Review



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*Review*

# Disinfection Byproducts and Related Compounds in the Coastal Zone: A Preliminary Compilation

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**Abstract:** Disinfection byproducts (DBPs) are formed whenever disinfectant ingredients are in contact with organic matter. Due to widespread increase in production and use of disinfectants and antiseptics, the presence of their components in coastal environments is increasing. However, very limited scientific literature is available in regard to the occurrence of DBPs and related compounds in the coastal zone and particularly in seawater. In this context, the aim of this preliminary compilation is to outline the main up-to-date findings and raise awareness on their increasing occurrence in coastal environments. Sampling and analysis of marine samples in the USA, Spain, China, Taiwan, Malaysia, Korea, Kuwait and Hong-Kong revealed the presence of several categories of DBPs and related compounds in coastal environments, with the highest levels being detected in urban areas, probably related to the nearby wastewater treatment plants. The investigations considered in this preliminary compilation, report the detection of DBPs and related compounds as well as the formation of toxic brominated and iodinated DBPs in high levels due to chlorination of wastewater, especially after use of seawater for flushing in toilets. New DBP groups have also been detected, namely halogenated phenolic DBPs and halopyrroles.

**Keywords:** DBPs; synthetic antimicrobials; personal care products; seawater; coastal environment

## 1. Introduction

Disinfection byproducts (DBPs) are frequently detected in environmental samples, due to the widespread use of disinfectants and the release of untreated and treated wastewater into the environment (McCance et al., 2018, Ojemaye et al., 2019, Riviera-Utrilla et al., 2013, Salimi et al., 2019). Increase of global sales of disinfectant chemicals has been estimated to reach 3% annually by 2050 (Massey et al., 2013), resulting in further increase of their DBPs that end up in costal environments. Synthetic antimicrobials are also frequently used in personal sanitizing and cleaning products and personal care products (PCPs) such as soaps, antiseptics, bath foams, and detergents namely parabens, triclosan (TCS) and triclocarban (TCC). Other ingredients include polyethoxylate alcohols (AEOs), polyethylene glycols (PEGs), phthalates and chlorides (Chen et al., 2020, Eganhouse et al., 1983, Farzana et al., 2020, Hayman et al., 2021, Kueh et al., 2008, Lee et al., 2019, Moeris et al., 2020, Smith et al 2015, Traverso-Soto et al., 2014). The occurrence of these pollutants in the coastal zone can have adverse effects on biodiversity balance, degrade water quality and constitute a human health risk via human exposure. Research has already revealed the possible cancer risks and reproductive problems related to DBPs (Dodds et al., 1999, Hrudey et al., 2015, King et al., 1996, Richardson et al., 2007, Feng et al., 2017, Hong et al., 2017, Krasner et al., 2009, Le Roux et al., 2017, Sun et al., 2009, Zhu et al., 2016).

In several countries, the use of seawater for domestic and public toilet flushing has been adopted in order to reduce freshwater demand. However, wastewater deriving from seawater contains higher concentrations of bromide and iodide ions, which are oxidized during chlorine disinfection, forming brominated and iodinated species of DBPs, which are toxic for the environment (Ding et al., 2013, Gong et al., 2014, Liu et al., 2014, Yang et al., 2013, Yang et al., 2015). Since wastewater treatment plants are located, in most cases, in coastal areas with commercial seashores and ports, further than

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environmental concerns, risks are posed also for swimmers, via dermal contact, inhalation and swallowing during swimming. While numerous research papers have dealt with DBPs and related compounds formation in drinking water, very limited scientific literature is available in regard to their occurrence in the coastal zone and in particularly in seawater. In this context, the aim of this preliminary compilation is to outline the main up-to-date findings and raise awareness on the increasing occurrence of DBPs and related compounds in coastal environments.

## 2. DBPs and related compounds (synthetic antimicrobials, personal care products) in the coastal zone

### *2.1. AEOs & PEGs in USA and Spain*

AEOs and PEGs concentrations detected in seawater were lower than 1 μg/L. Wastewater concentrations in the same area were 31.2 μg/L AEOs and 39.8 μg/L PEGs in the influent and respectively 0.16 μg/L and 1.6 μg/L in the effluent. (Lara-Martin et al., 2014, Wang et al. 2022). PEGs were present in both untreated and treated wastewater, probably because they are metabolites of AEOs, but they also frequently occur as constituents in a large number of personal care products (soaps, bathing foams, cleansing agents). Wastewater treatment as well as dilution in seawater seems to reduce their concentrations, however they are not completely eliminated from seawater, resulting in possible transformations as well as potential long-term environmental risks.

### *2.2. Phthalate esters and methyl-paraben in seawater in Southern China*

The average concentration of phthalate esters in seawater in Southern China Gulf was 3.27 μg/L, i.e. lower than global marine water quality criteria – MWQC. On the contrary, the average concentration of benzyl-butyl-phthalate in seawater was 1.18 μg/L, a value exceeding European MWQC, therefore indicating the need for preventive measures in regard to this type of marine pollution (Farzana et al., 2020).

Methyl-paraben was the main paraben species detected in seawater, with average concentration 4.87 ng/L (Chen et al., 2007, Penrose and Cobb 2022). According to NICNAS 2016 (National Industrial Chemicals and Assessment Scheme in Australia), the maximum environmentally acceptable concentration for methyl-paraben is 20 μg/L.

### *2.3. PCPs and chlorinated analogues in Taiwan and Malaysia*

PCPs and their chlorinated analogues were analyzed in a surface water stream receiving treated wastewater as well as in receiving seawater. The results are comparatively presented in Table 1. Their concentrations in stream water samples were significantly higher than those in the seawater samples, probably due to dilution. All compounds were present, both in stream water and in seawater, except for Cl-methyl-paraben, which was detected only in stream water. The highest levels of all compounds, except for Cl-methyl-paraben and triclosan, occurred in the sampling point located nearest to the urban area with a hospital (Chen et al., 2020, Nazifa et al., 2020). PCPs-were detected in wastewater, with aromatic compounds being more abundant in untreated was 100 ng/L, except for methyl-paraben and triclosan. The primary and secondary wastewater treatment procedures, including chlorination, resulted in the removal of most of these toxic pollutants, namely more than 95% for propyl-paraben, 73.9% for butyl-paraben, 84.0% for methyl-paraben and 53.1% for triclosan (Chen et al., 2020). Chlorination was not the main process in the treatment plant and the removal percentages due to chlorination were lower than 30%, except for butyl-paraben and ethyl -paraben, for which 132% and 47.1% removal percentages were reported respectively. In regard to transformation products, only 2Cl-propyl-paraben was detected in samples from all sampling locations at frequencies of detection ranging from 57.1 to 71.4%. The highest concentration was detected, as expected, in influent wastewater samples, and, after primary and secondary treatment processes, decreased by 23.8 times. Moreover, 2Cl-propyl-paraben was frequently detected in the river, a fact suggesting the existence of additional unknown sources contributing to its levels in the samples. Nevertheless, primary and secondary treatment removed 95.8% of 2Cl-propyl-paraben.

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Butyl-paraben levels were negligible. The concentrations of PPCPs ethyl paraben, methyl paraben and propyl paraben were ten times higher in wastewater compared to seawater. Triclosan levels ranged from thousands ng/L near the exit of the wastewater treatment plant to not detectable concentrations in seawater. Triclosan is more vulnerable in seawater than in freshwater due to photo degradation, especially in tropical zone directly receiving solar energy. Therefore, dilution and photo degradation in seawater result in not detectable levels of triclosan in the area studied.

**Table 1.** Range of average concentrations of PCPs and chlorinated analogues in surface water and seawater (nd: not detected) (Chen et al., 2020, Nazifa et al., 2020, Wee et al., 2019).

| **Compound** | **C(ng/L) in water stream** | **C (ng/L) in receiving seawater** |
|---|---|---|
| Butyl paraben | nd-71,1 | nd |
| Ethyl paraben | 503-1810 | 115-228 |
| Methyl paraben | 5.75-9400 | 2.25-1030 |
| Propyl paraben | 369-4810 | 25-314 |
| Triclosan | 1010-2560 | 3.14-20.28 |
| 2Cl-Methyl paraben | nd-13400 | 645-2180 |
| 2Cl-Propyl paraben | 299-1430 | 13.5-77.8 |
| Cl-Propyl paraben | nd-2480 | nd-2.20 |

The highest concentrations of PPCPs were detected in an area with width 1.5 m and depth 2 m, with 30 +/- 5 cm water in continuously sunny days. Water in this shallow channel mainly contained hospital wastewater.

PPCPs and TPs showed different chemical profiles in seawater and in channel water. The main compounds were ethyl-paraben, methyl-paraben, propyl-paraben, and triclosan, Cl-methyl-paraben, 2Cl- propyl-paraben and Cl-propyl-paraben. Their concentrations in the channel were significantly higher than in seawater, probably due to dilution. All compounds were detected both in seawater and in channel water, except for Cl-methyl-paraben, which was detected only in channel water.

Among all sampling points, the highest levels of PPCPs and TPs, except for Cl-methyl-paraben and triclosan, occurred in the urban are with the hospital.

Despite the fact that research in wastewater and natural water has shown seasonal variations for these pollutants due to higher photochemical and biological degradation during summer or during runoff in rainy periods (Borruli et al., 2019, Nam et al., 2014, Padhye et al., 2014, Pal et al., 2014), in the study of Chen et al., 2020, no such trends were observed. Taiwan and Malaysia have tropic and oceanic climate characterized by high temperatures and rainfalls throughout the year.

### *2.4. Parabens and antimicrobials in the coastal zone of Korea*

The concentrations of parabens, TCS and TCC in sediment samples from the coastal zone of Korea are presented in Table 2 (Lee et al., 2019).

Methyl-paraben was detected in all sediment samples that were tested, a fact revealing the range of sediment pollution in the coastal zone. The percentages of detection of ethyl-paraben, propyl-paraben and butyl-paraben were 36%, 34% and 26% respectively. Similar concentrations of methyl-paraben were measured in marine sediments in Florida, USA (Xue et al., 2016), Tokyo, Japan (Liao et al., 2013) and in Chinese river sediments (Peng et al., 2017, Liu et al., 2015). TCC concentrations were

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higher and TCS lower than those measured in Spain (Gorga et al., 2015) USA, (Katz et al., 2013), Italy (Casatta et al., 2015) and China (Peng et al., 2017).

Lee et al. (2019) reported the concentrations of parabens, TCS and TCC in sediments from Korean coastline. Methyl paraben was detected in all sediment samples, a fact revealing the range of coastal environment pollution in the area. The percentages of detection of ethyl paraben, propyl paraben and butyl paraben were 36%, 34% and 26% respectively. The levels of methyl-paraben in the sediments measured were similar to those in Florida (US) (Xue et al., 2016), Tokyo (Japan) (Liao et al., 2013) and the rivers Pearl and Jiangje of China (Peng et al., 2017, Liu et al., 2015). Methyl-paraben concentrations, in river sediments in China, however, have shown higher levels (Zhang et al., 2015, Huang et al., 2018). Ethyl and propyl paraben levels were lower compared to those in other countries.

In regard to the distribution of parabens in sediments, the highest levels occurred along the eastern coast of Korea, while the lowest in sediments of western part of Korean coastline. Pollution from parabens is directly related to wastewater release and to wastewater treatment plant effluents (Liao et al., 2013, Haman et al., 2015). However, the eastern coast of the country where higher levels of paraben occurred, had the lowest percentage of wastewater treatment operation (64.8%). In contrast, in the western part of Korea, the most wastewater treatment plants with a total function percentage (90%).

### *2.5. DBPs and related compounds in Kuwait seawater*

Smith et al. (2015) conducted measurements of DBPs and related compounds in seawater from Kuwait after phenomena of fish deaths associated with a release of non-treated wastewater. The results are presented in Table 3. The levels of phthalates and terpineol were particularly high, while marine pollution from methyl and propyl- parabens, chloroxylenol, dimethylphenol, biphenyl ether as well as bromoform was considerable. These results indicate the need for safeguarding coastal water quality from increased levels of DBPs and related pollutants contained in wastewater that eventually enter the sea.

**Table 2.** Concentrations of parabens and antimicrobials (ng/L) detected in coastal sediments in Korea (nd: not detected) (Lee et al., 2019).

| Compound | min | max | average |
|---|---|---|---|
| Methyl paraben | 0,13 | 11,2 | 2,3 |
| Ethyl paraben | nd | 0,08 | 0,02 |
| Propyl paraben | nd | 0,1 | 0,01 |
| Butyl paraben | nd | 0,07 | 0,02 |
| Benzyl paraben | nd | 0,06 | 0,01 |
| Heptyl paraben | nd | 0,02 | 0,01 |
| Triclosan | nd | 41 | 2,78 |
| Triclocarban | nd | 47 | 3,25 |

**Table 3.** DBPs and related compounds concentrations in seawater receiving wastewater (nd: not detected) (Al Gazali, Kuwait) (Smith et al., 2015).

| Compound | Concentration in seawater (ng/L) |
|---|---|
| Bromoform | 10.0-20.0 |
| Dimethyl-phenol | 20.0-80.0 |
| Chloroxylenol | nd-300.0 |
| Fluoranthene | nd-10.0 |

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| | |
|---|---|
| Triclosan | nd-20.0 |
| Methyl paraben | nd-200.0 |
| Propyl paraben | nd-160.0 |
| Terpineol | 4,800-12,000 |
| Biphenylether | nd-30 |
| Phthalates | 1,500-4,000 |

*2.6. DBPs and organic pollutants in Hong-Kong coastal zone*

Organic pollutants in Hong Kong coastal water occurred in very low levels. PAHs, TBTs, HCH and phenols were investigated, as well as nonylphenol (NP) and NP ethoxylates which are being frequently used in domestic cleaning agents, such as detergents, foams, and surface disinfectants.

NP was detected only in two locations; Deep Bay and Tolo Harbour, in low levels, while NP ethoxylates were additionally detected in Victoria Harbour. TBTs occurred generally in very low levels. Dioxines and furans were present in all samples of Hong Kong sediments, in low concentrations.

PAHs concentrations in marine sediments ranged from110-769 ng/g dw, while their maximum levels occurred in the center of Victoria Harbour. TBTs ranged among 0.60-10.7 ng/g dw, but they only occurred in less than 15% of the samples, while their maximum levels were also detected in Victoria Harbour. The levels of all above pollutants in Hong Kong sediments were much lower than their levels in other countries, such as Vietnam (Midorikawa et al., 2004), Japan (Baasansuren et al., 2004) and Malaysia (Sudaryanto et al., 2004).

HCH concentrations were very low, 0.14-0.45 ng/g, which was expectable due to restriction of some isomers of HCH in Hong Kong since 1991. Phenol was only detected in Tolo Harbour and in Inner Bay, where also NP and NP ethoxylates were detected were in values 122-2900 ng/g dw and 71-1680 ng/g dw respectively. Higher levels occurred in Jynk Bay and in Victoria Harbor, a fact denoting that the particular areas are more heavily affected from domestic release.

It can be considered that from all pollution sources, wastewater, industrial release and solid waste runoffs were transferred to treatment, while river water, rainwater and wastewater treatment effluents eventually entered seawater.

The levels of dioxins, furans and TBTs were comparable to those in seawater. In river water and in wastewater treatment plant effluent PAHs were not detectable, while in rainwater they were detected in trace concentrations These results indicate that the soil based releases could not be the main source of these compounds in the marine environment.

Moreover, phenols, NP and NP ethoxylates were found in the pollution sources in significantly higher levels than those of local seawater. Untreated wastewater contained the higher levels of phenols which are frequently found in domestic, commercial and industrial products. River water contained only trace levels of phenol, therefore it is not a considerable pollution source.

In river sediments and wastewater sludge, dioxins and furan were present in levels comparable to those in marine sediments. NP, NP ethoxylates, HCH, TBTs, seemed significantly accumulated in wastewater sludge, rather than the river sediments.

Recent research in the coastal zone of Hong-Kong (Feng et al., 2019) has focused on the identification of new categories of DBPs that are formed during chlorination of seawater-based wastewater. This kind of wastewater is produced from the use of seawater for toilet flushing, in an effort to reduce freshwater demand. However, the different properties of seawater compared to freshwater, can enhance the formation of different categories of DBPs. Further than trihalomethanes (THMs), haloacetic acids (HAAs) and haloacetonitriles (HANs), new categories include trihalophenoles (THPs) and halocarbazoles (HCZs), while the speciation of the compounds detected shifts to more brominated analogues and iodinated DBPs are also formed (Yang et al., 2013, Zhao et al., 2014, Xu et al., 2017, Grote et al., 2022 Liu et al., 2022).

Recent literature has reported that iodinated DBPs exhibited significantly higher developmental toxicity and growth inhibition compared to their brominated and chlorinated counterparts, and that

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halogenated phenolic DBPs exhibited significantly higher toxicity and growth inhibition than halogenated aliphatic DBPs (Yang et al., 2013, Liu et al., 2014). A new category of DBPs includes iodinated -trihydroxybenzosulfidic acids which belong to "iodinated phenolic DBPs", with higher toxicity and environmental persistence being of particular interest. According to the results of analysis performed by Feng et al. (2019), iodinated DBPs were not detected in the chlorinated wastewater effluents after use of seawater in toilets, and this was attributed to the fact that they could be formed at a very early stage and fully decompose thereafter, due to the reaction both with chlorine and chloramines. Halocarbazoles levels were also not detectable during this research. However trichlorophenol and tribromophenol were detected at levels up to 1000 ng/L in chlorinated saline wastewater effluent samples.

Yang et al. (2014) investigated another new category of DBPs, halopyrroles, and the factors that can affect their formation: chlorine dose, contact time, temperature and pH. Their research outlined that relatively low chlorine doses, 6 and 10 mg/L enhanced halopyrrole formation. Increase of chlorine dose from 10 mg/L to 20 mg/L, resulted in decrease of the levels of tetrabromopyrrole, tribromochloropyrrole, 2,3,4-tribromopyrrole and 2,3,5- tribromopyrrole by 92.2%, 88.9%, 79.9% and 97.3% respectively. The authors attributed this fact to decomposition of the pyrrolic ring due to chlorine. The concentrations of tetrabromopyrrole measured in chlorinated saline wastewater after chlorine dosages 6, 10, 15 and 20 mg/L were 0.74, 0.69, 0.14 και 0.08 mg/L respectively (Yang et al., 2014).

The precursors of halopyrroles were also investigatesd by Yang et al. (2014). Tetrapyrroles, with molecules containing four pyrrolic rings, were considered to be the major precursors. Tetrapyrroles are compounds of particular importance for the ecosystems, as they contain chlorophylls, aimins and porfyrins, essential metabolites for almost all living organisms, which for the same reason are present in effluent wastewater samples. Therefore, the basic hypothesis for the formation of halopyrroles is that, in presence of bromine and iodine, chlorine reacts with such pyrrole containing compounds, creating halopyrroles.

As tetrabromopyrrole (Figure 1) was the dominant compound of this category, its developmental toxicity was assessed, with the use of the species P. dumerilii. The results have shown that tetrabromopyrrole exhibited the maximum developmental toxicity as it was 460 and 8805 times more toxic than bromoform and bromoacetic acid. Taking into account that the concentrations of tetrabromopyrrole in the chlorinated saline water samples ranged at levels 1/10 to 1/20 of those of bromoform and bromoacetic acid respectively, the risk posed by tetrabromopyrrole for the marine environment is 10 to 100 times higher than that of bromoform and bromoacetic acid (Yang et al., 2014).

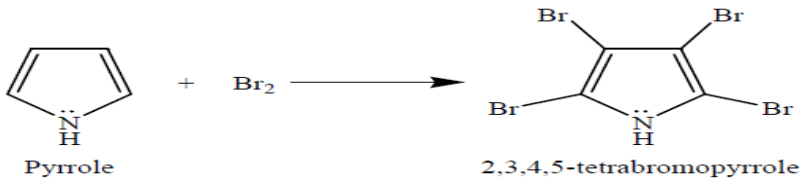


**Figure 1.** Formation of 2,3,4,5-tetrabromopyrrole (Sky of Chemistry, 2020).

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