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Foresight Look on the Disinfection By-Products Formation

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Abstract

In the water treatment industry, if there is a process that has attracted polemic discussion in terms of pros and cons disinfection has attracted the main part for its disinfection by-products (DBPs) formation. This work focuses on DBPs precursors, link among disinfection and DBPs, DBPs elimination, and study futures. During the last half-century, chlorination has been shown highly toxic to human health. Indeed, as a classical disinfectant, chlorine generates a bigger number of halogenated by-products than other disinfectants. Unfortunately, novel disinfection techniques and emerging pollutants in water can form fresh DBPs. DBPs surfacing lately are frequently with low levels and elevated poisoning. Further, as the oxidizing agent of the disinfectant increases, the formation of conventional DBPs is reduced, but more toxic DBPs emerge. Membrane processes, such as ultrafiltration and nanofiltration, depicted greater performance in eliminating organic matter if paralleled with traditional techniques. As a perspective, research should concentrate on physical processes such as distillation and/or solar disinfection, and filtration for better water treatment instead of injecting chemicals into water highly previously chemically polluted.

Subject Areas

Chemical Engineering & Technology

Keywords

Disinfection, Disinfection By-Products (DBPs), Organic Matter (OM), Chlorination, Ozonation, Water Treatment

1. Introduction

In the potable water treatment industry, disinfection has a key contribution to demobilizing pathogens in water [1] [2] [3] [4]. It considerably decreases the diffusion of various waterborne infectious diseases comprising typhoid and cholera, participating importantly in the safeguard of human health [5] [6] [7] [8]. Usual disinfection techniques comprise chlorination (chlorine and chloramines) [9] [10] [11], chlorine dioxide (ClO_2) [12] [13], ozonation [14], electrochemical advanced oxidation processes (AOPs) [15] [16] [17] [18] [19], and ultraviolet (UV) disinfection [20] [21] [22] [23]. More novel disinfection processes have also been suggested [24]-[31]. Nevertheless, the disinfection application is frequently joined by the generation of disinfection by-products (DBPs) which can induce additional public health troubles [32] [33] [34] [35].

DBPs were first proposed in 1974 [36] [37]. During water treatment, it was proved that chlorination can produce greatly toxic trihalomethanes (THMs) like chloroform [1] [12]. Further, high THMs concentrations in potable water were related to harmful reproductive outcomes [38]. Chlorination could generate additional poisonous DBPs like haloacetic acids (HAAs). Later, a bigger number of DBPs have been identified. Following chlorination or chloramination, haloamides, haloacetonitriles (HANs), and aldehydes also have been identified in drinking water [39] [40] [41]. In addition to chlorination, additional disinfection techniques also generate DBPs. The ozone disinfection method could form brominated organic and inorganic compounds (bromate, iodate, and chlorate) [42]. Among them, bromate is a suspected human carcinogen [43]. Brominated organic compounds like dibromoacetonitrile (DBAN) can also be produced throughout ozonation in the occurrence of high bromide levels [44], which could generate subchronic toxicity in rats [1]. UV disinfection could form nitrite if the water being treated holds nitrate [45].

This work focuses on the recent findings in DBPs precursors, link among disinfection and DBPs, DBPs elimination techniques, and study futures.

2. DBPs Precursors

DBPs precursors comprise both organic and inorganic matters and possess a fundamental contribution in generating such hazardous chemicals [1] [46] [47].

The main attention on precursors concentrated on natural organic matter (NOM), which is the primary organic precursor [1] [48] [49]. NOM was described as the complex matrix of organic material existing in natural water and possesses an evident impact on the disinfection process [50] [51] [52] [53]. It comprises humic substances [54] [55] [56] [57] and non-humic substances [58] [59] [60] [61] [62]. Humic substances are a crucial DBPs precursor and constitute an additional research interest [63]. The high molecular weight (MW) hydrophobic NOM fraction was more reactive with chlorine, while the low MW hydrophilic NOM fraction was more reactive to bromine and iodine [1] [63]. Fractionation of NOM depicted that the hydrophilic acid portion was the most

reactive precursor for THMs, but least reactive for HAAs [1]. Further, ClO_2 reacted with the humic fraction of NOM usually in the aromatic part of the molecule, and the carbonyls concentration considerably augmented with the reaction time between ClO_2 and carbonyl precursors [1]. The chlorination of NOM led to increasing in assimilable organic carbon (AOC) production via the oxidation and chlorine substitution on aromatic molecules [10] [12]. AOC was useful to microbial growth, inducing a hazard to the biological safety of potable water [1] [49]. Moreover, NOM can generate high levels of poisonous aromatic DBPs which were afterward transformed into aliphatic DBPs during chlorination/chloramination [1] [50] [52]. Traditional water treatment reduced most of the hydrophobic NOM with high MWs [63]. However, the low MW hydrophilic NOM was hard to remove, dominating residual organics [63]. Thus, it is fundamental to follow the reduction of the hydrophilic NOM part with low MW. AOPs [15] [31] [64] [65] [66] combined with biofiltration or biological activated carbon (BAC) [67] [68] [69] remains a more techno-economically practical choice to mineralize NOM [70].

Algogenic organic matter (AOM) [68] [71]-[76], a significant autochthonous organic derivative of algae, augmented the hazard of DBPs generation [77]. AOM was rich in nitrogen and protein; while, NOM was abundant in aromatic content [1]. Algae augmented dissolved (DON) in water and conducted to the augmentation of the possibility to form nitrogenous DBPs (N-DBPs) and other total organic halides (TOXs) [1]. In water, AOM could induce DBPs generation, taking up 20% - 50% of the DBP formation potential (DBPFP) in usual treatment circumstances [1]. The origin of AOM was divided into extracellular organic matter (EOM) intracellular organic matter and (IOM). EOM and IOM of algae are known to participate in the production of DBPs. EOM and IOM were mostly classified in low-MW (<1 kDa) and high-MW (>100 kDa) portions, possessing a significant contribution in the generation of carbonaceous DBPs (C-DBPs) and N-DBPs in both chlorination and chloramination [1]. EOM and IOM conducted to the bigger production of N-DBPs and haloaldehydes than NOM throughout chlorination, while the quantity of N-DBPs and C-DBPs produced from chloramination of EOM and IOM was much less than that from NOM. Juxtaposed with EOM, IOM had a bigger portion of total organic nitrogen, larger proportions of higher MW compounds, more hydrophobic contents, as well as higher fractions of free amino acids but lower fractions of aliphatic amines [1].

Soluble microbial products (SMPs) are one more type of precursors that could give rise to more dichloroacetic acid (DCAA) and N-DBPs. Further, they possess a bigger DBPFP than NOM [1]. In an investigation on *Chlorella* sp., it was proved that with elevating algae cultivation time in wastewater, the collection of SMPs increased the production of DBPs and the trend of DBP generation was as follows: chloroform > DCAA > trichloroacetic acid (TA) [1]. The majority of identified N-DBP precursors tended to be of low MW and low electrostatic

charge relative to most NOM. Consequently, it was not simple to eliminate them by traditional water treatment methods like coagulation [78] [79] [80] [81], while biodegradation and nanofiltration (NF) stay excellent options [82]-[87]. On the other hand, there were more contaminants from wastewater which may become DBPs precursors [7] [8] [88] [89] [90] [91] [92]. As an illustration, tertiary amine-containing pharmaceuticals or other quaternary amine-containing constituents of personal care products could work as N-nitrosodimethylamine (NDMA) precursors. A group of pharmaceuticals and personal care products (PPCPs) containing amine groups served as nitrosamine precursors during chloramine disinfection [93]. Moreover, phenols in raw water could lead to the generation of comparatively dangerous phenolic DBPs [57]. Biophysical and chemical processes also conduct to the formation of DBP precursors, leading to more DBPs generation in the reclaimed water [94] [95] [96] [97] [98].

The inorganic precursor bromide also magnetized awareness, as it conducted to the production of mixed bromochloro- and brominated DBP species throughout chlorination and chloramination [1]. Such chemicals are more carcinogenic and cytotoxic than their chlorinated counterparts [99] [100]. Bromide could be transformed into bromate throughout ozonation [100]. The occurrence of bromide also had an impact on the generation of iodo-DBPs (I-DBPs). As the reaction rate of HOI to IO_3^- influenced the production of I-DBPs, pre-oxidation procedures with powerful oxidants like ozone and ferrate were employed for controlling I-DBPs [101].

3. Link among Disinfection and DBPs

As a classical disinfectant, chlorine generates a bigger number of halogenated by-products than other disinfectants [44]. THMs, HAAs, and halonitromethanes (HNMs) are mostly formed throughout the chlorination. Chlorine injection could also produce nitrosamines, HANs, aldehydes and some aromatic DBPs in the occurrence of particular precursors. Further, pre-oxidation via chlorine could lead to chlorate formation [42]. With a view to ameliorating disinfection, numerous disinfectants comprising ozone, ClO_2, and chloramines have been utilized [1]. Implementing such agents diminishes the yield of the four regulated THMs, trihalogenated HAAs, and TOX; however, many priority DBPs produced from such chemicals could give rise to other troubles [102].

If juxtaposed with chlorination, chloramination usually forms lower levels of regulated DBPs [103]. However, chloramination could conduct to the generation of DBPs with greater toxicity, like HANs and HNMs, as well as NDMA, a kind of nitrosamine [104]. Furthermore, the fraction of Br-DBPs, which are more dangerous than their chlorinated analogs, was frequently bigger throughout chloramination than that throughout chlorination [105]. As a consequence, the formation of fresh DBPs has to be considered if substituting chlorine disinfection by chloramine [1].

Chlorine dioxide stays an efficacious disinfectant that forms fewer DBPs.

THMs generated via ClO_2 when juxtaposed to Cl_2 [106]. Nevertheless, ClO_2 conducts to the formation of HANs, aldehydes, and many inorganic DBPs like chlorites and chlorates. ClO_2 is commonly integrated with chlorine disinfection; in this context, ClO_2 oxidation prior to chlorination may decrease the generation of THMs and TOX [107]. However, in the occurrence of bromide, the part of Br-DBPs augmented after ClO_2/Cl_2 pre-oxidation. The ratio of ClO_2 has a considerable contribution to producing DBPs. With augmenting chlorine content in mixed oxidants, the generation of chlorite was diminished while more chlorate was produced. There is an optimum ClO_2: chlorine ratio which generates the smallest level of HANs and TCNM [108]. Consequently, it remains requested to set such a ratio following the water quality, so as to dominate the production of DBPs [1].

Ozonation could avert the formation of some DBPs [14]; however, pre-ozonation elevates the DBPFP of specific HAAs throughout coming chlorination [109]. If contrasted with chlorine disinfection, ozonation usually produced DBPs with bigger toxicity. Ozonation frequently formed carbonyl-containing by-products comprising aldehydes and short-chained carboxylic acids [1]. Further, bromate is produced by ozone in the existence of bromide. If ozonation is merged with BAC filtration, the generation of DBPs could be dominated to some extent via reducing precursors. BAC may efficiently decrease DBPFP, as well as the yields of DBPs such as N-nitrosamines, haloacetaldehydes (HALs), and haloacetamides (HAcAms), which were generated throughout ozonation [110] [111]. Taking into account the merits of this incorporation, O_3/BAC could be utilized as an alternative disinfection method to chlorination [1] [12] [14].

In summary, as the oxidizing agent of the disinfectant increases, the formation of conventional DBPs is reduced, but more toxic DBPs emerge. When adopting the disinfection technique, it has to be considered with the particular water quality to decide if fresh DBPs may be given rise [1].

4. DBPs Elimination

For dominating DBPs, several techniques work on eliminating precursors such as coagulation, membrane filtration, AOPs, and their merged methods [1].

As the most frequent and economically realizable methods, coagulation and flocculation were employed to reduce NOM from potable water [1] [49] [51] [112]. However, more efficacious and economical technologies stay requested to eliminate both NOM and organic matter (OM) due to water quality troubles [72] [81]. Many investigations have been dedicated to reducing DBP precursors by enhancing the action of coagulation [49] [50] [113]. Numerous fresh coagulants have been utilized in the water treatment industry [114] [115], like pre-hydrolyzed ferric and alum coagulants [116], Mg/Al hydrotalcite, dendrimers, hyperbranched polymers, carbon nanotubes, and cyclodextrins [1] [117]-[123]. Further, the bromide level could be decreased efficiently via enhanced coagulation [49]. On the other hand, a few coagulants like chitosan can participate in

producing N-DBPs [1].

Membrane processes depicted greater performance in eliminating NOM if paralleled with traditional techniques [85] [86] [87]. Membrane processes such as ultrafiltration (UF) and UF-nanofiltration (NF) can be efficacious in decreasing DBP precursors [124]. In addition, filtration could be integrated with ozonation to retain NOM [1].

For dealing with DBPs, AOPs involve ozonation, UV disinfection, oxidation with hydrogen peroxide (H_2O_2), activated carbon and their integrations [33] [34] [35]. Vacuum ultraviolet (VUV, 185 + 254 nm) reaches better performance than UV_{254} (only 254 nm) in decomposing HANs [1] [23]. Many hybridizations of UV with additional treatment technologies are applied such as UV-H_2O_2 founded AOP that was the most tried. UV/H_2O_2 possesses the capability to dominate efficiently nitrosamines [125]. UV/H_2O_2/micro-aeration techniques are performant in decomposing totally DCAA [126]. When juxtaposed with a downstream BAC filter, the UV/H_2O_2 method can reduce THM and HAAs formation greatly since BAC could efficaciously eliminate biodegradable DBPs [127]. UV may be also hybridized with Cl_2. Indeed, UV/Cl_2 is more performant than UV alone in dealing with I-THM generation [1]. Fresh AOPs comprising UV/PS, UV/TiO_2, UV/Cl_2, TiO_2/O_3, O_3/H_2O_2, and MnO_2/O_3 have been implemented for dominating the production of DBPs [1].

5. Study Futures

Tang *et al.* [1] focused on the research trends in terms of three aspects: 1) analytical techniques; 2) toxicity and health effects; 3) water quality standards and control methods.

Figure 1 illustrates the first year in which each DBP species given in the keywords appear in the literature. Since the first publication concerning DBPs were released in 1974 [36] [37], novel categories of DBPs have constantly been detected until these days. As shown in **Figure 1**, DBPs found later usually have a tendency to be more poisonous than DBPs of primitive researches.

Novel disinfection techniques and emerging pollutants in water can form fresh DBPs [11] [88]. DBPs surfacing lately are frequently with low levels and elevated poisoning [1] [5] [7] [8] [89].

Due to the influences on human health, lower limits on numerous DBPs have been decided. Such regulated DBPs are mostly established following classical chlorination. However, employing different disinfectants (like chloramine, ClO_2, and O_3) conducts to the emergence of a fresh set of DBPs with worse poisoning, which are famous as unregulated DBPs [112]. It is vital to add hazardous emerging DBPs to the water quality standards, which can participate in attaining the target of rendering potable water safer. To satisfy the requirements of regulations, several types of research have been performed on dominating DBPs. DBPs dominating techniques are mostly incubated in two sides: enhancing disinfection engineering and pre-treatment techniques. As it forms a few DBPs, UV

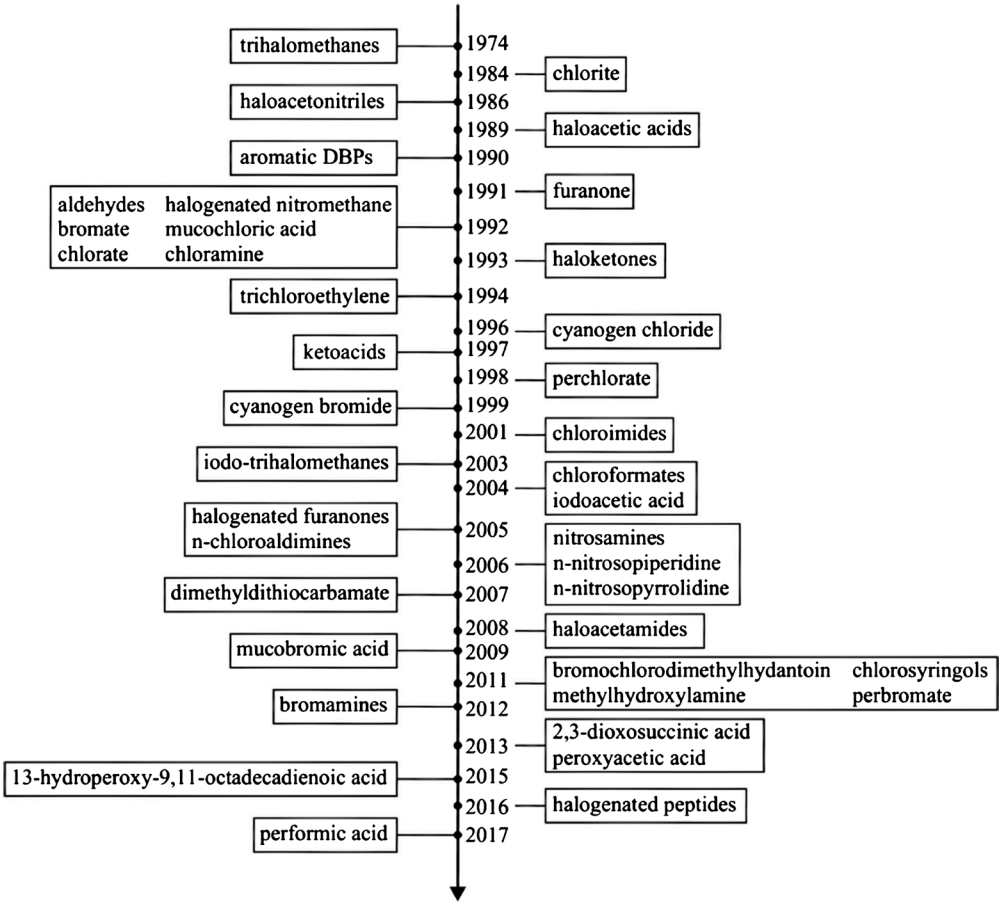

Figure 1. The historical review of DBPs from 1974 to 2018 [1].

disinfection is advocated to substitute classical disinfection processes. In addition, UV could be hybridized with ozonation. Indeed, Integrated O_3-UV AOP is more performant than either ozone or UV treatment alone and is efficient in eliminating OM [23]. Additional integrated methods like O_3/BAC, permanganate oxidation, and powdered activated carbon adsorption (PM-PAC) were found efficient in dealing with DBPs [14]. Integrating diverse disinfection processes is viewed as a crucial choice to ameliorate disinfection engineering in the next years. Further, several fresh disinfection processes came out like solar disinfection via photocatalysis that is an encouraging technique possessing the capacity to eliminate both microorganisms and DBP precursors [68]. On the other hand, many nanomaterials could be utilized as disinfectants thanks to their antimicrobial characteristics and decrease the risk of grave DBPFP through the classical disinfection process [128]. Aside from enhancing disinfection engineering [129], it is more vital to eliminate DBPs precursors. Coagulation stays the most broadly implemented and economical treatment process [130]. AOP remains an encouraging technology for removing precursors, in which UV, H_2O_2, and O_3 are employed commonly. Membrane processes should be adopted for safe potable water [15] [19] [64] [65] [118]. For distinct DBPs, several technologies may be merged to obtain satisfying elimination [1] [70] [131] [132].

6. Conclusions

In the water treatment industry, if there is a process that has attracted polemic discussion in terms of pros and cons disinfection has attracted the main part for its disinfection by-products formation. This work concerns DBPs precursors, link among disinfection and DBPs, DBPs elimination, and study futures. From this work, the main conclusions emerge.

During the last half-century, chlorination has been shown highly toxic to human health. Indeed, as a classical disinfectant, chlorine generates a bigger number of halogenated by-products than other disinfectants. Unfortunately, novel disinfection techniques and emerging pollutants in water can form fresh DBPs. DBPs surfacing lately are frequently with low levels and elevated poisoning. Further, as the oxidizing agent of the disinfectant increases, the formation of conventional DBPs is reduced, but more toxic DBPs emerge. Membrane processes, such as UF and NF, depicted greater performance in eliminating organic matter if paralleled with traditional techniques. As a perspective, research should concentrate on physical processes such as distillation, solar disinfection, and filtration for better water treatment instead of injecting chemicals into water previously highly-chemically polluted.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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