

Strategies for Organochlorine/Organobromine Removal from Aqueous Environment Using Nanotechnology: A Review

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ABSTRACT: Organohalogenes have been discovered since the 1930s and have been used for many applications ever since. The rapid development of industrial activities and reliance on organochlorine/organobromine compounds have further increased their production, ultimately leading to their leakage into our natural environment, where they circulate indefinitely. Exposure to these persistent organic pollutants (POPs) not only results in detrimental effects on human health, such as various cancers, nervous system damage, and liver damage, in addition to fetal and infant growth defects, but also affects fauna, such as bird populations, by depriving them of the ability to reproduce successfully, and farm livestock. The direct consumption of the latter or its derivatives will also lead to the bioaccumulation of POPs in the human body. The POP treatment methods discussed in this review include granular activated carbon (GAC) adsorption, magnetic nanospheres coated with polystyrene, hydrogen peroxide (H₂O₂), UV, as well as O₃ ozonation. The mechanisms, along with the advantages and drawbacks of these methods, were thoroughly discussed. Finally, challenges faced in reducing organochlorine/organobromine pollution were discussed, such as the lack of updates on water quality standards and the list of dangerous pollutants, and the failure to control illegal disposal issues.

KEYWORDS: Persistent organic pollutants (POPs), pesticides, adsorption, hydroxyl radicals

1. Introduction

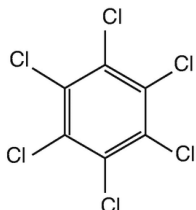
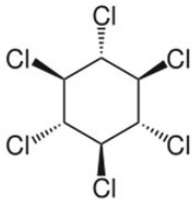
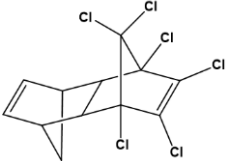
Organohalogenes are any class of organic compounds that contain at least one covalently bonded atom from group 17 of the periodic table. Currently, chemists have discovered 2,320 types of organochlorines, as well as 2,050 organobromines, 115 organoiodines, and 34 organofluorines [1]. Organohalogenes have widespread applications in many industries, including industrial, agricultural, and consumer products, among others. Table 1 shows a brief history of organochlorine compound development. The use of organochlorine agents as insecticides against pests such as lice, the Colorado beetle, and mosquitoes was first introduced by Paul Muller, a Swiss chemist and Nobel Prize winner, in the 1930s. Earlier discoveries of natural organobromine included the ancient Egyptian dye known as Tyrian Purple in 1909 and 3,5-dibromotyrosine, obtained through the hydrolysis of a coral-containing protein in 1913 [2]. The

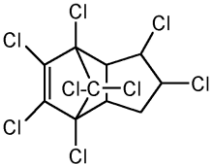
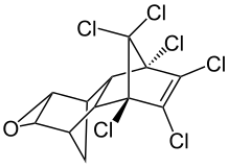
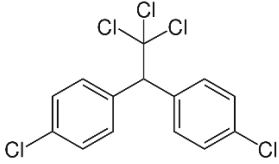
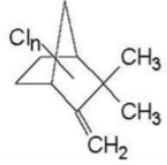
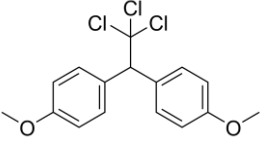
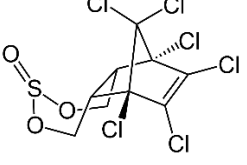
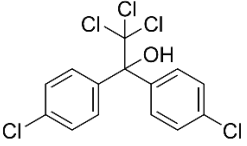
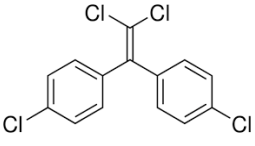
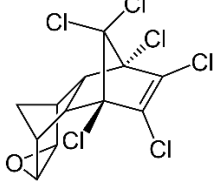
applications of organobromides were then expanded to items such as fire retardants [3], pharmaceuticals, fumigants, and biocides. However, some of these organohalogenes, such as polychlorinated biphenyls (PCBs), chlordanes (CHLs), and dichloro-diphenyl-trichloroethane (DDTs), are well-known environmental contaminants denoted for their persistence, bioaccumulation, and toxicity. While synthetic manufacturing of these chemicals has been limited and banned in some countries, organohalogenes still persist in the global ecosystem and pose a threat to life on Earth. Table 2 shows some major organochlorine pesticides, their chemical structure, and persistence. Considering all of the above, it is logical to deduce that continuous human exposure to these compounds will slowly but surely threaten human health. This review will focus specifically on organochlorine and organobromine, with further details on their applications, toxicity effects, and methods and efforts to deal with organohalogen pollutants, along with the challenges in achieving less pollution.

Table 1. Development of organochlorine compounds-a brief history [4].

Year	Incident
1939	DDT's insecticidal properties discovered in Switzerland by Paul Muller
1941-42	BHC's insecticidal properties discovered in France and UK
1945	Chlordane synthesized
1947	Toxaphene synthesized
1948	Aldrin, Dieldrin synthesized by Julius Hymen, USA
1948	Methoxychlor synthesized
1949	DDT residue detected in cow milk
1951	Eldrin synthesized
1954	Mirex introduced
1955	Dicofol introduced
1956	Endosulfan synthesized
1957	Teledrin synthesized
1958	Chlordecone synthesized
1962	The book 'Silent spring' by Rachel Carson (1962) attracts international attention towards ill effect of pesticide use
1968	Insecticide act passed in India to ensure safety from pesticides
1970	Trials against DDT appears in USA and Sweden

Table 2. Major organochlorine pesticides, their chemical structure and persistence [4].

No	Chemical name	Structure	Persistence in environment
1	Benzene hexachloride (BHC) (C ₆ H ₆ Cl ₆)		Highly persistent, half-life 3-6 years
2	Lindane (C ₆ H ₆ Cl ₆)		Highly persistent, half-life 15 months
3	Aldrin (C ₁₂ H ₈ Cl ₆)		Moderately persistent, half-life 4-7 years

No	Chemical name	Structure	Persistence in environment
4	Chlordane (C ₁₀ H ₆ Cl ₈)		Highly persistent, half-life 10 years
5	Diendrin (C ₁₂ H ₈ Cl ₆ O)		Highly persistent, half-life 9 months
6	Dichloro trichloroethane (C ₁₄ H ₉ Cl ₅) diphenyl (DDT)		Highly persistent, half-life 2-15 years
7	Toxphene (C ₁₀ H ₁₀ Cl ₈)		Moderately persistent, half-life 11 years
8	Methoxychlor (C ₁₆ H ₁₅ Cl ₃ O ₂)		Highly persistent, half-life 120 days
9	Endosulfan (C ₉ H ₆ Cl ₆ O ₃ S)		Moderately persistent, half-life 35-150 days
10	Dicofol (C ₁₄ H ₉ Cl ₅ O)		Moderately persistent, half-life 60 days
11	1,1 dichloro 2,2 bis (p-chlorophenyl) ethane (DDD)		Highly persistent, half-life 10 years
12	Endrin (C ₁₂ H ₈ Cl ₆ O)		Moderately persistent, half-life 1-12 years

2. Sources of Organochlorine and Organobromine

Dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCH) were deemed highly effective and less costly when used as pesticides, hence becoming a popular option among Chinese agricultural farmers to protect their crops between 1950 and 1983 [5]. Data analysis has shown that in the 1970s, China dominated the output of DDTs and HCHs globally, accounting for 60% of worldwide production [6]. Prior to the banning of synthetic production of DDTs and HCHs in 1983, the total production of DDTs and HCHs in China was about 0.4

million tons and 4.9 million tons, respectively [7]. However, residual DDT and HCH are still detected due to the continuous usage of dicofol and lindane, containing DDT and HCH respectively, in agricultural activities and malaria control in China [8].

On the other hand, the demand for organobromine is primarily driven by the growth in the pharmaceutical industry, in line with the growing needs for vital medicines such as anticonvulsants, antiseptics, and sedatives. Organobromine derivatives are also used to treat water in pools and spas, thanks to their ability to decrease the formation of algae and bacteria and odors in water. Moreover, the demand for organobromine peaked in 2014, recording a total amount of 844.1 KT in the global market, with fire retardants being the largest application. Other key factors for the sudden rise in demand include the enforcement of fire safety regulations and the expansion of electronic and automotive industries along with ship-building industries [9]. It is also worth mentioning that illnesses like pneumonia and cocaine addiction are treated with the assistance of drugs containing organobromine ingredients.

3. Fate of pollutant

The evaporation of organohalogens in the ocean will fall onto land surface either as a component of rainwater through wet deposition or simply through dry deposition. The deposited organohalogens may be deposited onto high mountains, rivers, lakes or just plain land surface. Other means of organohalogens entering the hydrological cycle include natural runoff from high mountains, emission/volatilization of pollutants and wastewater discharge in industrial factories, usage of fertilisers in agricultural activities through crop dusting, and some urban runoff. These organic compounds will enter the water cycle either through the river, through lakes or through groundwater infiltration which subsequently flows into aquifer. The river organic compounds flow into the ocean as the river reaches its end, while the organohalogen compounds in the aquifer enter the ocean through seawater intrusion into the aquifer. Hence, the fate of organohalogens is highly related to the natural water cycle [6].

4. Toxicity Effect of Organochlorine/Organobromine

4.1. Human health.

There are many toxic effects when organohalogens enter the body. Organochlorines are known to disrupt the human endocrine system and inhibit its function [10]. Endosulfan food contamination is reasonably plausible since it can persist in the environment long enough to bioaccumulate in plants and animals [11]. Endosulfan is neurotoxic and mainly affects the central nervous system, and it was found to be more likely to enter the body through inhalation rather than through direct skin contact. Moreover, studies have shown that organochlorine pesticides were responsible for an increased probability of getting lung, prostate, breast, and stomach cancer [12]. This carcinogenic characteristic is also shared by organobromine compounds such as Decabromobiphenyl, 2,2-bis(bromomethyl)propane-1,3-diol, and vinyl bromide. Organobromine compounds such as pentabromodiphenylether and 1,2,5,6,9,10-hexabromocyclododecane cause serious damage to the liver and thyroid gland in case of repeated exposure [13].

Additionally, it is possible for organochlorines to affect the pregnancy and infant feeding process. For example, β -HCH and DDT residues from organochlorine pesticides may bioaccumulate in the maternal body and be transferred to the fetus, causing detrimental effects to the thyroid hormone level of the newborn [14]. In China, other studies have reported

dangerous amounts of POPs inside the maternal body, with DDT contamination in human breast milk being fairly significant in Chinese populations in cities such as Beijing, Guangzhou, Hong Kong, Dalian, and Shenyang. Comparisons of data from 1998 to 2002 showed that coastal populations such as Guangzhou (DDE+DDT 2.13 $\mu\text{g/g}$ fat), Hong Kong (2.87 $\mu\text{g/g}$ fat), and Dalian (2.13 $\mu\text{g/g}$ fat) contain higher concentrations of DDT, raising the query on whether higher consumption of marine livestock plays a role or not, compared to Chinese populations in Shenyang (0.87 $\mu\text{g/g}$ fat) and Beijing (1.96 $\mu\text{g/g}$ fat) [15]. Other organobromine compounds that cause harm to the human fetus include Octabromodiphenylether and 2,4,6-tribromophenol [13].

4.2. Water.

Organochlorine pesticides are widely used in the agricultural sector throughout the world. However, Pimentel discovered that only 0.3% of the applied pesticides reach the target pest, while the remaining 99.7% flows into the environment [16]. POPs such as HCH and DDT can be easily transported through air and water. A study in 2006 reported that groundwater samples in Hyderabad were contaminated with DDT, β -endosulfan, and lindane, with contaminant concentrations exceeding the respective acceptable daily intake (ADI) values for humans [4]. Groundwater contaminants, such as 1,2,5,6,9,10-hexabromocyclododecane, 2,4,6-tribromophenol, and Tetrabromobisphenol A, flowing into aquatic systems may cause toxic effects to indigenous organisms [14].

4.3. Bird population.

Considering that agricultural threats contribute to 87% of the decline in bird populations, birds serve as suitable indicators for detecting environmental problems [17]. With POPs leaking into the environment, earthworms can easily take up these pesticides, leading to POP bioaccumulation in organisms up the food chain, specifically affecting bird populations in the area. DDT bioaccumulation impacts embryos through eggshell thinning, reduced chick hatchability, skeletal abnormalities, and compromises the function of the nervous and reproductive systems. Effects on adult birds include acute mortality, accumulative stress levels, reduced fertility, suppression of egg formation, and impaired incubation and chick-rearing behaviors [18]. Some wildlife bird predators experiencing local extinction include peregrine falcons, cormorants, and eagles [17]. Farm livestock is also impacted by organochlorine compounds, which can easily accumulate in fat-rich foods such as meat and milk due to their liposolubility [19]. Cattle consume these organochlorine compounds when drinking contaminated water or feeding on dried hay [4]. According to a study on milk and dairy product monitoring in India, HCH residue levels were lower than DDT levels, with DDT levels in butter being the highest among other products. Thankfully, all organochlorine pesticide levels in those products were within the limits set by the WHO/FAO [20]. Continuous exposure of honeybees to pesticides also affects the quality of honey, with the treatment of beehives with pesticides being the most direct route of honey contamination [21]. Wild animals, including the Greater Cane rat (*Thryonomys swinderianus*), which serves as a source of protein, are seriously affected by pesticide contamination, especially since the people of Ghana consider it a food source [22]. Therefore, humans are bound to take up these compounds in their bodies as they continue to consume meat, milk, and crops.

5. Treatment

5.1. Granular activated carbon.

An effective method for pesticide removal involves the use of granular activated carbon (GAC). The mechanism of this method is simple: place a suitable amount of GAC into the water and let the GAC adsorb the pesticide. In a 2010 experiment, Traylor K-81/B GAC was used in the public water supply system in Belgrade, where pesticide removal reached up to 99%, reducing the starting concentration from $2 \mu\text{g}/\text{dm}^3$ to just $0.02 \mu\text{g}/\text{dm}^3$ when virgin GAC was used [23]. Besides the high removal efficiency, the saturated GAC can be regenerated and reused. During regeneration, contaminants are transformed into sublethal by-products, and the sorption capacity of the carbon is re-established, thereby increasing the effective lifespan of the GAC and decreasing the costs of water treatment [23]. However, while the adsorption affinities for virgin and regenerated activated carbon in the experiment were similar, the pesticide concentration in the effluent from the regenerated GAC column was higher than the effluent from the virgin GAC. This is due to incomplete regeneration and surface deterioration during heating. Moreover, the pollutants are not actually destroyed in the process but transferred to another medium, corresponding to the transformation into sublethal by-products mentioned earlier. The adsorption efficiency also relates to the surface properties and porosity of GAC, as well as the chemical properties and geometry of the pollutants. The study showed that while HCHs were easily rinsed from the saturated column containing virgin GAC, DDT derivatives were more tightly bound to the GAC column, hinting that pesticide solubility in H_2O is in diametrically opposed correlation to the adsorption affinity of the GAC. The efficiency of regenerated GAC is also dependent on the rinsing efficiency [23].

5.2. Magnetic nanospheres coated with polystyrene.

Organochlorine pesticides can also be removed using Magnetic Nanospheres Coated with Polystyrene. The mechanism of this method is as follows: place a suitable amount of the sorbent ($\text{Fe}_3\text{O}_4@\text{PS}$) into the water and let it adsorb the pesticide. After the rapid adsorption of the pesticide onto the exterior surface of $\text{Fe}_3\text{O}_4@\text{PS}$, equilibrium on the exterior surface causes the pesticide molecules to enter the pores of the polystyrene and then be adsorbed by the interior surface of the polystyrene pores. In a 2014 experiment, $\text{Fe}_3\text{O}_4@\text{PS}$ was used and showed a removal efficiency of 93.3%, 96.2%, 96.5%, and 95.9% for lindane, aldrin, dieldrin, and endrin, respectively, in the treatment of actual water samples, with adsorption equilibrium achieved in less than 20 minutes [24]. Apart from the high removal efficiency, this technology performs better at removing organic pollutants when treating organochlorine pesticides at the ng/mL level compared to removal using activated carbon [25]. This may be due to the fact that the adsorption capacity of the chemical compounds using this method is relatively lower. In addition, an increase in treatment time does not linearly correlate to removal efficiency, due to an increase in the mass of pesticides adsorbed in the pores causing more diffusion resistance, thus decreasing the diffusion rate. It is noteworthy that due to the polystyrene coating on the outer surface, the magnetization of the Fe_3O_4 nanoparticles will be slightly reduced [24]. The concept of this method transferring the pollutant from one medium to another implies that an additional transformation process of the pollutant into other sublethal by-products will be required, similar to the GAC adsorption.

5.3. Hydrogen peroxide/UV.

Hydrogen peroxide is also a common substance used in Organochlorine Pesticide (OCP) removal. Table 3 shows the standard reduction potentials in an aqueous medium of various oxidizing agents. In a 2018 experiment, the removal of OCPs using H_2O_2 was determined with varying variables such as pH, temperature, H_2O_2 concentration, initial pesticide concentration, and total treatment time [26]. The mechanism of this method is as follows: The first stage of photolytic degradation, which is the most crucial part in determining the success of this method, involves the initial formation of hydroxyl radicals ($\bullet\text{OH}$) [27].

Table 3. Standard reduction potentials in aqueous medium of various oxidizing agents [28].

Oxidiser	Reduction reaction	E/V
Fluorine	$\text{F}_2(\text{g}) + 2\text{H}^+ + 2\text{e}^- \rightarrow 2\text{HF}$	3.05
	$\text{F}_2(\text{g}) + 2\text{e}^- \rightarrow 2\text{F}^-$	
Hydroxyl radical	$\text{OH} + \text{H}^+ + \text{e}^- \rightarrow \text{H}_2\text{O}$	2.80
Sulphate radical anion	$\text{SO}_4^- + \text{e}^- \rightarrow \text{SO}_4^{2-}$	2.60
Ferrate	$\text{FeO}_4^{2-} + 8\text{H}^+ + 3\text{e}^- \rightarrow \text{Fe}^{3+} + 4\text{H}_2\text{O}$	2.20
Ozone	$\text{O}_3(\text{g}) + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{O}_2(\text{g}) + \text{H}_2\text{O}$	2.08
Peroxodisulphate	$\text{S}_2\text{O}_8^{2-} + 2\text{e}^- \rightarrow 2\text{SO}_4^{2-}$	2.01
Hydrogen peroxide	$\text{H}_2\text{O}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow 2\text{H}_2\text{O}$	1.76
Permanganate (a)	$\text{MnO}_4^- + 4\text{H}^+ + 3\text{e}^- \rightarrow \text{MnO}_2(\text{s}) + 2\text{H}_2\text{O}$	1.67
Hydroperoxyl radical (a)	$\text{HO}_2 + 3\text{H}^+ + 3\text{e}^- \rightarrow 2\text{H}_2\text{O}$	1.65
Permanganate (b)	$\text{MnO}_4^- + 8\text{H}^+ + 5\text{e}^- \rightarrow \text{Mn}^{2+} + 4\text{H}_2\text{O}$	1.51
Hydroperoxyl radical (b)	$\text{HO}_2 + \text{H}^+ + \text{e}^- \rightarrow \text{H}_2\text{O}_2$	1.44
Dichromate	$\text{Cr}_2\text{O}_7^{2-} + 14\text{H}^+ + 6\text{e}^- \rightarrow 2\text{Cr}^{3+} + 7\text{H}_2\text{O}$	1.36
Chlorine	$\text{Cl}_2(\text{g}) + 2\text{e}^- \rightarrow 2\text{Cl}^-$	1.36
Manganese dioxide	$\text{MnO}_2 + 4\text{H}^+ + 2\text{e}^- \rightarrow \text{Mn}^{2+} + 2\text{H}_2\text{O}$	1.23
Oxygen	$\text{O}_2(\text{g}) + 4\text{H}^+ + 4\text{e}^- \rightarrow 2\text{H}_2\text{O}$	1.23
Bromine	$\text{Br}_2(\text{l}) + 2\text{e}^- \rightarrow 2\text{Br}^-$	1.07

(a) Circumneutral or weakly acidic medium; (b) Strongly acidic medium.

After the formation of highly reactive $\bullet\text{OH}$ radicals, they react with the organic compound (in this case, the OCP) through the abstraction of a hydrogen atom, addition to $\text{C}=\text{C}$ double bonds, or electron transfer. The reaction mechanisms depend on the nature and functional groups of the molecule, though the abstraction of a hydrogen atom is more likely, leading to the formation of the organic radical $\text{R}\bullet$ and subsequent rapid reaction with dissolved O_2 to form the peroxide organic radical $\text{RO}_2\bullet$ [29]. The organic radicals decompose through bimolecular reactions, giving rise to various types of degradation products. The safety of H_2O_2 utilization is enhanced by the lack of formation of treatment by-products, unlike chlorination and ozonation. Consequently, there is a minor concern about H_2O_2 pollution since a readily available natural purification system already exists. Not only does it slowly decompose in the presence of light, but H_2O_2 is also a metabolite of many organisms, and those organisms can decompose H_2O_2 into oxygen and water. H_2O_2 solubility in water allows the reaction to occur more easily. Moreover, by adjusting the conditions of the reaction (the aforementioned variables), H_2O_2 can often be made to oxidize preferred pollutants or even favor different oxidation products from the same pollutant. This attribute is extremely useful to achieve certain levels of specified organic matter in water. Other advantages of using H_2O_2 include commercial availability and easier on-site storage [26].

The disadvantages of this technology include the fact that H_2O_2 only works in acidic water, with the optimal pH in the experiment being 3, and does not perform any pesticide removal in neutral or alkaline conditions [26]. Additionally, this technology is only applicable to waters containing photosensitive compounds and with low levels of Chemical Oxygen

Demand (COD) [30]. On the other hand, the process may be inhibited if high levels of organic compound concentrations are found in the targeted wastewater for treatment. Moreover, the presence of high concentrations of H_2O_2 may have a scavenging effect on the hydroxyl radicals and, hence, may impair the effectiveness of the oxidation process. This is in line with the results from the experiment, whereby an increased H_2O_2 dose led to lower removal percentages [26]. Consequently, the initial H_2O_2 concentration must be carefully regulated to enhance removal efficiency. Furthermore, H_2O_2 is an expensive reagent that increases the total operating costs of the process [31]. Considering the importance of the first stage of photolytic degradation, the aforementioned variables related to the UV light source and wastewater properties must be carefully adjusted and monitored to maximize the efficiency of the process in line with the desired target.

6. Challenges

6.1. Outdated and 'country-specific' standards.

The standards and guideline values for POPs in surface water and groundwater in China do not align with worldwide standards. The existence of water quality regulations is to encourage and urge effective water treatment and maintenance technologies, and these regulations should reflect the most up-to-date information regarding any risks to mankind and the environment. The idea of 'country-specific' risks is absurd, as the risks mentioned are equally possible to occur at any place and at any time. Therefore, allowing orders of magnitude higher concentrations in China compared to other countries cannot be justified [6]. Moreover, the number of listed pollutants under the current groundwater quality standard in China is relatively few, particularly compounds related to organic pollution. Any studies based on this outdated standard would result in the negligence of contamination by other unlisted pollutants, as reflected in the groundwater pollution surveys in 2010 and 2015 [6].

6.2. Illegal disposal of POPs.

While the introduction and updating of water pollution prevention laws and discharge standards are crucial steps, their effectiveness hinges on the government or local authorities having sufficient manpower and funds to enforce these regulations and oversee related activities in the area. It will be in vain if there is a lack of resources dedicated to ensuring compliance. More efforts should be directed towards restraining the widespread problem of illegal wastewater discharge, beginning with discharge into important water sources and aquifers and then gradually extending control to more rural areas. Introducing stricter laws and imposing heavier punishments for violators of pollution laws, such as substantial fines or criminal charges, or a combination of both, can serve as deterrents [6].

4. Conclusions

The quality of our environment directly impacts the quality of life. The increasing global production and usage of organohalogen result in more discharge into aquatic environments, circulating in our hydrological cycle indefinitely. Exposure to these POPs not only poses detrimental effects on human health but also affects fauna, such as bird populations and farm livestock. Direct consumption of the latter or its derivatives can lead to the bioaccumulation of POPs in the human body. Effective POP treatment methods vary widely, with adsorption techniques emerging as a more economical option. However, these methods still require

additional transformation processes for pollutants to become less toxic. On the other hand, the use of H₂O₂ and O₃ in POP treatment is considered a more direct method of biodegrading POPs, though extensive monitoring of operational conditions is necessary to ensure success or maximize efficiency. Efforts to reduce POP pollution face challenges such as outdated water quality standards and the list of dangerous pollutants within those standards. Additionally, failure to address illegal disposal issues hampers these reduction efforts. In short, the combination of rapid industrialization and a lack of environmental regulatory oversight raises concerns for both mankind and the environment. Utilizing existing POP treatment technology is crucial in these reduction efforts, and further studies on technology improvement and identification of existing or yet-to-be-discovered organic pollutants will be necessary.

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Competing Interest

All the authors declared no conflict of interest.

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