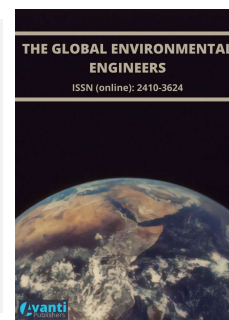




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Technologies for Halide Removal in Water Treatment – A State-of-the-Art Review

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ABSTRACT

Halides (X=Cl, Br, I) are naturally present in water, and halide concentrations can be high in water sources that are impacted by high salinity. Halides are also present in wastewater streams from various industrial operations such as pulp and paper, oil and gas, and mining. Drinking water guideline limits have been established for halides, and halide removal from water is important in several ways. Chloride concentration in water is more related to salinity, and its removal from water matters because of adverse health effects, water scarcity, corrosion, and industrial needs. In drinking water treatment, disinfection is essential to improve water quality and prevent the spread of water born pathogens. However, disinfectants also produce harmful disinfection by-products (DBPs) from precursors such as halides and natural organic matter (NOM) in the source water. Removing halides in the source water before disinfection is a preferred option to increase the disinfection efficiency and avoid forming more toxic DBPs. Some industrial-made isotopes are radioactive and carcinogenic, and iodide produces iodinated DBPs. Bromide removal is important because it produces brominated DBPs. Halides also affect AOPs and can cause more active radicals such as OH[•] and SO₄^{•-} to transform into less active radicals. This paper aims to comprehensively review the sources of halides, the chemistry, and interaction in forming DBPs, current regulatory limits and state-of-art removal technologies available, and their challenges.

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1. Introduction

Halide management is important in water treatment and desalination applications, specifically chloride, the most common precursor for disinfection by-product formation with natural organic matter (NOM). The existence of bromide and iodide during water purification with advanced oxidation processes can also produce hazardous disinfection by-products. Even with the removal of NOM from water, brominate can form, which is related to male infertile [1]. Some iodine isotopes have radioactive properties. This paper aims to provide a comprehensive review of the importance of halide removal, sources of halides in water, the chemistry and interaction in forming DBPs, current regulatory limits, and state-of-art removal technologies available. The removal technologies' effectiveness, advantages, and disadvantages are compared in depth. The challenges of halide removal are also discussed.

1.1. Importance of Halide Removal

1.1.1. Chloride

The majority of water on Earth is seawater (59%) and brackish water (22%) [2]. Seawater desalination for agricultural, domestic, and industrial water purposes has been increasingly used in arid and semi-arid regions such as the Middle East [3, 4], Spain, and Australia [4]. Seawater desalination for irrigation is critically involved with chloride removal because chloride constitutes more than half of the seawater salts. The other ions in seawater, such as Ca^{2+} , Mg^{2+} , and SO_4^{2-} , can be desirable for plant nutrition in agriculture [4]. Excessive chloride has significant negative impacts on crops and infrastructure lifetime. The corrosiveness of chloride ions has a far-reaching negative impact on infrastructure sustainability; chlorides corrode metallic pipes, concrete, electrodes used to extract metals from ores, heat exchangers, and Kraft recovery furnaces, amongst others [5-11]. Without chloride removal, significant operational disruptions and financial damages can occur to industries and communities [5, 12-14]. For example, bridge corrosion by chloride was estimated to cost USD 275.7 billion/per year and USD 123 billion in the USA in 1998 [12] and 2017, respectively [15]. Such infrastructure in contact with seawater or de-icing brines is highly prone to be corroded [16].

1.1.2. Iodide

Iodine can be produced via pharmaceutical industry wastes. Different isotopes of iodine are also produced within the plutonium and uranium fission reactions. Exposure to these iodine isotopes can lead to thyromegaly, leukemia, thyroid cancer, and metabolic disorders [17-20]. Moreover, in the context of nuclear energy systems, it is critical to ensure the safe handling and storage of radioiodine, such as ^{129}I and ^{131}I . Because of the long half-life of iodine (^{129}I = 15.7 million years), it is critical to regulate its discharge to waterways [18, 21] and minimizes human and environmental exposure [19, 20]. The most common chemical forms of I in nuclear wastes are iodide (I^-) and iodate (IO_3^-) [22-24], and iodide concentrations have been recorded to be up to 42 mg/L in leachate from radioactive waste [24].

Until 1998, 2360 kg of ^{129}I was released into seawater by Sellafield Ltd (UK) and at La Hague (France). In addition, from 1944-1972, plutonium production at the Hanford Site (Washington) released 266 kg of ^{129}I into the air [21]. The Chernobyl (Ukraine) and Fukushima (Japan) nuclear disasters released high amounts of iodine into the environment [25-27]. Water released from the Fukushima disaster contained 6.9-52 % ^{131}I . The Chernobyl disaster released 1,760 Petabecquerel (PBq) ^{131}I and other isotopes of iodine, including 1,040 PBq $^{132}\text{Te}/^{132}\text{I}$, 910 PBq ^{133}I , 250 PBq ^{135}I , and 25 PBq ^{134}I [28]. Digestion or inhaling of ^{131}I can result in thyroid diseases such as goiter, autoimmune thyroiditis, and thyroid cancer [25, 29, 30], and iodine concentrations >0.1 mg/L can irritate the eyes and lungs, and cause bronchitis, diarrhea, weight loss, and tremors [31].

Water discharges from mines for iodine [32], uranium [23, 24], gold, and silver [33, 34] contain iodine, and uncontrolled discharges to the environment need to be avoided [35].

Disinfection by-products (DBPs) associated with the chlorination of drinking water treatment contain iodide. Iodinated DBPs are more genotoxic and cytotoxic than chlorinated and brominated DBPs, and strict public health regulations for iodide are therefore in place [36, 37].

1.1.3. Bromide

Bromide affects the taste and odor of water [38]. It also produces brominated DBPs (Br_2 , HOBr, and OBr⁻) when it reacts to natural organic matter (NOM) [37, 38] and halo acetic acids (HAAs) [39]. The toxicity index of brominated DBPs is $>10^4$ times greater than that of chlorinated counterparts. Also, brominated DBPs are more reactive than their chlorinated counterparts (Table 1), aggravating the situation [36]. At elevated bromide concentration (500 $\mu\text{g/L}$), bromoform (CHBr_3) concentrations can be 70-174 $\mu\text{g/L}$ [39].

Bromide and iodide removal prior to disinfection can reduce highly toxic DBPs [38]. Furthermore, the absence of bromide and iodide makes applying other kinds of disinfectant feasible because the oxidation of the iodide/bromide would not be a problem [38]. In addition, water treatment's capital and operational cost would decrease [40].

1.2. Effect of Halides on the Advanced Oxidation Processes (AOPs)

Halides can affect the water treatment processes, including desalination. Two-step desalination with RO became mandatory in Australia to ensure no bromide was left [41]. The most concern of halides in the water treatment processes lay in the advanced oxidation processes.

Halides can affect AOPs by decreasing the activity of free radicals, altering kinetic reactions, and therefore affecting DBP formation. AOP in saline water can be human-made to avoid membrane biofouling in reverse osmosis [42]. Also, in seawater, AOP can happen naturally, in which solar radiation can make hydroxyl ($\text{OH}\cdot$) or dissolved organic matter ($\text{DOM}\cdot$) radicals in seawater. Then these radicals react to halide and make halide radicals, leading to AOP occurring naturally [43].

The water matrix should be considered when applying AOP [44]. AOP in high saline waters such as seawater has a different effect than wastewater, and considerations should be taken [44, 45]. The high chloride content in seawater could decrease the mass transfer of oxidants. This leads to lower disinfectant concentration in seawater than in drinking water [44]. Also, high salinity in seawater can change the solubility of gaseous oxidants. The effect is called the salt-in and salt-out effect [46]. Chloride accelerates AOPs (mostly from $\text{OH}\cdot$ and $\text{SO}_4\cdot^-$) but forms toxic chlorinated intermediates [47]. It suppresses mineralization rates and accumulates absorbable organic halides (AOXs) [48].

Elevated halide concentrations inhibit the degradation of free radicals acting as radical scavengers above a certain level [45]. Degradation of acid orange 7, benzoic acid, phenol, and coumarin with persulfate does not inhibit 0-0.2 mM bromide [45]. An increase of halide concentration leads to a change in the more active radicals ($\text{OH}\cdot$, $\text{SO}_4\cdot^-$, $\text{Cl}\cdot$) to less active radicals ($\text{Br}_2\cdot^-$, $\text{Cl}_2\cdot^-$, $\text{BrCl}\cdot^-$) [49-51], which may degrade more slowly, but the lower rate radical loss lead to reach higher radical concentrations [49, 50, 52, 53].

The effect of halide on various disinfectant materials of persulfate, hydrogen peroxide, UV, and Cl_2/HOCl is explained here.

Persulfate is one of the oxidants being used for disinfection. Persulfate is not suitable for advanced oxidation in highly saline water [47], especially for water with low pH [47, 54], because high chloride concentrations increase AOXs, which leads to increases in chlorinated DBPs and acute toxicity [47]. Unless halide concentrations in water are small, halide reduces persulfate removal efficiency [55-65]. For example, low concentrations of chloride and bromide enhance the degradation efficiency of AO7 with persulfate [54]. $\text{Cl}_2\cdot^-$ is the primary oxidant of acid orange 7 (AO7) in the presence of chloride [45, 54], while $\text{SO}_4\cdot^-$, $\text{Br}\cdot$, and $\text{Br}_2\cdot^-$ are primary oxidants for AO7 degradation with persulfate [45] in the presence of bromide. In acidic conditions, $\text{Cl}\cdot$ is formed from advanced persulfate oxidation, but $\text{Cl}_2\cdot^-$ is formed at neutral pH and plays a vital role in the degradation of AO7. In alkaline conditions, $\text{ClOH}\cdot$ is formed, which is not stable and is converted to $\text{OH}\cdot$, which also plays a vital role in the degradation of AO7 [54].

Hydrogen peroxide form bromate based on OH radical mechanism. Hydrogen peroxide reacts with bromide forming $\text{BrO}_2\cdot$ which reacts with hydroxyl radical, forming bromate [66]. However, at very high bromide

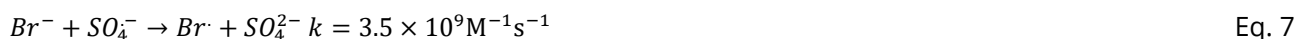
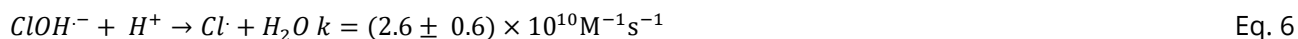
concentrations (e.g., 158 mg/L), OH radicals are scavenged (>90%) by halides to produce halide radicals [44, 67-69], and no bromate is formed [44]. The increases in concentrations of Br₂^{•-} and BrCl^{•-} are almost 3-4 times higher than concentrations of OH[•] [49, 52, 70]. Bromide in seawater is a scavenger for hydroxyl radicals, while chloride is not. Because bromide reacts with hydroxyl radicals to form BrOH^{•-}, which is transformed into BrCl^{•-} (efficiency 73%) and Br[•] (efficiency 3%). When chloride reacts with hydroxyl radicals, it forms ClOH^{•-}. However, it is not stable, and 99.98% of it converts to its initial form of OH[•] and Cl[•]. Bromide plays a key role in scavenger hydroxyl radicals [67, 71].

High salinity can affect the AOP and disinfection efficacy where UV radiation is used for disinfection because of decreases in the optical pathway of irradiation [44]. Alternatively, UV is the best disinfection method in high saline water because UV quenches chlorine and does not produce DBPs [42]. However, chloride has a negative impact on the H₂O₂/UV [72] and UV/TiO₂ advanced oxidations [73].

Ozone reacts with bromide in seawater to produce OBr^{•-} and HOBr at neutral pH values [74, 75]. OBr^{•-} and HOBr are transformed into BrO₃^{•-} in nonacidic conditions and low ammonia concentrations. Bromate is classified as carcinogenic by WHO and USEPA [42] and is the main reason for DBP formation [66].

Disinfectants chlorine and HOCl convert to each other depending on the pH aqueous. The application of chlorine and HOCl should be considered carefully when applying in different water matrices (e.g., bromide). With the application of chlorine in drinking water, chloroform is the primary concern for DBP formation, while in seawater, bromoform is more dominant than chloroform [44]. Chloroform forms with the application of chlorine for drinking water disinfection, while chlorine application in seawater forms bromoform majorly than chloroform [44]. Another concern in seawater with the application of chlorine is bromate formation. Because in seawater, chlorine oxidizes bromine to form BrO^{•-} and HOBr. This causes bromate and DBPs formation [44]. One of the examples is brackish water (with 35 mg/L Br₂), in which 85-100% of THMs formed contain bromine [44]. AOP with HOCl affects by the presence of bromide in the water matrix. For example, the reactivity changes with bromide in the AOP process with UV/HOCl [50, 53]. HOCl transforms to HOBr, and ClBr^{•-} and loses the hydroxyl radical [53], and with HOCl conversion to HOBr, the degradation of organic substrates increases [50, 53].

In saline waters, OH[•] and SO₄^{•-} scavenge the chloride and bromide. General reactions of halides that can inhibit or enhance the AOP are presented below (Eq. 1-Eq. 7) [52, 54, 73, 76, 77].



Zhang and Parker (2018) have developed a comprehensive computational modeling procedure for the kinetics of halide radicals [43]. The reactivity of halogen radicals with different organic substrates is shown in Table 1.

2. Sources of Halides in Water

2.1. Chloride

Seawater intrusion (3-3.8% NaCl) and de-icing of roads (20-30% NaCl/CaCl₂ de-icing brines) are the primary sources of chloride release into water and damage the infrastructures by their corrosion [16]. Other sources of

Table 1: Kinetic rate of X radicals and HOX/OX[•] with organic materials. X=Cl, Br, I.

	k ($M^{-1} s^{-1}$)	Reference
	Saturated aliphatic compounds	
Cl [•]	10^7 - 10^9	[78-81]
Cl ₂ ^{•-}	10^3 - 10^7	[82, 83]
Br [•]	10^4 - 10^8	[83]
Br ₂ ^{•-}	Below 10^6	[43]
I ₂ ^{•-}	Below 10^6	[43]
	Olefinic compounds	
All halogen radicals except I [•] and I ₂ ^{•-}	10^6 - 10^9	[83-85]
	Aromatic compounds	
Halogen radicals	$< 10^5$ - 10^{10}	[82-84, 86-90]
	Organosulfur species	
Halogen radicals	$< 10^6$ - 10^{10}	[82, 88, 91-93]
	organic substrates	
HOCl and HOBr rate constants	$< 10^{-2}$ - 10^7	[94-96]
OCl [•] , OBr [•]	Reacts slowly 10^{-2} - 10^7	
	Aromatic	
HOCl and HOBr	10^{-2} - 10^7	
	Amino substrate	
	10^1 - 10^6	
	Olefinic substrate	
	$\sim 10^0$ - 10^6	
	Organosulfur substrate	
	10^5 - 10^8	

chloride are the kraft pulping process which turns wood into wood pulp. Chloride comes from electrostatic precipitator catch or dust (ESP dust), which consists of 1-10% wt chloride [97]. Chloride would be released from ESP because chlorine is one of the process elements which enters the kraft pulping process with wood and other chemicals and will accumulate [98]. The boiler burns the black liquor containing organic and inorganic materials such as chloride [99]. Chloride concentration from the inland mill is 0.1-0.8% of the liquor dry solid, while for the coastal mill, chloride concentration could be as high as 3-5%, and in the closed process, chloride concentration could be even higher [99]. Mining water also could contain high chloride concentrations, depending on the geological location, dissolved metals, and deposit characterizations in contact with water. The mines that release a large quantity of chloride into water are radio-nucleolus uranium and coal mines [100]. However, they have a different chemistry from each other. In particular, coal mine water is usually neutral, with an elevated TDS, high electrical conductivity, and high chloride concentration, which will turn into saline mine water [100]. Other than that, if the water is in contact with the tailings containing chloride, called tailing water, it would have high chloride concentrations due to the leaching of chloride into water [100]. For example, Polish mine water drainage contributed 6,500 tonnes of chloride in the Wisla and Odra rivers, which disqualified the water from being suitable for drinking or agriculture. Also, this leads to increased corrosion of pipes [101]. Most of Upper Hunter Valley's open pits have saline mine water (chloride concentration from 3100-6580 mg/L), used chiefly for coal washing and dust suppression [102]. In Kent Coalfield (UK), chloride concentration was 318,000 tonnes (up to 5000 mg/L) in

mine drainage water [103]. If not addressed, this degree of salination caused by mined water would result in heavy penalties for the mining industries [101]. The produced water (the resultant water after the oil and gas activities) also contains high chloride concentrations [104-106]. Chloride concentration in produced water could reach 250,000 mg/L [105]. For example, chloride concentrations in the water produced from Louisiana and Texas platforms were reported to have 37,000-110,000 and 28,000-73,000 mg/L, respectively [107]. Josephine Brine Treatment in Pennsylvania had 80542 mg/L of chloride in the effluent. Moreover, the chloride concentration in Blacklick Creek was 55 000 and 98 000 mg/L [108].

The produced water from the extraction of shale gas (SG) and tight gas (TG), or conventional natural gas, is saline (>30 g/L chloride) and hypersaline (> 40 g/L chloride), respectively. Wastewater from hydraulic fracturing during oil and gas extraction contains very high levels of halide concentration and requires treatment processes precisely to remove halides to avoid toxic DBP formation [109]. However, coalbed methane (CBM) produced waters have less than 30 mg/L chloride ions [104]. Chloride concentration in the produced water from the oil field, gas field, steam gravity drainage (SAGD), and coal gas steam (CGS) reaches 152,750 mg/L, 2,300 mg/L, 4,800 mg/L, and 3,100 mg/L, respectively [110]. The coal seam water has a high concentration of sodium, chloride, and bicarbonate (salinity 1,500-10,000 mg/L) [111, 112]. Australian (QLD) coal seam water contains more chloride (500-2000 mg/L) than bicarbonate [111, 113]. The coal seam water in Queensland, Australia, has < 5910 mg/L chloride [114], whereas in New Zealand and the USA chloride concentration is lower, in the range of 49.3-146 mg/L [114]. Chloride concentrations in produced water and some coal seam gas water are presented in Table 2. Salt concentration becomes more critical when oil/gas operations occur onshore [105]. At a higher amount of chloride, corrosion and fracture in reactors and vessels will happen. Therefore, keeping chloride concentration as low as possible is necessary for feeding oil [115]. The offshore produced water will be released into the sea, which might endanger aquatic life [105]. If produced, water leaks to surface water could damage the ecosystem drastically [113].

Table 2: Chloride concentration in produced and seam gas water [104, 114, 116].

Produced Water (Coalbed Methane)		Produced Water (Shale Gas)				Reference
min	max	Min	Max			
0.7	70100	48.9	212700			[104]
Tight Gas		Conventional Natural Gas		Flowback Water (Marcellus Shale)		
min	max	Min	Max	min	max	
52	216000	1400	190000	32000	148000	[104]
Seam Gas Water						
Southern tenements (Surat) -WB - 56 wells		Moranbah area (Bowen) -WB - Blackwater Group coal formation - 58 wells				
min	max	Min	Max			
28	5870	1151	4500			[116]
Coal Seam Gas, Surat Basin, Queensland						
Field A (54 wells)		Field B (73 wells)		Field C (23 wells)		
min	max	Min	Max	min	max	
471	4390	875	2930	823	5910	[114]

2.2. Iodide

Human activities also can lead to iodine in the water, such as the liquid crystal display (LCD) polarizing films industry, which could release very high amounts of iodine/iodide in their wastewater, such as 10.2 g/L or (1-1.5 wt%) of I⁻ [20, 117, 118], with the flow rate of 100-300 m³/day [119]. Typical wastewater treatment plants cannot

treat the wastewater produced by the TFT-LCD industry due to the possibility of a biocidal effect on microorganisms [117, 118]. Nuclear fission is the other way that iodine (mostly iodide and iodate) enters the water [22, 118, 120]. Moreover, mining activities and produced water from gas production could also cause the release of iodine into waterways [31, 117, 121, 122]. A small spill of shale gas wastewater increased iodide up to 28 mg/L in the surface water [123]. Iodine was found in the Oko uranium deposit in Gabon. As a result of the fission reaction, the heavy elements (Kr, Xe, Rb, Cs, Sr, Ba, Mo) and I are very mobile [124]. The migration of ^{129}I in four uranium ore deposits of the Alligator Rovers region, Northern Territory, Australia, was reported by Fabryka-Martin *et al.* [125]. The weathering of the deposits contains iodine. In the case of Alligator Rovers region deposits, the -FeOH groups in ores, such as goethite FeO(OH), were likely the groups where iodide sorption occurred. The weathering of deposits resulted in a robust partitioning of ^{129}I in the groundwater, which depends on the distance to the ore place [125]. These iodine extractions lead to water contamination for the people who live near the mining site, which urged the local population to use the desalination plant treatment [122]. Another example is Southeastern Nigeria's abandoned mine sites, where iodide concentrations were found at 0.013-0.05 mg/L [126].

The last but not least way of entering iodine into waterways through human activity is the chemical wastes of x-ray, disinfection, and medical (nuclear medicine) dumps, because of iodine application in these industries [22, 120].

2.3. Bromide

Today, bromine is used in flame retardants [127] and biocide in various waters, including pool water, cooling water, industrial water treatment [127], and photo industries [127]. All of these industries would release bromide into the waters. Road salts and some fertilizers also bring bromide into water [128]. Textile industries that produce flameproof fabrics use brominated compounds, which would lead to bromide release into water [128].

Scrubbers in coal-fired power plants [128, 129], incinerations [130], mining activities [131], and gas extractions industries, including hydraulic fracturing in natural gas extraction, release a massive amount of brines containing a high concentration of halides [108, 109, 123, 127].

Coal-fired power plants release bromide into their wastewater within 2 pathways. One is coal that has bromide from 1-2 mg/L in sub-bituminous coal to bituminous coal with 20 mg/L [132]. Secondly, bromide salt is added to the flu gas to make mercury more soluble and more accessible to be removed. However, in the end, bromide would accumulate into wastewater of flue gas. Therefore, for making 1 MW of electricity, 0.23-2.8kg/day of bromide is released into the wastewater [128]. A coal fire plant has increased bromide in the Allegheny River-Pennsylvania up to 410 $\mu\text{g/L}$ [133].

In Switzerland, the chemical industry (50%) and municipal waste incarnation (20%) contributed most to bromide release into water [134].

Mining water from potash deposits in Saskatchewan, Canada, showed 28500 mg/L bromide [131].

Gas extractions produce flow backs and produce waters that are highly brine, and their bromide concentration would be very high. Josephine facility in Pennsylvania USA, a brine facility in Pennsylvania for hydraulic fracturing water treatment, showed had 601- 8,290 mg/L bromide with a flow rate of 155,000 gal/day [128]. The oil and gas industry (produced water) can release a significant amount of bromide into the water, such as 3070 mg/L in USA shale [131].

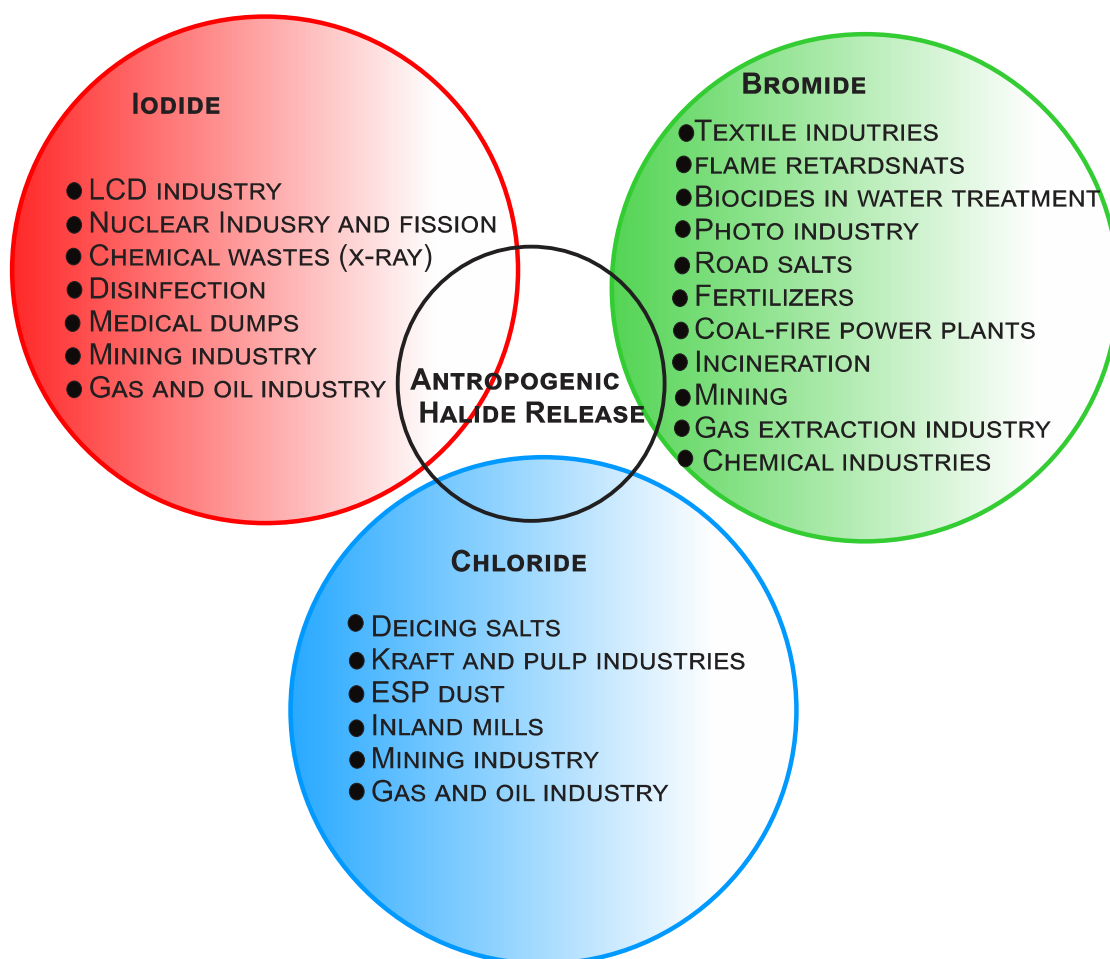
When bromide concentration is less than 0.1 mg/L, it was shown that the number of DBPs produced would be considerably lower [135]. Nonetheless, even bromide concentration from 50-100 $\mu\text{g/L}$ would lead to the formation of hazardous DBP [136].

The summary of bromide concentration in various water substrates has been presented (Table 3) [37, 40, 136-141].

Human activities that lead to excess halide release into the environment have been summarized below (Fig. 1).

Table 3: Bromide concentration in various water substrates.

Substrate	Bromide Concentration	Additional Information	Reference
Natural Waters	100–3000 µg/L	Br: I 10:1, Cl: Br 100-200	[37, 138-140]
Brackish Water (study)	1, 5, 10 mg/L		[136]
Freshwater	0.5 mg/L		[137]
Sea Water	65-80 mg/L	Cl: 18980 mg/L to over 23000 mg/L	[141]
Desalinated Water	1 mg/L		[137]
Moderate Bromide Concentration	0.5-1.2 mg/L		[40]

**Figure 1:** Summary of anthropogenic activities that lead to halide release into the environment.

3. Chemistry of Halides

Iodine to iodide conversion requires +0.54 V (standard reduction potential), while Cl_2/Cl^- standard reduction is +1.36 V, and Br_2/Br^- has the standard reduction potential of 1.09 V (Eq. 8, Eq. 9, Eq. 10). Also, the first ionization energy for Cl, Br, and I are 1251, 1140 kJ, and 1008 kJ/mol, respectively. This depicts that oxidizing power follows $\text{Cl}_2 > \text{Br}_2 > \text{I}_2$ [31].

The oxidation power of halogen radicals ($\text{X}\cdot$) gets higher than halogen gas (X_2). Reduction potentials for halide radicals are as follows $\text{Cl}\cdot$ (2.5 V), $\text{Cl}_2\cdot^-$ (2.2 V), $\text{Br}\cdot$ (2 V), $\text{Br}_2\cdot^-$ (1.7 V), $\text{I}\cdot$ (1.4 V), $\text{I}_2\cdot^-$ (1.1 V) [142].



Halogen gas ($X_2=Cl_2, Br_2, I_2$) reacts with water, produces hypohalous acid and halide, and makes the water acidic. Previously, the rate constants of halogen gas dissolution in water and other related halides reaction had been calculated (Table 4). Halogen gas reaction with water undergoes the reaction shown in Table 4 and Fig. 2 [118, 143, 144].

Table 4: Halid reactions and their rate constants.

Reaction Number	Reaction	K	Reference
		Rate Constant $M^{-1}s^{-1}$	
1	$H^+ + OCl^- \rightarrow HOCl$	5.0×10^{10}	[145]
2	$HOCl \rightarrow H^+ + OCl^-$	1.6×10^3	
3	$Cl_2 + Cl^- \rightarrow Cl_3^-$	2.0×10^4	[146]
4	$Cl_3^- \rightarrow Cl_2 + Cl^-$	1.1×10^5	
5	$Cl_2 + H_2O \rightarrow Cl^- + HOCl + H^+$	22.3	[143]
6	$Cl^- + HOCl + H^+ \rightarrow Cl_2 + H_2O$	18.2 ± 0.7	
7	$H^+ + BrO^- \rightarrow HBrO$	5.0×10^{10}	[145]
8	$HBrO \rightarrow H^+ + BrO^-$	7.9×10^1	
9	$Br_2 + Br^- \rightarrow Br_3^-$	9.6×10^8	[146]
10	$Br_3^- \rightarrow Br_2 + Br^-$	5.5×10^7	
11	$HBrO + Br^- \rightarrow Br_2OH^-$	5×10^9	[147]
12	$Br_2 + H_2O \rightarrow Br_2OH^- + H^+$	97 ± 7	[144]
13	$Br_2OH^- + H^+ \rightarrow Br_2 + H_2O$	2×10^{10}	[147]
14	$Br_2OH^- \rightarrow HOBr + Br^-$	5×10^9	
15	$I_2 + H_2O \leftrightarrow I^- + H^+ + IOH$	4.3×10^{-13}	
16	$I_2 + I^- \leftrightarrow I_3^-$	830	
17	$IOH \leftrightarrow H^+ + IO^-$	2.3×10^{-11}	
18	$IO^- + I^- + H_2O \leftrightarrow I_2OH^- + OH^-$	0.13	
19	$H_2O + I_2 \leftrightarrow H_2O \cdot I^+ + I^-$	1.2×10^{-11}	
20	$I_2 + H_2O \rightarrow OI^- + 2H^+ + I^-$	2.0×10^{-13}	
21	$I_2 + 2OH^- \rightarrow I^- + H_2O$	30	
22	$3OI^- \rightarrow 2I^- + 3IO_3^-$	10^{20}	

As mentioned above, in the halogen group going down from chlorine to iodine, the halogens' oxidative activity would reduce [31], but OX^-/HOX ($X=Cl, Br, I$) acts differently. For example, the rate constant of HOBr with halophenolates and phenolate is 3000 times more than the reaction of these organic materials to HOCl [95].

Chloride is highly soluble under all pH and E_h values unless high voltage is being applied to oxidize chloride to ClO_4^- even in the case of ClO_4^- would still be soluble. Therefore, it shows that removing chloride would be a challenge due to its high solubility [148].

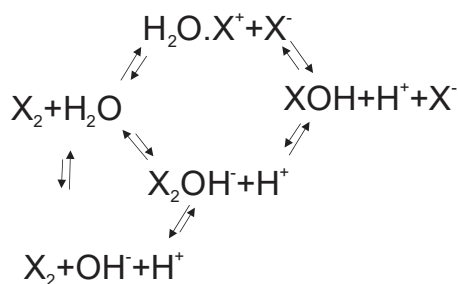


Figure 2: Halogen gas reaction with water [147].

Iodate and iodine are prevalent in low pH and high E_h values. In other conditions, iodide is common [149]. However, iodine in water will react and produce iodide and hypiodite (OI^-) [118]. Likewise chloride, bromide is also very soluble in various conditions of E_h -pH [148]. Therefore, bromide removal is also challenging because of its high solubility.

4. Limits and Regulations

A summary of halide concentrations in different water applications has been provided Table 5. More regulations are set for chloride than for the rest of the halides. Unfortunately, no regulations have been set for bromide in water. Because bromide is not considered toxic, industries do not need to report its concentration [128]. However, the safe concentration of bromide in water by various studies has been indicated (Table 5). Australian water regulation suggested having a bromide concentration below 0.1 mg/L because of bromate formation in the oxidation process. Specifically, they suggested that if bromide is less than 0.1 mg/L in water, it would contain less than 0.02 mg/L bromate, with Australian regulation alignment. However, they did not set any

Table 5: Various levels of halides regulations and suggestions for different applications.

-	Concentration	Regulated Organization	Comments	Ref.
Chloride	250 mg/L	Australian guidelines and WHO	drinking water	[150-152]
	500 mg/L	IEPA	surface water	[153]
	94 mg/L	NA.	below risk limits for the ecosystem	[154]
	< 105 mg/L	NA.	Water for agricultural application	[3]
	2000 mg/L	Qld. regulation	livestock consumption	[114]
	< 0.4% by mass of cement in acid-soluble, <0.15% by mass of cement in water-soluble environments	-	reinforced concretes in Europe and North America	[7]
Bromide	< 60 μ g/L	NA.	Low risk	[40]
	60-500 μ g/L	NA.	Moderate risk	
	> 500 μ g/L	NA.	High risk	
	< 50 μ g/L	NA.	Low risk	[38]
	110 μ g/L	NA.	Moderate risk	
	76-540 μ g/L	NA.	Moderate to high risk	
	~700 μ g/L	NA.	Very high v	
	> 100 μ g/L	NA.	Will cause bromate	[150]
Iodide/Iodine	4-18 μ g/L (Iodine)	WHO	USA drinking water	[155, 156]
	0.5 mg/L (Iodide)	Australian guidelines	drinking water	[150]

direct regulations for bromide itself [150]. When bromide concentration increases by 50 µg/L, the bladder cancer risk would increase by 10^{-4} - 10^{-3} due to THM formation [129].

The effect of halide on DBPs formation is significant. A summary that shows regulated DBPs with their risks is shown in Table 6. This signifies why halide removal matters.

Table 6: Summary of regulated DBPs during and their health issues.

Class of DBPs		Compounds	Health Effects	Occurrence	Genotoxicity	Carcinogenicity	WHO Guideline Value µg/L	USEPA µg/L	Australia µg/L	Ref.
Total THM	Trihalomethanes (THM)	Chloroform	Cancer, liver, kidney, and reproductive effects	low- to mid µg/L	-	+	200	Total THM 80	Total THM 250	[157-159]
		Dibromochloromethane	The nervous system, liver, kidney, and reproductive effects				60			[157-159]
		Bromodichloromethane	Cancer, liver, kidney, and reproductive effects	low µg/L	+	+	60			[157-159]
		Bromoform	Cancer, liver, kidney, and reproductive effects	low µg/L	+	+	100			[157, 159]
Trihalomethanes (THM)	Chlorodibromomethane	chromosomal aberrations or sister chromatid exchanges, hepatocellular tumors	low µg/L	+	+	100		-	[158, 160]	
Haloacetonitrile (HAN)	Trichloroacetonitrile	Cancer, mutagenic and clastogenic effects	sub- to low-µg/L levels	+					[157]	
	Dichloroacetonitrile	No data yet	sub- to low-µg/L levels	+		20			[158].	
	Dibromoacetonitrile	No data yet	sub- to low-µg/L levels	+		70			[158].	
Halogenated aldehydes and ketones	Formaldehyde	Mutagenic	sub- to low-µg/L levels	+	+	900		500	[157-159]	
Haloacetic acids (HAA)	Dibromoacetic acid	hepatocellular tumors, lung tumors, mesothelioma, Leukaemia	low- to mid µg/L	+	+		Total HAAs 60			[158, 161]
	Bromoacetic acid	No cancer studies performed	sub- to low-µg/L levels	+						[158, 159]
	Chloroacetic acid		sub- to low-µg/L levels	+	-			150		[158, 159]
	Dichloroacetic acid	Cancer and reproductive and developmental effects	low- to mid µg/L	+	+	50		100		[157-159]
	Trichloroacetic acid	Liver, kidney, spleen, and developmental effects	low- to mid µg/L	-	+	100		100		[157, 159]
Halocarbonyl compounds	Chloral hydrate	Skin allergy, carcinogenic,	low µg/L	+	+	10		100	[157-159, 162]	
Oxyhalides	Bromate	Renal tumors	sub- to low-µg/L levels	+	+	10	10	20	[158, 159, 163-166]	

5. Technologies for Removing Halides

5.1. Chloride

There are various technologies for removing chloride (desalination). Selection of a suitable process for chloride removal depends on the ease of operation, its effect on the environment/plant location, the amount of investment, and the cost of maintenance and operations [10, 167]. Various methods have been employed for chloride removals, such as chemical precipitation, electrochemical remediation technology, membrane technologies (non-thermal like RO and thermal like MD), adsorption, and ion exchange.

Chemical/electrochemical precipitation with UHLA produces insoluble minerals of double-layered Friedel salt. The disadvantage is that existing of other anions, including sulfate, decreases chloride removal efficacy [168-170]. Chloride precipitation with silver nitrate is expensive because the silver loss in this process is inevitable [171]. Produced waters with high chloride concentrations will use RO and thermally driven technologies such as MD for chloride removal [104]. ZDL will be used to recover brine for zero discharge pollutants [105].

Primary desalination treatments are happening with RO at 58.65%, MSF at 26.95%, and MED at 9.51% (Fig. 3). Maximum capacity of various technologies' desalination is shown in Fig. (3 and 4). Data related to the graphs are presented in Table 7 and Table 7. The CAPEX and OPEX of leading technologies for a typical treatment capacity are shown in Fig. (5). Here, we separated RO, MSF, MED, and ED as major desalination technologies and the rest as non-major desalination technologies. This category was made based on the technologies' capacities. From Fig. (5), RO has the highest OPEX and lowest CAPEX. Nonetheless, MED and MSF have higher CAPEX and lower OPEX. Therefore, selecting the technology should be based on preference and considering which outweighs the OPEX or CAPEX. Unfortunately, information about ED CAPEX was not found. On the other hand, the CAPEX of those technologies (MSF and MED) are higher than RO. CAPEX of RO is 1313, MSF is 1598, and MED is 1,860 US \$/m³/day (Fig. 5a). OPEX of RO could be varied from 0.2-12.99 US\$/m³, while other technologies costs are less comparatively. For example, MSF 0.2-1.75, MED 1.36-1.6, ED 0.015-1.05 US \$/m³ (Fig. 5b).

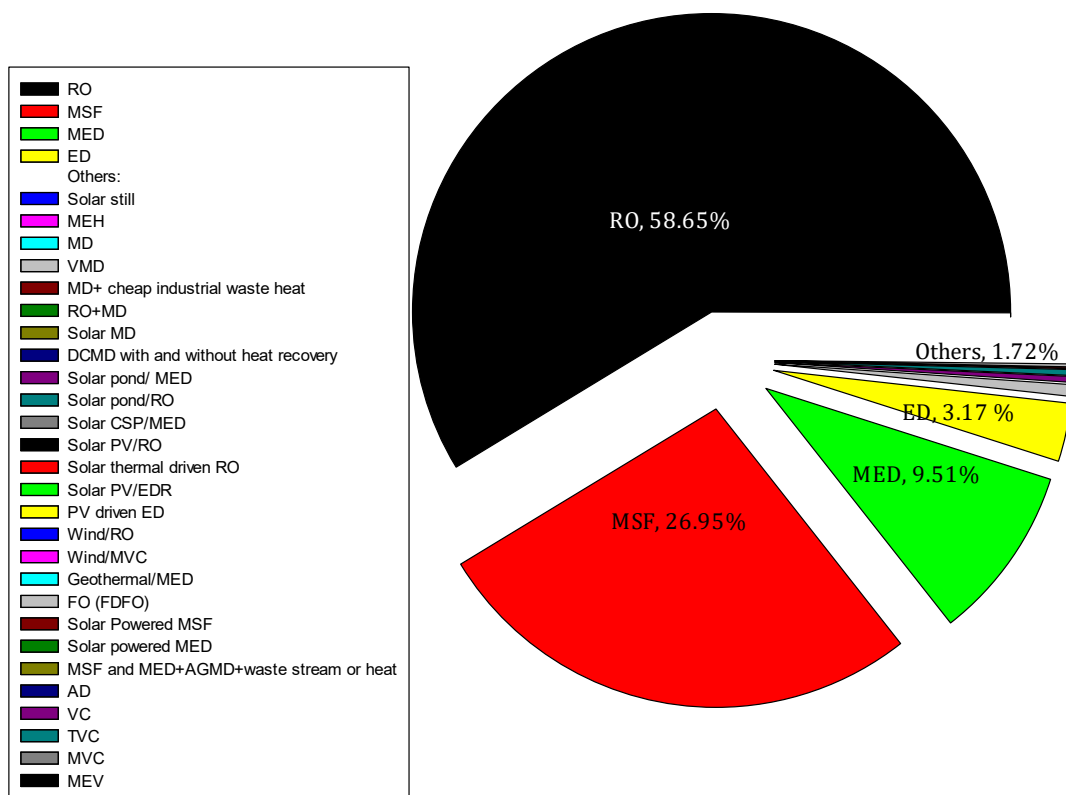


Figure 3: Maximum capacity of desalination plants details of data are presented in Table 8.

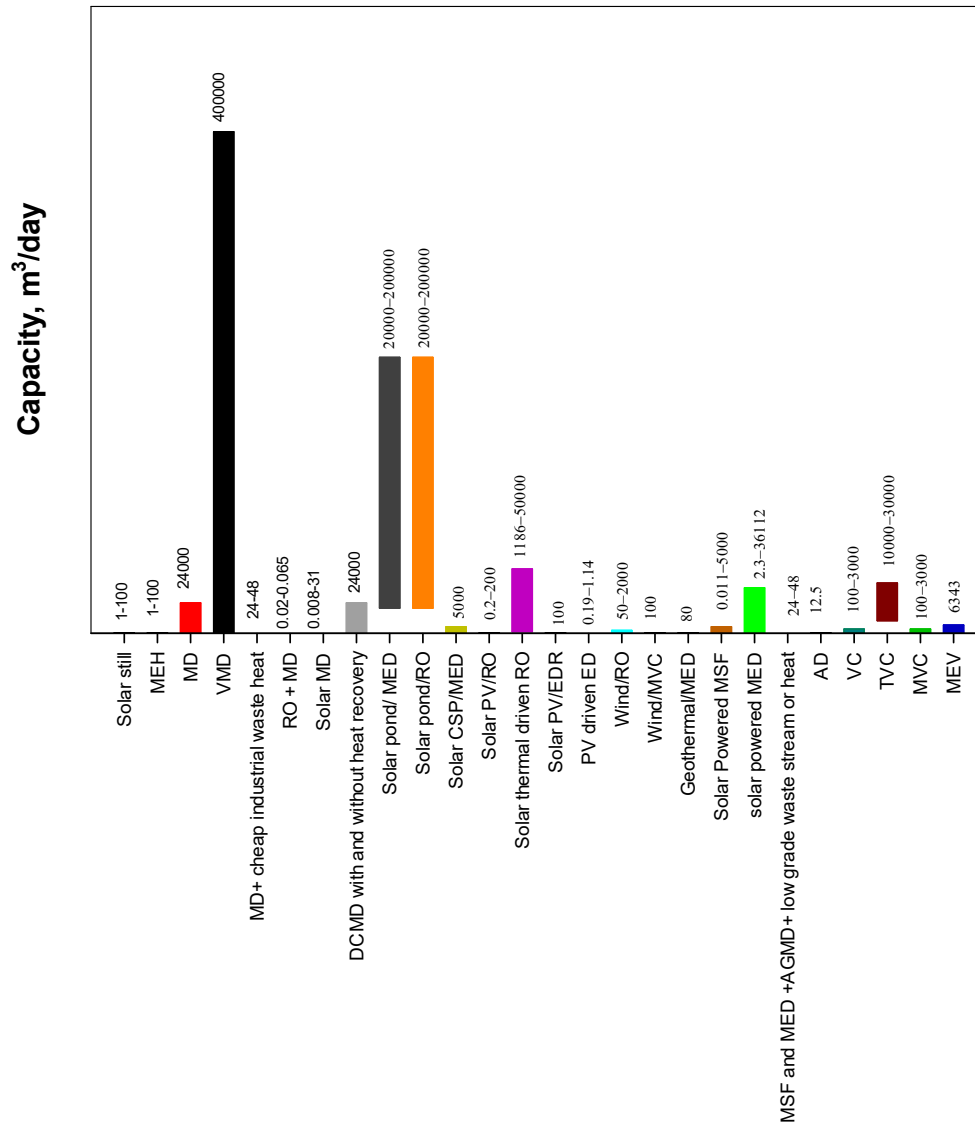


Figure 4: Capacities of other technologies (non-major technologies) for desalination/ chloride removal-related data are presented in Table 7.

Fig. (6) [172-185] presents desalination costs for significant technologies Fig. 5. Non-major desalination technologies costs are shown in Figure 6) [172, 174, 177, 178, 183, 184, 186-192]. Non-significant technologies that are hybrids of other technologies have less capacity than the major technologies; therefore, their applications are less than the major ones. They can be used in small-scale applications and are cheaper than the major technologies.

Due to the high importance of the leading desalination technologies (RO, MSF, MED, and ED), the amount of their energy consumption is presented in Table 8 [172, 174, 175, 179, 185, 186]. Some of these technologies work with electrical energy, and some need electrical and thermal energy. For example, RO and ED work with electrical energy, while MSF and MED work with electrical and thermal energy (more thermal basis than electrical basis energy consumer) (Table 8).

Adsorption and CDI are considered cheap technologies for chloride removal at low concentrations. The capacity adsorbents and CDI are presented (Table 9 and Fig. 7).

Table 7: Maximum capacity of desalination plants.

Desalination Plant	Capacity	Reference
RO	37 million m ³ /day	[174]
MSF	17 million m ³ /day	
MED	6 million m ³ /day	
ED	2 million m ³ /day	
Solar still	100 m ³ /day	[172]
MEH	100 m ³ /day	[183]
MD	24000 m ³ /day	
VMD	400000 m ³ /day	[175]
MD+ cheap industrial waste heat	48 m ³ /day	[187]
RO+MD	0.065 m ³ /day	[193]
Solar MD	31 m ³ /day	[189]
DCMD with and without heat recovery	24000 m ³ /day	[183]
Solar pond/ MED	200000 m ³ /day	[172]
Solar pond/RO	200000 m ³ /day	
Solar CSP/MED	5000 m ³ /day	
Solar PV/RO	200 m ³ /day	[189]
Solar thermal driven RO	50000 m ³ /day	
Solar PV/EDR	100 m ³ /day	[172]
PV driven ED	1.14 m ³ /day	[189]
Wind/RO	2000 m ³ /day	[172]
Wind/MVC	100 m ³ /day	
Geothermal/MED	80 m ³ /day	
FO (FDFO)	100000 m ³ /day	[184]
Solar Powered MSF	5000 m ³ /day	[189]
Solar powered MED	36112 m ³ /day	
MSF and MED +AGMD+ low-grade waste stream or heat	48 m ³ /day	[187]
AD	12.5 m ³ /day	[174]
VC	3000 m ³ /day	[172]
TVC	30000 m ³ /day	
MVC	3000 m ³ /day	
ME	6343 m ³ /day	[177]

Table 8: Main technologies used for desalination and their energy consumption.

Technology	Electrical Energy Needed (kWh/m ³)	Thermal Energy Needed (kWh/m ³)	Total Energy Needed (kWh/m ³)
RO	0.5-10	0	14.29
MSF	2.5-5	15.83-78.3	19.58-57.14
MED	2-21.5	40.3-63.9	43.21
ED	0.7-8	0	0.7-8

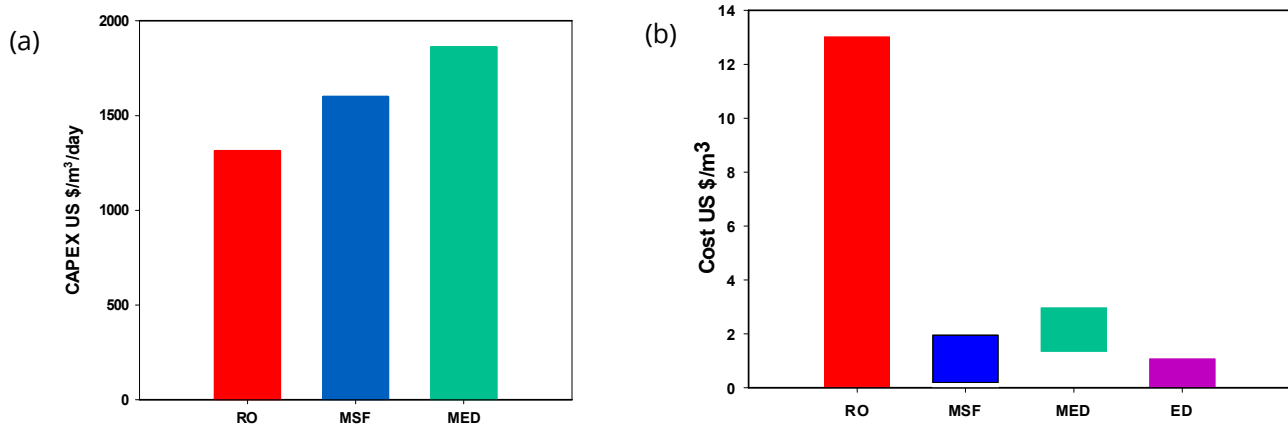


Figure 5: Main technologies for desalination (chloride removal); (a) CAPEX and (b) OPEX (for capacity of 37850 m³/day).

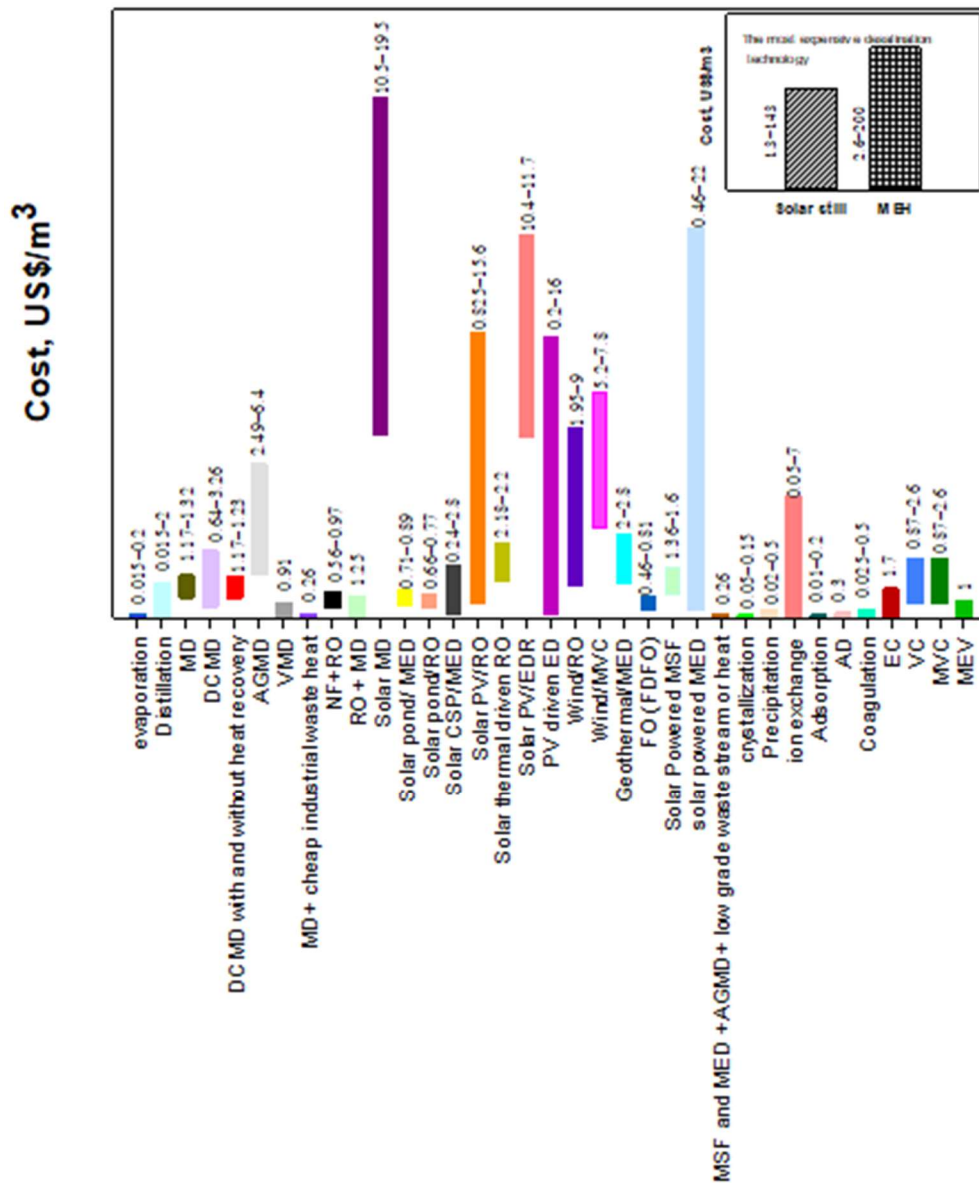


Figure 6: Cost of other technologies (non-main technologies) for desalination/chloride removal.

Table 9: Comparison of the maximum capacity of various adsorbents for chloride removal.

Adsorbent	Capacity (Q_m) mg/g	Reference
Cement (silicate hydrate (C-S-H))	21.27 mg/g (0.6 mmol/g)-172	[194-196]
Friedel's salt	70.91- 514.07 mg/g (2-14.5 mM)	[197]
Calcium kaolinite	3 m. equivalent (me)/100 g	[198, 199]
Natal soil	0.4 me%-1.0 me% (1.4 -3.5 mg/g)	[199]
Kent sand	more than 0.4 me% (more than 1.4 mg/g)	[199]
Gibbsite	approx. 2.48 mg/g	[200]
γ -alumina- modified with sodium oxide	11.25 mg/g	[201]
Goethite	50 μ mole/g	[200]
Tripoli	30 mg/g	[202]
Pozzolana	12.07 mg/g	[203]
zeolite	9.23 mg/g	[203]
Anthracite coal	98.559 mg/g	[204]
Dolomite	3.139 mg/g	[204]
Limestone	93.795 mg/g	[204]
Pozzolan	55.153 mg/g	[204]
FF	106 mmol/g (3758.02 mg/g)	[205]
FS	373 mmol/g (13223.97 m/g)	[205]
DI-60	105 mmol/g (3722.565 mg/g)	[205]
RH	49 mmol/g (1737.2 mg/g)	[205]
Activated carbon modified with carboxyl groups	3.082 mg/ g	[206]
Parthenium sp.	340000mg/g	[207]
Parthenium sp. leaf biomass and assistance of sugar	8000 mg/g	[208]

Other technologies which have been studied for chloride removal (selective technologies) are coagulation and electrocoagulation (UHLA-ultra-high lime aluminum technology), chemical precipitation, ion exchange, adsorption desalination (AD), and adsorption, which their pros and cons are presented in Table 10. Chloride removal by biological treatment is also less effective [223, 224].

5.2. Iodide

Generally speaking, iodide can be removed via various methods, such as ion exchange [149], adsorption [18, 22, 156, 225-243], membrane separation [244, 245], and chemical precipitation [246, 247]. Membrane-assisted technologies, electrochemical and adsorption as the leading technologies for removing iodide and bromide from water, have shown their removal efficacies, pros, and cons (Table 11 and Table 12).

I^- shown to co-precipitate with hydroxides such as Zr, U, and Th at pH 4, pH 8, and pH 7. In addition, iodide has shown a high affinity towards $Bi(OH)_3$ [19]. Other adsorbents for iodide are iron zero-valent, illite, activated aerogels impregnated with silver ions, and Mg-Al LDH/Mg-Al (NO_3) LDH [118, 119]. Alternative technologies for iodine removal are ion exchanges of Amberlite 400 and DOW 21K XLT [121], permeation, and chemical desorption (PCD) [117]. Some ion exchanges need iodide oxidation, forming iodate and then passing through the resin. It is clear that this process also deals with the oxidating agent, which is not very environmentally friendly [117].

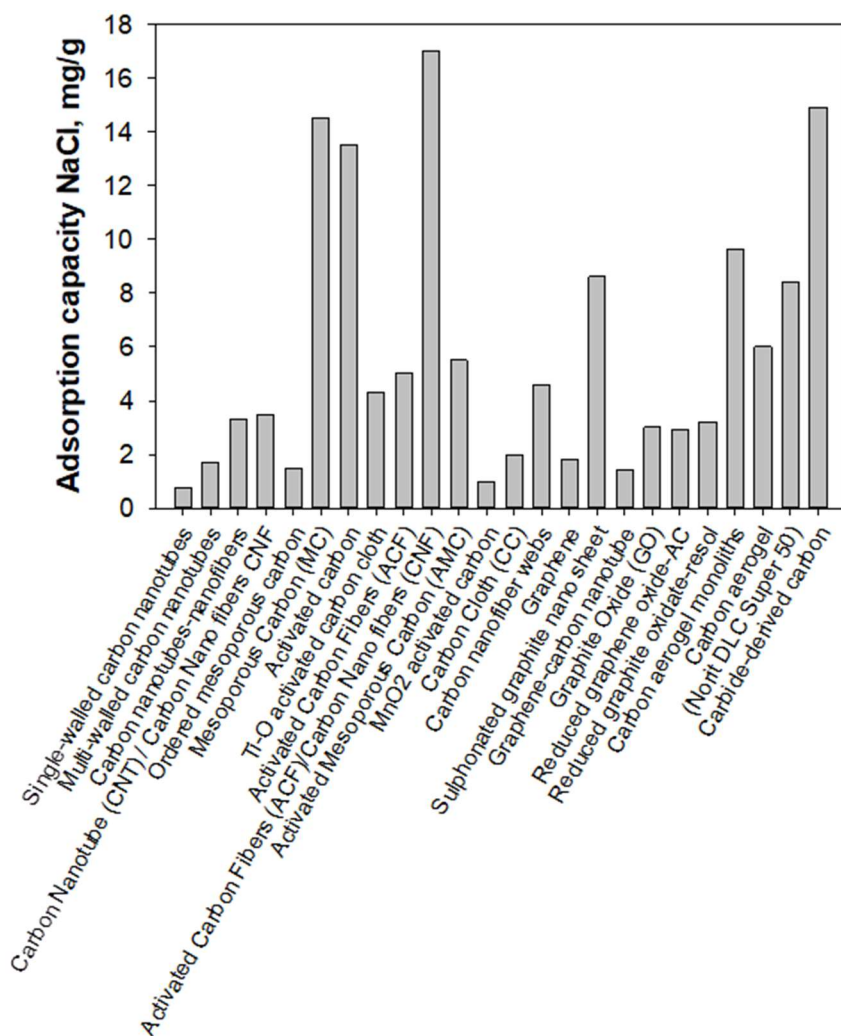


Figure 7: Capacity of CDI electrode for removing sodium chloride [209-222].

5.3. Bromide

Membrane desalination and electrochemical methods are energy extensive, and the membrane is prone to fouling and scaling [40]. Therefore, if these technologies are used just for bromide removal are more expensive than other technologies [40]. Electrochemical technologies such as electrolysis can oxidize bromide to HBrO , BrO^- and Br_2 , which would limit their application on a large scale due to the possibility of the formation of brominated DBPs [40]. Adsorption is another technology used for bromide removals, such as xerogels and hydrogels, with a removal efficiency of 9-80%. Nonetheless, xerogels and hydrogels are not effective in the presence of NOM for bromide removal. Because they have more affinity towards the organic than inorganic species [40], other adsorbents, such as aluminum chloride and LDH, but these adsorbents have limited application due to their efficiency decrease in the presence of other inorganic anions and organic species. Silver integrated into the adsorbent has the disadvantage of silver leaching into the water and is expensive [40].

6. Challenges of Halide Removal

6.1. Chloride

Chloride in chemical compounds such as NaCl is very stable and soluble in water [145-148]. The solubility of NaCl, the most commonly used chloride salt, is 35.7 g/100 g at 0 °C in water. When a chemical species (chloride) is very soluble in water, it tends to be in its ionic form (Cl^-) [353]. Chloride is considered an inert chemical that would not be removed by bioremediation techniques and could not be easily precipitated [354]. Conversion of chloride

to hydrochloric acid is not a good long-term option as it would be only changing chloride form [10]. Also, the oxidation of chloride to chlorine gas by various oxidants, such as potassium permanganate, is very dangerous for the workplace and could form DBPs [355]. Another challenge of chloride removal is that it is usually the less preferred species by the substrate in adsorption and ion exchange processes. Generally, the selectivity of an ion would get higher if its charge is much higher than its hydration radius (i.e., smaller ions with more charges have more selectivity than the bigger ions with smaller charges) [356]. For example, fluoride has a smaller hydrated radius with the same charge as chloride; therefore, fluoride would have more preference to be adsorbed than chloride [293]. The preference for chloride over bromide and iodide for removal is due to the hydration radius, as they have the same charge affinity [293, 357].

Table 10: Summary of other technologies just used for chloride removal.

Technology	Advantage	Disadvantage	Remarks	Reference	
Chemical precipitation	Silver nitrate	Can be used on an industrial scale	Precipitation with silver nitrate is expensive and is mainly used in hydrometallurgy processes. Silver loss in this process is inevitable.	Silver nitrate was used to remove chloride from hydrometallurgical solutions, and silver was regenerated again. The regeneration of silver was carried out by using zinc in the solution. Zinc would alter silver chloride to zinc chloride, regenerating silver. Subsequently, the metallic silver would contact acid nitrate to convert it to silver nitrate. Silver nitrate is considered suitable for hydrometallurgy processes.	[171]
	Copper slag	Can be used on an industrial scale	Copper slag is toxic (due to the application of zinc) and is suitable and limited to hydrometallurgy. It needs high acidity and high sulfate.	Copper oxide process is considered more environmentally friendly than copper slag with zinc because it does not need zinc to reduce copper (II) to (I), and zinc is not environmentally friendly.	[171, 336]
Membrane assisted technology	MD	High chloride rejection. It can reject 99–100% of salts (chloride) and produce water with very high quality (less than 10 mg/L). Feed water quality, and salt concentration, have minimal effect on the performance. It works with small /no pressure. The membrane used in this process does not need very good mechanical properties (fewer requirements on the membrane). It also has a large contact area per unit of equipment volume. Finally, it can also work in low temperatures, like when used in a vacuum. The process heat could be gained from renewable energies or industrial waste/ residual heat.	It is energy-intensive technology (more than RO and less than evaporation and crystallization). It has a lower permeate flux due to mass transfer resistance than RO. There are few designs made for membrane/ membrane modules. Water needs pre-treatment to avoid scaling and fouling of membranes. For example, fouling is possible if the membrane gets wetted using a hydrophobic membrane.	It is more suitable for brine (from desalination plants) treatment. Using plastic materials to avoid corrosion was suggested, especially at high chloride concentrations. The cost of this process has yet to be discovered and depends on the type of energy source.	[175, 186, 249, 250, 337]

(Table 10) Contd....

Technology		Advantage	Disadvantage	Remarks	Reference
Thermal	MSF	It is a robust technology that deals with low-quality water and adverse conditions. It produces high-quality water with less than 10 mg/L pollutants/TDS.	It needs high temperatures of 90-110 °C as primary energy. The final water should be mixed with cold water. It has low water recovery. Even after dilution, the high salinity of concentrate would be very high, even 15% higher than feed water.	2 nd , a desalination process in the world after RO. Today, the share of RO plants in desalination capacity in the world is 59%, while the thermally driven desalination methods have only 27%.	[172, 176, 186, 338]
	MED	It produces water with high quality of 10 mg/L of pollutants/TDS. Its advantage over MSF is that it is more flexible, less plausible to scale, and works at a lower temperature of 70 °C as primary energy.	This process is more suitable for limited-capacity plants because it works at a partial load of MSF.		[172, 186]
	Solar still	simple with less maintenance.	Low productivity. It needs large areas of land and is mainly suitable for low capacity.	In the pure thermal process, solar/dam needs 0.63 kWh/m ³ as electrical energy, and the thermal is 475.01 kWh/m ³ .	[174, 189]
	Evaporation ponds	It is cheap, the operation is simple, and it is used in industries. Also, there is a possibility of salt recovery.	It needs a large area of land.		[175]
	Evaporation and crystallization systems	The technology is available and is being used on an industrial scale.	It is an expensive technology, and CAPEX and OPEX are both high.	It needs new developments to decrease its energy consumption.	[175]
	VC (MVC, TVC)	It produces water with high quality, TDS almost 10 mg/L. It is relatively cheap, especially if it is combined with heat waste.	Its capacity is limited to small to medium-sized plants (100-30000 m ³ /day).	This technology is based on vapor compression, which can be achieved by mechanical or thermal vapor compression. MVC uses electricity, while TVC uses the steam jet for pressurizing the water. TVC has a higher capacity than MVC.	[172]
AD	It consumes a small amount of energy. It can also work with a low-temperature heat source or solar heat. It has low maintenance and low evaporative temperature, fouling, and scaling in the evaporators. Finally, it is cheap (low payable cost) if it uses the waste heat of renewable energies.	The data available for this technology is only known at the pilot or demonstration scale. Its economic aspect is questionable if it is not using heat from cheap sources, such as renewable energies or heat waste. Finally, the robustness of silica gel absorber beds has yet to be discovered.		[174, 249]	

Table 11: Summary of technologies for halide removal.

Technology	-	Advantage	Disadvantage	Remarks	Reference
Membrane assisted technology	RO	<p>Effective Br rejection >90.6% and iodide rejection >89%</p> <p>Can reject chloride with high concentrations up to 10000 mg/L.</p> <p>Recently, RO has improved its efficiency and reduced its energy consumption and footprint by</p> <p>Modifying membrane modules, using a semi-batch process, and combining RO with NF reduce energy consumption.</p> <p>Semi-batch RO has high to medium membrane recovery, depending on the feed water quality (recovery%: 55% seawater and 95% brackish water desalination).</p> <p>Two-stage RO will increase the water recovery.</p>	<p>Generally, it has a high cost,</p> <p>Concentrate disposal and management is tricky,</p> <p>Scaling and fouling concentration polarization is an accumulation of particulate and dissolved material near the membrane. This will affect the RO performance by higher salt flux and rejection decline, decrease the water flux because of the higher osmotic pressure, precipitation on the membrane surface would be more plausible, and cake formation on the membrane by the particulates. The osmotic pressure and viscosity would decrease RO performance.</p> <p>Semi-batch RO is known very little; data is available for only pilot and demonstration scales.</p> <p>The effect of high cross flow and brine circulation on membrane life is unknown.</p> <p>Two-stage RO needs high reagent dosage, which will increase the process expenses.</p>	<p>1st, the desalination process in the world.</p> <p>Rejections are as follows Cl>Br>I. This is because of the higher hydration energy of chloride, then bromide, and finally, the iodide.</p> <p>Electrostatic forces play the primary role in the rejection of halide ions.</p> <p>The removal of halides increases with increasing the pressure to the threshold limit. Above the threshold limit, the rejection of halides decreases.</p> <p>Spiral-wound and fine hollow fiber is the most commonly used membranes for RO.</p> <p>RO, with modular construction, has a small footprint. Also, semi-batch RO consumes lower energy (no energy recovery devices) and reduces footprint, fouling, membrane elements, and pre-treatment. Semi-batch RO has high crossflow velocity, higher water flux (more than 30%), and higher rejection of salts (90%) for the brackish and wastewater. Can produce water with a total TDS of 400-500 mg/L from seawater</p> <p>Increasing halogen content in the DBP molecule and substituting smaller chlorine-containing DBPs with larger molecular size bromine-containing DBPs achieve greater rejection by RO membrane.</p> <p>Poor rejection of low molecular weight uncharged compounds.</p>	[172, 175, 182, 186, 188, 248-252]

(Table 11) Contd....

Technology	-	Advantage	Disadvantage	Remarks	Reference
	FO	<p>It has simple technology. It is an osmotically driven process.</p> <p>This does not need hydraulic pressure or works with low pressure with lower energy consumption. Lower fouling potential due to lack of applied pressure. It has high feed water recovery and lesser pre-treatment needed.</p> <p>Good for desalination of highly saline water.</p> <p>It has high salt rejection, which would hardly be affected by the feed water contaminants.</p> <p>It can recover feed water with a wide range of 500-175000 mg/L as TDS.</p>	<p>Full-scale installation is limited, and more development is needed in FO membrane technology. It is more suitable as a pre-treatment.</p> <p>Recovery of clean water and draw solution regeneration would require a suitable combination of other technologies like RO—reverse diffusion of solute from draw to feed solution present.</p>	<p>FO, as pre-treatment, can reduce the cost of RO.</p> <p>The cost of FO/RO for desalination of seawater is lower than RO due to less electrical consumption. Its hybridization with RO is a perfect approach. FO/RO recycles 76% of water.</p> <p>FO costs depend on the application of the technology. For example, for FO draw solution recovery/reconcentration, the direct cost will be energy requirement.</p> <p>It can utilize waste heat sources for the regeneration of the draw solution.</p> <p>High iodide rejection 98.4% from LCD wastewater. MgCl₂ and cetyltrimethylammonium bromide as draw solution.</p> <p>Needs more studies for iodide and bromide removal.</p>	[175, 186, 249, 250, 253]
	NF	<p>Efficient, lower capital and operational costs compared to RO, lower working pressure, producing less concentrate compared to RO.</p>	<p>medium to high energy consumption.</p> <p>Needs pre-treatment has scaling and, fouling, brine disposal problems.</p> <p>It is not practical to remove chloride from mining water.</p>	<p>Membranes are charged, and electrostatic forces are important in the rejection part. Polyvalent ions are more readily rejected than monovalent ions, such as halides. The removal of halides increases with increasing the pressure to the threshold limit. Above the threshold limit, the rejection of halides decreases.</p> <p>Operating conditions are significant in the selectivity of halide rejection. Usually, chloride has more selectivity than iodide to be rejected due to the hydration energy. However, it was shown that if the operational pressure gets less than optimal conditions</p>	[250, 254-265]

(Table 11) Contd....

Technology	-	Advantage	Disadvantage	Remarks	Reference
				(pressure in which iodide has more rejection), iodide would have more selectivity over chloride. Tight NF-45 has similar mass transfer to RO properties and is a good option for halide rejection. NF membranes with higher molecular weight could reject higher amounts of halides.	
Electrochemical assisted	ED and EDR	<p>Tests in pilot scales show that it can have 80% bromide removal.</p> <p>ED, compared to RO, has lower energy, chemical consumption, pre-treatment requirements, operational costs, and water recovery. ED does not need pressure, despite RO.</p> <p>EDR has a higher membrane lifetime than RO.</p> <p>ED and EDR can produce high-quality water with TDS 150-500 mg/L (80-94% water recovery). ED can reduce chloride to 200 mg/L in brackish water (97-98% recovery).</p>	<p>They are not conventional technologies in drinking water treatment. For example, EDR has technical and economic limitations such as high capital costs and energy consumption.</p> <p>In ED, precipitant can build up on the concentrate sides because no current reversing exists despite EDR.</p> <p>ED is prone to scaling and fouling of the membrane</p> <p>Possibility of oxidation chloride to toxic chlorine gas, mainly if the stainless steel is used as an electrode. It has high CAPEX and OPEX.</p>	<p>Generally, ED and EDR are suitable technologies for halide removals. In this technology, hydraulic and electrical staging is used for halide removal.</p> <p>ED is cost-effective technology suitable for medium concentrations (NaCl up to 10 g/L) or TDS \leq 4000 mg/L.</p> <p>At higher TDS > 4000 mg/L is better to apply RO as a more cost-effective technology.</p> <p>EDR is better than ED in aspects of longer electrode lifetime and cleaner membranes due to reversing current.</p>	[7, 172, 175, 249, 250, 266-270]
	Electrolysis	<p>Conversion of halogen to the gaseous form can give disinfection properties.</p>	<p>In the case of bromide, the gas conversion can make hypobromous acid. Then hypobromous acid reacts with NOM in water and produces DBPs.</p> <p>Electrolysis was not used on a large scale and needed more studies to make its pilot scale more feasible.</p>	<p>It is based on oxidation and conversion of halide to halogen gas. pH and the species in water, such as carbon dioxide (a role in pH decline), can convert hypobromite to hypobromous acid.</p> <p>Therefore, a need to control pH and species in water. Electrode corrosion is an issue.</p> <p>Parameters such as current density, type of electrode, time of electrolysis, the ratio of halides in water, conductivity of water, and electrodes' distance will affect the process of halide removal and energy consumption.</p>	[271-275]

(Table 11) Contd....

Technology	-	Advantage	Disadvantage	Remarks	Reference
	CDI	<p>Does not need extra pressure despite RO. It has low pre-treatment, minimum scaling and fouling, low maintenance, low energy requirement, and more stability than RO.</p> <p>Regeneration is possible and easy by reversing the current polarity.</p> <p>More cost-effective than RO and EDR.</p>	<p>It needs more study to be implemented on a large scale, especially for brackish water.</p> <p>Q_e is small—for example, maximum sorption capacity (NaCl) of almost 18 mg/g of the electrode, approximately.</p> <p>Competitive anions can reduce chloride removal from water.</p> <p>Electrodes' efficiency needs optimization, in which limited data is available.</p>	<p>suitable for low salinity feed water sources (TDS < 15,000 mg/L).</p> <p>Activated carbon and carbon aerogels are electrode candidates for this technology.</p> <p>Removal efficiency of halides depends on the electrode properties such as pore structures in electrodes, surface area and chemical composition of the electrode (affinity towards halides), and electrostatic forces.</p> <p>Order of removal is $Cl^- < Br^- < I^-$ because the charge transfer in iodide is more than the other halides.</p> <p>Likewise, of other electrochemical technologies, the current density, electrode surface area, and conductivity of the solution impact removal and energy consumption. Polarity reversal will give self-cleaning of electrodes.</p> <p>This technology is suitable for brackish water treatment from mining and gas industries and recovery of iodide.</p>	[172, 175, 212, 249, 250, 276-285]
	MCDI	<p>Higher removal efficiency than CDI. This is because ion exchange membranes prevent adsorbed ions from leaving the electrode region.</p>	<p>Difficulties in scale-up. That is because we must find cheap aerogels to make them commercially applicable.</p> <p>Membrane scaling and fouling in MCDI would be possible.</p>	<p>More studies, especially for halide removal. Need more investigations to make it practical on a large scale.</p>	[286-288]
Adsorption	Adsorption- LDH	<p>It is a cheap, universal, fast, and flexible technology with low maintenance and is easy to design and operate. Regenerate the sorbent is possible.</p> <p>Useful for iodide and bromide removal from drinking water.</p>	<p>LDH materials, such as Mg-Al-Cl LDH, are affected by the exiting of other anions. For example, the removal of iodide decreased with increasing chloride concentrations.</p> <p>The existence of bicarbonate can reduce bromide removal, and sulfate and bicarbonate .</p>	<p>Depending on the adsorbent type, the adsorption capacity could be low to high.</p> <p>Applying metals in the LDH structures with affinity to halides such as Cu^{2+} would enhance the iodide/other halide removals.</p>	[194, 199, 202, 289-298]

(Table 11) Contd....

Technology	-	Advantage	Disadvantage	Remarks	Reference
			<p>can reduce iodide removal. Removal of chloride with adsorbents (like C-S-H) in the existence of other anions (such as nitrate, carbonate, bromate, and sulfate) would favor other anions.</p> <p>Existence of NOM and high alkalinity is not suitable for bromide removal.</p> <p>They can be as effective as commercial resins in the right place of usage.</p> <p>Adsorption depends on the pH and isoelectric of the adsorbent.</p> <p>halide leaching after adsorption is possible.</p> <p>The cost of the whole process considering continuous dosing adsorbent could be high due to finite adsorbent capacity.</p> <p>CO_3^{2-}, SO_4^{2-}, OH^-, and F^- have more preference for sorption than chloride in LDH and CLDH.</p>	Parameters such as calcination/non-calcination, time and temperature of calcination, type of metal being used in the system, and metal ratio can affect the LDH materials' sorption.	
	Coal and activated carbon	Cheap and high usage in water treatment processes.	Has low removal for iodide removal.	The oxidation of iodide can increase the removal efficiency of iodide to iodine.	[299]
	Silver-impregnated and silver chloride-impregnated activated carbon.	<p>Impregnation of silver into the carbon active has an extensive application in drinking water treatment.</p> <p>Cheap technology in according with no need for big infrastructures.</p> <p>Silver gives antibacterial properties.</p>	Silver leaching.	Applying a proper impregnation process could prevent silver from leaching under acidic conditions. For example, impregnating silver chloride instead of silver significantly reduces silver leaching in acidic conditions.	[300, 301]
	Silver impregnated Carbon Aerogels	Higher adsorption capacity than activated carbon for halides.	<p>High cost and not applicable on an industrial scale.</p> <p>Existing chloride would reduce bromide and iodide removal. Preferring</p>	Increasing silver concentration on the aerogel increases the macropores and mesopores.	[302-305]

(Table 11) Contd....

Technology	-	Advantage	Disadvantage	Remarks	Reference
			for halide removal is $Cl^- > Br^- > I^-$. This is because the larger ion size, like iodide, is disadvantaged. NOM and competing anions reduce the halide removal efficiency.	By activation and carbonization, the surface area increases. This increases the iodide and bromide removal efficiency. Therefore, carbonization and activation of aerogels are highly suggested.	
	Soil	Cheap and available.	Limited functionality, especially in complex water matrices, has a low capacity for iodide and chloride removal. Other anions and organic materials can reduce halide removal significantly. No study for the bromide, and it does not have the capability for bromide removal.	Iron oxide in the soil showed better iodide removal than in soils that contain aluminum oxide. For example, iodide has better removal efficiency with hematite than kaolinite. However, chloride is better removed in soils that have aluminum minerals.	[306-316]
Coagulation and electrocoagulation	Coagulation with aluminum	Cheap and conventional water treatment method. Capable of being pilot scale. Coagulation with UHLA is used for chloride removal and can be used on an industrial scale. UHLA's aluminum source can be from the aluminum electrode (EC), sludges containing aluminum, or alum. UHLA with alum has advantages such as compact instrumentation, simplicity, automation, and versatility. It produces less sludge over chemical coagulation and has smaller footprints. Chloride removal efficiency with UHLA 84%.	No studies for iodide with Coagulation with alum. No studies for iodide and bromide removal with UHLA. Coagulation with alum is sensitive to pH and other existing ions and humic acids on bromide removal. In UHLA, chloride removal would be less with the existence of other anions, such as silica and sulfate, and it is suitable for chloride concentrations higher than 100mM. Finally, aluminum in the feed should be monitored. Alum alone is not efficient for chloride removal, while when calcium salt is added would be very effective (UHLA). In the case of EC, aluminum electrodes are costly.	Conventional coagulation, sedimentation, and filtration are unsuitable for halide removal and higher molecular weight, hydrophobic fractions of natural organic materials. Only low molecular weight, hydrophilic fractions are removed. Increases the halide-to-organic ratio, leading to increased halide incorporation in halogenated DBPs. Coagulation alone needs a high dosage of aluminum chloride if there are competitive ions and organic materials in water. UHLA for chloride removal (alum and calcium salt chemical coagulation) Cl-chemical coagulation needs Ca:Al:Cl ratio of 10:0:4: 1 (for Freidel's salt). In UHLA technology and the application of EC, cathode passivation is plausible because calcium salt precipitation on the cathode is possible, which can be avoided by applying stainless steel.	[113, 169-171, 298, 317-321]

(Table 11) Contd....

Technology	-	Advantage	Disadvantage	Remarks	Reference
			<p>Expensive for iodide and bromide removal.</p> <p>Because iodide and bromide concentrations are lower than chloride.</p> <p>However, it is more economical for chloride than other halides and resin's function is sensitive to the water conditions such as alkalinity (bicarbonate) and competing ion concentrations in MIEX®.</p> <p>Lower capacity for bromide sorption after regeneration MIEX®.</p>	<p>Strong anion-exchange resin with polystyrene functional groups can remove iodide and bromide.</p> <p>Amine groups, especially quaternary amine functional groups in the resins, are highly capable of removing iodide and bromide.</p>	
Resins		<p>This method is good for the low concentration of chloride. It would be comparatively cheap because of lower power consumption, minimal brine production, and low capital cost. Therefore, it is still cheaper in low and high salinity, even if we consider the chemicals needed to regenerate resins.</p> <p>It is selective towards chloride.</p> <p>The capacity of the anion resins (for chloride) is 1-1.3 q/L for the strong base anion resins.</p>	<p>Metal incorporated into the resins is susceptible to metal leaching.</p> <p>Weak base resins (WBA) need acidic conditions for removing chloride. Ion exchangers are made of toxic materials, which is suggested for lower chloride concentration.</p> <p>Generally, ions with a smaller hydrated radius and higher valence have a higher permeability in an ion exchange membrane than ions with a larger hydrated radius and lower valence. Therefore, $I^- > NO_3^- > Br^- > Cl^- > SO_4^{2-} > F^-$.</p> <p>Specifically, in strong base resins I^-, NO_3^-, Br^-, ClO_4^-, SO_4^{2-}, HPO_4^{2-}, CO_3^{2-} have preference for sorption more than chloride.</p> <p>Weak base resins OH^- has the most preference for sorption.</p> <p>Regeneration of IX columns is costly.</p>	<p>Lack of studies on iodide removal with resins because most studies are about bromide removal.</p> <p>Need to find a cheaper resin.</p> <p>Regeneration cost of resins could be decreased; as a result, the cost of comprehensive technology, by applying core-shell arrangement in the resins. Shallow shell ion exchanges, which are the ones that provide core-shell arrangements, have the advantage of regenerating more quickly than normal ones. These resins also reduce the volumes of rinsing and increase running times.</p>	[113, 191, 293, 298, 322-335]

6.2. Iodide

Ion exchangers have limited applications for removing iodide from water because of their limited removal efficiency and cost [358]. Some natural minerals and sediments have been shown with iodide/iodine removal capacity, but their removal rates needed to be increased, too [20, 22].

Technologies which have been routinely employed to remove iodide (RO, NF, ED/EDR, electrolysis, CDI, resins) are expensive, and adsorption, which is cheaper (except silver-contained adsorbents) is not practical all the time

such as activated carbon [37, 247]. Another challenge of iodide removal is that most of the adsorbents for iodide removal possess slow reaction kinetics. In a nuclear disaster, it would be desirable to have an adsorbent to quickly and effectively remove iodide from water [17].

6.3. Bromide

Bromide removal is challenging and is less effective with common water treatment technologies (coagulation/flocculation) [158]. It is expensive to be removed by membrane-assisted technologies such as RO and NF. Bromide oxidation is also not a good strategy to remove bromide from water, especially if there is any NOM in water, then brominated DBPs would be produced [37]. Another challenge of bromide removal is that bromide adsorption on adsorbents or resins in the presence of NOM or alkalinity would decrease. NOM could block the adsorption sites, and bromide removal would decrease [40, 135]. For example, MIEX® showed removal of 49% and 20% for alkalinity in water 11 mg/L and 95 mg/L, respectively [135].

Table 12: Technologies for iodide and bromide removal and their efficiencies.

	Material Type (membrane/ adsorbent)	Bromide (mg/L)	Iodide (mg/L)	Competitive Anions	Type of Water	Removal Br %	Removal I %	pH	Temperature (°C)	Ref.
RO	Composite Polyamide	64.4-71	-	Yes	Seawater and natural water	99.34-99.8	-	7.9-8	20-25.1	[312]
	NS		0.0358 - 0.0511	Yes	Seawater and natural water	-	89	7.1-7.6	26.1-38.5	[339]
	Thin-Film Composite (TFC) ® Polyamide	51.3	49.9	Yes	Natural water, oilfield water	90.7	92	6	11±1	[340]
	Polyamide Membrane	3995	6345	No	seawater	99.6	99.4	6.8	25	[341]
NF	Polyamide TFC [NF-90 (200 Da)	1030	1	Yes	Synthetic water	94-97	84-91	7.8 - 9.8	20	[254]
	NF-90	51.3	49.9	Yes	Natural water	80	78.3	6	25	[340]
	NF70-2540 (Polyamide)	-	719.5-2538	No	Synthetic water		56.785-80	-	20	[252, 257] [261]
	ESNA	51.3	49.9	Yes	Natural water	-	55.6	6	25	[340]
ED/EDR	Mark III to Mark IV type	0.5-1.2		Yes	Natural water	75-80	-	6.5-7.3	25	[266]
	AR204-SZRA and CR67- HMR	0.185	-	Yes	Natural water	72	-	7.6	25	[342]
	DE81	-	1.2 - 12.7	No	Synthetic water	-	92-97	-	25	[343]
Electrolysis	-	0.200	-	Yes	Natural water	>99	-	-	-	[273]
	-	0.461		Yes	Synthetic water	79	-	~7.5	-	[275]

(Table 12) Contd....

	Material Type (membrane/ adsorbent)	Bromide (mg/L)	Iodide (mg/L)	Competitive Anions	Type of Water	Removal Br %	Removal I %	pH	Temperature (°C)	Ref.
CDI	-	~75	~30	Yes	Natural water	62.5	77	7.5-9.1	23	[279]
	-	191.8	304.56	No	Synthetic water	0.108 mmol/g	0.154 mmol/g	-	25	[280]
Adsorption	Mg-Al-(NO ₃) LDH	-	342	Yes	Natural water, wastewater	-	59	9.2	25	[119]
	Mg-Al (molar ratio of 4, calcined at 500°C) LDH	100	100	No	Synthetic water	94	96.5	-	30	[344, 345]
	Zn-AILDH	-	330	No	Synthetic water	-	60	7	-	[346]
	Fe ₂ O ₃ ·Al ₂ O ₃ ·xH ₂ O	120	-	Yes	Synthetic water	80	-	-	22	[347]
	Sub-bituminous coal	-	1.3	Yes	Synthetic water	-	46	6.2	25	[348]
	Merck activated carbon	-	1269	Yes	Synthetic water	-	21	7	-	[289]
	Silver-impregnated activated carbon (20 wt % of Ag)	-	0.127	Yes	Synthetic water	-	98	-	20	[349]
	Silver-impregnated activated carbon	-	1.0 – 200.5	No	Synthetic water	-	3.10-153.7 (µmol/g)	5,7,8	-	[350]
	Activated silver-doped aerogels	0.15	0.15	No	Synthetic water	5.78 (µmol/g)	5.03 (µmol/g)	7	25	[302, 304, 305, 351]
Resins	MIEX®	0.076-0.900	-	Yes	Synthetic water, Natural water	17-83	-	6.6-8.1	15-26	[322-325, 328]
	MIEX®+Alum	0.0137-0.540	-	Yes	Natural water	0-94	-	6.1-8.2	20,25	[324, 326]
	Indion-850	2876.4	5.1	Yes	Synthetic water	66.6	57.5	-	25	[352]
	Duolite A-113	3403.7	5.3	No	Synthetic water	54	45.3	-	25	[327]
	DOWEX MARATHON MSA					90	-			[322]
Coagulation	Aluminum coagulation	0.2-2	-	Yes	Synthetic water and natural water	30.4-98.8	-	6-8	-	[317, 318]
Soil	Udic Ferrisols	-	4	No	Synthetic water	-	57.5	5.05	25	[314]

7. Conclusion

A state-of-the-art review on halide removal has been carried out. This review highlights the importance of halide removal, the sources of excessive halide concentrations in water, and the technologies available for halide removal in water treatment. The review also presents how halide presence affects oxidation processes such as AOP through scavenging active radicals, especially in saline water. In AOPs, bromide scavenges the hydroxyl and sulfate radicals. Bromide also affects the ozonation and chlorination processes and will lead to the formation of bromoform and bromate. However, chloride would affect sulfate radicals. Bromide plays a key role in oxidation in high saline waters such as seawater with high chloride concentrations. The ClOH^\cdot is not stable and transforms to OH^\cdot and Cl^\cdot . Increasing halide concentration formation of BrCl^\cdot and Br_2^\cdot is more likely. The best strategy to use AOP in high-saline waters is the application of UV. It does not have much efficiency, but DBPs production is less likely. Also, removing iodide and bromide from water is as important as chloride because they will produce DBPs in the oxidation process.

Chloride concentration in water is highly regulated compared to other halides, such as bromide and iodide, which can cause environmental and health issues due to more toxic DBPs formation. The halides in water can be engineered to enhance the AOP. Halide management is very critical as unwanted DBPs can be produced. One of the techniques to have less dangerous DBPs in water is to remove their main formation precursors, bromide and iodide, prior to any water purification or oxidation.

Using adsorbents and ion exchange to remove halide could be advantageous with a fast kinetic rate, and there is ample scope for further research and development. However, the presence of other ions in water poses a challenge to its removal. For example, resins and adsorbents' active sites could be clogged by the associated natural organic matter in water, making iodide and bromide removal less effective. There needs to be further studies on affordable adsorbent/ion exchanger development with high affinity and fast kinetic reaction. This will make adsorption a more potent technology that can be used in water purification at lower costs. Electrochemical capacitive deionization is another potential technology that can be studied to selectively target bromide removal from source waters undergoing disinfection to reduce brominated DBP formation.

Alternate treatment methods, such as using biomass and industrial wastes for halide removal, have yet to be investigated thoroughly. Flora such as *Lolium rigidum* (forage grass in southern Australia), *Aaicennia marinia*, *Avicennia*, *Aegialitis* and *Aegiceras*, Fourwing saltbushes, and low Cl^- excluder plant K51-40 should be further researched as natural halide management techniques. However, these plants could have a low kinetic rate of removal. However, case studies have shown that some flora, such as *Corymbia maculate* Hook, *Eucalyptus botryoides* Smith, *E. tereticornis* Smith, and *E. occidentalis* Endl, can be used for rehabilitation of post-mining lands in NSW, Australia. However, they were not used for water treatment.

Halides are removed as total dissolved solids by membrane-assisted technologies with less selectivity. For example, current commercial membrane processes are suitable for bromine removal. However, much of its energy is utilized for the removal non targeted dissolved solids in water instead of selectively removing bromide. When applying these technologies, the costs should be considered along with the environmental outcomes. Integrating renewable energies such as geothermal and waste heat with membrane technologies could reduce the total operating costs. The research in this area seems bright. Membranes can be engineered to reduce their fouling and selectivity. For example, the designed cross-linked matrix of polymers and engineered nanoparticles could be widely applied in future membranes.

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