



Ultrasonic as Green Chemistry for Bacterial and Algal Control in Drinking Water Treatment Source



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Abstract

This paper studied the enhanced effects of ultrasonic as non-chemical oxidant on traditional coagulation of raw water with alum for microorganisms' removal. The treatment processes were studied using ultrasonic frequencies; 20, 40 and 60 KHz at different time intervals namely 15, 30, 45 and 60 minutes. The study revealed that removal percentage of Total Coliform, Faecal Coliform and Faecal Streptococcus ranged between 5% - 46%. There was a positive correlation between ultrasonic intensity, sonication time and bacterial removal. There are clear morphological changes in the algal organisms without cell disruption especially green algae due to the release of photosynthetic pigments "Chlorophyll". No changes of chlorophyll "a" content were detected. Pre-treated samples with 20, 40, and 60 KHz ultrasonic frequencies for 60 min decreased the alum dose by 6.7 %, 13.3 % and 20 % respectively.

Keywords : Ultrasonic; Total coliform; Faecal Coliform; Faecal Streptococcus; algae; Chlorophyll "a"; Coagulation; Alum.

Introduction:

"Water is life"; actually, water is the most plentiful compound on Earth and is vital natural resource for human health and essential to life. It is most important that, the water which people drink and use for other purposes must be **clean water**. The presence of microbial pathogens in source waters, drinking water and recreational water bodies is a global problem. Microbial contamination of drinking-water contributes to disease outbreaks and background rates of disease in developed and developing countries worldwide [1].

Control of waterborne disease is an important element of public health policy and an objective of water suppliers. Waterborne diseases caused by the consumption of contaminated water

can affect a large number of people in a short time [2, 3]. The World Health Organization (WHO) has reported that two million deaths related to such diseases occur worldwide each year, primarily in individuals under 5 years of age. In addition, approximately 663 million people continue to lack access to improved drinking water sources [4]. Significant sources of water pollution include human sewage and animal waste poured into water distribution systems and surface water [5, 6].

Algal blooms are a major worldwide water treatment concern due to their potentially harmful effects on humans and livestock. Algal blooms can cause release of toxins which in high concentrations are a serious health hazard for humans and animals. Cyanobacterial blooms adaptation of rapid cell growth causes higher rates of nutrients and oxygen

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consumption [7, 8, 9, 10, and 11]. From a water treatment perspective filter clogging can occur due to presence of diatom species such as *Melosira* sp. Green species increase turbidity and chlorophyll "a", and cyanobacteria produce taste and odor issues related usually to toxin release. Green algae and cyanobacteria are the two main algal divisions responsible for increased chemical demand required to treat a water supply [12, 13, and 14].

Raw water is not safe to drink, so it is treated. All the action taken to make sure that drinking water is potable is called **water treatment**. Meeting the goal of clean, safe drinking water requires a multi-barrier approach that includes: protecting source water from contamination, appropriately treating raw water, and ensuring safe distribution of treated water to consumers' taps [15]. Pre-oxidants (such as ozone, chlorine dioxide, chlorine, or permanganate) are required to enhance coagulation as they have the capacity to inactivate microorganism's cells [16, 17, 18, 19, 20 and 21]. Historically, chlorine has been the most widely used and the principal oxidant employed in water treatment plants can reduce water borne diseases [22, 23, and 24].

Processes for removal of different groups of organisms and pollutants from water include chlorination (Oxidant and disinfectant), coagulation, flocculation and sedimentation; and filtration. Most drinking water must be treated with disinfectants in order to kill or inactivate naturally occurring organisms that can potentially cause illness and diseases. This process is called disinfection. The use of chlorine (Cl_2) as a water disinfectant has come under scrutiny because of its potential to react with natural organic matter (NOM) and form chlorinated disinfectant by-products (DBPs) of public health concern during the chlorination process [25, 26]. Within this context, NOM serves as the organic DBP precursor, whereas bromide ion (Br^-) serves as the inorganic precursor. More than 600 DBPs have been identified to date [27, 28]. Treatment strategies generally available to water systems exceeding drinking-water standards include removing DBP precursors and using alternative disinfectants for primary and/or secondary (distribution system) disinfection [29, 26].

There is a trend within the water treatment industry to develop and employ more environmentally responsible technologies to help reduce the impact of chemicals in effluent waters and reduce water consumption in the process [30].

Advanced oxidation processes (AOPs) are being deployed to solve challenging drinking water quality problems. Ultrasound use was classified as a non-chemical strategy to control algal growth when documented by a large metropolitan utility company in the U.S. Midwest. Ultrasound is an AOP that has been investigated as a possible technology for advanced water treatment [31, 32, and 33]. Ultrasonic applications for water treatment include cell disruption, sterilization, dispersion of solids, extraction of plants, anaerobic digestion, water remediation, and sewage. The ability of ultrasound to break cell membranes found application in biological research, for example, for the extraction of lipids, proteins or enzymes from cells. Also now a day's ultrasound is widely used as a chemical-free way of water treatment. Main advantage of ultrasound usage in this field is that it is relatively cheap, extremely easy to install and operate [34, 35, 36, and 37].

So, the main aims of this study are: to study

- 1- The effect of ultrasonic on inactivity Bacteria and Algae of Raw water.
- 2- Improvement of coagulation efficiency without chlorine addition.

Materials and Methods

1. *Sampling site description:* Water samples were collected during the study from the intake of Shoubra Al-Khamah water treatment plants (Fig. 1).
2. *Water quality:* Physico-chemical, bacteriological and biological parameters were carried out according to APHA (2017) [38].
 - 2.1. *Physico-chemical characters:* Temperature and pH were measured using pH-meter model (Janway 3150) while turbidity measured using Hach 2100 DN Turbidimeter equipment according to Nephelometric Method # 2130 B.
 - 2.2. *Algae samples and counting:* Samples for biological analysis were also brought to laboratory and enumeration of phytoplankton was carried out. Diatoms, green algae, blue green algae and total algae were counted by Olympus compound microscope BX-53 in a Sedgewick-Rafter Counting chamber.
 - 2.3. *Determination of Chlorophyll "a" using UV-VIS Shimadzu Spectrophotometer Model (UV-1800) according to Method no. 10200 H.*

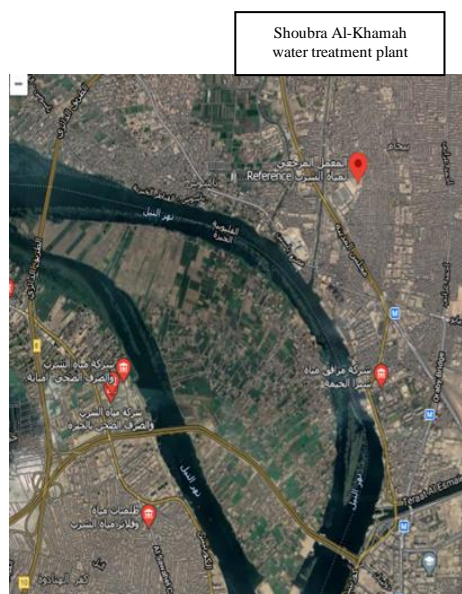


Fig. (1) Sampling Site

2.4. **Bacteriological Examination:** Media of bacterial culture used in different bacterial groups was Merck Products.

2.4.1. Detection and Enumeration of Total Coliform by Membrane Filter Technique (Method no. 9222 B.).

2.4.2. Detection and Enumeration of Faecal Coliform by Membrane Filter Technique Method (Method no. 9222 D).

2.4.3. Detection and Enumeration of Faecal Streptococcus and Enterococcus Groups by Membrane Filter Technique (Method no. 9230 C).

2.5. **Ultrasonic Equipments:**

➤ 20 KHz was 3510E-DTH BRANSON.

➤ 40, 60 KHz was LBS2 10 Lt.

3. **Water treatment processes:**

Experiments were carried out using 1% Aluminium Sulfate soln. (Panreac Aluminium Sulfate 18-hydrated PRS), through **Jar Test** [39].

- **Jar Test:** Coagulation and flocculation were conducted via the "Jar test" procedure. The apparatus used "Phipps & Bird" consists of multiple stirrer fitted with 6 paddles. This multiple stirrer is equipped with a speed regulator.

For the jar test one litter sample of raw Nile water were placed into jars and test coagulant added in

rising dosages. While stirrer set to flash mixing at a stirring speed 200 r.p.m. for 1-minute after which, the speed was further reduced in stepwise order down to 30 r.p.m. for another 15-minutes. Then the jars were then left 30-minutes to allow settling the formed flocs. This was followed by sample siphoning from the supernatant solution into clean containers. Characterization of water following coagulation was carried out for turbidity determination, Bacteriological examination and residual phytoplankton counts.

Results and Discussion:

The samples were sonicated at 20, 40 and 60 KHz for 15, 30, 45 and 60 mins. for each ultrasonic frequency. To examine the effect of ultrasonic on the viability of bacteria and algae; the following experiments were carried out.

1. Bacteriological Examination:

The biocidal effect of sonication at different frequencies using membrane filter technique is shown in Figs 2–4. The results recorded at 20, 40, and 60 kHz during different time of sonication namely 15, 30, 45 and 60 mins indicate a decline in cell numbers. Figs. 2, 3 and 4 showed a very remarkable reduction in bacterial count which is ranged between 5 % - 46 % for total coliform, faecal coliform and faecal Streptococcus. The lowest removal was detected for 20 KHz frequency at 15 mins. sonication while the highest removal was detected after sonication with 60 KHz for 60 mins.. Increase in bacterial removal was recorded when time of sonication and frequency increase.

Previous studies using low frequencies indicate a drop in cell numbers and increasing ultrasonic exposure time resulted in increased inactivation of bacteria [40, 41, and 42].

2. Effect of Ultrasonic on Morphological Characters and Chlorophyll "a" Content of Algae:

When comparing algae of raw water (Fig. 5) with algae in figures 6, 7 and 8, we found that; there are clear morphological changes in the algal organisms without cell disruption especially green algae due to the release of photosynthetic pigments "Chlorophyll a".

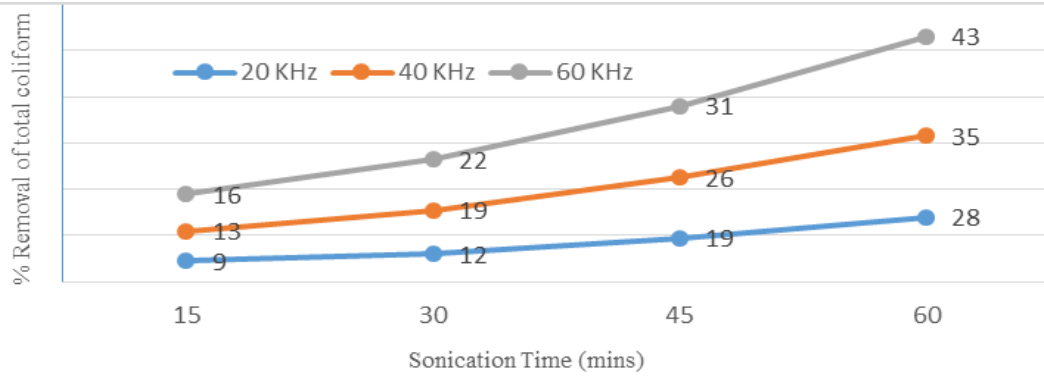


Fig. (2) Effect of Different Ultrasonic Frequencies at Different Time on Total Coliform Bacterial Removal

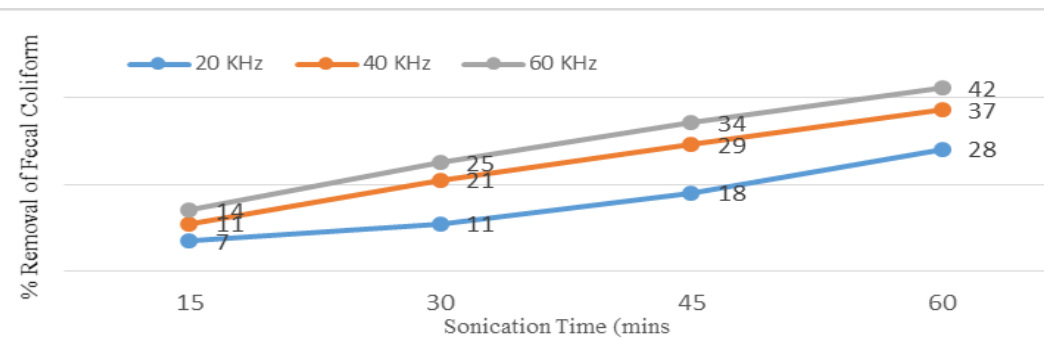


Fig. (3) Effect of Different Ultrasonic Frequencies at Different Time on Fecal Coliform Bacterial Removal

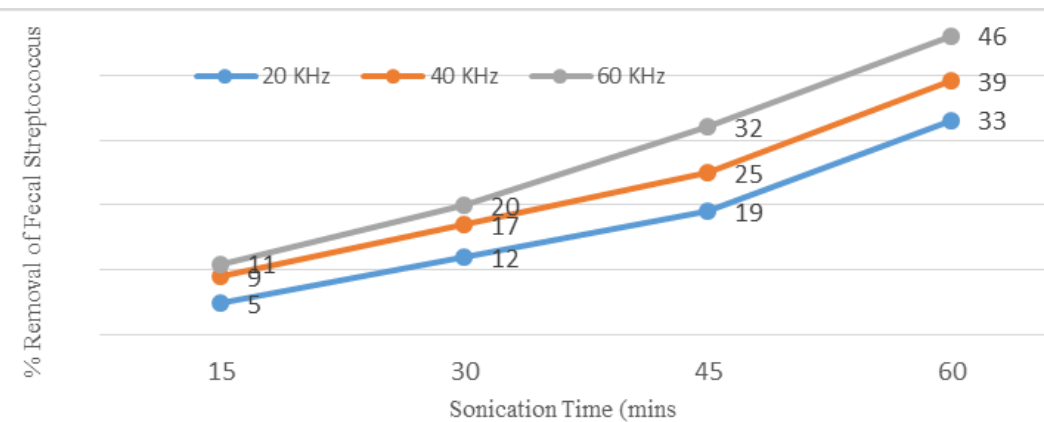


Fig. (4) Effect of Different Ultrasonic Frequencies at Different Time on Fecal Streptococcus Bacterial Removal

On the other hand; no remarkable removal or changes of chlorophyll "a" content was detected.

The ability of ultrasound to break cell membranes found application in biological research, for example, for the extraction of lipids, proteins or enzymes from cells [34]. Simon and Helliwell, 1998 [43], found that, without cell disruption, only a quarter of the

potential chlorophyll "a" was able to be extracted by an optimal method. Diane et al. 2013 [44], found that, Ultrasonic irradiation caused more impact to photosynthetic activity compared with cell removal for the colonial / unicellular algal species *Scenedesmus subspicatus*, and *Microcystis aeruginosa*.

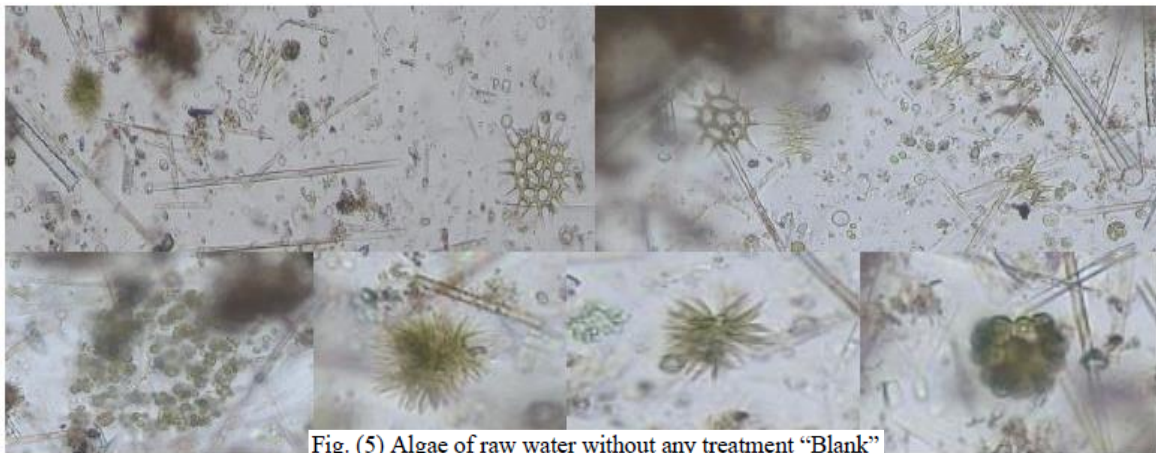
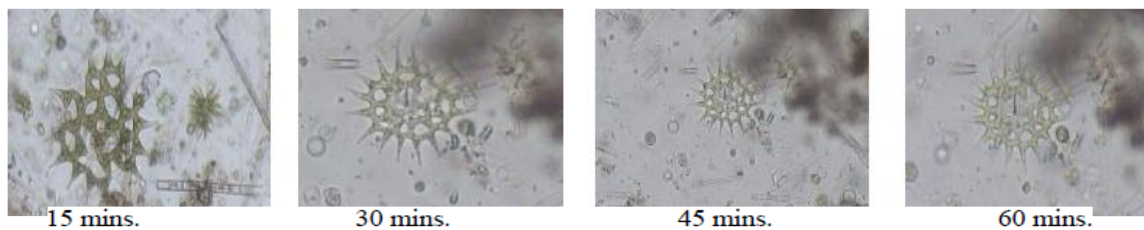


Fig. (5) Algae of raw water without any treatment "Blank"



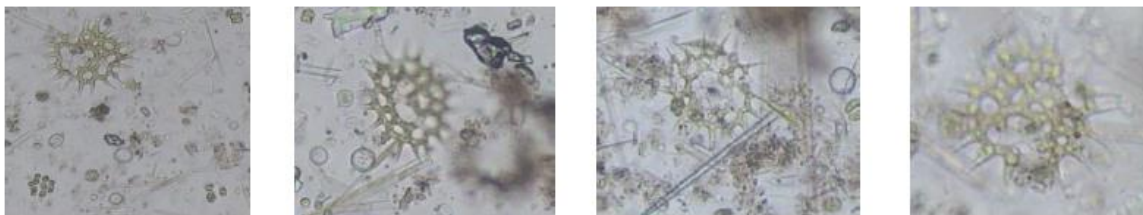
15 mins.

30 mins.

45 mins.

60 mins.

Fig. (6) Algae sonicated with 20 KHz.



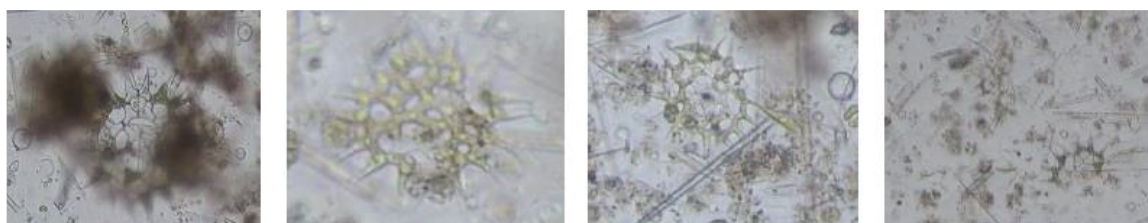
15 mins.

30 mins.

45 mins.

60 mins.

Fig. (7) Algae sonicated with 40 KHz.



15 mins.

30 mins.

45 mins.

60 mins.

Fig. (8) Algae sonicated with 60 KHz.

3. *Effect of Pretreatment with Ultrasonic on the performance of Alum for Water Treatment:*

Table (1) shows that, the optimal dose of alum was 30 mg/L which reduce the algal count by 84.6 % from the raw water algae content without any sonication. Mouchet and Bonnely 1995 [45], found that the removal rate of Nile water algae in the alum

clarified water at Cairo plants was 85% in the absence of chlorine. Also, Salwa, et al. 2002 [46], found that removing Nile water algae clarified with alum was 79%. Qiaohui, 2011 [47], stated that 98% of algae was removed with alum coagulation.

Pre-treated samples with 20, 40, and 60 KHz ultrasonic frequencies for 60 min were used to study the effect of ultrasonic on the efficiency of alum. The performance of alum was detected by

Table (1) Determination of Optimal Aluminum Sulphate Dose
Raw Nile Water: - Total Algal Count = 15620 Org./ml
Turbidity = 8.5 NTU

Alum Doses mg/L	18	22	26	28	30	32
Parameters						
Total Algal Count Org./ml	6427	5446	4157	3601	2400	2648
% Removal of Algae	58.6	65.1	73.4	76.9	84.6	83.1
Turbidity NTU	5.9	5.3	4.7	3.9	2.7	2.8
% Removal of Turbidity	30.6	37.6	44.7	54.1	68.2	67.1

Table (2) Effect of Ultrasonic Frequencies on Alum Dose for Algae and Turbidity Removal
Raw Nile Water: - Total Algal Count = 15620 Org./ml
Turbidity = 8.5 NTU

Treatment Parameters	30 mg Alum	20 mg/L Alum +			22 mg/L Alum +			24 mg/L Alum +			26 mg/L Alum +			28 mg/L Alum +			30 mg/L Alum +		
		20 KHz	40 KHz	60 KHz	20 KHz	40 KHz	60 KHz	20 KHz	40 KHz	60 KHz	20 KHz	40 KHz	60 KHz	20 KHz	40 KHz	60 KHz	20 KHz	40 KHz	60 KHz
Total Algal Count Org./ml	2400	6599	6012	6605	5064	4918	3318	4321	3750	2018	3172	2199	2098	2298	2515	2151	2315	2699	2425
% Removal of Algae	84.6	57.8	61.5	57.7	67.6	68.5	78.8	72.3	76	87.1	79.7	85.9	86.6	85.3	83.9	86.2	85.2	82.7	84.5
Turbidity NTU	2.7	5.4	5.1	4.6	4.9	4.7	3.2	4.5	4.1	2.2	3.3	2.1	2.6	2.2	2.4	2.1	2.1	2.6	2.5
% Removal of Turbidity	68.2	36.5	40	45.9	42.4	44.7	62.4	47.1	51.8	74.1	61.2	75.3	69.4	74.1	71.8	75.3	75.3	69.4	70.6

turbidity and algae count removal. Sonication of water had very remarkable effect on alum dose and improve the algal removal. Table (2) shows that, sonicated water with 20, 40 and 60 KHz decreased the alum dose by 6.7 %, 13.3 % and 20 % respectively. Anca, and Mariana, (2013) [48], found that, the dose of aluminium sulphate, decreases 8 times at the use of ultrasound waves. The results obtained in this study was consistent with the work of Zhang et al. (2006) [49] and Yitao et al. (2019) [33]. No effect had detected on pH and electric conductivity (EC) for water samples before and after sonication with different frequencies of ultrasonic at different times.

Conclusions

- A positive relationship was found between ultrasonic frequencies, sonication time and bacterial inactivation.
- When the results for the algae examination of sonicated water were compared to those for the raw water samples, it can be concluded that different ultrasonic frequencies at different time exposure cause clear morphological change in the algal cells without cell disruption.
- Ultrasonic frequencies at different time exposure no remarkable in chlorophyll "a" content was detected.
- Ultrasonic is a very effective and promising non-chemical oxidant to improve the efficiency of conventional coagulant "Alum" and decrease the optimal dose.

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