



Removal of algae from raw water by ultrasonic irradiation and flocculation: A pilot scale experiment

Guozhong Yu*, Chengmei Zhao, Bing Liu, Qingfei Li and Hui Gao

School of Urban and Environmental Science, Xinyang Normal University, Xinyang, 464000, China

*Corresponding Author email : ygz1966@163.com

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Abstract

Using Chl *a* removal rate as index, a 28 kHz/900w ultrasonic cleaning machine was applied to testify algal removal by ultrasonic irradiation from raw water of a pool, where *Microcystis aeruginosa* colonies is absolutely dominated with temperature being over 20°C, and the irradiation lasted for 5 min. PAC was used as flocculant at the dose of 60 mg l⁻¹, jar tests were done to investigate the Chl *a* removal by flocculation. The results showed that ultrasound raised the water temperature instantly but did not lead to a regular pH change pattern in all the treated samples. Ultrasound could remove more than 90% of Chl *a* from raw water with temperature over 31°C, but less than 20% of Chl *a* or even increased Chl *a* concentration in some samples with temperature lower than 31 °C. Compared with the algal removal effects by direct raw water flocculation with PAC, ultrasound did not enhance markedly the flocculation effects on algae removal, which was not in agreement with the findings reported. The reason might be due to morphology and characteristics of natural algae differed greatly from that of algae cultured in laboratory.

Key words

Ultrasonic irradiation, Algal removal, PAC flocculation, Pilot experiment, Raw water

Introduction

In recent years, ultrasound has been used to treat algae, especially Cyanobacteria in water. Since it can integrate advanced oxidation, burning and supercritical in one process with simple operation, no secondary pollution (Mao *et al.*, 2006) and harm to aquatic life (Chu *et al.*, 2008). This technology has been regarded as a reliable, safe and environment friendly water algal control process (Nakano *et al.*, 2001; Ahn *et al.*, 2003; Song *et al.*, 2005; Zhang *et al.*, 2006a; Chu *et al.*, 2008; Joyce *et al.*, 2010). Ultrasound removes algae by mechanism of ultrasonic cavitations, such as high pressure, shock wave, acoustic streaming and shear force, which could disrupt the vesicle, inactivate enzymes (Shu *et al.*, 2008), damage antenna complexes in cell or even fracture the cell (Zhang *et al.*, 2006b). Frequency, intensity and treatment time of ultrasonic wave were thought to be important factors for algal control. Higher ultrasonic frequencies removed more algal (Zhang *et al.*, 2006a; Joyce

et al., 2010). 5 min for ultrasonic duration was applied by some researchers and found it could cause gas vesicles to collapse (Lee *et al.*, 2002; Zhang *et al.*, 2006a; Shu *et al.*, 2008; Dong *et al.*, 2008), effectively damage antenna complexes (Zhang *et al.*, 2006b) and inhibit microcystins formation and release (Lee *et al.*, 2002; Zhang *et al.*, 2006a). However, ultrasound treatment of 5 min was found to cause cell rupture in blue-green algae and thus increase extracellular microcystins (Dong *et al.*, 2008; Zhang *et al.*, 2009). Considering energy being exhausted, lower power and suitable frequency ultrasonic technology on algae control has become a research target.

However, most successful ultrasound work on algal control was performed under laboratory conditions where the treated algae were cultured. It is well known that the characteristics of some algal species (especially *Microcystis aeruginosa*) cultured in laboratory differs to some extent from that of the ones living in nature which may play a

critical role in algal control during water treatment processes. The risks concerning the release of Cyanobacterial toxins by ultrasonic irradiation has shown contrary views (Lee *et al.*, 2002; Ma *et al.*, 2005; Ross *et al.*, 2006; Dong *et al.*, 2008) and it was also found that ultrasound selectively controlled algal species (Chu *et al.*, 2008). In view of the above reasons, it could be inferred that this technology is not well established for full scale testing (Colucci *et al.*, 2010). So, it is necessary to investigate its practical efficiency although few *in situ* experiments have showed effective Cyanobacteria bloom control (Ahn *et al.*, 2003; Ding *et al.*, 2009). This paper investigated the effectiveness of algal control by ultrasound and compared the effects of PAC flocculation on algal removal from raw water and effluent of the operator.

Materials and Methods

Materials : XR28 ultrasonic cleaning machine with 28 kHz/900W and inner tank size 500 × 380 × 220 mm (Zhangjiagang Xinren Ultrasonic Equipment Co., Ltd, Zhangjiagang, China) was used. TJ6 series coagulation mixing instrument (Hengling Science & Technology Co., Ltd, Wuhan, China) was used for jar tests. pH and water temperature were determined by pH meter (Meixiang Instrument Co., Ltd., Shanghai, China). M-50 water filter (Shanghai Pharmaceutical Glass Factory), 0.45 µm Whatman GF/C with Φ47 mm, 2XZ-1 rotary vane vacuum pump (Zhejiang Huangyan Vacuum Pump Factory, China), TD5A desk centrifuge with Maximum speed of 5000 rpm (Shanghai Chemical Machinery Plant Co. Ltd., China) and UV/Vis spectrophotometer (Shanghai Jinghua Science & Technology Company, Ltd., China) etc., were used to measure Chl *a* concentration of sample. For Chl *a* estimation 90% acetone solution, 1% suspension of MgCO₃ and 1mol HCl, were prepared in laboratory. Polymeric aluminum chloride (PAC) was offered by a local water supply works of China.

Water samples : The tested raw water was drawn from a man-made shallow pool, where rain water is the only source of water. The experiments were conducted intermittently from 20th August to 27th October 2011. During this period, *M. aeruginosa* was significantly dominant over other algal species and always emerged in the form of amorphous aggregates. Therefore, it was very difficult to count its number clearly under microscope. A few species of Chlorophyta such as *Scenedesmus obliquus*, *Chlorella* etc., and few diatom as well as flagellate were also found, but their number was small during this period. When the temperature of raw water dropped to 15.4°C, the dominance of *M. aeruginosa* was gradually replaced by few species of Chlorophyta such as *Chlorella*. Few *M. aeruginosa* cells emerged in water at temperature below 15°C.

Analytical methods : 10 l of raw water from the pool was brought to the laboratory and mixed well. The pH and temperature were measured and 6 l of mixed water was poured into XR28 tank and ultrasonicated immediately. 5 min ultrasound exposure was applied in this experiment although there were contrary reports about microcystins released during ultrasound treatment. The pH and temperature of the treated water were measured immediately.

Jar tests were done with the TJ6 series coagulation mixing instrument. One litre each of raw water and effluent of XR28 tank were poured into two jars respectively. PAC at the dose of 60 mg l⁻¹ was added into each jar, which was determined in previous investigations (Zhao *et al.*, 2011) and flocculated. The flocculation was operated under conditions of rapid mixing of 150 rpm for 2 min followed by slow mixing of 40 rpm for 15 min, then sedimentation for 30 min. The supernatant from each jar was used for Chl *a* estimation.

Chl *a* concentration was used as algal biomass index and determined following the acetone extraction methods given by Jin *et al.* (1990). Since 0.45 µm filter was easily clogged when *M. aeruginosa* dominated the water, 250ml sample of raw water and effluent of XR28 tank and the two supernatant, respectively, was used for Chl *a* estimation. The average of the three measured values was regarded as the final concentration of each sample. Chl *a* removal (%) represented algal control effects by ultrasonication or flocculation.

Results and Discussion

The temperature and pH change in the water samples before and after ultrasound treatment is given in Table 1. Compared with raw water, 5 min of ultrasound treatment instantly raised the temperature in all the treated samples. When the temperature of raw water was lower than 20°C, the ultrasound treatment could raise temperature over 20°C.

However, the ultrasound treatment did not lead to a regular pattern change of pH in all the water samples, pH increased in some samples while decreased in others. This result was inconsistent with the previous reports where pH decreased after 5 min of sonication (Ahn *et al.*, 2003; Zhang *et al.*, 2006a), and the reason might lie in the pool system differs greatly from the cultured *M. aeruginosa* system in laboratory. The cultured system in laboratory is single, where ultrasound can easily cause the acidic intracellular and extracellular algal polysaccharides released into water to decrease pH. The pool is open to nature, where complex reactions occurred due to ultrasound and led to the irregular change in pH. Surely, to natural water, further studies are needed to come to a conclusion whether ultrasound treatment increases or decreases the pH value.

Table 1 : Temperature (°C) and pH of water samples before and after ultrasound treatment

Date	20 th Aug	29 th Aug	1 st Sep	11 th Sep	19 th Sep	20 th Sep	22 nd Sep	27 th Sep	9 th Oct	27 th Oct
Tem ^b	25.3	30.9	31.7	21.8	17.1	17.5	18.4	24.2	26.9	15.4
Tem ^a	29.2	33.7	33.8	26.4	20.2	23.9	22	26.4	28.6	20.6
pH ^b	6.93	8.19	8.31	7.19	6.93	8.05	7.94	6.91	6.49	6.45
pH ^a	6.89	8.31	8.34	7.41	6.89	7.86	8.09	7.54	7.04	7.23

Tem^b, pH^b - temperature and pH of samples before ultrasound treatment; Tem^a, pH^a - instant temperature and pH in effluents from ultrasound treatment unit

Table 2 : Chl *a* concentration (µg l⁻¹) in raw water samples and its removal by ultrasound treatment unit

Date	20 th Aug	29 th Aug	1 st Sep	11 th Sep	19 th Sep	20 th Sep	22 nd Sep	27 th Sep	9 th Oct	27 th Oct
Chl <i>a</i> ¹	382.2	423.15	277.55	395.8	81.9	436.8	341.25	368.55	259.35	286.65
Chl <i>a</i> ²	316.7	423.15	27.3	518.7	109.2	419.4	313.95	450.45	232.05	259.35
Removal(%)	17.1	0	90.2	-31	-33.3	4	8	-22.2	10.5	9

Chl *a*¹ - Chl *a* concentration in raw water samples; Chl *a*² - Chl *a* concentration in effluents from ultrasound treatment

Table 3 : Chl *a* concentration (µg l⁻¹) of raw water and its removal by direct flocculation and the process combined ultrasound with flocculation

Date	20 th Aug	29 th Aug	1 st Sep	11 th Sep	19 th Sep	20 th Sep	22 nd Sep	27 th Sep	9 th Oct	27 th Oct
Chl <i>a</i> ¹	382.2	423.15	277.55	395.8	81.9	436.8	341.25	368.55	259.35	286.65
Chl <i>a</i> ³	559.65	13.65	27.3	368.55	27.3	40.95	40.95	54.6	13.65	9.1
Removal(%)*	-46.4	96.8	90.2	6.8	66.6	90.6	88	85.1	94.7	96.8
Chl <i>a</i> ⁴	259.35	13.65	13.65	436.8	68.25	81.9	27.3	54.6	54.6	9.1
Removal (%)**	32.1	96.8	95	-10.3	16.6	81.2	92	85.1	78.9	96.8

Chl *a*³ - Chl *a* concentration in effluents from flocculation only; Chl *a*⁴ - Chl *a* concentration in effluents from the process combined ultrasound with flocculation; Removal(%)* - removal of Chl *a* concentration by flocculation only; Removal (%)** - removal of Chl *a* concentration by ultrasound and flocculation

The removal of Chl *a* content from raw water by ultrasound treatment is given in Table 2. The maximum removal of Chl *a* from raw water samples was observed on 1st September. While on other days, Chl *a* removal was much lower and was zero on 29th August. On 11th, 19th and 27th September, Chl *a* concentration increased over 22% after ultrasonication. Therefore, ultrasound treatment did not remove the living algae remarkably in the present study, which is inconsistent with the previous studies (Nakano *et al.*, 2001; Ahn *et al.*, 2003; Song *et al.*, 2005; Zhang *et al.*, 2006a; Chu *et al.*, 2008; Joyce *et al.*, 2010). However, to a great extent our result corresponds well with the previous report of Wang *et al.* (2005) that showed 9 min of ultrasound irradiation removed 25% of *M. aeruginosa*.

The lower Chl *a* removal in the present study might be due to following reasons: In some algae, especially *M. aeruginosa*, the morphology in pure or single culture system differs markedly from that in natural waterbody. Under natural environment stresses, it is well known *M. aeruginosa* cells often aggregate together to form many amorphous

colonies, in which single cell is wrapped up by extracellular matters secreted by alga cells. Extracellular matters play a very important role in preventing cells being wrapped up against outside attack, prey or destroy e.g., grazing from *Ochromonas sp.* caused morphological and physiological changes of the alga (Yang *et al.*, 2008). However, cells cultured under laboratory conditions often have single and less extracellular matter covering each cell, and are easily attacked, preyed or destroyed (Zhang *et al.*, 2007; Yang *et al.*, 2008). Colonization of *M. aeruginosa* cells might be a mechanism for field *M. aeruginosa* to effectively prevent it against harm from ultrasound, which affirmed to a certain extent the finding that *M. aeruginosa* cells were minimally damaged by free radicals from ultrasonic irradiation (Ahn *et al.*, 2003).

Thus, ultrasonic irradiation of 5 min was not enough to destroy the cells' defensive mechanisms or kill them. Ahn *et al.* (2003) and Ding *et al.* (2009) found that increased ultrasonic treatment (1 hr and 3 days respectively) could effectively inhibit algal growth.

As short term exposure of ultrasound could not completely inhibit or destroy all the cells, 5 min ultrasound irradiation actually stimulated Chl *a* concentration in water samples with optimal temperature for cell growth on 11th, 19th and 29th September. Once the irradiation was stopped, *M. aeruginosa* immediately recovered its growth (Ahn *et al.*, 2003).

The maximum Chl *a* removal observed on 1st September might be due to the higher water temperature this time. Higher temperature had already done harm to *M. aeruginosa* cells in the pool, so less strong living cells showed in the raw water sample, and ultrasonic irradiation could easily damage them. It implies that longer irradiation time may be beneficial for ultrasound to destroy natural *M. aeruginosa* cells, otherwise, the cells will set up mechanisms to prevent them from ultrasonic harm, and thus, ultrasound will lose its function on *M. aeruginosa* control. This study suggests that it should be very prudent to apply the results of cultured *M. aeruginosa* removal by ultrasound in laboratory into practice although some researches achieved higher removal in shorter irradiation time (Lee *et al.*, 2002; Hao *et al.*, 2004; Shu *et al.*, 2008; Ding *et al.*, 2009).

The Chl *a* removal from raw water samples and effluents of XR28 tank by PAC flocculation respectively are given in Table 3. Both the processes showed better flocculation results under raw water temperature over 30°C or below 16°C. On the other hand, the process of ultrasound combined with PAC flocculation did not show its advantage over flocculation only at all due to lower Chl *a* removal, so, ultrasound did not greatly enhance flocculation for algal control, which is also inconsistent with the earlier reports (Lee *et al.*, 2002; Zhang *et al.*, 2006b; Zhang *et al.*, 2009). After flocculation, the maximum increase of Chl *a* concentration in the sample on 20th August was due to the fact that 25°C was approximately optimal for *M. aeruginosa* cells growth, which was beneficial to forming light and loose floc by PAC, so the sample used for Chl *a* estimation contained numerous cells and led to Chl *a* rising in the sample.

Generally, the suitable water temperature for *M. aeruginosa* cell growth varies between 15 and 30°C. When water temperature is over 30°C or below 15°C, *M. aeruginosa* cell's activations are inhibited and other algae gets chance to dominate the water. Even more, the alga will sink to the water column bottom so that few *M. aeruginosa* cells can be observed in water phase. Under such conditions, intervention from *M. aeruginosa* cells in flocculation is weakened. Thus, by flocculation it is possible to remove more algae, and ultrasound treatment is not required at all (Table 3). Thus the pilot scale study could not deny completely that 5 min of ultrasound irradiation could inhibit *M. aeruginosa* cell growth to a certain extent. But due to low

Chl *a* removal rate by ultrasound process, it was difficult to identify the exact way by which algae were controlled in this experiment.

The results of algal removal in this study is not in agreement with the previous findings either by ultrasound only or the combination of ultrasound with PAC flocculation. For the *M. aeruginosa* dominating natural water, more bench or full scale investigations should be carried out to clarify whether ultrasound can enhance flocculation on this species.

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References

- Ahn, C.Y., H.P. Myung, H.J. Seung, S.K. Hee, Y.J. Kam and M.O. Hee: Growth inhibition of cyanobacteria by ultrasonic radiation: Laboratory and enclosure studies. *Environ. Sci. Technol.*, **37**, 3031-3037 (2003).
- Chu, Z.S., P. Yan, F.Z. Shuo and X.C. Jin: Algal control by ultrasonic radiation and its risks to the aquatic environment. *Acta Scientiae Circumstantiae*, **28**, 1335-1339 (2008).
- Colucci, L.A.: Ultrasonic Algal Control Literature Review. City of Austin Watershed Protection, **7**, 1-5 (2010).
- Ding, Y., P.P. Yue, H.Y. Li, X.Q. Xiao, Y.H. Li. and W. Wu: Parameters optimization of ultrasound algae removal technology and bloom removal study in Taihu Lake. *J. Southeast Uni. (Nat. Sci. Edition)*, **39**, 354-358 (2009) (in Chinese).
- Dong, M.Y., J.L. Qiao, G.Q. Wang and Z. Wang: Study on microcystin release and degradation by ultrasonic. *Wat. Purific. Technol.*, **27**, 21-23 (2008) (in Chinese).
- Hao, H.W., M.S. Wu, Y.F. Chen, J.W. Tang and Q.Y. Wu: Algae bloom control by ultrasonic irradiation at 20 kHz and 1.7 MHz. *J. Environ. Sci. Hlth., Part A*, **39**, 1435-1446 (2004).
- Jin, X.C. and Q.Y. Tu: Standard on lake eutrophication survey. 2nd Edn., China Environmental Science Press, Beijing, China. (1990) (in Chinese).
- Joyce, E.M., X.G. Wu and T.J. Mason: Effect of ultrasonic frequency and power on algae suspensions. *J. Environ. Sci. Hlth., Part A: Toxic Haz. Sub. Environ. Eng.*, **45**, 863-866 (2010).
- Lee, T., K. Nakano and M. Matsumara: A novel strategy for cyanobacterial bloom control by ultrasonic irradiation. *Wat. Sci. Technol.*, **46**, 207-215 (2002).
- Ma, B.Z., Y.F. Chen, H.W. Hao, M.S. Wu, B. Wang, H.G. Lv and

- G.M. Zhang: Influence of ultrasonic field on microcystins produced by bloom-forming algae. *Colloids and Surfaces B: Biointerfaces*, **41**, 197-201 (2005).
- Mao, Y.H., J.Y. Li and Y.J. Liu: Technology and application of water treatment by ultrasonic wave. *Ind. Water Treat.*, **26**, 10-13 (2006) (in Chinese).
- Nakano, K., T.J. Lee and M. Masatoshi: *In situ* algal bloom control by the integration of ultrasonic radiation and jet circulation to flushing. *Environ. Sci. Technol.*, **35**, 4941-4946 (2001).
- Ross, C., S.V. Lory and P. Valerie: Toxin release in response to oxidative stress and programmed cell death in cyanobacterium *Microcystis aeruginosa*. *Aqua. Toxicol.*, **78**, 66-73 (2006).
- Shu, T.G., B.L. Yuan and S.R. Wang: Studies on the removal of *Microcystis aeruginosa* by low-power ultrasonic. *J. Huaqiao Uni. (Nat. Sci.)*, **29**, 72-75 (2008) (in Chinese).
- Song, W.H., T. Terri, R. Kathleen and E.O. Kevin: Ultrasonically induced degradation and detoxification of microcystin-LR (cyanobacterial toxin). *Environ. Sci. Technol.*, **39**, 6300-6305 (2005)
- Wang, B., G.M. Zhang and H. Wang: Removal of *Microcystis aeruginosa* by ultrasound. *Tech. Equip. Environ. Pollu. Contr.*, **6**, 46-49 (2005) (in Chinese).
- Yang, Z., F.X. Kong, X.L. Shi, M. Zhang and H.S. Cao: Grazing effect of *Ochromonas* sp. on morphological and physiological characteristics of *Microcystis aeruginosa*. *J. Lake Sci.*, **20**, 403-408 (2008) (in Chinese).
- Zhang, D.L. and G.Z. Yu: Treatment of *Microcystis aeruginosa* in eutrophic raw water with NaOCl and H₂O₂. *J. Henan Uni. (Nat. Sci.)*, **37**, 245-248 (2007) (in Chinese).
- Zhang, G.M., P.Y. Zhang, B. Wang and H. Liu: Ultrasonic frequency effects on the removal of *Microcystis aeruginosa*. *Ultrason. Sonochem.*, **13**, 446-450 (2006a).
- Zhang, G.M., P.Y. Zhang, H. Liu and B. Wang: Ultrasonic damages on cyanobacterial photosynthesis. *Ultrason. Sonochem.*, **13**, 501-505 (2006b).
- Zhang, G.M., B. Wang, P.Y. Zhang, L. Wang and H. Wang: Removal of algae by sonication-coagulation. *J. Environ. Sci. Hlth., Part A: Environ. Sci. Eng. Toxicol.*, **41**, 1379-1390 (2006).
- Zhang, G.M., P.Y. Zhang and M.H. Fan: Ultrasound-enhanced coagulation for *Microcystis aeruginosa* removal. *Ultrason. Sonochem.*, **16**, 334-338 (2009).
- Zhao, C.M., D.L. Zhang, M.G. Niu, H. Gao, B. Huang and G.Z. Yu: Removal of *Microcystis aeruginosa* from a raw water with an up-flow bioreactor. *J. Xinyang Norm. Uni. (Nat. Sci. Edn.)*, **24**, 525-528 (2011) (in Chinese).