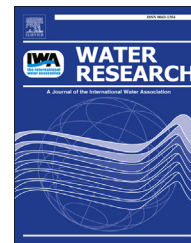


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To add or not to add: The use of quenching agents for the analysis of disinfection by-products in water samples



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ABSTRACT

The formation of disinfection by-products (DBPs) is a public health concern due to their potential adverse health effects. Robust and sensitive methods for the analysis of DBPs, as well as appropriate sample handling procedures, are essential to obtain accurate, precise and reliable data on DBP occurrence and formation. In particular, the use of an appropriate quenching agent is critical to prevent further formation of DBPs during the holding time between sample collection and analysis. Despite reports of decomposition of DBPs caused by some quenching agents, particularly sulphite and thiosulphate, a survey of the literature shows that they are still the most commonly used quenching agents in analysis of DBPs. This study investigated the effects of five quenching agents (sodium sulphite, sodium arsenite, sodium borohydride, ascorbic acid, and ammonium chloride) on the stability of seven different classes of DBPs commonly found in drinking waters, in order to determine the most appropriate quenching agent for the different classes of DBPs. All of the quenching agents tested did not adversely affect the concentrations of trihalomethanes (THMs) and haloacetic acids (HAAs), and thus are suitable for quenching of disinfectant residual prior to analysis of these DBPs. Ascorbic acid was found to be suitable for the analysis of haloacetonitriles (HANs) and haloacetones (HKs), but should not be used for the analysis of chlorite. Sodium arsenite, sodium borohydride, and ascorbic acid were all acceptable for the analysis of haloacetaldehydes (HALs). All of the quenching agents tested adversely affected the concentration of chloropicrin. A 'universal' quenching agent, suitable for all groups of DBPs studied, was not identified. However, based on the results of this study, we recommend the use of ascorbic acid for quenching of samples to be analysed for organic DBPs (i.e. THMs, HAAs, HANs, HKs, and HALs) and sodium sulphite for analysis of inorganic DBPs. Our study is the first comprehensive study on the effects of quenching agents on the stability of DBPs involving a wide range of DBP classes and quenching agents.

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1. Introduction

In drinking water treatment, disinfectants react with organic and inorganic materials in the source waters, producing disinfection by-products (DBPs). Since the first discovery of DBPs in the early 1970s, over 600 species of DBPs have been identified (Richardson, 2011). The most commonly found and abundant organic DBPs in drinking waters are the trihalomethanes (THMs) and the haloacetic acids (HAAs); less abundant DBPs include the haloacetonitriles (HANs), halo ketones (HKs), haloacetaldehydes (HALs), and the oxyhalide anions (i.e. bromate, chlorite, and chlorate) (Richardson, 2011; Weinberg et al., 2002). Some species of DBPs are known to be toxic, carcinogenic, and/or mutagenic, and have been associated with adverse human health effects such as bladder and colon cancers (Richardson et al., 2007). Due to the potential adverse health effects associated with DBPs, the concentrations of some DBPs in drinking waters are regulated in some countries. For example, the United States Environmental Protection Agency (US EPA) regulates the total concentration of the four species of THMs at $100 \mu\text{g L}^{-1}$, the total concentration of five species of HAAs (HAA5) at $80 \mu\text{g L}^{-1}$, and the concentration of bromate at $10 \mu\text{g L}^{-1}$ (Richardson, 2011).

Studies on the formation and occurrence of DBPs in waters have been carried out to provide further understanding on the mechanisms of DBP formation and the fate of DBPs in drinking water distribution systems, to ultimately inform management and control strategies for DBPs in water treatment. In these studies, accurate, precise and reliable measurements of DBPs are critical. Therefore, robust and sensitive methods for the analysis of DBPs, as well as appropriate sample handling procedures, are essential. In particular, the use of an appropriate quenching agent is critical to ensure quality in the analytical process. For accurate determination of DBP concentrations, the reactions between the disinfectant and the organic and inorganic materials in the sample must be terminated, to prevent the additional formation of DBPs during the holding time between sample collection and analysis. This is achieved by the addition of a quenching agent, generally a reducing agent (reductant) that reacts with the disinfectant (an oxidant) in a redox reaction, making the disinfectant unavailable for further reactions with organic and inorganic materials in the water sample. The quenching agent is added in excess to water samples, typically at 120% of the stoichiometric ratio of the reaction with the disinfectant (Reckhow and Singer, 1990).

The properties of an ideal quenching agent include: rapid and complete reaction with residual disinfectants, chemically inert towards the DBPs or other analytes of interest, negligible interfering effects in the analysis of the DBPs or other analytes of interest, and non-detectable signal for the compound or its reaction products (Urbansky, 1999). Some quenching agents that are commonly used prior to analysis of DBPs include aqueous solutions of sodium sulphite (Na_2SO_3), sodium thiosulfate ($\text{Na}_2\text{S}_2\text{O}_3$), ascorbic acid, ammonium chloride (NH_4Cl), sodium arsenite (NaAsO_3), and sodium metaarsenite (NaAsO_2). Sodium borohydride can also be used, however, its use is less common. Except for ammonium chloride, these quenching agents react with disinfectants in a redox reaction.

Ammonium chloride is a quenching agent specific for chlorine. It reacts with hypochlorous acid (HOCl) or hypochlorite anion (OCl^-) to form chloramine (Equation (1)), which theoretically does not form the regulated DBPs, the THMs and HAAs (Malliarou et al., 2005; Munch and Hautman, 1995):



In several studies, different quenching agents have been used for different groups of DBPs because some quenching agents have been found to cause decomposition of some DBPs (e.g. Hua and Reckhow, 2007; Diehl et al., 2000). In standard methods for the analysis of total organic halogen (TOX) (Standard Method 5320B) and THMs (Standard Method 5710 B), sodium sulphite is used as the quenching agent, while ammonium chloride is used in the US EPA standard method for the analysis of HAAs (EPA Method 552.2) (Clesceri et al., 1998). Trehy and Bieber (1981) found that dihaloacetonitriles can readily undergo dehalogenation reactions with sodium sulphite. Bauman and Stenstrom (1989) showed that significant reduction in TOX levels could result from dechlorination with aqueous sodium sulphite solutions. Croué and Reckhow (1989) reported that chloropicrin, trichloroacetonitrile, and dibromoacetonitrile can be rapidly destroyed in chlorinated drinking waters with small doses of aqueous sodium sulphite solutions, which means that sodium sulphite should not be used as a quenching agent when analysing for these compounds. Munch and Hautman (1995) stated that sodium sulphite promotes the decomposition of HANs, 1,1-dichloropropanone, 1,1,1-trichloropropanone, and chloropicrin, and suggested the use of ammonium chloride as an alternative quenching agent. Ascorbic acid has been reported to be a favourable alternative to sodium sulphite and sodium thiosulfate as a quenching agent for analysis of DBPs (Serrano and Gallego, 2007; Urbansky, 1999). Reports have indicated that ascorbic acid does not cause decomposition of DBPs, except for chloral hydrate, which is also degraded by other quenching agents (Urbansky, 1999).

Despite reports of decomposition of DBPs caused by some quenching agents, particularly sodium sulphite and thiosulfate (Munch and Hautman, 1995; Bauman and Stenstrom, 1989; Croué and Reckhow, 1989; Trehy and Bieber, 1981), a survey of the literature shows that sodium sulphite and sodium thiosulfate are the most common quenching agents used in the analysis of DBPs, especially for analysis of TOX and THMs. Various studies have used different quenching agents for the same group of DBPs, but there is little indication as to why a particular quenching agent was selected for a particular group of DBPs. There are also contradictions in the reported effects of quenching agents on the stability of DBPs. For example, Urbansky (1999) reported that sodium sulphite caused the decomposition of chloral hydrate, yet EPA Method 551.1 recommends the use of sodium sulphite as the quenching agent for samples to be analysed for chloral hydrate (Munch and Hautman, 1995). The inconsistencies in the choice of quenching agent and the contradictions that have been observed in published reports signify the need for a comprehensive study on the effects of quenching agents on the stability of DBPs, in order to determine the most suitable quenching agents for the different classes of DBPs.

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