Water Science & Technology

Water Science & Technology Vol 90 No 1, 363 doi: 10.2166/wst.2024.205

Relationship between chlorine decay and nanobubble application in secondary treated wastewater

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ABSTRACT

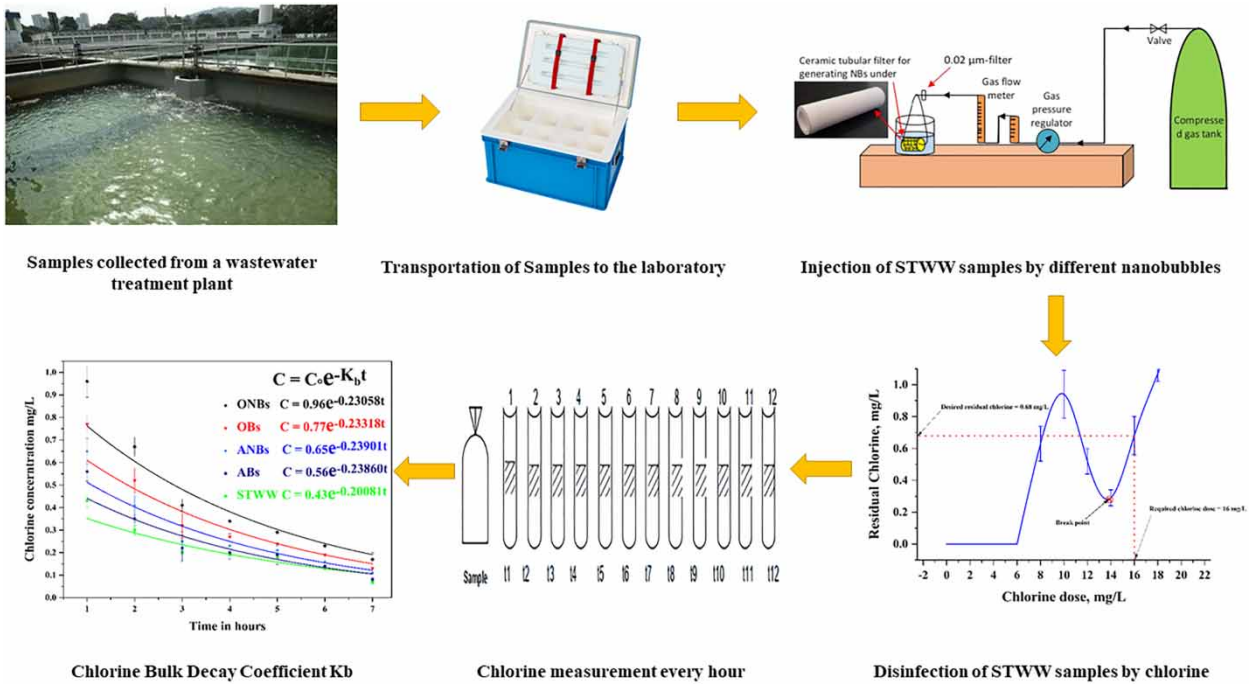
There has been numerous research on the uses of treated wastewater that needs chlorine disinfection, but none have looked at the impacts of injecting nanobubbles (NBs) on the decomposition of residual chlorine. Gas NB injection in treated wastewater improves its properties. The kinetics of disinfectant decay could be impacted by changes in treated wastewater properties. This paper studies the effect of various NB injections on the residual chlorine decay of secondary treated wastewater (STWW). It also outlines the empirical equations that were developed to represent these impacts. The results show that each type of NBs in treated wastewater had a distinct initial chlorine concentration. The outcomes demonstrated a clear impact on the decrease of the needed chlorine quantity and the reduction of chlorine decay rate when utilizing NB injection for the STWW. As a result, the residual chlorine will remain for a longer time and will resist any microbiological growth under the application of NBs on treated wastewater. Moreover, NBs in secondary treated effluent reduce chlorine usage, lowering wastewater disinfection costs.

Key words: chlorine decay kinetics, disinfection, nanobubble application, residual chlorine, secondary treated wastewater

HIGHLIGHTS

- The nanobubble (NB) injection in the secondary treated wastewater reduced the required chlorine dose.
- Different types of NBs have different bulk chlorine decay.
- Bulk chlorine decay for different NBs has been determined.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Alternative sources of water are required to ensure the protection of the environment and water resources, as well as to make socioeconomic activities sustainable due to the rise in water requirements for numerous applications and the shrinking water supply, which are exacerbated by the results of climate change (Abdella Ahmed *et al.* 2022; Fico *et al.* 2022). Reusing and recycling water is increasingly being viewed as a crucial approach to lessening water shortages and overcoming water contamination (Nixdorff *et al.* 2021). Reclaimed water can be restored from wastewater, which is considered an inexhaustible reused water resource, encouraging the conversion of wastewater treatment facilities into technologies for recovering water supplies in accordance with the circular economy idea. Reclaimed water is a term for the utilization of treated wastewater. It has been applied and controlled globally, mostly in China, Australia, European Union (EU), Spain, Portugal, Israel, and the USA (Truchado *et al.* 2021). The EU research on reusing water identified the ideal methods and minimum requirements for managing water for reused water quality (Costa *et al.* 2021). Reclaimed water is accompanied by a microbiological risk due to waterborne diseases. Especially, microorganisms contained in reclaimed water offer a significant risk to human health when they penetrate skin contact, ingestion, and inhalation without suitable treatment and management methods (Cao *et al.* 2021; Wallmann *et al.* 2021). Lately, specific rules for reclaimed water usage have been introduced by the EU for irrigation uses, bolstering the recommendations for irrigation with reclaimed wastewater, and encouraging the proper usage of reclaimed water despite numerous barriers, in order to reduce harm to the environment and public health. Worries were expressed regarding the specifications needed to provide adequate reclaimed water disinfection from wastewater treatment plants to the application point. Pathogens must be controlled, and reclaimed water must be disinfected to assure its safety. Ozone, chlorine, and ultraviolet (UV) radiation are some of the traditional ways used to disinfect water and treated wastewater. The disinfection process using chlorine is one of the most used. It has the advantages of being relatively affordable, simple to use, and effective at killing bacteria, with the added benefit of staying in the system for a very long time (Kinani *et al.* 2022). The physical properties of the network and system components, such as dead-end sections, tank shape, pipe substance, and age, all influence the rate at which chlorine degrades. Other water quality factors that influence chlorine degradation include temperature, initial chlorine content, organic matter, iron concentration, and frequency of rechlorination (Costa *et al.* 2021).

Even though there has been research on the issue of chlorinating water for drinking, further study is required to control the quality of reclaimed water effectively and run its supply networks (Costa *et al.* 2021). Previous research on the reduction of

residual chlorine in treated wastewater showed that, as compared to drinking water, the interaction between chlorine and treated wastewater demonstrated significant features due to the fact that treated wastewater contains more organic matter and chemicals than drinking water. Treated wastewater conducts electricity over 20 times better than drinking water and contains over twice as much organic content. Additionally, drinking water has lower levels of dissolved organic carbon, dissolved organic nitrogen, and assimilable organic carbon than reclaimed water does (Kang & Ahn 2022). The amount of free chlorine necessary for reclaimed water (chlorine demand) must be determined, taking into account two factors: (i) the efficiency of the wastewater treatment and (ii) the amount of residual chlorine needed to ensure the required microbial quality for the safe utilization of reclaimed water (Shekhawat *et al.* 2020).

Maintaining an acceptable residual disinfectant concentration to stop microbial reproduction is still challenging since reclaimed water contains larger concentrations of organic matter and ammonia than drinking water does. The optimization of chlorine dosages required to achieve reclaimed water disinfection needs, while reducing disinfection byproduct (DBP) production and encouraging safe water reusing, hence requires a greater understanding of the mechanisms of chlorine degradation in reclaimed water. Chlorine degrades more quickly in reclaimed water pipes than it does in pipes carrying drinking water, making it difficult to keep the residual chlorine content constant throughout the distribution system. In addition, there has not been as much research on the chlorine decay of reclaimed water as there has been on drinking water. Due to the complex and variable water quality of reclaimed water, traditional drinking water chlorine decay models are not relevant. Understanding the dynamics of chlorine deterioration and predicting the amount of chlorine in pipe systems are crucial. A proper model of chlorine degradation is therefore needed. Both the decay of pipe walls and bulk materials contribute to the total decomposition of chlorine in a pipe system. High concentrations of pollutants in bulk water have a bigger impact on taking in the remaining chlorine in treated wastewater (Wang *et al.* 2019).

Over the last 20 years, several wastewater treatment methods have been developed, and each one has an impact on the amount of chlorine that remains in the treated wastewater. One such method is the treatment of wastewater with nanobubbles (NBs). NBs are microscopic gas voids within liquids smaller than 1 μm in diameter (Fan *et al.* 2019). Compared to larger bubbles, they have special qualities. They have a large specific surface area, which increases the area of contact between liquid and gas (An *et al.* 2019). They are also known as surface NBs because they can remain stable on surfaces (Senthilkumar *et al.* 2018). Additionally, the large specific surface area of NBs promotes mass movement, chemical reactivity, and physical adsorption at the gas–liquid interface (Shi 2022). Their small buoyancy causes them to stay in the liquid for a longer period of time and rise to the surface very slowly (Kalogerakis *et al.* 2021). By shrinking the bubbles and increasing their internal pressure, they increase the effectiveness of the gas mass transfer (Azevedo *et al.* 2019). The shock waves generated by the NBs' burst led to the production of hydroxyl radicals (Farid *et al.* 2022). Hydrophobic surfaces have been shown to attract one another thanks to NBs (Ahmed *et al.* 2018). In addition, NBs possess a number of additional advantageous qualities, such as strong stability against coalescence, mechanical strength, and exceptional thermal stability (Phan *et al.* 2021; Soyluoglu *et al.* 2021; Wu *et al.* 2021; Levitsky *et al.* 2022). Nanotechnology has the ability to solve some of the biggest problems that current wastewater treatment systems are now facing (Ahmed *et al.* 2023). NB-based technologies for treating wastewater require less energy than more conventional techniques (Shen *et al.* 2022).

As a result, the primary focus of this work is on the bulk degradation of chlorine under applications of different types of NBs in treated wastewater. A unique method based on pushing compressed gases over a tubular ceramic membrane was used to create NBs. Many different types of cavitation, including particles, optical, hydrodynamic, and acoustic cavitation, are commonly used in the traditional methods of NB production. The rigidity, intricacy, and high energy consumption of these methods are just a few of its flaws. This strategy, however, is simple, efficient, and scalable (Ahmed *et al.* 2018). This returns to the ability of NBs — as a natural ecofriendly process – to enhance the treated wastewater characteristics, which may affect the bulk chlorine decay.

The relationship between chlorine degradation and NBs has never been studied in earlier research. The novelty of the study is to provide empirical formulas that relate the type of NBs, the time of injection, and the amount of initial chlorine to bulk chlorine decay. We can therefore use these formulas to forecast the rate of chlorine degradation at any given time in waste treatment plants that employ the NB approach as an advanced treatment technique.

2. MATERIALS AND METHODS

2.1. Sampling site

Undisinfected samples of secondary treated wastewater (STWW) were collected from the discharge of a secondary settling chamber at the Al Kawlah wastewater treatment plant (WWTP), located in Sohag, Egypt, in order to study the effects of

NB injection on residual chlorine decay in STWW. Domestic sewage influent was treated using conventional sedimentation and activated sludge processes, respectively. This WWTP satisfies the discharge requirements of the Egyptian code in order to reuse treated wastewater (ECP 501/2015). In accordance with the classification of the treatment level (ECP 501/2015), the grade of each tested sample was D. The undisinfected STWW samples were meticulously encased and immediately delivered in an ice cooler to the laboratory. When the sample arrived at the laboratory, it was kept at 4 °C in a refrigerator in the dark to eliminate the changes in its constituents.

2.2. Disinfection of the STWW by chlorine

The STWW sample is disinfected by adding chlorine in the form of sodium hypochlorite. The recommended procedure for calculating the amount of chlorine needed to achieve an acceptable concentration of residual chlorine after 30 min of contact with wastewater was performed in accordance with standard methods for examining water and wastewater (Rice & Bridgewater 2017). The required chlorine dose, as shown in Figure 1, is 16 mg/L depending on the results of the breakpoint chlorination test, which corresponds to a remaining chlorine dose of 0.68 mg/L and is considered to meet the requirements of the (ECP 501–2015).

2.3. Generation of NBs

Pressurized air and pure oxygen were directly injected through a ceramic membrane tube with 100 nm-sized pores and 0.1 wave aberration function (WAF) to create air NBs (ANBs) and oxygen NBs (ONBs) in STWW samples (Khaled Abdella Ahmed *et al.* 2018; Zhang *et al.* 2021). Air bubbles (ABs) and oxygen bubbles (OBs) were produced using the same pressurized gases by injecting them directly into the wastewater samples instead of using the ceramic membrane. The gases were released with the laboratory temperature set at 25 °C, at a pressure of 50 PSI (345 KPa), and a flow rate of 1 L/min.

2.4. Experimental design

The STWW was divided into four samples (1, 2, 3, and 4) of 3 L each. For 120 min, each sample was subjected to the generation of ONBs, ANBs, OBs, and ABs. For the purpose of comparing the results, a blank was prepared using the same STWW but without the injection of any bubbles or NBs.

After the injection period, the samples underwent a thorough agronomic and physico-chemical investigation to ascertain the effects of the four different bubbles on the features of the sample. Moreover, all four samples, as well as the blank sample, were chlorinated with the required chlorine dose. Considering that all of the used containers were cleaned to remove any chlorine demand before starting to chlorinate any samples. As part of the cleaning process, they were washed in deionized water, soaked in deionized water that had been super-chlorinated overnight, then deionized water was used for cleaning, after which it was allowed to dry. By keeping the samples (250 mL) incubated at set temperatures in dark bottles, it is possible to measure the rate of chlorine decay. One bottle was used for each measurement, with the

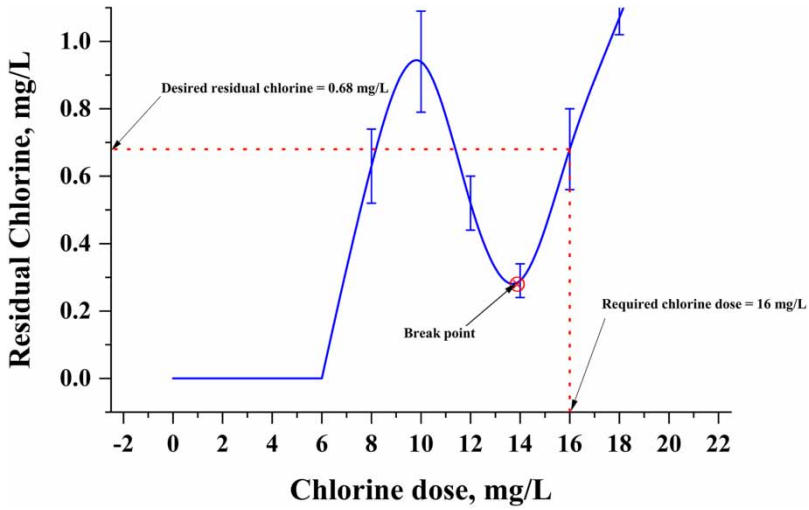


Figure 1 | Breakpoint chlorination.

number of bottles prepared depending on the length of the trial. The DPD powder (*N*,*N*-diethyl-p-phenylenediamine) colorimetric method was used to calculate the level of free chlorine present in each sample (4500-Cl G APHA standard method) (Rice & Bridgewater 2017). Duplicate measurements were taken for each spot sample, which helped to reduce error and served as an important quality assurance measure. After an hour, the initial residual chlorine concentration measurements for all samples were taken until the chlorine concentration value was nearly zero. All sample levels of chlorine were monitored regularly (every 60 min). A curve fitting program (the origin program) was used to process the measurement data, and an exponentially decreasing curve was created to determine the chlorine decay coefficient (K_b), which exclusively depicts the interaction of chlorine with the water body.

2.5. Laboratory analyses

The Standard Procedures for the Evaluation of Wastewater and Water were used to analyze each sample (Rice & Bridgewater 2017). For the recently analyzed samples, the pH, temperature, and dissolved oxygen concentration were measured using Orion Star A329 multiparameter meters. To assess the total dissolved and suspended solids (TDS and TSS) gravimetrically, 50 mL aliquots of each were dried at 180 and 103–105 °C, respectively. Whatman 42 filter paper was used to examine the soluble ions in the samples. The orthophosphate concentration was tested by UV–Vis spectroscopy with the stannous chloride method. The closed reflux method was used to assess the chemical oxygen demand (COD). The biological oxygen demand at 5 days (BOD_5) was calculated utilizing the 5-day BOD test. Iodometric analysis was used to calculate the amount of sulfur. Oil and grease were identified by the partition gravimetric method. The persulfate method was used in order to measure the total Kjeldahl nitrogen concentration. The macro-Kjeldahl method was used in order to measure the amount of organic nitrogen. The ammonia electrode method was used to calculate ammonia. Using the fluorogenic substrate approach, *Escherichia coli* was tested.

3. RESULTS AND DISCUSSION

3.1. Characteristics of the STWW samples after NB injection

The results are detailed in Table 1. The temperature and pH of the used blank sample, which had a pH of 8.2 and a temperature of 25 ± 0.3 °C, were raised by the continuous injection of ONBs and ANBs, which also significantly raised the concentrations of dissolved oxygen DO concentration and TSS. In addition, the NBs decreased sulfide, oil and grease, and orthophosphate, while also lowering COD and BOD_5. Total Kjeldahl nitrogen, organic nitrogen, and ammonia decreased as a result of the strong oxidizing action of ONBs but increased as a result of the high nitrogen content of ANBs. *E. coli* levels

Table 1 | The physico-chemical characteristics of STWW samples after NB injection

Parameter	Unit	Blank sample	AB samples	ANB samples	OB samples	ONB samples
Temperature	(°C)	25.2	25.4	27.8	25.5	28
pH	–	8.2	8.5	8.7	8.32	9.86
DO	(mg/L)	4.8	7.86	12.5	18.1	42
TSS	(mg/L)	14	12	10	8	6
TDS	(mg/L)	630	625	630	625	630
Orthophosphates	(mg/L)	0.52	0.49	0.44	0.47	0.42
COD	(mg/L)	78	68	57	46	29
BOD_5	(mg/L)	23	18	13	10	6
Sulfide	(mg/L)	0.3	0.28	0.26	0.24	0.21
Oil and grease	(mg/L)	9	9	8	7	6
Total Kjeldahl nitrogen	(mg/L)	9.4	14	16.2	8.1	7.8
Organic nitrogen	(mg/L)	4.2	4.6	4.8	3.7	3.4
Ammonia	(mg/L)	5.2	10.2	11.3	4.6	4.4
E. coli	(MPN/100 mL)	45,000	10,400	7,900	4,600	2,400

dropped by 94.66% as a result of the ONB injection, indicating excellent disinfection. Additionally, the Zetasizer Nano ZS apparatus and dynamic light scattering were used to analyze the bubble size distribution and zeta potential (ZP). The results have been previously indicated by Khaled Abdella Ahmed *et al.* (2018).

3.2. Finding the order of reactions

For every sample, the sequence of the kinetic reaction between chlorine and STWW was calculated using the findings of the residual chlorine monitoring. Three curves were graphed for each sample as follows: the first curve shows the residual chlorine content (C) as a function of time. In the second curve, Ln(C) was plotted versus time. The third curve was a time-versus-1/(C) plot. The data typically follow a straight line if the selected curve is accurate. The first, second, or third curves, respectively, stand in for zero, first, or second order. R^2 was calculated for each curve to determine the accuracy of the data representation (García-Ávila *et al.* 2020). Following the above-mentioned methodology, the kinetic reaction order is established for each sample. As an illustration, the adjustment coefficient R^2 for the STWW sample is 0.88404 for a zero-order reaction (Figure 2(a)), 0.92228 for a reaction of the first order (Figure 2(d)), and 0.77823 for a reaction of the second order (Figure 2(g)). A first order is therefore approved. The exact same process was performed on all samples, and the results demonstrated the existence of a first-order response in the context of the present study as shown in Figure 2 and Figure 3, which supports the

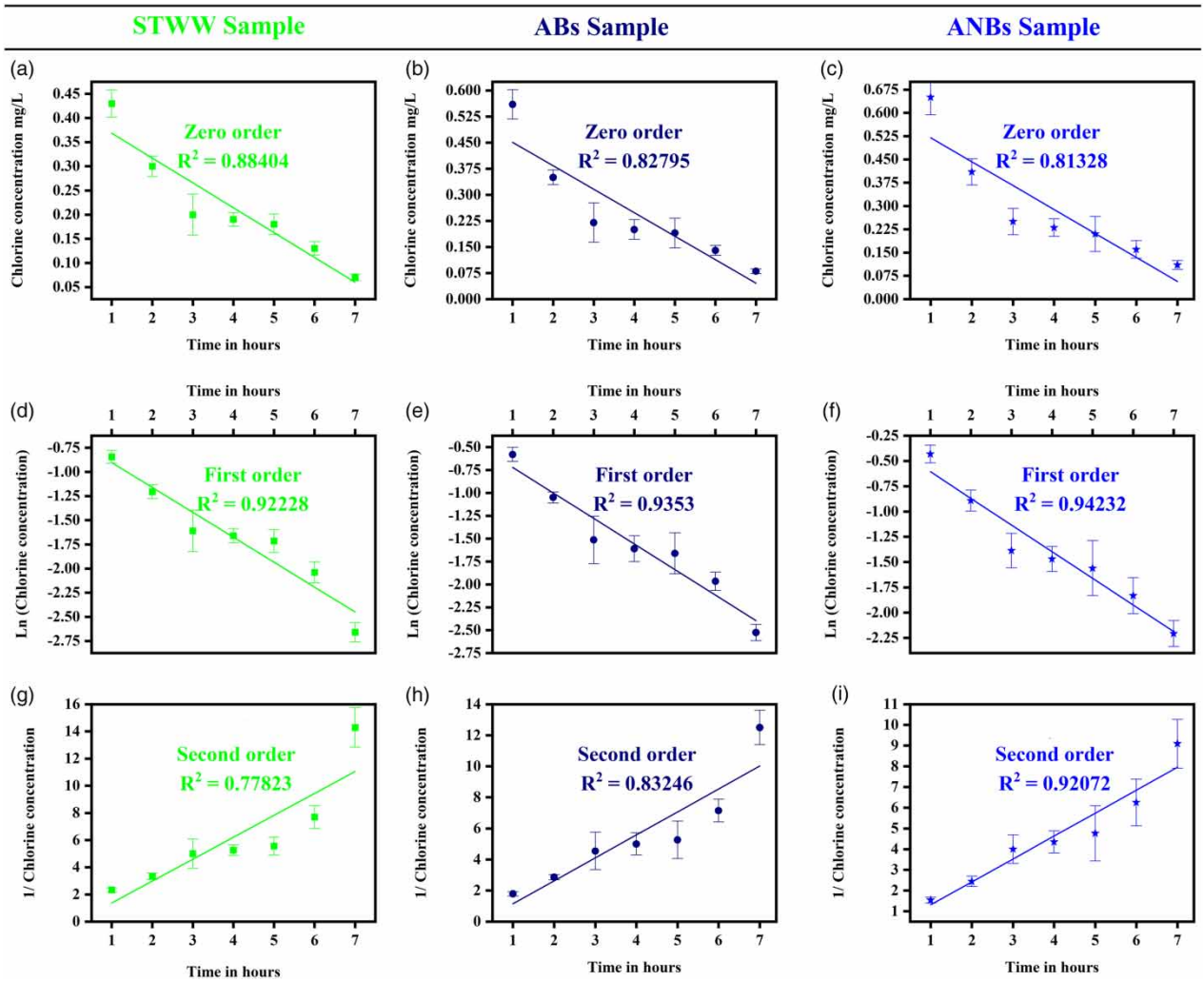


Figure 2 | Reaction order of chlorine decay for STWW, AB, and ANB samples. (a–c) Charts of chlorine content versus time. (d–f) Semi-log of chlorine content versus time. (g–i) Chlorine inverse content versus time.

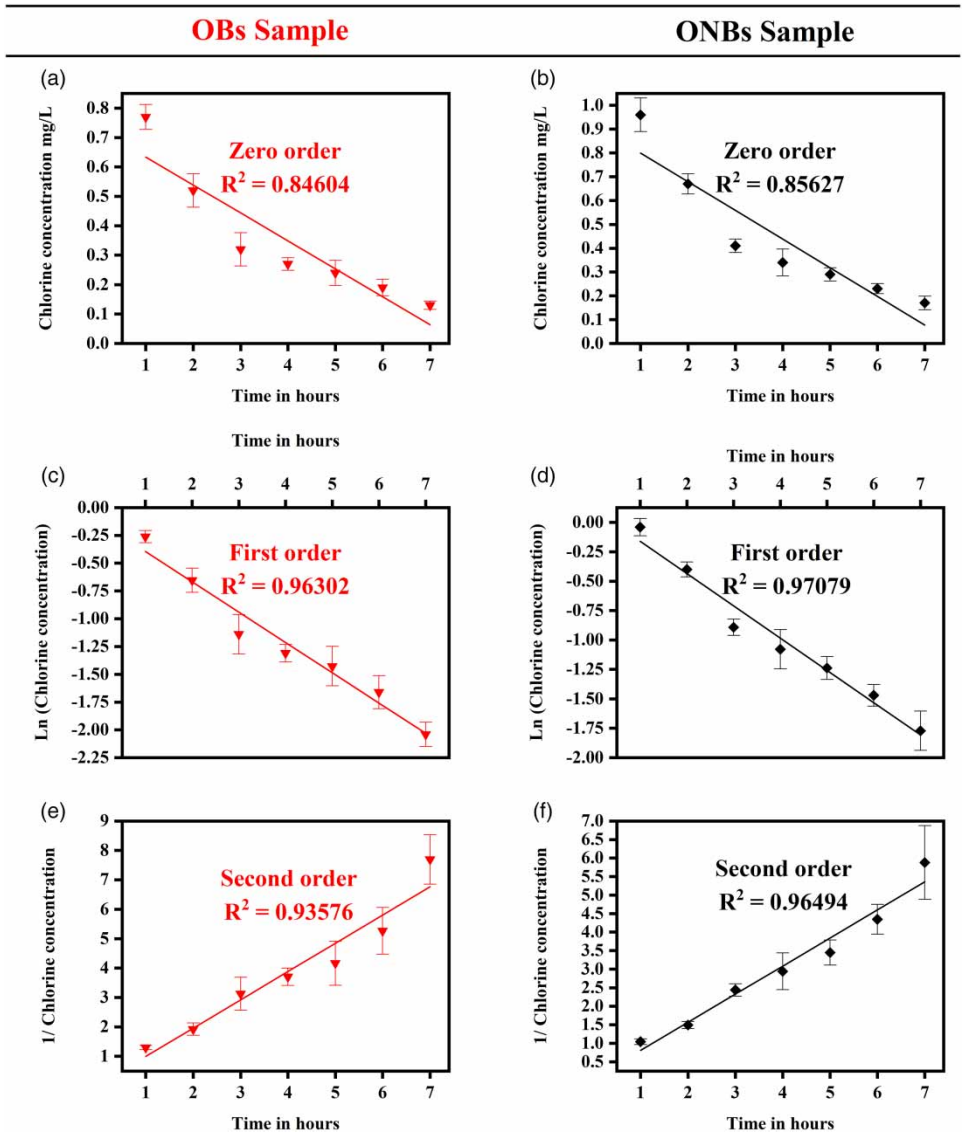


Figure 3 | Reaction order of chlorine decay for OB and ONB samples. (a) and (b) Chlorine content versus time. (c) and (d) Semi-log chlorine content versus time. (e) and (f) Chlorine inverse content versus time.

dynamics of the chlorine interaction with STWW for all samples.

$$C = C_0 \ e^{-K_b t}$$

where K_b is the chlorine decay coefficient, expressed in 1/h, and C is the quantity of chlorine at time t. C_0 is the initial chlorine content.

3.3. Calculation of the bulk decay coefficient K_b

Following confirmation that the order of reaction was first order for all samples, the chlorine measurements were displayed on a time-based graphic. The reaction coefficient K_b for every specimen was then obtained after an exponential adjustment, as depicted in Figure 4. According to the equations shown in Figure 4, the bulk decay coefficient (K_b) is equivalent to 0.20081 h^{-1} for the STWW sample, 0.23860 h^{-1} for ABs, 0.23901 h^{-1} for ANBs, 0.23318 h^{-1} for OBs, and 0.23058 h^{-1} for ONBs. The equation's negative sign indicates that the amount of chlorine has gradually decreased over time. Although

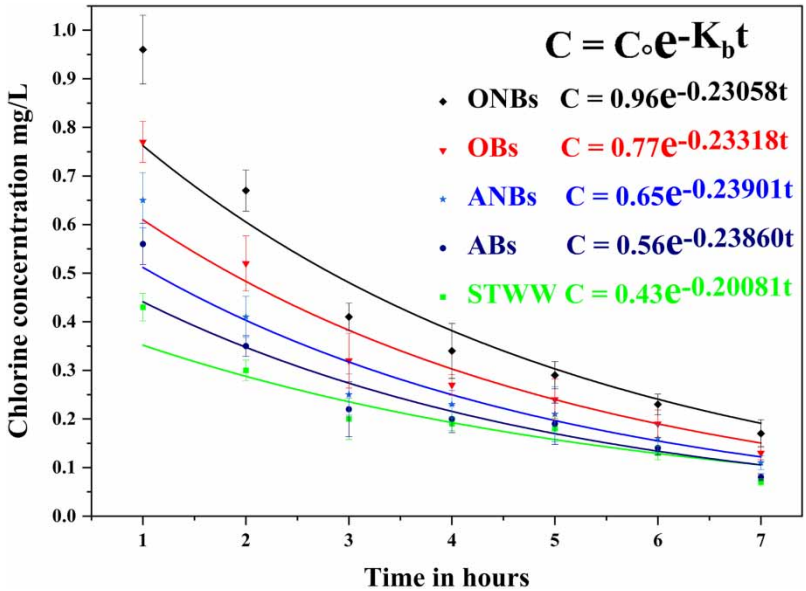


Figure 4 | Chlorine bulk decay coefficient K_b for STWW, injected STWW with ABs, ANBs, OBs, or ONBs.

all samples were chlorinated simultaneously with the same initial chlorine dose of 16 mg/L, this figure makes it obvious that the chlorine concentration after 1 h varies for each sample. After an hour, STWW, AB, ANB, OB, and ONB samples had chlorine concentrations of 0.43, 0.56, 0.65, 0.77, and 0.96 mg/L, respectively. The ONB sample has less organic content due to its higher oxidation content than other samples, raising the DO of the samples from 4.8 to 42 mg/L, which causes this variation because the presence of organic materials affects how chlorine reacts with STWW. Additionally, the variations in the K_b values for the five samples were attributed to the variations in organic matter in each sample, which led to the variations in the initial residual chlorine concentration, which was taken into consideration an hour after chlorination. The main reason why the organic matter decreased is that the reactive oxygen species, such as hydroxyl radicals, were generated thanks to the NB collapse in the treated wastewater, and that is why the NBs had better results in prolonging the chlorine compared to traditional coarse bubbles. These reasons are thought to be the primary determinants of K_b values. Although it is anticipated that the K_b for the ONB sample will be lower than that of the STWW sample, the figure shows the opposite, which is explained by the varied initial residual chlorine concentration for the two samples.

Overall, the application of NBs in wastewater treatment can lead to an increase in the lifetime of residual chlorine, lowering the required chlorine dose. This could have economic and environmental effects. In other words, reduced chlorine doses can result in cost-savings, increased operational efficiency, and lower operating costs. Environmentally, this can prevent the creation of DBPs caused by chlorine reacting with organic materials, which can be hazardous to both human health and the environment.

4. CONCLUSION

The research examined and discussed the effect of different types of NB injections in STWW leading to the following key conclusions:

- The quantity of chlorine necessary for the chlorination process is reduced when NBs are injected into secondary treated effluent.
- The first-order model equation accurately describes chlorine degradation in all of the investigated STWWs under different NB injections.
- The residual chlorine decays more quickly in the bare STWW samples than in those that had received different NB injections.
- The rate of residual chlorine degradation is significantly influenced by the initial residual chlorine concentration.
- The degradation rates of residual chlorine were inversely correlated to the initial residual chlorine concentration.

• The variation in the residual chlorine decay constants is due to differences in organic matter in each sample.

NBs contributed to the purification process of STWW, as evidenced by the reduced need for chlorine in disinfection.

Therefore, it is suggested that NB technology can be employed more successfully for advanced wastewater treatment, especially when employing chlorination as the disinfection method. Based on this paper, additional study is required to determine the relationship between the chlorine decay of STWW and the injection of NBs, especially when it comes to the timing of the injection, the method used to create the NBs, their diameter, ZP, types, and other characteristics.

AUTHORS' CONTRIBUTIONS

A.K.A.A. conducted data curation, formal analysis, methodology, investigation, resource management, validation, and visualization, and wrote the original draft. They also reviewed and edited the manuscript. M.S. contributed to the inquiry, conceptualization, formal analysis, methodology, project administration, resource management, validation, and visualization, and wrote a draft. They also reviewed and edited the manuscript. O.N. and T.A.-W. were involved in conceptualising, drafting, reviewing, and editing the manuscript.

HUMAN ETHICS

The manuscript does not report studies involving human participants, human data, or human tissue.

CONSENT FOR PUBLICATION

All authors have agreed to the publication of the paper.

DATA AVAILABILITY STATEMENT

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

CONFLICT OF INTEREST

The authors declare there is no conflict.

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First received 22 April 2024; accepted in revised form 31 May 2024. Available online 14 June 2024