Roles and Knowledge Gaps of Point-of-Use Technologies for Mitigating Health Risks from Disinfection Byproducts in Tap Water: A Critical Review

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Abstract

Due to rising concerns about water pollution and affordability, there is a rapidly-growing public acceptance and global market for a variety of point-of-use (POU) devices for domestic uses. However, the efficiencies and mechanisms of POU technologies for removing regulated and emerging disinfection byproducts (DBPs) are still not systematically known. To facilitate the development of this field, we summarized performance trends of four common technologies (i.e., boiling, adsorption, membrane filtration, and advanced oxidation) on mitigating preformed DBPs and identified knowledge gaps. The following highest priority knowledge gaps include: 1) data on DBP levels at the tap or cup in domestic applications; 2) certainty regarding the controls of DBPs by heating processes as DBPs may form and transform simultaneously; 3) standards to evaluate the performance of carbon-based materials on varying types of DBPs; 4) long-term information on the membrane performance in removing DBPs; 5) knowledge of DBPs' susceptibility toward advanced redox processes; 6) tools to monitor/predict the toxicity and diversity of DBPs formed in waters with varying precursors and when implementing different treatment technologies; and 7) social acceptance and regulatory frameworks of incorporating POU as a potential supplement to current centralized-treatment focused DBP control strategies. We conclude by identifying research needs necessary to assure POU systems protect the public against regulated and emerging DBPs.

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Background

Chlorine is the most widely-applied disinfectant at drinking water treatment plants (DWTPs) due to its cost-effectiveness. However, chlorination inevitably produces halogenated disinfection byproducts (DBPs) via reactions involving halides (bromide and iodide) and natural organic matter (NOM). During disinfection at centralized DWTPs, NOM reacts with chlorine and can result in halogen substitution into NOM and then formation of DBPs, such as trihalomethanes (THMs) and haloacetic acids (HAAs) (Jiang et al., 2020; Pan and Zhang, 2013; Richardson and Postigo, 2015). In addition, because maintaining chlorine residual (usually less than 4 mg/L as chlorine or chloramine as chlorine) (USEPA, 2006) in water distribution systems is critical to prevent the regrowth of microbial pathogens, halogenated DBPs can also continue to form as water travels to homes and businesses where water is used for drinking, cooking, and hygiene (e.g., baths, showers), hence exposing humans to DBPs via multiple routes (ingestion, inhalation, and dermal passage). Although THMs and HAAs are among the most detected and regulated DBPs in chlor(am)inated tap water, emerging DBPs such as halogenated aromatic DBPs and nitrogenous DBPs are also detected, and they have attracted increasing concerns because they are more toxic than currently regulated DBPs (Li and Mitch, 2018; Liu and Zhang, 2014; Plewa Michael et al., 2008; Yang and Zhang, 2013). In the absence of regulations on emerging or non-regulated DBPs, common point-of-use (POU) or household water treatment (HWT) devices—which were originally installed to remove unaesthetic tastes (e.g., salts) or odors (e.g., geosmin and chlor(am)ines) by homeowners, restaurants, and building and businesses managers—might also serve to treat regulated and non-regulated DBPs. However, despite studies on using POU treatment for regulated DBPs, little is currently known about the performance of POU technologies in mitigating non-regulated DBPs.

The global POU device market is growing rapidly and is estimated to be valued at 34.1 billion US dollars by 2025 (PRNewswire, 2017). Reverse osmosis (RO) dominates the POU device market share (43%). In 2019, over 18 million POU units were sold in China alone, and 80% of those contained RO cartridges and at least one activated carbon (AC) pretreatment module. Although sediment filters and carbon block (CB) filters can be stand-alone POU systems and have estimated values of ~7.5 billion US dollars by 2025, they are sometimes also equipped in RO systems to pretreat particulates, odors, and residual chlor(am)ines. The rapid POU device market growth is largely due to increasing concerns toward water pollution, a desire to improve aesthetics, and rising affordability of the devices, especially in the Far East, Middle-East and other developing countries. However, while POU systems are often certified and designed to remove legacy, regulated, and mostly non-polar pollutants (e.g., volatile organics measured by gas chromatography), few POU certification protocols examine removal of unregulated and polar DBPs. These unregulated DBPs express the majority forcing agents for toxicity (Stalter et al., 2016a). These DBP classes include iodinated DBPs (Dong et al., 2019; Wagner and Plewa, 2017), nitrosamines (NAs) (Dai

and Mitch, 2013; Wagner et al., 2012), halonitromethanes (HNMs) (Plewa et al., 2004), haloacetonitriles (HANs) (Wei et al., 2020; Zhang et al., 2018a), haloacetamides (HAMs) (Plewa et al., 2008), haloacetaldehydes (HALs) (Jeong et al., 2015), halogenated aromatic DBPs (Pan and Zhang, 2013; Zhai and Zhang, 2011) and even nonhalogenated aromatic DBPs (Jiang et al., 2020; Zhang et al., 2020).

To facilitate the development of this area and raise recognition of the importance of POU treatment, we herein summarize some exemplar studies of DBP removal by four common POU systems (Figure 1), including heating/boiling, AC adsorption, membrane filtration, and advanced oxidation process (AOP). In addition, this study identifies some knowledge gaps that inhibit further understanding of POU technologies. Along with needs for in-depth exploration, the review intends to highlight the roles of decentralized POU technologies in supplementing utility-scale, system-wide DBP control strategies.

Knowledge Gap #1: We lack data on the occurrence of many regulated and emerging yet unregulated DBPs within homes.

Although many studies have surveyed DBP occurrence at DWTPs and in distribution systems, very few studies have reported DBPs at the tap within homes, and even fewer have reported DBPs at the cup after POU treatment. Epidemiologists realize the need to survey DBPs in homes to determine human exposure levels, but such surveys are rarely performed (Arbuckle et al., 2002; Calderon, 2000; Hamidin et al., 2008; Hrudey et al., 2015; Krasner et al., 2016). In-home measurements are also needed to develop and tailor POU technologies for balancing and mitigating health risks (Li and Mitch, 2018). To the best of our knowledge, only regulated DBPs have been measured in tap water in limited epidemiological studies aiming to link their exposures with adverse outcomes (Jeong et al., 2012; Legay et al., 2010; Parvez et al., 2019; Wang et al., 2019). Those limited data of regulated DBPs at taps within homes and businesses showed significantly different DBP concentration trends compared with those leaving DWTPs or in distribution systems, probably because of continued reactions of chlorine with DBP precursors within premise plumbing (Charisiadis et al., 2015; Dion-Fortier et al., 2009; Li and Mitch, 2018; Saetta et al., 2021; Salehi et al., 2020; Symanski et al., 2004). The role and impact of plumbing systems on regulated and emerging DBPs remain an under-studied area wherein many processes and components (e.g., ion exchange, copper pipes, and biofilms) are all likely to extend residence times and affect DBP formation.

In one example, a year-long case study surveyed two classes of regulated (THMs and HAAs) and one class of emerging DBPs (i.e., HALs) in a business building daily, weekly, monthly, and seasonally before and after a typical POU system (Figure 2). The POU system consisted of a polypropylene cotton filter, two granular activated carbon (GAC) filter cartridges, plus a boiler in sequence (Wang et al., 2019). Figure 2 illustrates the temporal variations of DBPs. Results showed large seasonal DBP variations at the tap, and most target DBPs were higher in summer when water temperature was also higher. The removal of THMs and HAAs by POU generally decreased with time or facility aging, but the control of HALs was always highly efficient (~100%), suggesting that some emerging DBPs are

more vulnerable than regulated DBPs to the selected POU system. However, this survey was conducted at a single location and did not consider prior DWTP operations and other DBPs. As such, the study may not represent the conditions at other locations with different centralized water treatment operations, water usage patterns, or other types of DBPs.

We recommend future research to collect more occurrence data on broader classes of DBPs at the taps (kitchens, showers) across different types of homes, buildings, and commercial POU systems. Providing sufficient occurrence data will inform people about the relative risks of different classes of DBPs at the tap, either with or without POU devices. Furthermore, once POU performance knowledge expands, it becomes possible to tailor a specific technology to supplement existing DBP control strategies, which are mostly based on data from DWTPs and distribution systems currently.

Knowledge Gap #2: Uncertainties remain regarding the control of DBPs by domestic heating, boiling, and cooking processes.

Heating or boiling for DBP control is the most-studied method among all POU processes (Xiao et al., 2020). POU devices such as boilers, brewers, saucepans, and microwave ovens heat tap water for preparing foods and beverages. Additionally, domestic thermal water systems produce hot water for bathing, showers, and other hygiene activities. Because inhalation and dermal contact are important exposure routes for volatile and hydrophobic (or nonpolar) DBPs (Chowdhury et al., 2020), chemical transformations in these heating systems are also important. The effects of boiling on different DBP classes in chlor(am)inated waters have important implications for DBP exposure assessment in epidemiologic studies. Failure to directly measure residential DBP concentrations or adequately capture spatial variability of DBP concentrations in exposure assessments will lead to inaccurate findings from epidemiologic studies.

Overall, boiling can reduce total organic halogens (TOX, an indicator of organic halogenated DBPs) in chlor(am)inated waters by 40–60% (Liu et al., 2015; Pan et al., 2016; Pan et al., 2014). Thermal-accelerated transformation and volatilization are the two mechanisms responsible for DBP removals. Figure 3 illustrates the pathways by which heating or boiling affects the levels and speciation of DBPs in chlorine-containing tap water: (a) DBP formation through the thermal cleavage of large halogenated NOM molecules (Wu et al., 2001); (b) DBP formation via heating-accelerated reactions between DBP precursors (e.g., NOM and thermally-unstable DBPs) and chlorine residual (Liu et al., 2020b; Liu et al., 2015; Pan et al., 2014; Yan et al., 2016); (c) DBP transformation from halogenated to nonhalogenated via thermohydrolysis, and DBP conversion from nonvolatile to volatile via decarboxylation (Hua and Reckhow, 2008; Krasner and Wright, 2005; Ma et al., 2017; Pan et al., 2014; Shi et al., 2017; Zhang and Minear, 2002); and (d) DBP removal via volatilization (Leuesque et al., 2006; Ma et al., 2017).

The DBP removal efficiency of heating/boiling varies significantly depending on compound thermal stability and volatility. For nonvolatile and thermally-stable DBPs (e.g., some halogenated aromatic DBPs), concentrations may increase during the heating/boiling. It has been reported that four brominated aromatic DBPs increased by 25–90% in both simulated

and real tap water samples after boiling for 10 min (Liu et al., 2015; Pan et al., 2014). Pan et al. (Pan et al., 2014) even identified several new polar halogenated aromatic DBPs with one or more carboxylic groups attached to the benzene ring (e.g., 2,4,6-tribromo-5-hydroxyisophthalic acid). These large DBPs may undergo thermal decarboxylation to form relatively small aromatic DBPs (e.g., 2,4,6-trihalophenol) or THMs and HAAs. Consequently, it was not surprising that the levels of dihalo-HAA also increased after boiling (Chowdhury et al., 2010; Pan et al., 2014).

Nonvolatile and thermally-unstable DBPs (e.g., halogenated aliphatic acids) are mainly subjected to thermohydrolysis and decarboxylation during boiling (Pan et al., 2014; Urbansky, 2001; Zhang and Minear, 2002). Zhang and Minear (2002) reported that trihalo-HAAs (X_3CCOOH) might be decarboxylated to form THMs even at ambient temperature. Urbansky (2001) reported that trihalo-HAAs were partially dehalogenated to form dihalo-hydroxyl HAAs ($X_2C(OH)COOH$). A new group of polar aliphatic acids with "carbon–carbon double bond" (e.g., halobutenedioic acid) was identified to be thermally unstable and easily decomposed (Pan et al., 2014).

Volatile and semi-volatile DBPs (e.g., THMs, haloketones, certain HALs, HANs, and HAMs) are either volatilized directly or hydrolyzed to form THMs and then volatilized after 5 min of boiling (Krasner and Wright, 2005; Ma et al., 2017; Shi et al., 2017; Wu et al., 2001). As shown in Figure 3, many volatile halogenated DBPs (e.g., THMs, HALs, HANs, and HAMs) are produced via degradation of their precursors or hydrolysis of other DBPs. Because these volatile DBPs can be pulled out of the boiler with the aid of household ventilators during heating/boiling, the boiling process can be regarded as a "dehalogenation" process.

Because the fates of different DBP species under heating/boiling vary significantly, it is difficult to measure all DBPs individually; rather, it is better to analyze the overall toxicity of DBP mixtures. Based on earlier studies of DBP mixtures in chlorinated tap waters (Liu et al., 2015; Pan et al., 2014), 5 min boiling reduced the cytotoxicity (against the Chinese Hamster ovary cells) of a simulated tap water by 77% (Pan et al., 2014), and decreased the developmental toxicity (against the embryos of the polychaete *Platynereis dumerilii*) of two real tap waters by 53% and 57% (Liu et al., 2015).

Interestingly, recent studies showed that adding ascorbate (i.e., vitamin C) (Liu et al., 2020a), lemon slices (rich in Vitamin C) (Liu et al., 2020b), or tea before boiling could reduce the toxicity of DBP mixtures in chlorinated tap waters. The mechanism is mostly attributed to quenching of chlorine residual, which inhibits formation of halogenated DBPs. Separately, washing lettuce with chlorinated water produced substantial DBPs (Komaki et al., 2018; Lee and Huang, 2019; Shen et al., 2016). In addition, Liu et al. (Liu et al., 2020a) found that adding 2.5 mg/L sodium carbonate (i.e., soda powder) before boiling could further lower the toxicity of DBP mixtures, which is attributed to the alkaline-aided hydrolysis and dehalogenation of preformed DBPs. Therefore, boiling with certain food additives might enhance the detoxification of chlorinated water. In contrast, other commonly-used food cooking additives may trigger reactions between chlor(am)ine and DBP precursors and form relatively more toxic DBPs. For example, iodized table salts

containing I⁻ can be oxidized by chlor(am)ine to generate HOI, which subsequently reacts with organic matter in the dissolved food and forms iodinated DBPs (I-DBPs) (Pan et al., 2016; Yan et al., 2016). Considering iodinated DBPs have higher toxicity than regulated DBPs, the formation of I-DBPs during cooking should be systematically investigated (Dong et al., 2019; Ersan et al., 2019).

Overall, there are a variety of formation and transformation processes co-occurring during boiling, and future studies may attempt to evaluate the toxicity changes more systematically throughout the treatment process instead of taking a snapshot before and after treatment. The challenge and labor involved in analyzing each DBP in water, to the authors' opinion, could be avoided by focusing on the TOX and toxicity evaluation during common heating and cooking practices.

Knowledge Gap #3: We lack standards to better evaluate the performance of carbon-based materials and absorbability of most of DBPs.

Adsorption is the second most-studied process used in POU devices. Of all activated carbon (AC) materials, carbon block (CB) is the mostly used form of AC in POU. Carbon block consists of finely meshed AC particles (0.045 to 0.18 mm), which are formed together with binders to create a porous ceramic-like material that is heated, molded, and extruded in a tubular form with radial flow from the outside into the inside portion of the tube (Lau et al., 2005). Although the contact time for CB and water is often short (< 60 seconds), it is adequate to remove many organic micropollutants, tastes, odors, pathogens, and particulates from tap water (Anumol et al., 2015; Sublet et al., 2003; Wu et al., 2017). The recommended treatment lifespan before CB cartridges need replacement is on the order of 5000 to 10,000 liters. Chloroform removal is currently used as the standard to certify CB performance (NSF-International, 2019). Because POU systems are often operated only periodically, organic micropollutants have time to diffuse into AC pores, which therefore frees up surface sites for rapid adsorption in the next cycle (Kim et al., 2019). In addition, CBs are often used as pretreatment devices for RO systems because CB quenches chlorine residuals that could damage polymeric membranes (Wu et al., 2017).

In one of few thorough studies using POU devices to remove DBPs, six of eleven POU devices removed TOX by > 60%. The selected devices containing CB did not show uniform performance in removing absorbable organic halogen (AOX), but they generally exhibited higher removals for absorbable organic bromine (AOBr) than absorbable organic chlorine (AOCl) (Figure 4). Another study reported significant removals of THM, HAA, 3-chloro-4(dichloromethyl)-5-hydroxy-2(5H) furanone (known as MX), and TOX by CB, but it lacked data on HALs, HANs, and HNMs (Xiao et al., 2020). Moreover, CB is not effective in removing inorganic DBPs such as bromate or chlorate. In addition, results show that although two types of GAC modules demonstrated complete removal of HALs and HANs, the THM and HAA removals were relatively poor and gradually worsened over time (Wang et al., 2018a). The evidence suggests that POU devices are more efficient in treating emerging DBPs than regulated DBPs, but it also requires frequent replenishment or replacement of the AC cartridges.

In another study, while the combination of technologies made it difficult to attribute DBP removal to AC or ion exchange resin in a POU pitcher, the selected adsorbents removed ~90% of THMs and 50–70% of HAAs (Levesque et al., 2006). Given that most cation exchange resins are designed to remove divalent hardness ions while only a small portion of POU systems utilize anion exchange resins to remove nitrate and anionic pollutants, it is suggested that the selected pitcher contained anion exchange resins. In DWTPs, a powder activated carbon (PAC) system usually has a contact time of 5 to 30 min, and they have been shown to adsorb not only DBP precursors, but also preformed THM effectively; however, PAC was largely ineffective in removing HAAs (Tung et al., 2006). Moreover, the operational conditions and AC used in DWTPs are markedly different from those used in POU systems. In a review article, researchers summarized the removal of several regulated DBPs using AC (Figure 5); however, the article lacks data for emerging DBPs (e.g., HAL, HAN, HNM). Given that NSF International/American National Standards Institute(ANSI) standards only use chloroform as a surrogate (NSF-International, 2019; 2020), it may be reasonable to imply that DBPs with higher polarity than chloroform (i.e., log D_{ow} < 1.97) are more difficult to remove to levels > 95% (i.e., certification requirement for chloroform).

Given the lack of controlled or field data on DBP removal (Levesque et al., 2006), there is a need for this type of applied research. A challenge is the proprietary source and characteristics of adsorbents used in carbon block, activated carbon, ion exchange, or mixed sorbent materials (Smith et al., 2010). Among the neutral non-charged THM species, chloroform is more polar (log K_{ow} = 1.97) and removed less efficiently than bromoform, which has a higher octanol-water partition coefficient (log K_{ow} = 2.38) (Fischer et al., 2019). Thus, while chloroform is a reasonable surrogate for neutral DBPs, it is perhaps not a reasonable indicator for the effectiveness assessment of acidic or polar moieties. In the absence of experimental data, the best indicator across broad groups of DBPs is probably the pH-corrected octanol-water partition coefficient (log D_{ow}). In addition to the types of adsorbents, a comprehensive future study may examine the size and surface properties of those commonly-sold adsorbents in POU systems to better understand the range of materials.

Nowadays, the performance of AC filters can be evaluated at a manufacturer's laboratory by following NSF/ANSI standards such as Standard 42 for aesthetic parameters and Standard 53 for health-based chemicals (NSF-International, 2019). NSF/ANSI Standard 61 covers leaching of chemicals from materials that come into contact with water and includes removal performance of four THMs. NSF/ANSI 53 addresses systems designed to remove specific substances, but for DBPs it uses only chloroform as a surrogate. For POU device certification, the test requires reducing an influent chloroform concentration of 0.300 mg L^{-1} to a maximum permissible concentration of 0.015 mg/L (i.e., > 95% removal). In some tests, other DBPs (e.g., HAAs, HAN, haloketones) are measured as "incidental contaminants / emerging compounds" and can be requested to be measured. However, most of the certificate standards consider only aliphatic DBPs, which are known to be less toxic than aromatic DBPs (Li and Mitch, 2018; Liu and Zhang, 2014; Yang and Zhang, 2013). As such, there is also a need to revisit those certification standards to cover a broader range of DBPs.

Knowledge Gap #4: We lack long-term data on membrane performance in removing DBPs from tap water.

Among water treatment technologies, nanosized membranes such as nanofiltration (NF) and RO are often deemed omnipotent for removing pollutants. Compared to other physical-chemical-biological treatment approaches, membrane filtration features advantages like high efficiency, little chemical addition, and ease of operation. Therefore, membrane processes have been gradually accepted and extended to POU devices in recent decades. Because RO performs better than NF for desalination and rejection of micropollutants including DBPs (Doederer et al., 2014; Wang et al., 2018b; Yang et al., 2017), specifically nitrosamines (Fujioka et al., 2013), RO is deployed more frequently in POU products. However, because RO and NF are usually composed of the same type of material (i.e., polyamide), they are very similar except for pore size, and sometimes NF is termed "low-pressure RO."

In general, DBPs rejection efficiencies rank as follows: Cl- ≈ Br- ≈ I-HAAs for the compounds with the same number of halogens, and tri- > di- > mono-DBPs for the compounds with identical halogen types. Among DBPs, the rejection ranks as follows: HAAs   HALs ≈ HMs for compounds with identical halogenation types and degrees. Some environmental and operating factors may play important roles on the DBPs rejections during RO operation, including pH, ionic strength, membrane material and age, operating pressure, and water matrix. For example, polyamide RO membranes performed better for HAAs removal than cellulose acetate RO membranes (Agus and Sedlak, 2010; Kimura et al., 2003). Overall, rejection of DBPs dosed into real tap water was higher than DBPs dosed into ultrapure water (Chen et al., 2021b; Wang et al., 2018b), suggesting that laboratory experimental results would not overestimate the treatability of DBPs in real tap water using RO membranes. HAAs are the group of DBPs mostly studied with RO so far, followed by THMs, NAs, and HALs. However, not many studies have been done for other types of DBPs, thus demanding further investigation.

The mechanisms responsible for membrane filtration of chemical compounds are usually size-exclusion and electrostatic charge repulsion. However, because many DBPs are low in molecular weight and neutral in charge, some DBPs can penetrate membranes. With regard to membrane-micropollutant interactions, recent studies calculating mass balances after measuring DBPs in RO permeate and RO retentate revealed that membrane trapping/sorption also contributes to DBP loss during RO operation (Chen et al., 2021b; Fang et al., 2020). Certain small and neutral DBPs (e.g., monochloroacetaldehyde) permeated RO readily, thus bringing the highest risks (Chen et al., 2021b). In contrast, almost all negatively-charged HAAs were rejected > 90% by RO, thus imposing little risks to finished water. Some fractions of THMs (typically small, neutral, and hydrophobic DBPs) were retained in the RO membrane (Figure 6) and were not easily flushed out, thus posing chronic risks to water quality if they are eluted out later. Future studies should verify the long-term performance of RO in treating DBPs and explore novel methods to prevent micropollutant permeation during prolonged operation.

Notably, POU membrane systems often include prefilters and CB prior to the membranes. However, previous studies that assess DBP removal by membrane RO systems rarely

differentiate DBP adsorption on CB from membrane rejection. As such, there is perhaps a research need to evaluate the impacts of CB (e.g., a habit without frequent replacement) on the longevity of membrane-based POU systems.

High energy consumption and low water recovery are the major impediments for extensive adoption of RO in large-scale engineering applications but are less critical for domestic implementation. Typical RO system operation usually requires elevated pressure provided by a booster pump. To reduce energy and water wastes, earlier studies evaluated multi-stage RO process (Wang et al., 2018b) and flexible RO process (Chen et al., 2021b) as innovative designs to improve energy efficiency and enhance water recovery while eliminating DBPs. Applying a five-stage RO in this study achieved > 74.6% HAA rejections while recovering 87.0% of water. To balance the needs of water quality, water quantity, energy, and investment cost, a four-stage RO was recommended (Wang et al., 2018b). For flexible RO, 80.8% removals of 19 selected DBPs were obtained while holding water recovery constant at 80%. Designing a practical and sustainable RO system could be a future research direction.

While recognizing the robustness of RO in eliminating pollutants, we acknowledge that one inevitable drawback of RO-treated water is its lack of nutrients like calcium, fluoride, and trace elements in finished water, resulting in a long-time debate about the suitability of drinking RO-treated water (Sedlak, 2019). To the authors' opinion, this is a question of balancing nutrients and pollutants. For areas with enough nutrients coming from foods, the nutrients in water are negligible and therefore people might focus on reducing pollutants. In contrast, for areas with insufficient or nonreplaceable nutrient sources, people can focus on gaining more nutrients within acceptable pollutant levels. Regardless, we should be very cautious of labeling RO-treated water as toxic or deficient water. Water itself is not toxic or deficient; it is the levels of nutrients that matter. One way of bypassing this debate may be to add specific nutrients into RO-treated water.

Knowledge Gap #5: We lack knowledge of DBPs' susceptibility toward numerous advanced redox processes.

Advanced oxidation processes (AOPs) (Miklos et al., 2018) and advanced reduction processes (ARPs) (Vellanki et al., 2013; Xiao et al., 2017) span a wide variety of technologies potentially usable for water and wastewater treatment, but many of them are inapplicable to POU-relevant conditions. UV irradiation via low-pressure ultraviolet (LPUV) lamps that emit germicidal wavelengths (254 nm) are already used in DWTPs for microbial (bacteria, virus, oocyst) inactivation; therefore, it is likely safe and usable. Recently, interest is growing in incorporating LPUV or light emitting diodes (LEDs) that emit germicidal wavelengths (260–280 nm) into POU systems (Beck et al., 2017; Linden et al., 2019; Woo et al., 2019). Although integrating photocatalysts (e.g., TiO_2) to aid in destruction of trace organics is possible, issues surrounding catalyst leaching and recovery limit photocatalyst use in POU products (Kim et al., 2019).

Overall, two mechanisms (direct photolysis and indirect oxidation associated with hydroxyl radicals) are responsible for removal of DBPs in photo-dependent UV POU systems.

In direct photolysis, DBPs absorb emitted photons and undergo direct photolysis if their absorption spectra overlap with the emission spectra of the irradiation. The direct photolysis fluence-based rate constants (k) under LPUV irradiation (UV_{254}) ranges from 0 to 1.32×10^{-2} cm^2 mJ^{-1} for diverse DBPs. The k generally follows the trend of $k_{NA} \gg k_{THNM} > k_{THM} > k_{HAA} > k_{THAL} > k_{HAN} > k_{HAM}$. NAs have the highest k due to their high molar absorptivity at 254 nm (ε_{254}, 1,518–4,597 $M^{-1}cm^{-1}$) (Lei et al., 2020). For halogenated compounds, k generally follows the trend as I-DBPs > Br-DBPs > Cl-DBPs. Chloroform and chloral hydrate (CH, i.e., hydrated form of trichloroaldehyde) have no UV absorbance at 254 nm and therefore cannot be photodegraded via direct photolysis. NAs can be degraded by half under a UV_{254} dose of 165 mJ cm^{-2} ($E_{50\%}$) while I-DBPs including iodinated THMs and TIAA and some Br-DBPs including tribromoacetic acid, tribromonitromethane, and dibromochloronitromethane, can be degraded by half under a UV_{254} dose of 500 mJ cm^{-2}. In contrast, other DBPs are barely removed by UV_{254} doses below 500 mJ cm^{-2} (Figure 7). HAA photolysis under medium pressure UV generally follows the same trends as with LPUV (Wang et al., 2020a). Moreover, vacuum UV (VUV) can initiate multiple types of radicals and trigger degradations of DBPs such as I-DBPs and halophenols (He et al., 2021). The photon doses used in DWTPs are often on the order of 10 to 100 mJ cm^{-2}. As such, under a typical UV_{254} dose of 100 mJ cm^{-2}, only some NAs and iodinated DBPs can be degraded. However, because people may prolong the irradiation time conveniently at home and significantly enhance the light dosage (e.g., 10,000 mJ/cm^2), a POU system containing UV process may not be impractical.

Radical-induced reactions in various AOPs are reported to be robust in removing trace organic contaminants. The second-order reaction rate constants of $HO^{\bullet}$ with DBPs range from ~10^6 M^{-1} s^{-1} to ~10^9 M^{-1} s^{-1}. Specifically, NAs and I-DBPs generally have higher reaction rate constants than Br- and Cl-DBPs ($k_{I-DBPs} > k_{Br-DBPs} > k_{Cl-DBPs}$) (Chuang et al., 2016; Landsman et al., 2007; Mezyk et al., 2006; NIST) (Figure 7). In comparison, $SO_4^{\bullet-}$ reacts with haloacetonitriles at 2.43–7.48×10^7 M^{-1} s^{-1}, iodoacetic acid at 3.4×10^9 M^{-1} s^{-1}, and NDMA at 1.5×10^{10} M^{-1} s^{-1} (Gilbert et al., 1974; Hou et al., 2017; Xiao et al., 2016). Note that in POU treatment process, a combination of residual chlorine with UV becomes a UV/chlorine AOP, hence providing an extra benefit. However, the reaction rate constants of chlorine radicals with DBPs are seldomly reported in literature. Only one model has predicted the rate constant of $Cl_2^{\bullet-}$ with NDMA to be 3.0×10^5 M^{-1} s^{-1} (Velo-Gala et al., 2019). The lack of the chlorine radical-induced reaction kinetics impedes prediction of preformed DBP fates.

Advanced redox processes requiring addition of chemicals (e.g., H_2O_2, sulfite, iron, etc.) are less likely to be used in POU systems, partially because their residuals are hard to recover without leaching and partially because they might form more DBPs in the presence of DBP precursors (Wang et al., 2020b). Although certain new POU systems can produce H_2O_2 or chlorine in-situ electrocatalytically prior to UV irradiation (Barazesh et al., 2015; Radjenovic and Sedlak, 2015; Zodrow et al., 2017), more efforts are needed to validate their ability to destroy, instead of form, regulated and emerging DBPs to convert scientific concepts into commercializeable products. Similarly, adsorption coupled with electrolysis (*e.g.*, a cathode consisting of GAC) combine electro-sorption and reductive dehalogenation for more robust DBPs abatement (Ao and Liu, 2017; Liu et al., 2016; Liu and He, 2020;

Radjenovic et al., 2012). These reagent-free technologies hold promise for POU users, although some potential improvements remain: (i) improve the selective adsorption of GAC to DBPs; (ii) balance the electricity consumption and DBPs removal efficiency; and (iii) develop sustainable and non-toxic electrodes. Notably, several recent studies have reported the formation of nitrite (a regulated carcinogenic compound) (Han and Mohseni, 2020) and H_2O_2 (a compound linked to aging and inflammation) (Chen et al., 2021a) during VUV irradiation of tap water, suggesting that certain AOPs/ARPs even without additives may impose risks to drinking water quality.

Besides understanding the fates of preformed DBPs in AOPs (or ARPs), another important point to understand is the fate and transformation of DBP precursors, such as organic chloramines and aromatic halogenated intermediates. If possible, it is necessary to identify the most toxic intermediate byproducts during AOP/ARP treatment. However, given that halogenated DBPs are often dehalogenated (Huang et al., 2020; Wang et al., 2017) along with unknown products during photolysis process, it is more important to evaluate the overall toxicity changes from a panoramic view rather than snapshots of selected conditions.

Knowledge Gap #6: We lack tools to monitor DBPs and predict refractory DBPs under different POU treatments.

Monitoring DBPs and predicting DBP susceptibility in POU systems may enable better understanding and increase POU implementation. Because some DBPs can be produced within POU systems (e.g., NDMA from anion exchange, but not cation exchange, resins (Krasner et al., 2013)), users gain confidence if people understand temporal DBP variations. Future studies that design in-line and/or real-time DBP sensors/equipment and deploy them in home to monitor DBPs changes would be beneficial. In addition, it would also be useful to monitor the continuing reactions of chlorine with materials used in premise piping and POU systems.

Some companies have developed automatic online analytical tools for THMs (e.g., Parker-Hannifin, Foundation Instruments) (Iii et al., 2015; Ranaivo et al., 2011; Saetta et al., 2021). However, this equipment is currently more of a research tool and is too costly for integration into POU systems. Because THMs represent only a small portion (often < 30%) of the TOX (Chen et al., 2020), there are also desires to develop an on-line TOX analyzer in the future to enable more frequent monitoring of total DBPs (Bu et al., 2018; Chen et al., 2020; Weinberg et al., 2006; Zhang et al., 2018b). In the future, to be more practical and perhaps meaningful to the perceptions of the general public, a simple bioassay paper-like strip could be developed too. (Wujcik et al., 2016; Yoon et al., 2019).

Given the diversity of DBPs detected so far, quantitative structure-activity relationship (QSAR) analysis may serve as a robust tool in the future to predict the treatability of DBPs with POU units and to predict the DBPs' associated toxicity (Chen et al., 2015; Qin et al., 2019). For example, one study considered the homogeneous surface diffusion model to simulate breakthrough of neutral organic pollutants by CB and concluded that including axisymmetric flow is important to improve simulation of experimental data for three representative VOCs (Kim et al., 2019). Advancements in CB design would

be greatly enhanced through a broader range of such modeling and validation studies, which could be used to optimize contact times, AC particle pore size distributions, and potentially additional additives (e.g., ion exchange binding sites) suitable for removing ionic DBPs. Additionally, the models are expected to consider cyclical on-off operations, which likely play a critical role in maximizing AC surface sites available for adsorption during subsequent periods of hydraulic flow. However, we still currently lack a pool of data that can initiate modeling and/or estimation of CB's performance. In addition, QSAR models rely highly on descriptors to establish correlations between compounds and performance (Chen, 2011). As such, the type and number of descriptors needed for developing QSAR models is sometimes debatable (Chen et al., 2015). In applied engineering areas, a persistent challenge for predicting DBP removal balance between ease-of-use and reliability.

It is also desirable to have predictions or a reporting of molar absorptivities at specific wavelengths other than 254 nm. Emerging technology using LEDs in POU systems opens the opportunity to use single or multiple wavelengths of light for pathogen disinfection or activation of oxidants (Beck et al., 2018; Beck et al., 2015; Beck et al., 2017). There are few computation tools currently available to predict molar absorptivity of chemicals across the wavelengths of UV-A, UV-B, and UV-C (e.g., 220 to 400 nm) that may be employed in next-generation POU systems enabled by LEDs.

A priori prediction of radical reaction rates with organic DBPs is not widely available. Recently, prediction models for micropollutant degradation are emerging(Li and Crittenden, 2009; Minakata et al., 2014), which consider chlorine radicals (Minakata et al., 2017; Zhang et al., 2021), sulfate radicals (Xiao et al., 2015; Ye et al., 2017), hydroxyl radicals (Jin et al., 2015; Luo et al., 2017; Minakata et al., 2009), hydrated electrons (Li et al., 2019), superoxide/hydroperoxyls (Nolte and Peijnenburg, 2018), or other reactive oxygen species that may be produced by UV, electrocatalysis or electrolysis processes within POU systems. With these advances, AOPs and ARPs could be more strategically developed to target specific classes of DBPs using different mechanisms.

Similarly, in the absence of data on NF and RO rejection of DBPs in low pressure under-the-sink membrane devices, there is a need to be able to predict the separation potentials of newly-identified DBP based only upon their chemical structure and expected concentrations in tap water. Once we know the relative efficiency among thousands of DBPs and recognize which DBPs are hard to remove, people can then focus on developing tailored technologies and tools to remove the new DBPs.

Knowledge Gap #7: We lack consensus on social equity, acceptance of social responsibility, and regulatory acceptance of including POU as part of a holistic DBP control strategy.

Previously, people often believed that POU devices were mostly designed to meet the water quality needs for less-developed areas or for only pathogen controls. However, DBPs are carcinogens inevitably present in developed areas; therefore, limiting their human exposure is important to all people. Centralized DWTPs can remove DBP precursors and minimize

DBP formation. However, residual disinfectants will continue to be used to protect against acute health risks associated with pathogen intrusion into pipe networks, and the presence of DBPs in tap water is an unavoidable consequence. In this context, to the authors' opinion, communities will increasingly rely on decentralized water treatment to "polish" water closer to the point of use to align its water quality with intended use (i.e., fit for purpose water treatment). Because POU technologies at the tap, household, apartment building, and commercial building scale are already widely distributed and they are achieving many fit-for-purpose aims, what lacks is evidence that POU systems are providing sustained protection against DBPs associated with adverse health effects. Thus, POU systems are already "socially accepted"; in many cases, aesthetics and perceived safety of POU-treated water are driving the growth of POU markets.

To assure sustained performance, someone must take responsibility for maintaining and replacing consumables in POU devices. Some POU vendors have mechanical (e.g., cumulative volume treated, flow meters, pressure sensors) or electrical (e.g., operational time, light sensors) indicators, and the use of internet- or WiFi-connected devices is increasing to indicate when maintenance or consumable replacement is needed. These alerts are important in ensuring timely maintenance. However, POU devices, especially those with such advanced features, come at an economic cost and may only be affordable by middle- to upper-class citizens, or they may place a disproportional economic burden on less affluent households. The United Nations views access to clean water as a human right, and many communities fear that decentralized and POU devices could create disparities in access to the "cleanest" water.

Finally, drinking water regulations are established to balance health and economic risks. For some pollutants (e.g., arsenic), POU devices play a permissible role in regulatory compliance for small communities (e.g., fewer than a few hundred residents) (Gurian and Small, 2002; Petrusevski et al., 2002; Slotnick et al., 2006). Thus, there is a precedent for regulators approving POU to aid in compliance. Regulations often take many years to establish. However, a shorter path may be obtained by having public-private partnerships set performance and treatment goals that are independent of regulatory requirements. For example, this is the approach used by NSF International when approving POU devices based on their removal of chloroform (NSF-International, 2019). Thus, identifying a limited number of DBP surrogates (e.g., perhaps four chemicals) that are widely occurring in tap water and representative of different classes of DBPs could be beneficial when evaluating and approving POU technologies. This approach would be similar to the use of trace organic surrogates for evaluating potable reuse treatment systems (Dickenson et al., 2009).

Concluding remarks

DBPs are a special type of pollutant formed mostly in water supply system and therefore they need a control strategy that differs from other pollutants occurring in raw sources. POU device implementation has been growing rapidly, so that the roles of POU devices in mitigating DBPs deserve more attention. In this study, we have summarized some information on DBPs susceptibility toward four common POU technologies, and gained seven major knowledge gaps of concerns (Table 1).

In order to address the knowledge gaps, research needs are suggested in Table 1. Specifically to each of the seven gaps, among the more important research activities include the following: 1) monitoring types and levels of DBPs in homes before and after POU treatments to identify specific DBPs most resistant to treatment and which DBP exposure control overall health risks; 2) assessing the cumulative/collective influence of reactant precursors, products, and toxicity of DBPs formed during water heating processes ; 3) revisiting current POU certification standards to cover more DBPs with varying properties for commonly used adsorbents in POU device; 4) evaluating the long-term performance, reusability, and replacement needs of membrane modules, plus quantifying the fate of DBPs in both permeate and brine; 5) comparing performance of AOPs & ARPs on degradation of DBPs in different water matrices and influence on overall toxicity of the treated water; 6) developing easy-to-use methods and/or models to detect or predict DBPs in tap water be it with or without use of POU devices; 7) identifying the uniqueness of each class of DBPs (varying toxicity, varying chemical properties that influence removal) to understand roles of each stakeholder from water source protection, centralized municipal drinking water treatment, to POU deployment by individuals. Overall, through collective efforts and more in-depth explorations, we believe that the DBPs issue can be relieved remarkably by including POU as a part of the DBP control strategy.

Acknowledgements

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Figure 1.
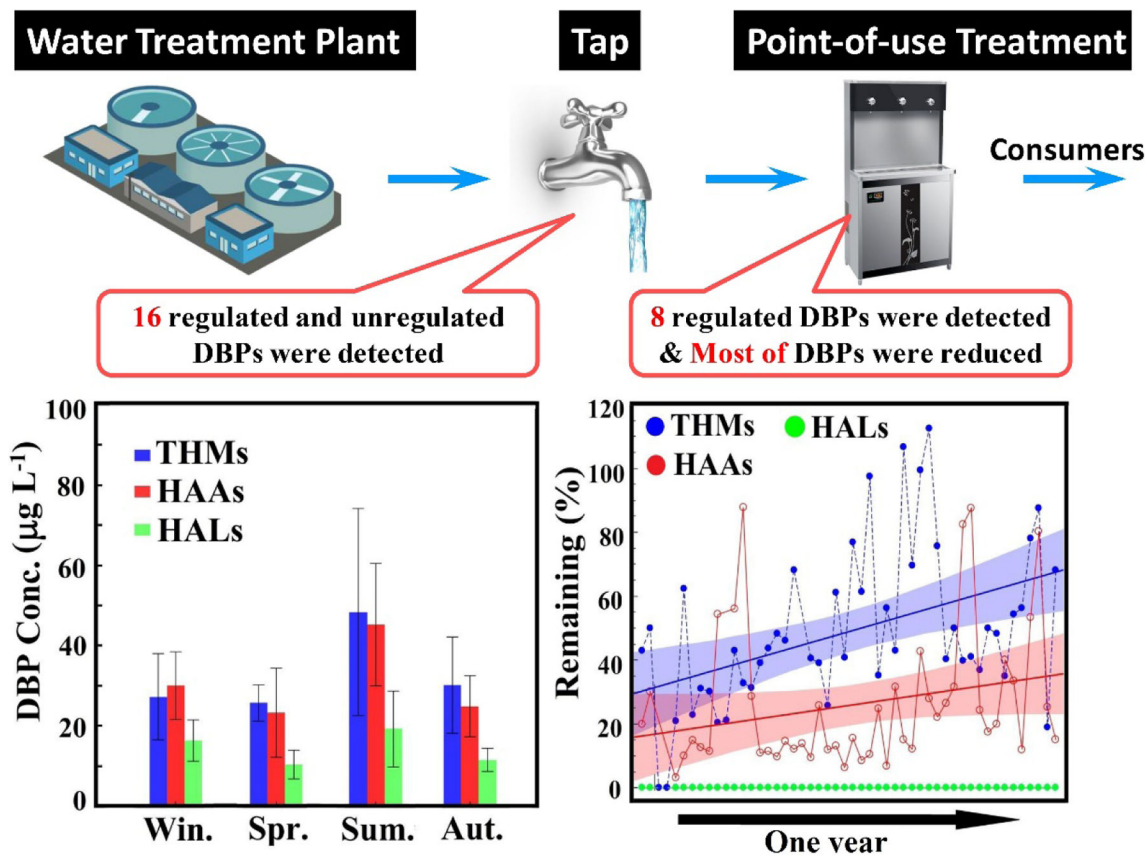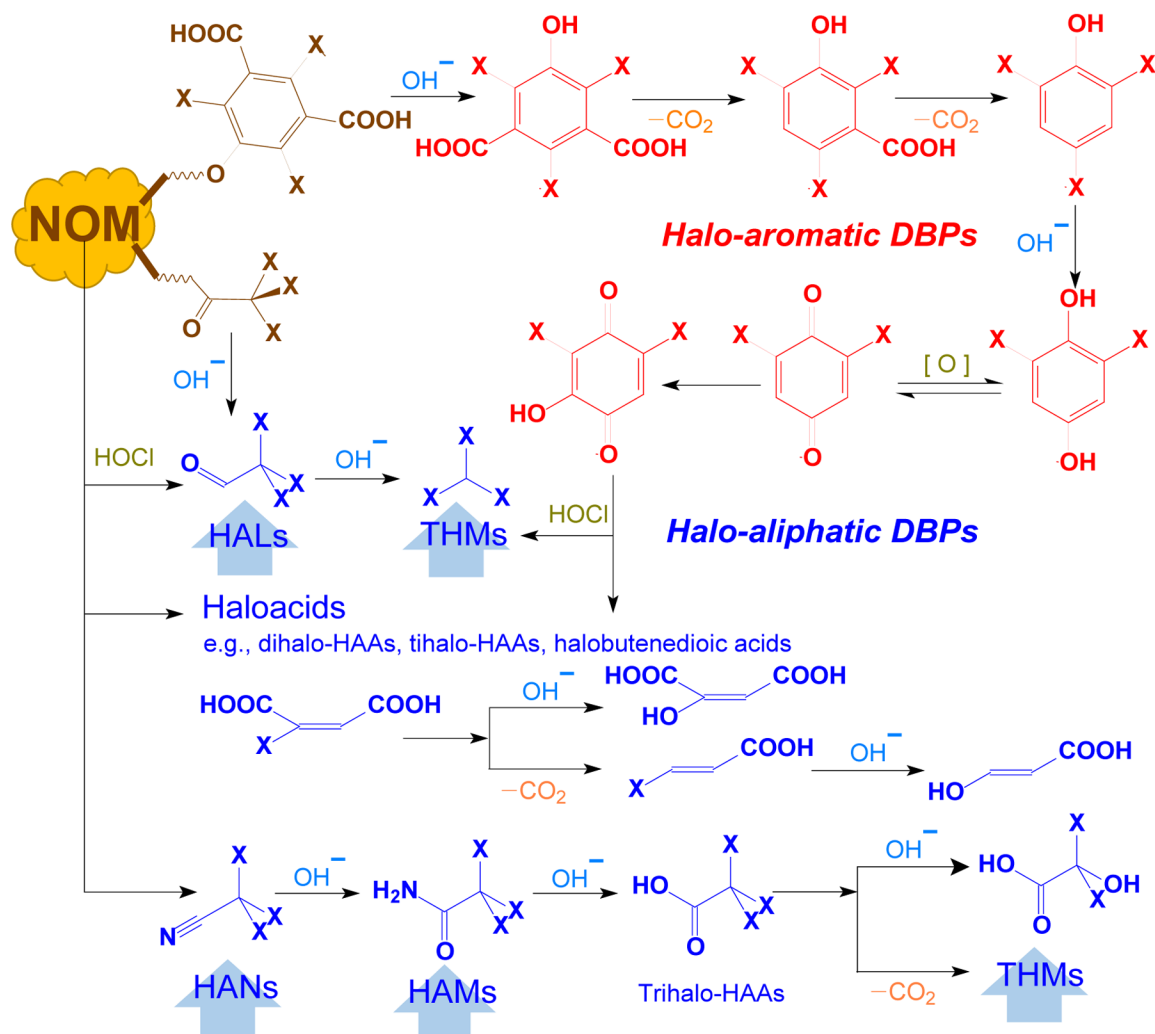A summary of POU treatment technologies for DBP mitigation

Figure 2.
Typical DBP occurrence at the tap and after treatment by a POU device in a one-year long survey (Reproduced from (Wang et al., 2019) with permission from Elsevier)
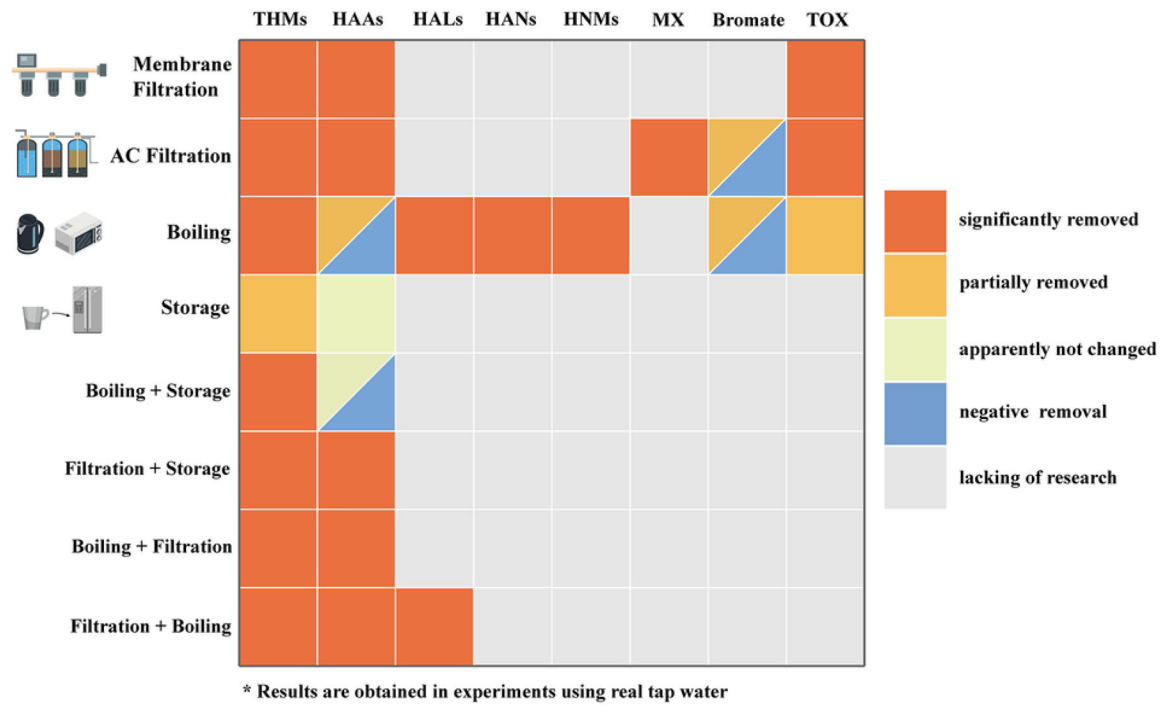
Figure 3.
The formation and conversion mechanisms of halogenated DBPs during boiling/heating of chlorinated tap waters (Hua and Reckhow, 2008; Jiang et al., 2020; Krasner and Wright, 2005; Pan and Zhang, 2013; Pan et al., 2014; Urbansky, 2001; Wu et al., 2001; Zhai and Zhang, 2011; Zhang and Minear, 2002).
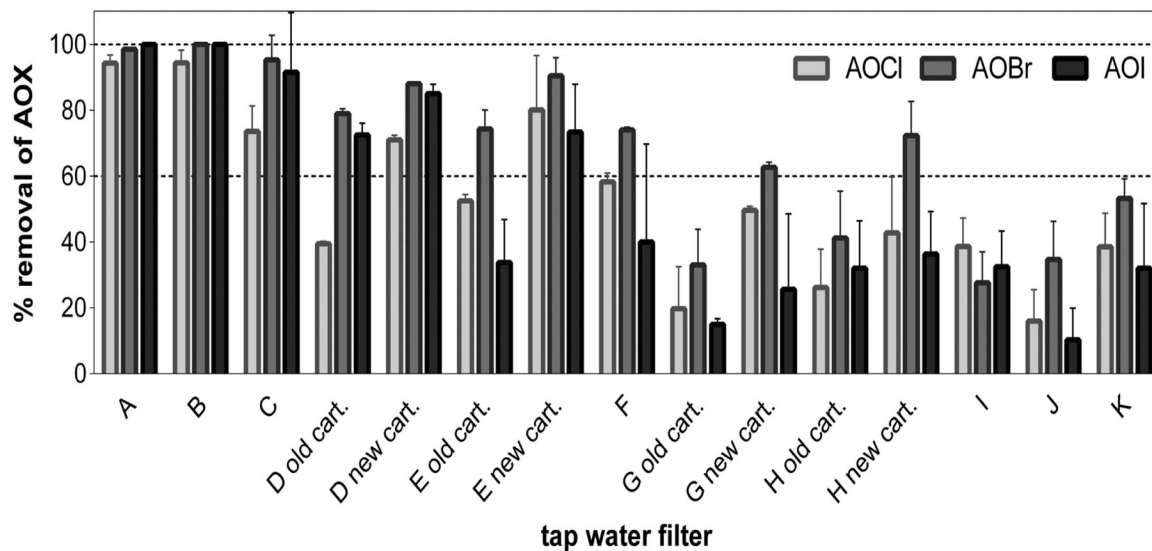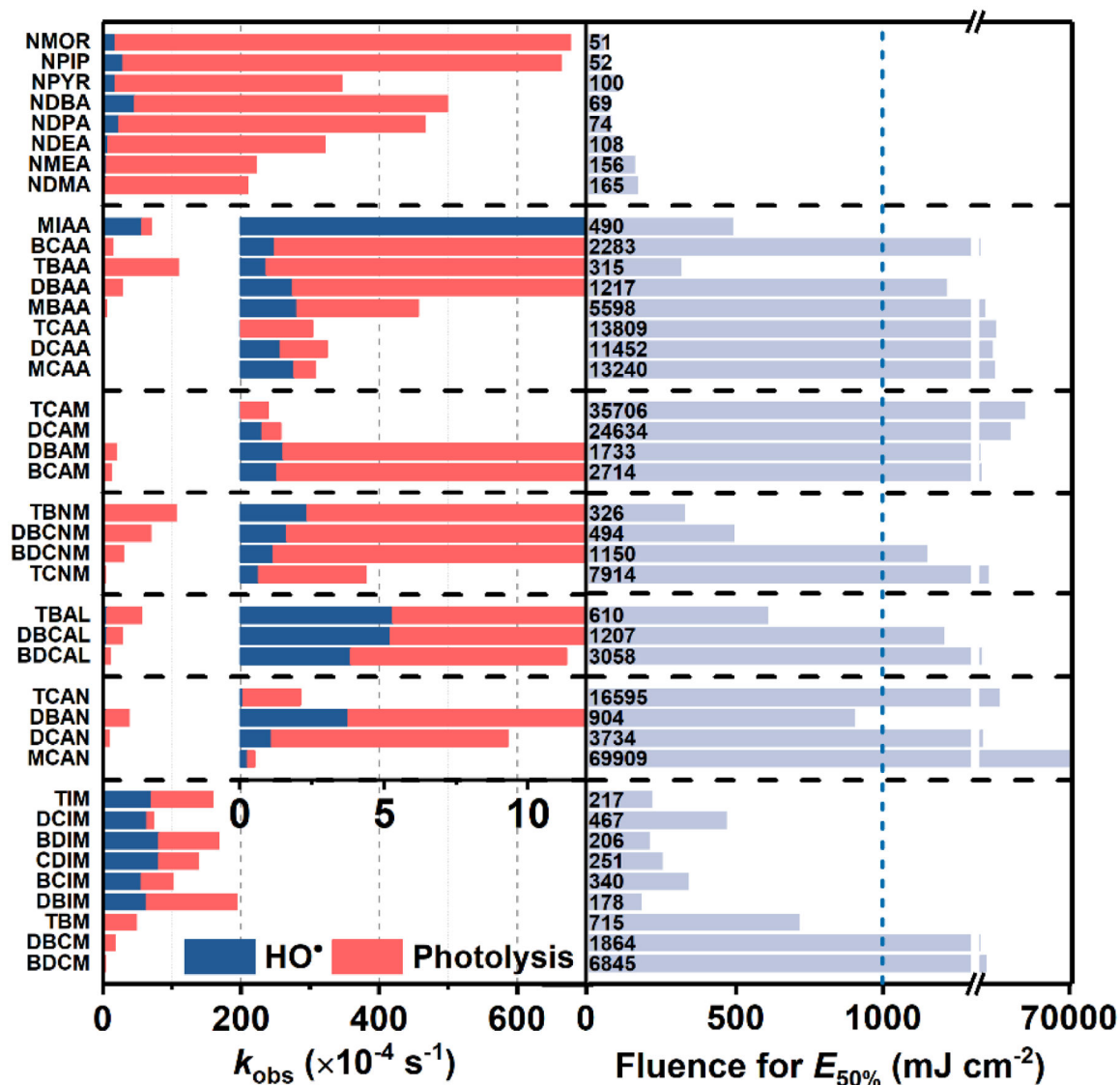
Figure 4.
AOX removal by various water filters (A: reverse osmosis; B: ceramic outer shell with granular activated carbon and activated alumina core; C, E, F: ceramic outer shell with granular activated carbon core; G: activated carbon and ion-exchange resin; D, H, J, K: carbon block; I: activated alumina without carbon). For filters D, E, G, H, results are given before (old cart.) and after filter cartridge replacement (new cart.). Displayed is the arithmetic mean ± standard deviation (n = 2–6) (Reproduced from (Stalter et al., 2016b) with permission from the Royal Society of Chemistry)

Figure 5.
POU treatment effectiveness on DBP removal. (Reproduced from (Xiao et al., 2020) with permission from IWA)
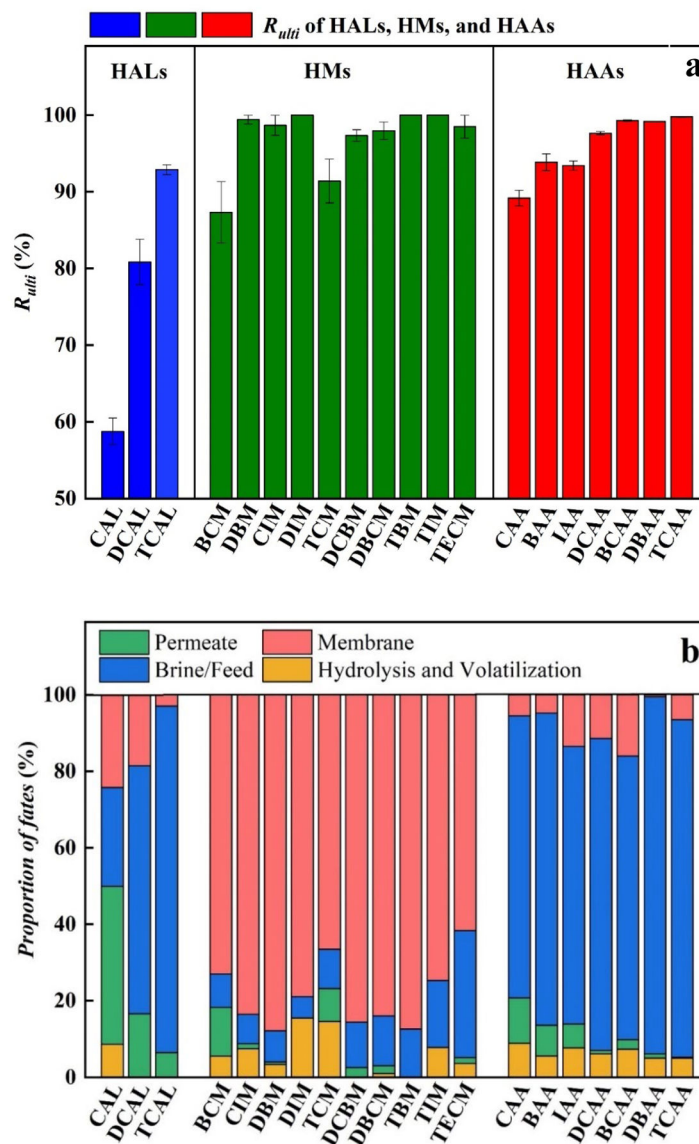
Figure 6.
a) DBP rejection using RO and b) DBP fates during RO operation. (Reproduced from (Chen et al., 2021b) with permission from Elsevier)

Figure 7.
The observed rate constants of DBPs induced by $^\bullet$OH and LPUV direct photolysis in UV/H_2O_2 AOP, and the fluence required to degrade half of the DBP concentration ($E_{50\%}$) when $I_0 = 5.0$ mW cm^{-2}, pH = 7.0, and [$^\bullet$OH]$_{ss}$ = 9.83 × 10^{-13} M. The inset enlarges the bars from MIAA to MCAN. Reproduced from (Lei et al., 2020) with permission from Elsevier)

Table 1.

Knowledge gaps and research needs for DBP control using POU technologies

	Knowledge Gaps		Research needs
1	Unknown DBPs exposure levels to human	•	Monitor types and levels of DBPs in homes before and after POU treatments
		•	Identify DBPs most resistant and toxic under various exposure routes
		•	Examine influence of plumbing materials and system operations on DBPs in homes
		•	Evaluate long-term performance of POU devices
		•	Develop database for DBPs' properties, occurrences, toxicities, and susceptibilities
		•	Design POUs tailored to treat different classes of DBPs
2	Uncertainty in boiling/heating/cooking processes	•	Assess toxicity changes along treatment process
		•	Identify key agents, mechanisms, and intermediate products along process
		•	Test thermal susceptibility of emerging DBPs
		•	Compare performance of various heating devices & develop models to predict their treatability
		•	Quantify emerging DBPs exposures during showering and design POU tailored for it
		•	Evaluate roles of commonly-used household chemicals on DBPs formation or transformation
		•	Evaluate impacts of aqueous disinfectant on food processing
3	Inadequacy in adsorption standard and operation	•	Revisit certification standards to cover more DBPs with varying properties
		•	Assess long-term performance, reusability, and replacement needs of adsorbents
		•	Test performance of commercial adsorbents on broader ranges of emerging DBPs
		•	Design system tailored for treatment of ionized and polar DBPs
4	Lacking data on membrane long-term performance	•	Assess long-term performance, reusability, and replacement needs of membrane
		•	Quantify DBP removals by membranes made of novel materials
		•	Evaluate impacts of water matrix and pretreatment unit on membrane performance
		•	Explore fates of DBPs in both permeate and brine
		•	Design water and energy-saving RO unit
5	Unknown performance and risks for advanced oxidation or reduction processes	•	Assess and balance toxicity changes along treatment process
		•	Identify forcing agents/mechanisms/intermediate products along process
		•	Compare various AOPs & ARPs in POUs on DBPs degradation in different water matrices
		•	Develop and validate QSAR models to predict reaction rate constants of DBPs
6	Lacking tools to monitor DBPs variation	•	Develop easy-to-use bioassay test strips
		•	Develop QSAR models to predict DBPs property/toxicity/susceptibility
		•	Develop models to estimate POU performance
		•	Simplify models and descriptors without compromising reliability
		•	Design easy-to-use online TOX analyzer
		•	Apply advanced equipment to detect new DBPs
7	Concerns from social and regulatory aspects	•	Emphasize the uniqueness of DBPs issue and inevitability of POU in solving the issue

Knowledge Gaps	Research needs
	• Assign holistically the roles of each stakeholders from water source protection to DWTP to POU
	• Evaluate lifecycle costs of POU devices
	• Evaluate long-term and sustained performance of POU on DBPs removal
	• Assess social and economic burdens and benefits if POU is included into water security strategy
	• Develop public-private partnership and set performance and treatment goals