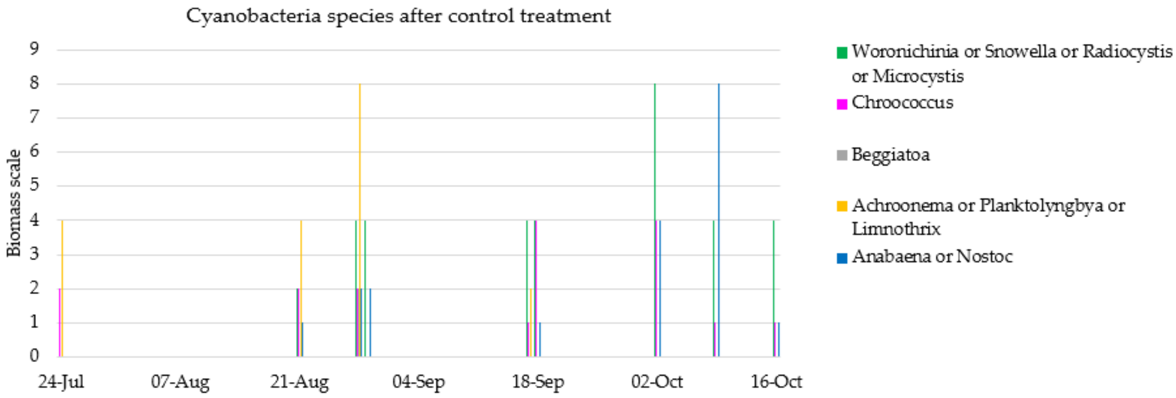
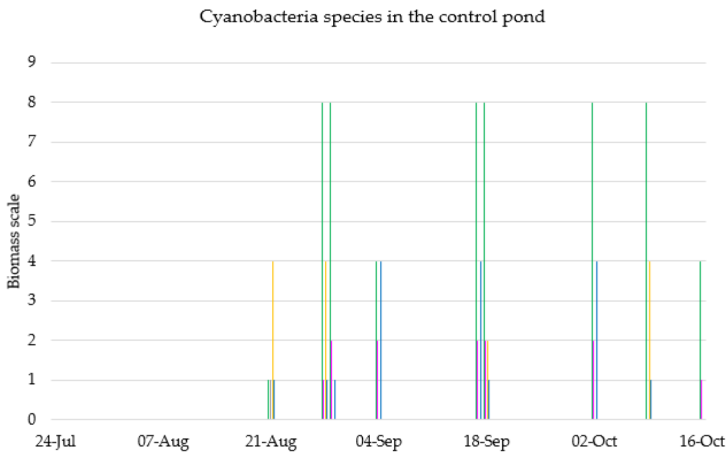
middle bar is the mean value and the top and lower bars show the standard deviation. The dotted line is the contact filter treatment process and the solid line is the control treatment process.)



Supplementary Figure 4. Ammonium content at different stages along the two treatment processes (mg/l) (The middle bar is the mean value and the top and lower bars show the standard deviation. The dotted line is the contact filter treatment process and the solid line is the control treatment process.)





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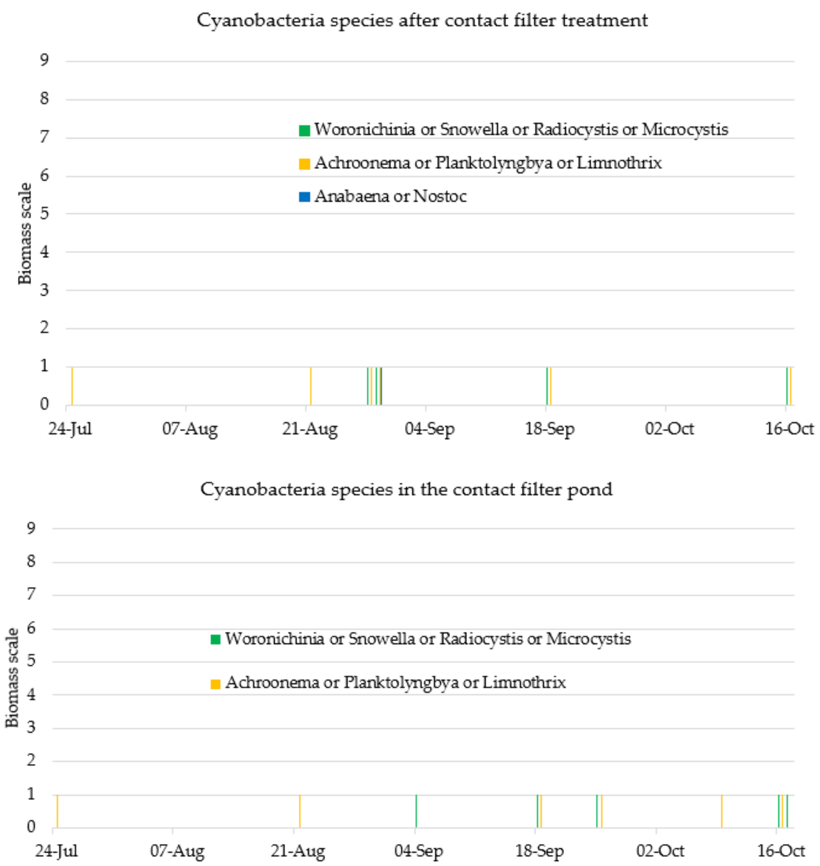
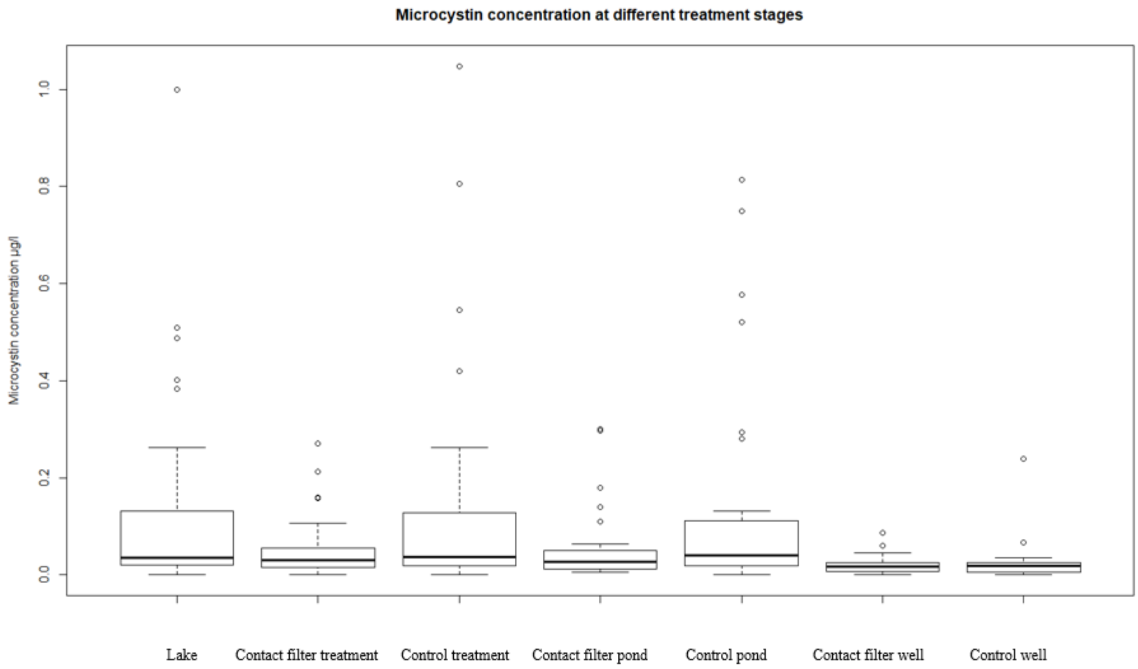


Supplementary Figure 5. Changes of algae types in the two treatment processes, the biomass scale is the area covered by algae i.e. 1= 10%, 2=20% and so on.



Supplementary Figure 6. Microcystin concentration at different treatment processes