Disinfection Byproducts

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Disinfection byproducts (DBPs) are formed by the action of oxidant-disinfectant chemicals, mainly chlorine derivatives (chlorine, hypochlorous acid, chloramines, etc.), that react with natural organic matter (NOM), mainly humic substances. DBPs are usually refractory to oxidation, mainly due to the presence of halogen compounds so that advanced oxidation processes (AOPs) are a recommended option to deal with their removal.

catalytic ozonation | photocatalytic ozonation | disinfection by-products | natural organic matter | humic acids | drinking water | chlorination

1. Introduction

Safety of drinking water has always been a concern for humankind from at least 500 BC when natural materials were used to purify water [1]. However, it was not until the end of the Middle Age when significant steps in the treatment of water were noticed. In fact, the discovery of the microscope at the end of the 16th century was, years later in 1850, the milestone to know the reason of pandemic problems associated to the use of water. With the aid of microscopes the presence of pathogens in drinking water was discovered and the use of disinfectants would become soon later. Specifically, in 1854 chlorine was first used in London to remove bad odors coming from sewers. Although at that time the disinfectant power of chlorine was not yet well known, the role of contaminated water to spread pandemic illnesses had already been confirmed with the cholera epidemic of London [2]. In 1879 chlorine was first used as a disinfectant also for sewage and in 1903 in drinking water treatment plants [3].

Since then, chlorine has been widely used for some other operations in drinking water treatment such as taste and odor removal, keeping safe water distribution systems, biofouling control, and color removal, to cite a few. For more than 70 years, chlorine was used with great success as a water disinfectant. However, water chlorination began to be questioned after the US National Organics Reconnaissance Survey [4] that revealed the presence of halogenated compounds in 80 USA drinking water plants in 1975. This survey was based on previous studies of Rook [5] and Bellar et al. [6] in 1974 sponsored by the United States Environmental Protection Agency (USEPA) that initiated a work on the analysis of contaminants in rivers such as the Mississippi River at New Orleans [7]. In these works, six main halogenated compounds were identified: Four trihalomethanes ($CHCl_3$, $CHCl_2Br$, $CHClBr_2$, and $CHBr_3$), CCl_4 and 1,2-dichloroethane [8]. Soon after, many more halogenated compounds, both of volatile and non-volatile nature, were detected in chlorine disinfected water and municipal wastewater secondary effluents. These compounds since then called disinfection byproducts (DBPs), were not previously present in the untreated waters.

2. Issues Related to DBPs Toxicity

As it was reported in the 1970s the reason for applying alternative disinfectants to chlorine was the potential toxic character of THMs and other halogenated organics found in finished chlorinated drinking water [9]. Specifically, the US National Cancer Institute in 1976 published that chloroform was carcinogenic in rodents [10]. Soon after, epidemiological studies suggested some relation between chlorinated drinking water and the occurrence of bladder, colon, and rectal cancer [11][12]. Since then, many studies on the evaluation of safety and hazard of DBPs have been reported [13]. According to DeMarini [14], at present, 20 out of 22 DBPs are rodent carcinogens, more than 100 genotoxic and 1000 water samples have been found to be mutagenic. It has been shown that brominated DBP are more carcinogenic than the chlorinated ones [15] and genotoxicity and cytotoxicity decrease in the following order for halogenated DBPs: Iodinated > brominated > chlorinated [16]. Generally, every DBP evaluated is genotoxic [14]. Comparing chlorinated and ozonated DBPs, the former are more genotoxic than the latter, at least, with *Salmonella* [17]. Regarding the way of DBPs exposure, some works [18] have reported higher cancer risk with the inhalation/dermal way than with oral intake. From inhalation or dermal way, volatile DBPs go directly to the blood stream, bypassing the liver, and once in the bladder they can be activated by some mutagen. From oral intake, DBPs go first to the liver where they could be detoxified. This is particularly important in swimming pool water and bath spa water where, in addition, many nitrogen containing DBPs (N-DBPs) have been found likely due to urine present in these waters. N-DBPs like nitrosamines have been found even more cytotoxic and genotoxic than their corresponding halogenated organics [19].

3. Concluding Remarks and Future Challenges

DBPs, specifically THMs, were discovered more than 40 years ago but, still, there is much research on their nature, since many recent papers deal with the founding of new DBPs. Already in 1980 about 400 DBPs were identified in chlorinated fulvic acids. At present, only a few DBPs are regulated with a maximum level concentration (MCL): Total THMs, total haloacetic acids (HAAs), bromate and chlorite. Nonregulated DBPs form different family groups: Halogenated compounds, ketones, aldehydes, and nitrosamines to quote the most representatives with the first ones as the most abundant formed in drinking water treatment (DWT). The halogenated compounds, (mainly chlorinated but also brominated and iodinated compounds) constituted by far the main family of DBPs from water treatment plants (WTP). Today, many of these compounds have been classified as rodent carcinogens, genotoxic, and mutagenic. This reveals the importance in the development of new analytical methods to quantify also nonregulated DBPs or surrogate parameters to fulfil future regulation in order to produce healthy drinking water.

Both DBPs precursors and DBPs have been treated with different oxidation processes where advanced oxidation processes, AOPs, are the most representative. Because of the double way of ozone reactions in water, ozone has attracted the interest of many researchers. Natural organic matter (NOM) and bromide ion are the main DBPs precursors treated with AOPs. In the last year, about 40 and 20 works were published on this matter with AOPs and ozone processes, as oxidants, respectively. On the whole, precursors react fast with ozone due to the presence of aromatic rings with hydroxyl substituents groups in humic and fulvic acid macromolecules. However, regarding DPBs removal classical ozone AOPs, such as O_3/UVC or O_3/H_2O_2, have been applied in a few cases to mainly remove a few HAAs (dichloroacetic and trichloracetic acids) and some N-nitrosamines. On the contrary, much more work has been done with ozone free processes such as Fenton, UVC/H_2O_2, and photocatalytic oxidation during the last 25 years. In these works, HAAs are the main DBPs studied. In general, these processes due to the formation of hydroxyl radicals reduce DBP concentrations but high oxidant doses are needed.

Most of the works dealing with catalytic and photocatalytic ozonation are lab-scale studies about catalyst properties, catalytic activity and optimization of operating conditions but much less work has been done to go in deep in the stability and reutilization of the catalysts in long term experiments. These studies are crucial to take a step forward to pilot scale studies with the best catalytic systems mainly for DBP formation potential removal. In addition, the use of natural radiation or environmentally friendly light emitting diodes (LEDs) should be prioritized in photocatalytic ozonation studies towards sustainable processes. However, comprehensive economic and environmental assessments are also required to balance the benefits of precursors or DBPs removal from drinking water in order to draw the best strategy from economic, environmental and health priorities.

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