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Two-step chlorination: A new approach to disinfection of a primary sewage effluent

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ABSTRACT

Sewage disinfection aims at inactivating pathogenic microorganisms and preventing the transmission of waterborne diseases. Chlorination is extensively applied for disinfecting sewage effluents. The objective of achieving a disinfection goal and reducing disinfectant consumption and operational costs remains a challenge in sewage treatment. In this study, we have demonstrated that, for the same chlorine dosage, a two-step addition of chlorine (two-step chlorination) was significantly more efficient in disinfecting a primary sewage effluent than a one-step addition of chlorine (one-step chlorination), and shown how the two-step chlorination was optimized with respect to time interval and dosage ratio. Two-step chlorination of the sewage effluent attained its highest disinfection efficiency at a time interval of 19 s and a dosage ratio of 5:1. Compared to one-step chlorination, two-step chlorination enhanced the disinfection efficiency by up to 0.81- or even 1.02-log for two different chlorine doses and contact times. An empirical relationship involving disinfection efficiency, time interval and dosage ratio was obtained by best fitting. Mechanisms (including a higher overall Ct value, an intensive synergistic effect, and a shorter recovery time) were proposed for the higher disinfection efficiency of two-step chlorination in the sewage effluent disinfection. Annual chlorine consumption costs in one-step and two-step chlorination of the primary sewage effluent were estimated. Compared to one-step chlorination, two-step chlorination reduced the cost by up to 16.7%.

1. Introduction

Sewage disinfection aims at providing protection for humans from exposure to pathogenic waterborne microorganisms (Jacangelo and Trussell, 2002). Chlorine, which indicates free chlorine in this study, is extensively used as a disinfectant in municipal sewage treatment plants due to its effectiveness, low cost and ease of application (Lee and von Gunten, 2010; Drinan and Spellman, 2012; Shannon et al., 2008). In addition to inactivating pathogenic microorganisms, chlorine reacts with various reducing inorganic ions (e.g., sulfide and ferrous), ammonia and effluent organic matter present in sewage effluents. Chloramines are generated from the reaction of chlorine with ammonia. At low Cl_2:N mass ratios (<5:1), the dominating species of chloramines is monochloramine (White, 2011).

Various factors can influence disinfection efficiency, including disinfectant type, disinfectant concentration, contact time, reaction condition, and microorganism species. Generally, disinfection efficiency can be expressed as: $-\log(Nt/No) = k\int_0^T Cdt$, where k is the inactivation rate constant of a disinfectant, C is disinfectant concentration at time t, and Nt/No is the ratio of remaining microbial concentration at time t to the initial microbial concentration (Haas and Karra, 1984; Crittenden et al., 2012). The disinfectant concentration and contact time can be quantified together as the Ct value, which is used to determine the compliance with water regulations for inactivation of microorganisms (U.S. EPA, 1991). Fecal coliform bacteria live in the intestinal tracts of humans and other warm-blooded animals. They are the most common microbial contaminants in natural waters and wastewaters. *Escherichia coli*, a main species in the fecal coliform group,

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is commonly used as an indicator organism to assess the effectiveness of water disinfection (Hu et al., 2005; Quek et al., 2006; Janjaroen et al., 2013).

For a particular disinfection unit in a sewage treatment plant, increasing the chlorine dose may increase the disinfection efficiency, but this will consume more chlorine and increase the operational cost, as well as cause the formation of more halogenated disinfection byproducts (DBPs) in the chlorinated sewage effluent (Rebhun et al., 1997; Fabbricino and Korshin, 2005; Krasner et al., 2009; Sun et al., 2009; Sedlak and von Gunten, 2011; Tang et al., 2012; Ding et al., 2013; Gong and Zhang, 2013, 2015; Richardson and Ternes, 2014; Gong et al., 2016; Cai et al., 2016) and possible adverse effects to the ecosystem of the receiving water body (Watson et al., 2012; Yang and Zhang, 2013, 2014; Liu and Zhang, 2014). Thus, it is of great importance to develop and optimize the sewage disinfection process with eco-friendly and cost-effective features.

Recently, Verma et al. (2013) reported that, for the same chlorine dose, a two-step addition of chlorine (two-step chlorination) showed a greater disinfection efficiency than a one-step addition of chlorine (one-step chlorination). The result indicated that two-step chlorination had the potential to reduce the chlorine dose for the same level of *E. coli* removal. However, this study of step-wise chlorination was still at a preliminary stage, as only one condition was reported (i.e., a chlorine dose of 2.5 + 2.5 mg/L as Cl_2 was applied with a 5-min time interval, for a total contact time of 20 min) and only one effluent sample from a secondary sewage treatment plant was collected and tested. Two other studies (Graham and Hong, 2001; HKDSD, 2015a) also reported similar tests with preliminary results. However, it has remained unclear whether two-step chlorination can also perform better for a primary sewage effluent than one-step chlorination and whether the better performance of two-step chlorination was reproducible. Additionally, it has remained unclear what the mechanisms are for the potentially higher disinfection efficiency of two-step chlorination. Such mechanisms should be of great scientific value.

Accordingly, this study aimed to compare the disinfection efficiencies of two-step chlorination and the commonly used one-step chlorination of a primary sewage effluent. Furthermore, the two-step chlorination process of the primary sewage effluent was optimized in terms of time interval (the time period between the first and second steps of chlorination) and dosage ratio (the mass ratio of chlorine dosed at the first step to the second step). The mechanisms for the observed greater disinfection efficiency of two-step chlorination were also explored.

2. Materials and methods

2.1. Chemicals and reagents

All the chemical solutions were prepared from chemicals of reagent grade or higher. Ultrapure water (18.2 MΩ•cm) was provided by a Cascada I Laboratory Water Purification System (PALL). A NaOCl stock solution was prepared by diluting a reagent grade sodium hypochlorite solution (Allied Signal). A preformed monochloramine stock solution was freshly prepared by mixing of the NaOCl stock solution and an ammonium chloride solution at a chlorine to ammonium mole ratio of 0.8:1. Both the NaOCl stock solution and the preformed monochloramine stock solution were standardized by the DPD colorimetric method (APHA et al., 2012). Thioacetamide and sodium thiosulfate were purchased from Sigma–Aldrich.

2.2. Collection and characterization of effluent samples from a primary sewage treatment plant

A total of 19 undisinfected effluent samples were collected from a primary sewage treatment plant over a 9-month period from May 7, 2015 to February 15, 2016, where each sample was collected on a given day. The sewage treatment plant has a treatment capacity of 2.0×10^6 m^3/d and is one of the largest primary sewage treatment plants in the world. Each collected sample was transferred immediately to the laboratory in an ice cooler to minimize changes in its constituents. A major portion of the sample was used immediately for conducting disinfection, and the remaining sample was used for characterization. Dissolved organic carbon (DOC) and ammonia were measured with a total organic carbon analyzer (Shimadzu, Japan) and a flow injection analysis system (8500 Series, Lachat, USA), respectively. Chemical oxygen demand (COD) and total suspended solids (TSS) were obtained from the Hong Kong Drainage Services Department (HKDSD, 2015b, 2016), and they were determined following the Standard Methods (APHA et al., 2012). The ultraviolet (UV) absorbance was measured at 254 nm with a 1-cm quartz cuvette by using a spectrophotometer (Lambda 25, Perkin Elmer, USA).

2.3. Disinfection of the primary sewage effluent

Initially, a preliminary test was conducted to choose appropriate disinfection conditions which were capable of meeting the sewage effluent discharge standard. Aliquots of a sewage effluent sample were chlorinated by dosing 1.0–6.0 mg/L NaOCl as Cl_2. After a 15- or 30-min contact time, each aliquot was dechlorinated with 105% of the requisite stoichiometric amount of 0.10 M $Na_2S_2O_3$. Two representative chlorination scenarios (i.e., dosing 4.0 mg/L of NaOCl as Cl_2 for a 30-min contact time and dosing 6.0 mg/L of NaOCl as Cl_2 for a 15-min contact time) for one-step chlorination were selected for further comparison as they could fulfill the disinfection goal (Fig. S1 and Supplementary Information (SI)).

To compare the disinfection efficiencies of two-step chlorination and one-step chlorination, aliquots of an undisinfected primary sewage effluent sample were chlorinated. For one-step chlorination, 100-mL aliquots of the sewage effluent sample were dosed with 4.0 mg/L of NaOCl as Cl_2 for a 30-min contact time; for two-step chlorination, 100-mL aliquots of the sewage effluent sample were chlorinated with the same total chlorine dose (4.0 mg/L as Cl_2), but the dose was split into two portions at different ratios (first step: second step = 1:1, 3:1, 5:1 or 8:1). The two portions of each ratio were dosed at different time intervals (first step at 0 s, and second step at 5, 10, 19, 38, 75, 150, 300 or 600 s). It should be mentioned that different dosage ratios and time intervals were examined to study their effects on the disinfection efficiency. After disinfection with thorough mixing for a total 30-min contact time, the chlorine residual in each aliquot was dechlorinated with 105% of the requisite stoichiometric amount of 0.10 M $Na_2S_2O_3$. To further compare the disinfection efficiencies of two-step chlorination and one-step chlorination, the experiments above were repeated except that a total chlorine dose of 6.0 mg/L as Cl_2 and a total contact time of 15 min were used.

To study the mechanism of two-step chlorination, the disinfection efficiencies of one-step chlorination and one-step chloramination of the primary sewage effluent were compared. Aliquots of an undisinfected primary sewage effluent sample were disinfected by dosing 4.0 mg/L chlorine or monochloramine as Cl_2. After disinfection with thorough mixing for a 30-min contact time, the chlorine residual in each aliquot was dechlorinated with 105% of the requisite stoichiometric amount of 0.10 M $Na_2S_2O_3$.