



Factors affecting the formation of iodo-trihalomethanes during oxidation with chlorine dioxide



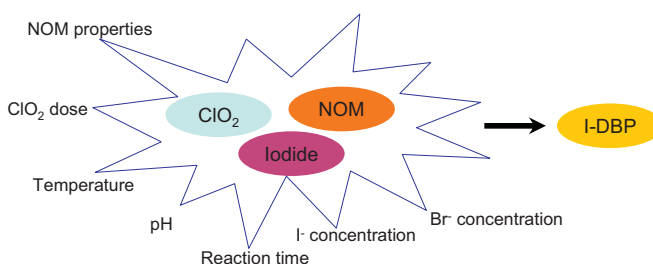
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HIGHLIGHTS

- Factors affecting I-THM formation during oxidation using ClO_2 were evaluated.
- CHI_3 yields correlated moderately with SUVA values ($R^2 = 0.79$).
- CHI_3 yields were the highest at I^-/ClO_2 molar ratio of 1–2.
- CHI_3 was predominant even in the presence of high concentrations of bromide.
- CHI_3 yields were the highest at pH 8.

GRAPHICAL ABSTRACT



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ABSTRACT

Effects of water characteristics, reaction time, temperature, bromide and iodide ion concentrations, oxidant doses, and pH on formation of iodinated trihalomethanes (I-THM) during oxidation of iodide-containing water with chlorine dioxide (ClO_2) were investigated. Among the water samples collected from ten water sources, iodoform (CHI_3) was the predominant I-THM and trace amount of chlorodi-iodomethane (CHClI_2) was occasionally found. CHI_3 yields correlated moderately with specific UV absorbance (SUVA) ($R^2 = 0.79$), indicating that hydrophobic aromatic content were important precursors. Longer reaction time led to continued formation of CHI_3 . I-THM containing bromide was also found in waters containing both bromide and iodide, but CHI_3 was dominant. The formation of CHI_3 was higher at 25°C than 5°C and 35°C . CHI_3 formation showed an increase followed by a decrease trend with increasing ClO_2 doses and iodide concentrations and the highest yields occurred at iodide to ClO_2 molar ratios of 1–2. pH 8 resulted in the highest CHI_3 formation. It should be noted that a high iodide concentration was spiked to waters before adding ClO_2 and the results may not reflect the formation yields of iodinated THMs in real conditions, but they provide information about formation trend of I-THM during oxidation of ClO_2 .

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1. Introduction

Disinfection by-products (DBPs) in water systems are of great concern due to their potential adverse health effects [1]. Alternative disinfectants, such as chlorine dioxide (ClO_2), are becoming popular due to the reduced formation of many DBPs compared with

chlorination [2–4]. Meanwhile, its biocidal efficiency is equal or superior to chlorine over a wide pH range [5–8]. ClO_2 can be also applied for color removal, algal removal, and taste and odor control [9–11].

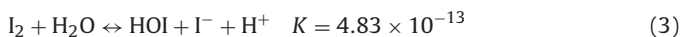
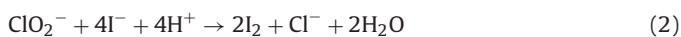
In iodide-containing waters, ClO_2 can react with iodide to form hypiodous acid (HOI), which reacts with natural organic matter (NOM) to form iodinated DBPs, including iodo-trihalomethanes (I-THMs), as the following equations show [12–15].



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Table 1
Water characteristics.

	DOC, mg/L	UV ₂₅₄ , cm ⁻¹	SUVA, L mg ⁻¹ m ⁻¹	Br ⁻ , mg/L	NO ₂ ⁻ -N, mg/L	NO ₃ ⁻ -N, mg/L	NH ₄ -N, mg/L	Total, N mg/L	DON, mg/L
SRNOM	3.0	0.12	4.0	<0.01	<0.02	<0.02	<0.02	0.06	0.06
S1	5.4	0.13	2.4	0.03	0.21	0.94	0.09	1.76	0.52
S2	2.0	0.05	2.6	0.08	0.16	1.56	<0.02	1.77	0.03
S3	2.7	0.08	3.0	0.03	0.12	1.24	0.24	2.29	0.69
S4	1.7	0.04	2.3	0.09	0.35	1.61	0.02	2.06	0.08
S5	1.6	0.04	2.6	<0.01	0.05	1.31	<0.02	2.04	0.66
S6	1.1	0.05	4.5	0.12	0.24	1.61	0.06	2.18	0.26
G1	1.3	0.02	1.6	0.43	0.49	2.60	<0.02	4.81	1.70
W1	7.7	0.11	1.4	<0.01	0.76	4.90	0.40	16.48	10.51
W2	7.8	0.04	0.5	0.09	0.69	2.80	0.13	–	–



I-THMs include iodoform (CHI₃), chlorodiiodomethane (CHCl₂I), bromodiiodomethane (CHBr₂I), dichloriodomethane (CHCl₂I), dibromiodomethane (CHBr₂I), and bromochloriodomethane (CHBrClI). I-THMs are more cytotoxic and/or genotoxic in mammalian cell assays than the brominated and chlorinated analogs [15,16] and also cause taste and odor problems [16–18]. The organoleptic threshold concentration of iodoform (CHI₃) lies between 0.03 and 1 μg/L, which is the lowest value of all of the I-THMs [15].

Many factors affect the formation of DBPs, including reaction time, pH, temperature, disinfectant concentration, bromide ion concentrations and precursor properties [19,20]. Some results on I-THM formation using ClO₂ are available in the literature [21]. CHCl₂I and CHI₃ were detected during oxidation of iodide-containing waters using ClO₂ [22]. Maximum concentrations of CHI₃ occurred at pH 8 [20,23]. I-DBP formation increased then leveled off with increasing ClO₂ doses. CHI₃ formation was enhanced with increases in iodide concentrations, but was suppressed at higher iodide concentrations [23]. In the presence of bromide, the results about I-DBP were inconsistent from the literature. Jones et al. [22] reported the equal amount of yields of CHCl₂I and CHI₃ detected during oxidation of iodide-containing waters using ClO₂. However, Hua and Reckhow [3] found the presence of three I-THM (CHCl₂I, CHClI₂ and CHI₃) and CHI₃ was the dominant specie. Trace amount of iodate was also observed. Ye et al. [23] reported that the formation of CHI₃ was not affected by low levels of bromide, however, mixed bromo-iodo-species were not measured in this study. Water properties also affected I-DBP formation. The formation of CHI₃ had three times difference from the oxidation of two raw waters with differed DOC concentrations under the same oxidation conditions [23]. However, how water properties other than DOC affected I-DBP formation remained unknown. Meanwhile, the effect of reaction time and temperature on I-DBP formation was also not found from the literature.

Therefore, the first objective of the study is to investigate the effects of water characteristics on the formation of I-THM during oxidation with ClO₂ in the presence of iodide. Seven surface waters, one groundwater and two wastewater effluents were used. The second objective is to investigate the effects of reaction time, temperature, pH, ClO₂ doses, iodide ion concentrations, bromide ion concentrations on I-THM formation. A synthetic water prepared from Suwannee River reverse osmosis (RO) NOM isolate (purchased from the International Humic Substances Society (IHSS)) and a real water collected from the source water of a drinking water treatment plant were used.

2. Materials and methods

2.1. Reagents

Individual I-THM standards including CHCl₂I, CHClI₂, CHClBrI, CHBrI₂, CHBr₂I, CHI₃ were purchased from Orchid Cellmark (Westminster, BC, Canada). Potassium iodide (KI) was obtained from Sigma. A solution of ClO₂ was prepared from gaseous ClO₂ by slowly adding dilute H₂SO₄ to a sodium chlorite (NaClO₂) solution according to the method described in the Standard Method [24]. The concentrations were determined using diethyl-p-phenylene diamine (DPD) ferrous titration [24].

2.2. Sample collection

A NOM stock solution was prepared by dissolving an aliquot of Suwannee River RO NOM isolate (Cat. No. 1R101N, International Humic Substances Society) into ultrapure water and filtering it with 0.45-μm membrane, hereby referred to as SRNOM. Seven water samples, including 6 surface water (S1 to S6) and 1 ground water (G1) were collected and they were source water for six water treatment plants. The other two samples were secondary effluents from two wastewater treatment plants (W1 and W2).

After collection, samples were shipped to the laboratory and kept at 4 °C until use. All the samples were filtered through 0.45-μm membranes to remove particles. The concentrations of dissolved organic carbon (DOC), total nitrogen, ammonia, nitrite, nitrate and bromide, and the UV absorbance at 254 nm (UV₂₅₄) were measured. Specific UV absorbance (SUVA) was calculated as UV₂₅₄ divided by DOC. The water characteristics were listed in Table 1. The iodide concentrations in the samples were not analyzed. But the background levels should be much lower than the spiked iodide concentrations in the test (in the range of 30–300 μM).

2.3. Experimental procedures

Experiments were carried out in amber glass vials with Teflon-faced septa. All samples were buffered to pH 7.0 with 5 mM phosphate buffer. A desired amount of ClO₂ was applied to the samples and the samples were stored headspace-free in the dark at room temperature (22 ± 1 °C). After appropriate reaction time, samples were quenched with ascorbic acid and extracted immediately with methyl tert-butyl ether (MTBE). The extracts were then subjected to DBP analysis by gas chromatography. All of the DBP formation tests were run in duplicate.

A baseline ClO₂ dose of 100 μM was applied to SRNOM and S1 solutions (3 mg/L DOC) buffered and reacted in the dark for 24 h. An orthogonal matrix experimental design was employed where reaction time, pH, ClO₂ doses, temperature, or iodide ion concentration was varied one parameter at a time from the baseline condition: ClO₂ dosages (10, 30, 50, 100, 200 and 400 μM, corresponding to 0.68, 2.03, 3.38, 6.8, 13.5 and 27 mg/L), pH values (5, 6, 7, 8, and

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