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Waste Water Disinfection During SARS Epidemic for Microbiological and Toxicological Control

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Objective To evaluate the disinfection of wastewater in China. **Methods** During the SARS epidemic occurred in Beijing, a study of different disinfection methods used in the main local wastewater plants including means of chlorine, chlorine dioxide, ozone, and ultraviolet was carried out in our laboratory. The residual coliform, bacteria and trihalomethanes, haloacetic acids were determined after disinfection. **Results** Chlorine had fairly better efficiency on microorganism inactivation than chlorine dioxide with the same dosage. Formation of THMs and HAAs does not exceed the drinking water standard. UV irradiation had good efficiency on microorganism inactivation and good future of application in China. Organic material and ammonia nitrogen was found to be significant on inactivation and DBPs formation. **Conclusion** Chlorine disinfection seems to be the best available technology for coliform and bacteria inactivation. And it is of fairly low toxicological hazard due to the transformation of monochloramine.

Key words: Wastewater disinfection; Chlorine; Chlorine dioxide; Ozone; Ultraviolet; Disinfection by-products

INTRODUCTION

For a long period, few wastewater plants in China have operated their disinfection facility in an appropriate way. Some plants' disinfection facilities have remained in a stage of blueprint designing[1]. The SARS outbreak between the end of 2002 and early 2003 prompted the government and environmental protection bodies to seriously consider the installation and operation of disinfection systems in wastewater treatment plants. On May 4, 2003, the State Environmental Protection Administration and the National Ministry of Construction issued a joint decree requiring all wastewater treatment plants to implement disinfection using chlorine, UV or ozone.

In order to give a guide for disinfection in Beijing wastewater plants during SARS epidemic, the research group carried out a study to investigate the disinfection efficiency with different disinfectants. It was focused on the relationship between disinfectant dosage and hygiene indices, between disinfectant dosage and disinfection by-product (DBPs) formation and the influence of other factors such as ammonia nitrogen (NH_3-N) and organic matters (COD). The optimal parameters for wastewater disinfection were determined on the basis of integrated results.

MATERIALS AND METHODS

Raw Water

During the SARS epidemic from April to June 2003, the Beijing Waste Water Group Company provided our laboratory with the final settle tank effluent of four waste water plants as raw water sample for research purpose. Among these plants, Gaobeidian Wastewater Plant was the largest plant in Beijing and much research work was focused on its effluent. Considering possible existence of SARS virus in the water, all the research work was carried out under strict protection. The water quality is shown in Table 1.

Disinfection Methods

The popular disinfection methods using chlorine, chlorine dioxide, UV and ozone were chosen in this study. Sodium hypochlorite solution (analytical reagent, A. R.) was diluted to 300 mg/L as free chlorine disinfectant. Chlorine dioxide was made by the reaction of sodium chlorite (A. R.) and hydrochloric acid (A. R.). The pure gas of chlorine dioxide was absorbed in pure water. The solution concentration was determined and diluted to 300 mg/L before disinfection. Ozone was produced by an ozonizer with air source. According to wastewater

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disinfection rule, the contact time of disinfectants was 30 min. Moreover, UV disinfection time was chosen as 8, 12, 20, and 40 s. The power of UV lamp was 5 mW/ (cm^2. s).

TABLE 1

Raw Water Quality

Raw Water	Date (in 2003)	pH	COD (mg/L)	NH_3-N (mg/L)
Gaobeidian	5.26	---	36.3	0.614
	5.30	---	45.1	3.03
	6.2	7.67	41.2	3.8
Beixiaohe	5.30	6.98	48.4	18.9
Qinghe	5.30	6.93	46.6	5.58
Fangzhuang	5.30	7.11	46.8	31.5

Waste Water Disinfection Indices and Standard

The discharge standard of pollutants for municipal wastewater treatment plant（GB18918-2002）requires that the fecal coliform count in discharge should be below 1000 CFU/L for reuse. However, during the SARS epidemic our colleague was restricted inside the campus and unable to buy the culture media or other reagents for fecal coliform count.

SARS coronavirus could exist for 2 days in the hospital wastewater, domestic wastewater and no-chlorine-contained drinking water[2]. In this paper, chlorine and chlorine dioxide were used to inactivate the SARS coronavirus, coliform 8099 and coliphage f_2 were added in the same domestic sewage. The concentrations of three microorganisms were 10^5 $TCID_{50}$/L, 10^6 CFU/L and 10^5 PFU/L. The results showed that the resistance of SARS virus to disinfectants in wastewater was weaker than that of coliform and coliphage f_2. All SARS viruses could be inactivated in 30 minutes at 20℃ with more than 0.5 mg/L residual free chlorine or 2.19 mg/L residual chlorine dioxide left.

According to the study above, coliform could be a good reference to SARS virus inactivation. In wastewater treatment practice, the ratio of total coliform count to fecal coliform count was about 5:1. So the microbiological criterion in our disinfection research was set as below 5000 CFU/L total coliform count. This criterion was more strict than the standards in most states of the USA (total coliform count≤1000 CFU/100 mL, fecal coliform count≤ 200 CFU/100 mL).

Total bacteria count was also introduced in this investigation as a means of quality control and also as a reference for microorganism inactivation.

DBPs are formed by the reaction of disinfectants with organic matters in the water. Chlorinated DBPs are the most important by-products since they have carcinogenic toxicity. Haloacetic acids (HAAs) and trihalomethanes (THMs) are the most important and popular kinds among all chlorinated by-products. Thus, these two DBPs were determined in this investigation as indices of toxicology.

One of the THMs, trichloromethane is included in the list of selective control index in discharge standard of GB18918-2002. Its maximum mean concentration per day is 300 μg/L. HAAs are believed to have a higher carcinogenic hazard, but they are not included in this standard because their analysis is more complex. Two kinds of HAAs are included in the sanitary standard for drinking water quality. The maximum concentration levels of dichloroacetic acid and trichloroacetic acid are 50 μg/L and 100 μg/L separately.

Analytical Methods

The analytical methods in this investigation are listed in Table 2.

TABLE 2

Analytical Indices and Methods

Indices	Methods	Note
Chlorine concentration[3-4] (mg/L)	DPD-ferrous Titrimetric Method	Free Chlorine, Monochl-oramine, Dichloramine And Dichloramien Could Be Determined Respectively
Chlorine Dioxide Concentration[5] (mg/L)	Five Step Iodometric Method	Chlorine Dioxide, Free Chlorine, Hypochlorite, Chlorite, Chlorate Could Be Determined Respectively
Ozone Concentration (mg/L)	Iodometric Method	
Total Bacteria Count[6] (CFU/mL)	Pour Plate Method	Incubation With Nutrient Agar at 37℃ for 24 h
Total Coliform Count[6] (CFU/L)	Membrane Method	Incubation With Endo Culture Medium at 37℃ for 24 h
Trihalomethane /THMs[4,6] (μg/L)	Head Space Gas Chromatography	Shimadzu GC-17A Gas Chromatographer With ECD Detector and HP-5 Capillary Column
Haloacetic Acids /HAAs[4,7](μg/L)	Chromatography With Micro-Extraction and Methylation	Shimadzu GC-17A Gas Chromatographer With ECD Detector and HP-5 Capillary Column

DATA AND RESULTS

The investigation was carried in two steps. In the first step, chlorine disinfection efficiency on four wastewater plants' effluent was evaluated in order to guide the practice against the latent SARS virus. In the second step, several popular disinfection processes were compared by their inactivation efficiency and DBPs formation in order to give recommendations for future reconstruction.

Chlorine Disinfections

Since chlorine disinfection was applied in these wastewater plants at that time, it was urgent to determine the chlorine dosage in response to the SARS outbreak, and to focus our major research efforts on chlorine disinfection. After chlorine addition, the configuration and concentration of residual chlorine in the water were affected by concentrations of ammonia nitrogen (NH_3-N) and organic matters, especially the former. Due to the fairly high NH_3-N concentration, the chlorine dosage did not exceed the breakpoint, and the residual chlorine usually existed in the form of monochloramine.

Coliform Inactivation

Actually, the disinfectant in wastewater was not free chlorine but monochloramine. The total coliform count concentration in all these effluents was 10^5-10^6 CFU/L, that was as high as that of usual effluent reported. The data are shown in Fig. 1.

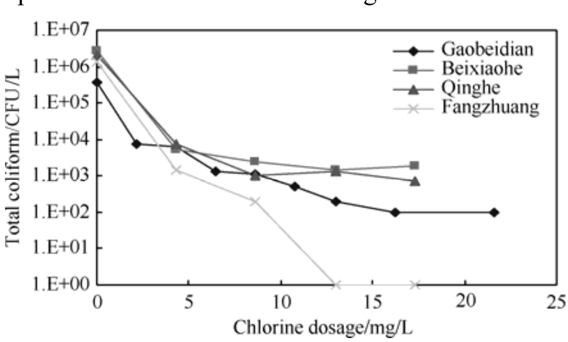

FIG. 1. Coliform inactivation curves by chlorine.

The inactivation curves could be divided into two periods. The first period was logarithmic inactivation when a large amount of microbes were killed quickly. With the addition of 2 mg/L chlorine, the total coliform count value decreased by 2-3 log. With the addition of 5 mg/L chlorine, the coliform concentration in all plants was less than 5000 CFU/L, that was set for safe criteria in this investigation. As the dosage increased, the coliform concentration decreased slowly and the curves turned to be flat, that was called platform or tailing off period. The platform phenomena existed in virtually all inactivation curves except for that of Fangzhuang Plant and the residual coliform was about 10^3 CFU/L. It was hard to inactivate the residual coliform further. Dosage more than 20 mg/L chlorine was needed for less than 100 CFU/L residual coliform concentration.

Platform period may be explained by the survival of a resistant sub-population as a result of protection by interfering substances (suspended matter in water), clumping, or genetically conferred resistance.

Microbe Inactivation

The shape of microbe inactivation curves was similar to that of coliform inactivation curves. A logarithmic period was followed by a platform period, shown in Fig. 2.

With the addition of 4 mg/L chlorine, the total bacteria count decreased by 2 log. As chlorine dosage increased, the curves turned to be plat. In the platform period, the residual microbes were between 10 to 100 CFU/mL.

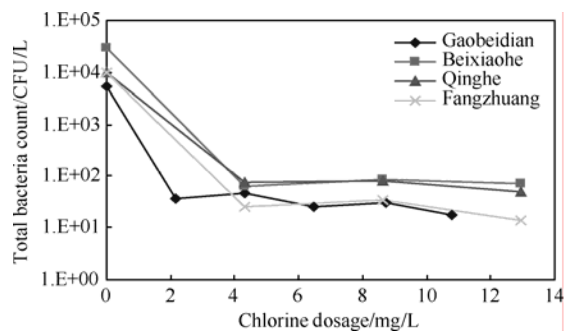

FIG. 2. Bacteria inactivation curves by chlorine.

Comparison of Disinfection Processes

For future reconstruction, a comparison of chlorine (Cl_2), chlorine dioxide (ClO_2), ozone (O_3) and ultraviolet irradiation (UV) disinfection processes was carried out with the effluent of Gaobeidian Wastewater Plant. Hygiene indices such as total coliform count and total bacteria count and toxicological indices such as trihalomethanes and haloacetic acids were used to assess different disinfection efficiencies.

Colifom Inactivation Comparison

The inactivation curves of Cl_2, ClO_2, O_3, and UV are shown in Fig. 3.

Chlorine and chlorine dioxide had virtually the same efficiency on coliform inactivation. With the addition of about 10 mg/L of Cl_2 and ClO_2, the coliform in waste water was killed by 3 log. With a fairly long irradiation time of 12 s and a high UV irradiation dose of 60 mW/(cm^2·s), the residual colifom was less than 5000 CFU/mL. Ozone has been reported as the most powerful disinfectant, however, its inactivation efficiency was poor. This phenomenon may be attributed to the consumption by organic matters for ozone's high oxidative potential.

Bacteria Inactivation

The result of bacteria inactivation was some what different from the result discussed above, as shown in Fig. 4. The shape of bacteria inactivation curves was similar to that of coliform inactivation. The efficiency of chlorine dioxide was lower than that of chlorine, which was opposite to former reports.

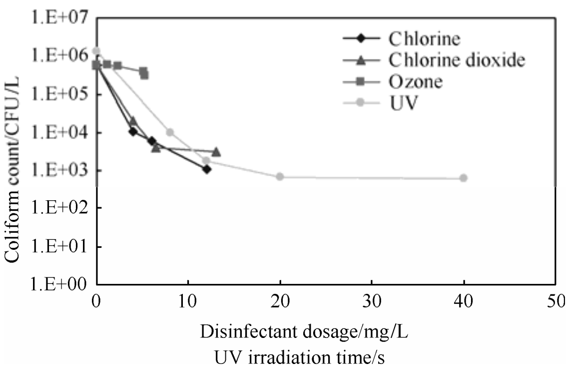

FIG. 3. Comparison of disinfection efficiency by coliform inactivation.

Note: ClO_2 dosage calculated as Cl_2; UV irradiation power: 5 mW/ (cm^2·s).

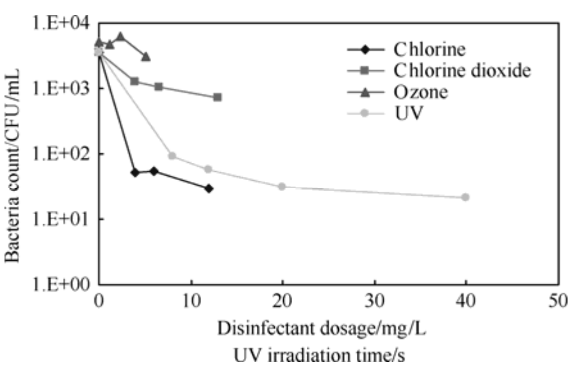

FIG. 4. Comparison of disinfection efficiency by bacteria inactivation.

Note: ClO_2 dosage was calculated as Cl_2; UV irradiation power was 5 mW/ (cm^2·s).

DBP Formation

Since ozone and UV disinfection did not form the halo-substituted disinfection by-products, the DBP formation was compared between chlorine and chlorine dioxide. As shown in Fig. 5, chlorine dioxide disinfection formed much less HAAs and THMs than chlorine.

HAAs formation was in direct proportion to chlorine dosage in the range of 3-7.5 mg/L. As Cl_2 dosage increased further, HAAs formation turned to be stable and the maximum HAAs concentration was 23.43 μg/L of dichloroacetic acid (DCAA) and 86.56 μg/L of trichloroacetic acid (TCAA). However, HAAs formation by ClO_2 was about 3 μg/L and did not increase with the dosage. HAAs formation by ClO_2 was 3.4%-15.0% of that by Cl_2.

THMs formation by both disinfectants did not increase greatly with dosage. The maximum THMs yields by Cl_2 were 28.28 μg/L of $CHCl_3$ and 0.86 μg/L of $CHBrCl_3$. THMs formation by ClO_2 was 27.0%-49.2% of that by Cl_2.

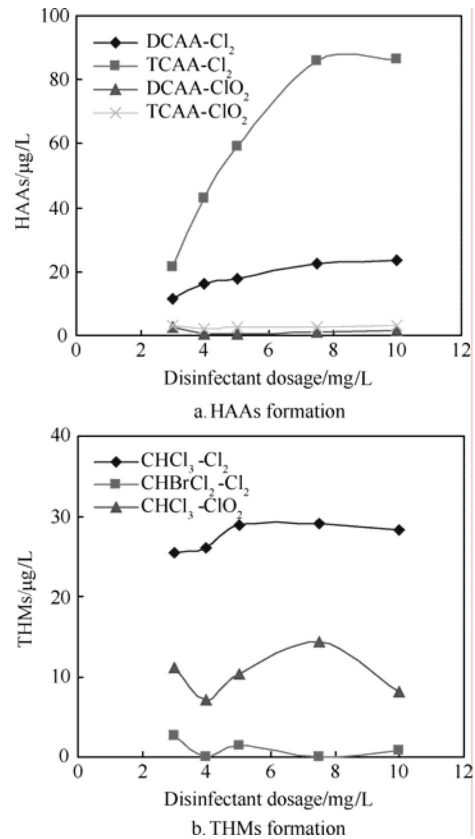

FIG. 5. Comparison of DBP formation with Cl_2 and ClO_2.

Note: ClO_2 dosage was calculated as Cl_2.

DISCUSSION

Chlorine Disinfection

The concentration and characteristics of residual chlorine were affected by the concentrations of ammonia and organic matters, especially the former. Since the ammonia nitrogen concentration was fairly high, the residual chlorine existed in the form of monochloramine according to the breakpoint-chlorination curve. The inactivation velocity of monochloramine was slower than that of free chlorine, but its thorough inactivation efficiency was high during a fairly long period.

All plants' curves of chlorine inactivation could be divided into two periods. The first period of lower than 5 mg/L chlorine addition could be regarded as a log inactivation period. It had about 99.5% inactivation efficiency and looked linear in log scale. The Ct product more than 150 min·mg/L could basically achieve the total coliform criterion of 5000 CFU/L. In the following platform period, the amount of residual coliform remained fairly stable and decreased slowly with the increase of chlorine concentration. However, the amount of residual coliform was still more than 100 CFU/L even if as high as 20 mg/L of chlorine was added. That is to say, advanced treatment was needed if the effluent was required to be lower than 3 CFU/L coliform, which is required in the recycling water quality standard for urban miscellaneous water consumption.

The inactivation curves of bacteria seemed alike to that of total coliform. The total bacteria count was about 10 times of total coliform count. Although the index of total bacteria count was not set in the standard, it could be helpful for wastewater disinfection.

Moreover, the phenomenon of platform phase in the curve of inactivation may be important in the practice of waste water disinfection since there was no obvious reduction of coliform amount although the chlorine dosage was increased greatly. The phenomenon could be explained by the survival of a resistant sub-population as a result of protection by interfering substances (suspended matter in water), clumping, or genetically conferred resistance.

Choice of Disinfection Process

According to our research, chlorine disinfection seems to be a good process for coliform and bacteria inactivation. Other research have also testified that chlorine disinfection has a better efficiency against SARS coronavirus than chlorine dioxide[2]. Moreover, chlorine disinfection is the cheapest and most popular technology in China.

Chlorine dioxide has almost the same efficiency on coliform inactivation as chlorine with the same dosage. However, its efficiency on bacteria inactivation is poorer than chlorine. Other research has proved that chlorine dioxide has a better inactivation efficiency than chlorine. Therefore, pilot test for disinfection process choice should be carried out before application.

UV irradiation has a good efficiency on coliform and bacteria inactivation. With a high dosage of 60 $mW/(cm^2 \cdot s)$, the residual total coliform was lower than 5000 CFU/L, suggesting that its application in China will increase greatly with the improvement of China's economy. Much work should be done to optimize the parameters of UV disinfection.

Ozone disinfection had a poor efficiency in this research, which might be attributed to the apparatus' poor absorbance in the water. Moreover, high concentration of organic matters in wastewater might greatly disturb ozone due to its high oxidative potential.

DBP Formation

Since the concentration of organic matters is high in the effluent of wastewater treatment plant, it seems that chlorine disinfection can form a high concentration of disinfection by-products. However, the concentrations of HAAs and THMs detected in 30-minute-chlorination even did not exceed the requirements in drinking water standard, which may be attributed to the existence of high concentration of ammonia nitrogen. Chlorine and ammonia nitrogen react quickly to form low-activity monochloramine, indicating that monochloramine can greatly decrease the formation of DBPs.

As far as other disinfection processes are concerned, DBPs formed by chlorine dioxide are 10% lower than those formed by chlorine. Ozone and UV do not form THMs and HAAs. These alternative disinfection processes will exhibit more and more advantages with the improvement of DBPs standards in the future.

CONCLUSION AND RECOMMENDATION

Chlorine inactivation curves could be divided into log inactivation period and platform period. The phenomenon of platform period may be explained by the survival of a resistant sub-population as a result of protection by interfering substances (suspended matters in water), clumping, or genetically conferred resistance. It may be important in the practice of wastewater disinfection. When total coliform count of 5000 CFU/L is set as the effluent criteria, 5 mg/L chlorine to be added is a fairly safe dosage.

Chlorine disinfection seems to be the best available technology for coliform and bacteria inactivation. Chlorine dioxide has almost the same efficiency on coliform inactivation as chlorine. However, its efficiency is lower on bacteria inactivation than chlorine. UV irradiation has a good efficiency on coliform and bacteria inactivation. However, ozone disinfection has a poor efficiency on organic matter's disturbance in wastewater.

As far as DBPs formation is concerned, disinfection of high concentration chlorine even did not form THMs and HAAs exceeding the drinking water standard. This may be explained by the existence of high concentration of ammonia nitrogen and its transformation of free chlorine to monochloramine. Chlorine dioxide forms less than 10% of THMs and HAAs than chlorine. Ozone and UV do not form THMs or HAAs. In general, all wastewater disinfection processes have a fairly low hazard of DBPs. The alternative disinfection processes will show advantages with the improvement of DBPs standards in the future.

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