

Effect of Wastewater from the in-water Cleaning Process of Ship Hull on Marine Organisms - A Review

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Over the past decade, there has been global expansion in the advancement of underwater cleaning technology for ship hulls. This methodology ensures both diver safety and operational efficiency. However, recent attention has been drawn to the harmful effects of ship hull-cleaning wastewater on marine animals. It is anticipated that this wastewater may have various impacts on a wide range of organisms, potentially leading to population- and ecosystem-relevant alterations. This concern is especially significant when the wastewater affects functionally important species, such as aquaculture animals and habitat-forming species living in coastal regions, where underwater cleaning platforms are commonly established. Despite this, information on the ecotoxicological effects of this wastewater remains limited. In this mini review, we discuss the adverse effects of wastewater from in-water cleaning processes, as well as the current challenges and limitations in regulating and mitigating its potential toxicity. Overall, recent findings underscore the detrimental effects posed by sublethal levels of wastewater to the health status of aquatic animals under both acute and chronic exposure.

Keywords: In-water cleaning, Wastewater, Antifoulant, Ecotoxicity, Marine animal

Introduction

The adhesion and accumulation of various aquatic organisms on submerged ship hulls, known as biofouling, result in substantial economic losses when ships remain in the water for extended periods (Konstantinou and Albanis, 2004). This is primarily due to the increased hydrodynamic resistance of vessels, leading to heightened fuel consumption, higher exhaust emissions, increased operational costs, and reduced system efficiency (Schultz et al., 2011; Lindholdt et al., 2015). In addition, enlarged fouled hull can directly affect the vessel's stability and structural integrity. The biofouling process is complex, involving dynamic shifts from the formation of thin biofilms to the aggregation of macrofouling communities, along with intricate interactions between fouling organisms and environmental factors (Raeid et al., 2019). Previous research has indicated that even the slime layer within a biofilm

can significantly alter vessel performance (Schultz et al., 2011). In addition, there is considerable concern regarding the potential environmental impact on local and endemic biodiversity, as shipping can inadvertently transport invasive alien species (Davidson et al., 2009). Consequently, significant progress has been performed in the development of diverse antifoulants aimed at diminishing biofouling, many of which are appended into antifouling paints.

A diverse range of substances, such as metals, biocides, booster biocides, pigments, solvents, and their combinations, have been widely employed as antifoulants in antifouling paints to prevent biofouling on ship surfaces (Yebra et al., 2004; Thomas and Brooks, 2010; Soroldoni et al., 2017). Antifoulants are essential in reducing friction, preventing corrosion of fittings, and decreasing fuel consumption in vessels by preventing the settlement of marine organisms on the ship's surfaces (Konstantinou and Albanis, 2004). In the past, organotin compounds such as tributyltin (TBT) were widely used

in antifouling paints (Omae, 2003). However, due to their persistence in water, significant toxicity to non-target organisms, and the unpredictable reproductive effects they caused (such as imposex identified in several mollusks), the global use of organotin in antifouling compounds was banned in 2008 (Alzieu, 2000; Evans et al., 2000). As an alternative, copper and metal-based antifouling agents such as copper pyrithione (CuPT) and zinc pyrithione (ZnPT) have been widely used as booster biocides in antifouling compounds (Yebra et al., 2004). Moreover, various other biocides or booster biocides, such as chlorothalonil, 4,5-dichloro-2-n-octylisothiazolin-3-one (DCOIT, Sea-Nine 211), dichlofluanid, diuron, 2-(tert-butylamino)-4-(cyclopropylamino)-6-(methylthio)-1,3,5-triazine (Irgarol-1051), 2,3,5,6-tetrachloro-4-methylsulphonyl (TCMS) pyridine, thiocyanatomethylthio-benzothiazole (TCMTB), and zineb, have been developed to diminish various fouling organisms, effectively preventing biofouling. However, these compounds are recognized as significant sources of aquatic pollutants because they have been consistently detected in the water column, sediments, and organisms, and are associated with toxicity (Yebra et al., 2004; Thomas and Brooks, 2010; Park et al., 2016; Chen and Qian, 2017; Amara et al., 2018). Although certain booster biocides have shown relatively low toxicity to non-target organisms due to their photo-degradable characteristics and short half-lives in aquatic ecosystems (Jacobson and Willingham, 2000; Sakkas et al., 2007), consistently reported potential adverse effects on numerous aquatic organisms have raised public concern. It is predicted that the synthesis, application, sale, and combining of biocides and booster compounds will significantly increase, resulting in elevated release and accumulation in aquatic environments (Torres and De-la-Torre, 2021).

Regular hull cleaning is essential for maintaining vessel performance and efficiency, as antifouling measures have limitations in preventing biofouling. By removing biofouling organisms, the process not only ensures optimal performance but also extends the lifespan of the ship, reduces fuel consumption, and minimizes the environmental impact by reducing emissions (Lindholdt et al., 2015). Additionally, regular in-water hull cleaning helps prevent the spread of invasive species, protecting delicate marine ecosystems and preventing damage to coastal infrastructure (Davidson et al., 2009). Several methods are available for cleaning ship hulls (Song and Cui, 2020). Traditional methods include manual scraping and brushing, which are effective but labor-intensive and time-consuming. Another common method is high-pressure water jetting in dry-dock, which employs powerful jets of water to remove fouling. Alternatively, underwater cleaning methodologies

have recently expanded worldwide (Song and Cui, 2020). In detail, mechanical brushes and scrapers can be attached to remotely operated vehicles (ROVs) or autonomous underwater vehicles (AUVs) for automated cleaning, reducing the need for human divers, ensuring diver safety, and minimizing the risk of hull damage. Additionally, there are environmentally friendly methods such as using specialized coatings to prevent marine growth or employing ultrasound technology to deter fouling without harming marine life.

Among the various methodologies for in-water cleaning, it is anticipated that the use of robots will expand, replacing the need for divers equipped with brushes to enter the water. While notable advancements in in-water cleaning technologies have been accomplished, insufficient interest has been directed towards capturing waste materials during the cleaning process, developing treatment technologies for removing toxic substances, and establishing standardized methodologies for the management of wastewater treatment on a global scale. Moreover, surveys on the potential wastewater toxicity in non-target animals are still lacking. Addressing this absence is pivotal for establishing agreement and regulations for the management of wastewater treatment. Therefore, it is imperative to consistently publish reports on wastewater toxicity in non-target animals. This will contribute to understanding the risks to marine organisms and support the management of effluent from hull-cleaning wastewater.

Measured components in hull-cleaning wastewater samples

In general, metals and several antifoulants were highly detected in wastewater samples. Since the 1980s, tin-based biocides have been strictly regulated, leading to the development of several alternative Cu and Zn-based antifouling formulations as possible replacements for TBT. These formulations are often combined with organic "booster" biocides to enhance their efficacy. This is because Zn and Cu have been extensively used as crucial antifouling components. For example, measurement of metal concentrations of particulate hull-cleaning wastewater collected from four ship hulls showed significant metal contamination, with Fe concentration being the highest at 55 mg l⁻¹, followed by Cu (8 mg l⁻¹) and Zn (4.7 mg l⁻¹) (Soon et al., 2021b). Concentration ranges for the four metals, Zn, Cu, Ba, and Fe in the aqueous phase of the wastewater were reported as 222~3820 µg l⁻¹, 63.7~365 µg l⁻¹, 30.3~86.6 µg l⁻¹, and 0.41~0.84 µg l⁻¹, respectively (Soon et al., 2021b). Of eight metals analyzed, Zn and Cu were the most

abundant metals in wastewater samples, with Zn measured over 4.5 mg l^{-1} and Cu over $24 \text{ } \mu\text{g l}^{-1}$ (Kim et al., 2024; Lee et al., 2024a, 2024b). The concentrations of these metals measured in previous studies far exceeded typical environmental levels found in Korean marine environments (Cu: $9.39 \text{ } \mu\text{g l}^{-1}$, Zn: $45.79 \text{ } \mu\text{g l}^{-1}$, Lee et al., 2018) and surpassed ecosystem protection criteria for metals in Korean seawater environments (Cu: $3.0 \text{ } \mu\text{g l}^{-1}$, Zn: $34 \text{ } \mu\text{g l}^{-1}$, Ministry of Oceans and Fisheries, 2018).

Although there is limited information on the measured concentration of organic biocides, recent reports showed that CuPT and ZnPT were the dominant compounds in wastewater samples, with CuPT measured over $123 \text{ } \mu\text{g l}^{-1}$ and ZnPT over $19 \text{ } \mu\text{g l}^{-1}$ (Kim et al., 2024; Lee et al., 2024a, 2024b). The notably higher concentration of CuPT compared to ZnPT can be attributed to the transchelation of ZnPT into CuPT (Thomas, 1999; Grunnet and Dahllöf, 2005). However, additional data on the chemical composition of wastewater and debris are needed to clarify their toxic effects on non-target organisms, as the underwater cleaning regions near coastal areas can continuously leach and release numerous toxic substances into the surrounding environment over time. Understanding the precise composition of the chemicals found in wastewater is essential for managing chemical toxicity based on their specific characteristics.

Contamination pathways

Antifoulant components can naturally be leached from ship hulls into the aquatic environment. Alternatively, a combination of these compounds can be released as crude debris or paint particles during in-water cleaning activities (Torres and De-la-Torre, 2021). Large amounts of a wide range of paint particles and debris in terms of shape, volume, and size are discharged directly into aquatic environments during physical cleaning activities, including jet spraying (high-pressure water blasting), grit blasting, and manual scraping. As a result, the primary components of paint particles, including metals and booster biocides, have been consistently detected in harbors and coastal areas (Thomas et al., 2002; Turner, 2010; Soroldoni et al., 2017). For example, around 75% of the biocides detected in marinas are estimated to originate from paint particles discharged during physical cleaning activities (Thomas et al., 2002). Metals, especially Cu and Zn, leaching from paint particles, have been identified as major sources of metal contamination in dock, harbor, and estuary sediments (Turner, 2010; Soroldoni et al., 2017; Torres and De-la-Torre, 2021). Particles of paint, varying in size and containing relatively high concen-

trations of metals (up to milligrams per liter), are directly released during the cleaning process of ship hulls (Soon et al., 2021a, 2021b). During the in-water cleaning process, despite the raw wastewater including crude debris being treated with automatic filtration within the platform, a substantial number of particles remained. This suggests that the current filtration system is experiencing certain mechanical or functional limitations probably due to the overload of wastewater and the limited volume of the filtration system. It is worth noting that nano-sized particles or plastics discharged from the filtration system can directly cause mortality and have detrimental effects on aquatic organisms. They can act as potential vectors, facilitating the entry of toxicants into their bodies (Moore, 2006; Mattsson et al., 2015). Unidentified chemicals, apart from metals and biocides that are routinely analyzed in wastewater samples, could also have contributed to the overall toxicity. As a result, releasing this filtered wastewater into seawater may lead to direct exposure to potentially harmful compounds and allochthonous communities that detach from the hull and enter the marine environment.

Effects of hull-cleaning wastewater on marine organisms

The ecotoxicological effects of paint particles and their components, such as metals and booster biocides, have been consistently studied in non-target marine animals (Bao et al., 2011; Guardiola et al., 2012; Chen and Qian, 2017; Amara et al., 2018). Previously, the significant toxicity of solid samples, such as paint fragments generated from dry-dock cleaning, has been highlighted (Singh and Turner, 2009; Rees et al., 2014). The deposit-feeder *Arenicola marina* exposed to paint particles demonstrated the accumulation of Cu through leachates (Turner et al., 2008). Direct ingestion of paint particles was confirmed in the bivalve *Mytilus edulis* with highlighting mechanism of particle discrimination (Turner et al., 2009). Ingestion of paint particles and subsequent lethal effects have consistently been reported in several benthic organisms, including the copepod *Paracalanus parvus* sl (Hyun et al., 2022), copepod communities (Molino et al., 2019), the benthic tanaid *Monokalliapseudes schubarti* (Soroldoni et al., 2020), the epibenthic amphipod *Hyaella azteca* (Soroldoni et al., 2020), the mysid *Neomysis awatschensis* (Lee et al., 2024a), the intertidal ragworm *Hediste diversicolor* (Muller-Karanassos et al., 2021), and the common cockle *Cerastoderma edule* (Muller-Karanassos et al., 2021). The accumulation of paint particles and their components can disrupt general metabolism, reduce detoxification capacity,

and decrease energy reserves, thus affecting the organism's ability to maintain homeostasis. Indeed, these animals play a crucial role in aquatic food webs by serving as intermediate prey species and food sources. Although they are non-target organisms for anti-foulants, they can be consistently exposed to paint particles and their components. This exposure occurs because their habitats are often near coastal regions, where hull cleaning procedures are commonly conducted. However, there is still limited information

available on the potential harmful effects of hull-cleaning wastewater and particles, particularly on non-target organisms.

Recently, studies have been conducted to measure the potential effects of the supernatant of wastewater collected from dry-dock cleaning or underwater cleaning procedures on several marine organisms (Table 1). Samples collected from dry-dock cleaning procedures clearly demonstrated detrimental effects on marine animals, and *in silico* modeling supported their toxicity (Soon et

Table 1. Studies on the effects of wastewater samples collected from cleaning in dry-docs or in-water cleaning procedures (updated from Lee et al., 2024b)

	Methodology	Test organism	Endpoint	Reference
Dry-dock cleaning	Hydroblast	Fish (embryo)	√ Mortality √ Developmental malformation √ Transcriptome analysis	Choi et al., 2020
	Hydroblast	Modeling and prediction	√ LC50 values for Cu and Zn retrieved from ECOTOX database	Soon et al., 2021a
	Hydroblast	Copepod	√ Egg hatching rate √ Mortality	Hyun et al., 2022
In-water cleaning	Diver cleaning with a hard brush	Modeling and prediction	√ LC50 values for Cu and Zn retrieved from ECOTOX database	Soon et al., 2021b
	Diver cleaning with a soft sponge or a hard brush	Microalgae	√ Growth √ Chlorophyll concentration √ Population composition	Lim et al., 2023
	Underwater robot cleaning	Copepod	√ Mortality √ Development √ Fecundity √ Transcriptome analysis	Park et al., 2023
	Underwater robot cleaning	Mysid	√ Ingestion of paint particle √ Mortality √ Enzymatic activity √ Chronic toxicity on growth and molting √ Reproduction √ Multigenerational toxicity	Lee et al., 2024a
	Underwater robot cleaning	Rotifer	√ Mortality √ Individual growth and life span √ Fecundity √ Enzymatic activity √ Population growth	Kim et al., 2024
	Underwater robot cleaning	Polychaete	√ Mortality √ Burrowing activity √ Cholinergic system √ Enzymatic activity	Lee et al., 2024b
	Underwater robot cleaning	Fish (embryo)	√ Mortality √ Developmental malformation √ Transcriptome analysis	Shin et al., 2023

al., 2021a). For example, exposure of olive flounder (*Paralichthys olivaceus*) embryos to the supernatant of wastewater from high-pressure water blasting resulted in significant malformations, including pericardial edema, spinal curvature, and tail fin defects, accompanied by strong transcriptional changes (Choi et al., 2020; Shin et al., 2023). These findings suggest that both the aqueous and particulate phases of wastewater from underwater cleaning procedures have significant toxic effects on aquatic organisms. The use of robots in underwater cleaning procedures has expanded to mitigate the disadvantages of dry-dock cleaning, which is an expensive and labor-intensive process. Employing robots ensures divers' safety and enables effective and frequent cleaning, even under extreme water conditions (e.g., low visibility, high pressure, and/or strong currents).

Cleaning, securing wastewater and particle debris, and disposing of pollutants are relatively straightforward when ship hull cleaning is conducted in dry docks. However, securing wastewater samples, including both crude and nano-sized particles, using in-water robot cleaning remains challenging. Even with automated filtration within the cleaning system, the wastewater still contained a variety of particles (Kim et al., 2024; Lee et al., 2024a). Furthermore, management regulations for treating and disposing of wastewater and debris from underwater cleaning processes have not yet been established. In addition, it is important to acknowledge that a considerable number of organisms can be released during the cleaning process, potentially posing a threat to the indigenous community (Hopkins and Forrest, 2008; Woods et al., 2012). Therefore, wastewater samples collected from in-water robot cleaning are of concern due to their potential ecotoxicity in the aquatic environment. Currently, several studies have documented the adverse effects of wastewater, including particles or supernatants collected from a commercial underwater cleaning robot, on marine organisms (Table 1). The ingestion of paint particles acutely induced toxicity and further detrimental effects on the development, fecundity, and transcriptome of the copepod *Tigriopus japonicus* (Park et al., 2023). Subsequently, the effects of dissolved compounds following the complete removal of suspended solids from the paint particles have reported in several marine animals. For example, the marine mysid *Neomysis awatschensis*, exposed to filtered hull cleaning wastewater, exhibited significant mortality with oxidative imbalance and inhibition of feeding rate, growth retardation, an extended intermolt duration, and diminished reproductive rate across generations (Lee et al., 2024a). Exposure of the monogonont rotifer *Brachionus manjavacas* to filtered hull cleaning wastewater resulted in detrimental effects on survival,

lifespan, fecundity, and population growth, accompanied by significant induction of oxidative stress (Kim et al., 2024). In the marine polychaete *Perinereis aibuhitensis*, treatment to filtered hull cleaning wastewater induced dose-dependently decreased burrowing activity and acetylcholinesterase activity with significant fluctuation in oxidative status (Lee et al., 2024b). Taken together, these reports will contribute to our understanding of whether wastewater derived from in-water hull-cleaning processes have detrimental effects on the molecular and biochemical systems, individual physiology, and population dynamics of non-target animals.

Challenge and limitation

In the past, guidelines for managing the removal of fouling organisms to minimize their transfer have been suggested by the Marine Environment Protection Committee and the International Maritime Organization (MEPC, 2011). However, regulations and policies for the management of wastewater and debris derived from underwater cleaning procedures remain limited, despite their emergence. Recent articles reviewed in this manuscript clearly demonstrate that completely removing toxic compounds dissolved in wastewater during in-water cleaning remains a challenge (Tamburri et al., 2020), highlighting the urgent need for advancements in removal technologies. Existing in-water cleaning systems primarily target the removal of fine particles but do not effectively address the problem of direct leakage or bypass from filtration systems. As a result, there is a risk of releasing wastewater with dissolved toxic compounds into the marine environment. Several researchers and institutions have suggested pore sizes for adequate filtration (Morrisey et al., 2015; Scianni and Georgiades, 2019). However, recent publications reviewed here focuses attention on that simply focusing on completely removing substances is inadequate. Additional wastewater treatment must prioritize the complete removal of dissolved toxicants in addition with establishment of the optimal pore size for wastewater filtration. Achieving additional filtration platforms and/or near-nanosized filtration in the cleaning platform is anticipated to be impractical due to the rapid penetration of a significant amount of wastewater including crude debris into the filtration system. It is important to note that the wastewater remaining after mechanical filtration should experience additional treatment processes (e.g., chemical removal, microbial degradation, and/or sludge formation). Finally, regular toxicity tests, along with chemical composition analysis, should be consistently conducted on wastewater released from

underwater cleaning process using non-target animals, as some of these animals serve as food sources for both animals and humans. The use of robots for underwater cleaning, as well as in the aquaculture industry, shows promise. Research on alleviation technologies and strategies for the management of wastewater reclamation, along with the establishment of regulations for wastewater release during the in-water cleaning process of ship hulls, is of utmost importance to protect aquatic environments.

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