Journal of Environmental Science and Engineering B 12 (2023) 249-256
doi:10.17265/2162-5263/2023.06.001

A Study on the Composition of Wastewater Produced during Ship Biofouling Management

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Abstract: The IMO (International Maritime Organization) recognized the problem of invasive species invasion and adopted the "International Convention for the Control and Management of Ships' Ballast Water and Sediments" in 2004, which came into force on September 8, 2017. In 2011, the IMO approved the "Guidelines for the Control and Management of Ships' Biofouling to Minimize the Transfer of Invasive Aquatic Species" to minimize the movement of invasive species by hull-attached organisms and required ships to manage the organisms attached to their hulls. Invasive species enter new environments through ships' ballast water and hull attachment. However, several obstacles to implementing these guidelines have been identified, including a lack of underwater cleaning equipment, regulations on underwater cleaning activities in ports, and difficulty in accessing crevices in underwater areas. The shipping industry, which is the party responsible for understanding these guidelines, wants to implement them for fuel cost savings resulting from the removal of organisms attached to the hull, but they anticipate significant difficulties in implementing the guidelines due to the obstacles mentioned above. Robots or people remove the organisms attached to the hull underwater, and the resulting wastewater includes various species of organisms and particles of paint and other pollutants. Currently, there is no technology available to sterilize the organisms in the wastewater or stabilize the heavy metals in the paint particles. In this study, we aim to analyse the characteristics of the wastewater generated from the removal of hull-attached organisms and select the optimal treatment technology. The organisms in the wastewater generated from the removal of the attached organisms meet the biological treatment standard (D-2) using the sterilization technology applied in the ships' ballast water treatment system. The heavy metals and other pollutants in the paint particles generated during removal are treated using stabilization technologies such as thermal decomposition. The wastewater generated is treated using a two-step process: (1) development of sterilization technology through pretreatment filtration equipment and electrolytic sterilization treatment and (2) development of technology for removing particle pollutants such as heavy metals and dissolved inorganic substances. Through this study, we will develop a biological removal technology and an environmentally friendly processing system for the waste generated after removal that meets the requirements of the government and the shipping industry and lay the groundwork for future treatment standards.

Key words: Biofouling, ballast water treatment system, filtration, sterilization, wastewater.

1. Introduction

Invasive aquatic species present a major threat to the marine ecosystems, and shipping has been identified as a major pathway for introducing species to new environments. The problem increased as trade and traffic volume expanded over the last few decades, and in particular with the introduction of steel hulls, allowing vessels to use water instead of solid materials as ballast. The effects of the introduction of new species have in many areas of the world been devastating. Quantitative data show the rate of bio-invasions is continuing to increase at an alarming rate. As the volumes of seaborne trade continue overall to increase, the problem may not yet have reached its peak. However, the Ballast Water Management Convention, adopted in 2004, aims to prevent the spread of harmful aquatic organisms from one region to another, by establishing standards and procedures for the management and control of ships' ballast water and sediments. Biofouling is also considered one of the main vectors for bioinvasions

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and is described as the undesirable accumulation of microorganisms, plants, algae and animals on submerged structures (especially ships' hulls). Studies have shown that biofouling can be a significant vector for the transfer of invasive aquatic species. Biofouling on ships entering the waters of States may result in the establishment of invasive aquatic species which may pose threats to human, animal and plant life, economic and cultural activities and the aquatic environment [1, 2].

However, the following obstacles were identified in implementing this guideline.

There are (1) lack of underwater cleaning facilities, (2) regulations related to underwater cleaning activities in ports, and (3) difficulty in accessing niche areas from underwater. The shipping industry, which is a stakeholder in the guidelines for managing hull attached organisms, wants to implement this guideline due to fuel cost reduction due to the removal of organisms attached to the hull, but many difficulties are expected in the implementation of the guideline due to the above obstacles. The wastewater transported after removing organisms attached to the hull by a robot or a person from the water contains various species of organisms and waste particles. However, there is currently no technology for sterilizing organisms contained in wastewater or for stabilizing heavy metals such as paint particles [3-6].

In this study, the optimal treatment technology is to be selected by removing the hull adhering organisms and analyzing the properties of the generated wastewater. The organisms contained in the wastewater from which the adherent organisms have been removed satisfy the biological treatment standard (D-2) by using the sterilization technology applied to the ballast water treatment system. Paint and particulate pollutants generated together during removal contain heavy metals and are treated using stabilization techniques such as thermal decomposition. We will develop bioremoval technology that meets the requirements of the government and shipping industry and an eco-friendly treatment system for by-products after removal and present the basis for future treatment standards.

2. Methodology

Various marine organisms attached to the hull of the ship are removed by the cleaning robot and sent to the storage tank. The stored wastewater is sent to the treatment device as shown in Fig. 1.

Observation of Samples Using Phase-contrast Microscopy

Fig. 2. is a phase-contrast microscopy (Carl Zeiss AXIOSKOP 50 DIC Trinocular F. Microscope) image of the substances present in ship ballast water. In this image, it was observed that the ballast water contains particulate matter of various sizes. The size of paint particles observed in ship ballast water under the microscope ranged from 50 micrometers to a maximum of over 350 micrometers.

Observation of Samples Using Scanning Electron Microscopy

To analyze the morphology of particles in ship ballast water, ship hull wash water samples were filtered through glass fiber filters. The filtered filters were thoroughly dried in a dry oven at 70 ℃. Subsequently, scanning electron microscopy (CLARA, TESCAN, Brno, Czech Republic) was employed for observation and imaging, with an acceleration voltage of 20 keV and magnifications ranging from 1,000 to 5,000 times. Additionally, the Energy Dispersive Spectrometer (EDS) functionality of the scanning electron microscope was utilized to analyze the elemental composition of the surfaces.

2.1 Biofouling Wastewater Test Method

2.1.1 Samples

Natural seawater, contaminated water, discharged water provided by the host organization were analyzed. For natural seawater, clear seawater was collected from a place away from the hull. Contaminated water was collected immediately after the first physical brush work was performed while the cleaning robot moved against the biofouling of the hull. As for the discharged water, the inflowing contaminated water passed through the filter and the outgoing water was collected.

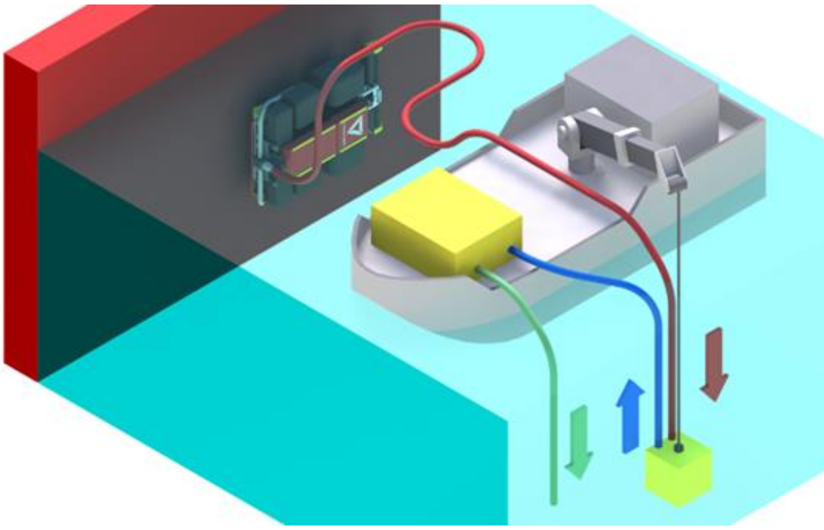


Fig. 1 Sampling location of hull fouling treatment wastewater.

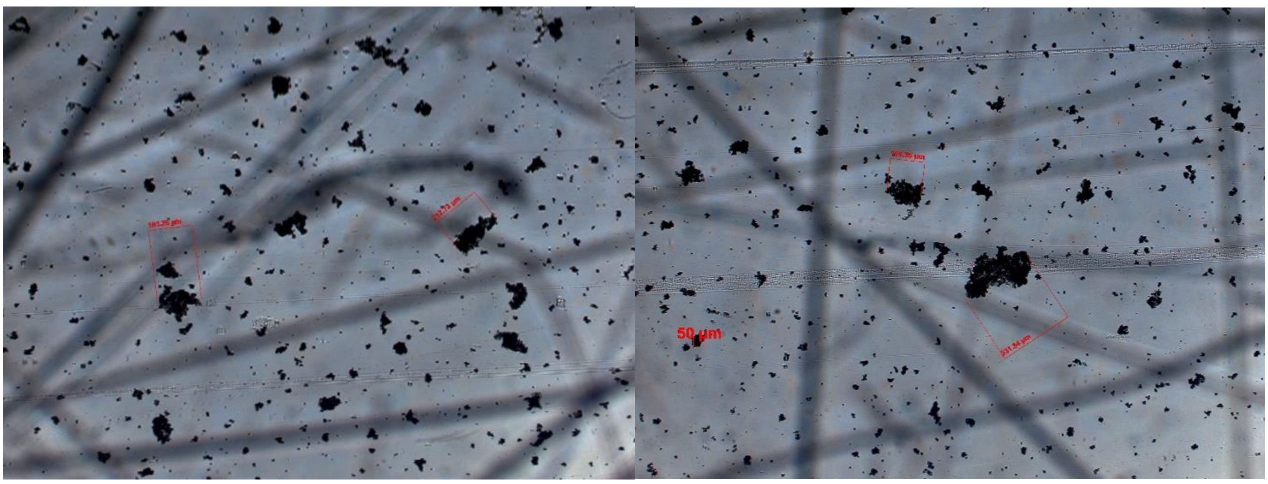
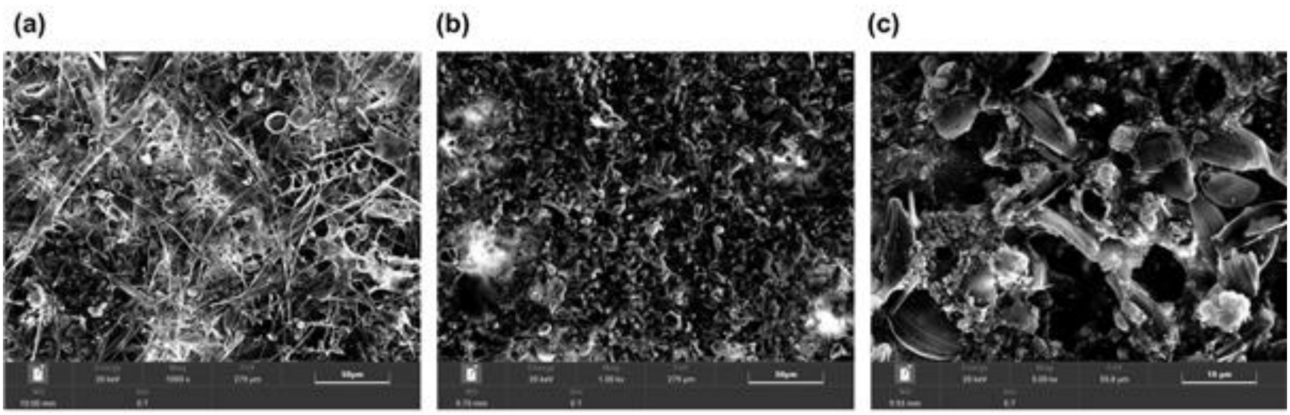


Fig. 2 Micrographs of biofouling wastewater and size of removed paint particles.



(a) (b) (c)

Fig. 3 Scanning electron microscope photo of a filter that filtered port seawater and a filter that filtered hull washing wastewater. (a) Port seawater sample 1,000 times, (b) Hull washing wastewater 1,000 times, (c) Hull washing wastewater 5,000 times.

Table 1 Biofling wastewater analysis item.

	Analysis item
General items (14 items)	Particle size, salinity, pH, conductivity, alkalinity, TSS(Total Suspended Solids), NO_3-N, NO_2-N, NH_4-N, PO_4-P, silicate, TOC (Total Organic Carbon), DOC(Dissolved Organic Carbon), POC(Particulate Organic Carbon)
Inorganic items (18 items)	Al, Cr, Mo, Cu, Cd, Pb, Sb, Si, Be, Co, Ag, Mn, B, Fe, Zn, Ni, As, Sn
Organic items (49 items)	1,1-Dichloroethene, Methylene chloride, trans-1,2-Dichloroethene, 1,1-Dichloroethane, Bromochloromethane, Chloroform, 1,1,1-Trichloroethane, 1,1-Dichloropropene, Carbon tetrachloride, Benzene, 1,3-Dichloropropane, Trichloroethene, 1,2-Dichloropropane, Dibromomethane, Bromodichloromethane, cis-1, 3-Dichloropropene, Toluene, trans-1, 3-Dichloropropene, 1,1,2-Trichloroethane, Tetrachloroethene,Dibromochloromethane, 1,2-Dibromoethane, Chlorobenzene, Ethylbenzene, (m+p)-Xylene, o-Xylene, Styrene, Bromoform, Isopropylbenzene, 1,1,1,2-Tetrachloroethane, 1,1,2,2-Tetrachloroethane,1,2,3-Trichloropropane, Bromobenzene, n-Propylbenzene, 2-Chlorotoluene, 4-Chlorotoluene, tert-Butylbenzene, 1,2,4-Trimethylbenzene, sec-Butylbenzene, p-Isopropyltoluene, 1,3-Dichlorobenzene, 1,4-Dichlorobenzene, n-Butylbenzene, 1,2-Dichlorobenzene, 1,2-Dibromo-3-chloropropane, 1,2,4-Trichlorobenzene, Hexachlorobutadiene, Naphthalene, 1,2,3-Trichlorobenzen

2.1.2 Number of Analyzes

Each sample was provided three times and the analysis items are as follows. The samples were subjected to three repeated tests, and the results were presented as the average.

2.1.3 Sample Observation Using a Phase-contrast Microscopy and Scanning Electron Microscope

Particle size and characteristics in ship hull wash water were analyzed using two types of microscopes.

3. Results and Discussions

3.1 General Items

Table 2 presents an analysis of the general characteristics of natural seawater, raw water, and treated water. The Total Suspended Solids (TSS) concentration was higher in the effluent water compared to natural seawater, with the results of the third analysis confirming this trend. Among nutrient pollutants, nitrate nitrogen, phosphate, and silicate showed higher concentrations in the effluent water than in natural seawater. Organic carbon components such as Total Organic Carbon (TOC), Dissolved Organic Carbon (DOC), and Particulate Organic Carbon (POC) exhibited concentrations in the effluent water similar to those in natural seawater.

3.2 Particle Size Distribution

Natural seawater had a particle size distribution mostly within 10 μm. The analysis of polluted water showed different results in three tests. In the first test, particles were distributed evenly within 31 μm, in the second test, similar to natural seawater, particles were distributed mostly within 10 μm. In the third test, particles were distributed within 100 μm, with the highest concentration of particles between 5~40 μm. Discharged water had a particle size distribution mostly within 5 μm.

3.3 Organic Items (VOC (Volatile Organic Compounds))

Table 3 shows the analysis results for organic compounds (VOCs). As a result of analyzing 49 items of VOCs, most of them were not detected.

3.4 Inorganic Items (Heavy Metals)

Table 4 shows the results of the analysis for inorganic substances (heavy metals). Among the heavy metals, the concentrations of Al, Cu, Cd, Sb, Si, Co, Mn, Fe, Zn, Ni, and Sn were higher in discharged water than in natural seawater. It was confirmed that the concentration

Table 2 Result of general items measurement data.

Item	Unit	The first round			Second round			Third time			Average		
		Seawater	Contaminated water	Discharged water	Seawater	Contaminated water	Discharged water	Seawater	Contaminated water	Discharged water	Seawater	Contaminated water	Discharged water
Salinity	PSU	31.1	31.2	28.9	27.6	29.7	13.8	27.0	29.0	28.9	28.6	30.0	23.9
pH	-	8.1	7.6	8.0	7.8	7.6	7.6	7.9	7.6	7.8	7.9	7.6	7.8
Conductivity	mS/cm	48.7	48.8	45.6	44.2	46.4	23.3	42.2	45.1	44.9	45.0	46.8	37.9
Alkalinity	mg/L $CaCO_3$	116	116	113	105	109	66.6	102	108	105	108	111	95
TSS	mg/L	4	106	2	9	172	6	40	412	118	18	230	42
NO_3-N	mg/L	0.007	0.038	0.072	0.012	0.056	1.12	0.125	0.062	0.052	0.048	0.052	0.415
NO_2-N	mg/L	N.D.	N.D.	N.D.	N.D.	N.D.	0.018	0.021	0.057	0.019	0.021	0.057	0.019
NH_4-N	mg/L	0.084	0.112	0.065	N.D.	N.D.	0.057	0.256	0.807	0.145	0.170	0.460	0.089
PO_4-P	mg/L	0.012	0.008	0.075	N.D.	0.027	N.D.	0.041	0.393	0.030	0.027	0.143	0.053
Silicate	mg/L	0.029	0.128	0.191	0.233	0.373	2.26	0.661	0.522	0.543	0.308	0.341	0.998
TOC	mg/L	1.90	13.9	1.90	1.67	6.38	2.54	4.63	6.27	3.39	2.73	8.85	2.61
DOC	mg/L	1.61	7.93	1.56	1.64	3.37	2.31	1.74	1.84	2.11	1.66	4.38	1.99
POC	mg/L	0.29	6.01	0.34	0.02	3.01	0.23	2.90	4.43	1.29	1.07	4.48	0.62

Table 3 Result of organic items (VOC) measurement data.

Item	Unit	The first round			Second round			Third time		
		Seawater	Contaminated water	Discharged water	Seawater	Contaminated water	Discharged water	Seawater	Contaminated water	Discharged water
1,1-Dichloroethene	μg/L	N.D.	1.42	N.D	N.D.	2.00	N.D.	N.D.	N.D.	N.D.
Methylene chloride	μg/L	1.2	0.16	0.21	N.D	N.D.	N.D.	N.D.	3.31	N.D.
trans-1,2-Dichloroethene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,1-Dichloroethane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Bromochloromethane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Chloroform	μg/L	0.15	0.16	0.29	N.D.	N.D.	1.92	N.D.	N.D.	1.63
1,1,1-Trichloroethane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,1-Dichloropropene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Carbon tetrachloride	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Benzene	μg/L	N.D.	N.D.	N.D.	0.17	0.15	0.3	N.D.	N.D.	N.D.
1,3-Dichloropropane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Trichloroethene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	1.96
1,2-Dichloropropane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Dibromomethane	μg/L	0.04	0.01	0.03	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Bromodichloromethane	μg/L	0.36	0.36	0.40	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.

Table 3 to be continued

cis-1,3-Dichloropropene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Toluene	μg/L	N.D.	N.D.	N.D.	N.D.	0.23	1.01	N.D.	N.D.	N.D.
trans-1,3-Dichloropropene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,1,2-Trichloroethane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Tetrachloroethene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Dibromochloromethane	μg/L	0.11	0.20	0.10	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,2-Dibromoethane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Chlorobenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Ethylbenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
(m+p)-Xylene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
o-Xylene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Styrene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Bromoform	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Isopropylbenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,1,1,2-Tetrachloroethane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,1,2,2-Tetrachloroethane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,2,3-Trichloropropane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Bromobenzene	μg/L	0.26	0.18	0.18	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
n-Propylbenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
2-Chlorotoluene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
4-Chlorotoluene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
tert-Butylbenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,2,4-Trimethylbenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
sec-Butylbenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
p-Isopropyltoluene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,3-Dichlorobenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,4-Dichlorobenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
n-Butylbenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,2-Dichlorobenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,2-Dibromo-3-chloropropane	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,2,4-Trichlorobenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Hexachlorobutadiene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
Naphthalene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
1,2,3-Trichlorobenzene	μg/L	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.

Table 4 Result of inorganic items (heavy metal) measurement data.

Iteam	The first round			Second round			Third time			Average		
	Seawater	Contaminated water	Discharged water	Seawater	Contaminated water	Discharged water	Seawater	Contaminated water	Discharged water	Seawater	Contaminated water	Discharged water
Al	1.04	1.06	7.38	0.83	2.47	12.5	1.35	0.78	1.47	1.07	1.44	7.12
Cr	0.22	0.22	0.55	0.68	0.74	0.45	0.72	0.43	0.68	0.54	0.46	0.56
Mo	13.3	32.9	16.3	12.5	13.7	6.83	11.7	13	13.5	12.5	19.87	12.21
Cu	4.2	1 080	18.3	2.17	53.4	12	1.51	99.1	18.3	2.63	76.25	16.2
Cd	0.03	3.21	0.33	0.05	0.05	0.16	0.04	0.24	3.99	0.04	1.17	1.49
Pb	0.2	0.42	0.11	N.D.	0.19	0.06	0.01	0.04	0.02	0.11	0.22	0.06
Sb	0.12	0.18	0.19	0.14	0.12	0.3	0.15	0.11	0.17	0.14	0.14	0.22
Si	109	897	193	161	309	920	546	474	479	272	560	531
Be	0.25	N.D.	N.D.	0.14	N.D.	0.15	0.03	0.2	0.02	0.14	0.2	0.09
Co	N.D.	1.51	0.56	0.06	0.46	0.64	0.03	0.14	4.97	0.05	0.7	2.06
Ag	1.49	1.85	1.75	1.36	1.35	0.63	1.22	1.34	1.34	1.36	1.51	1.24
Mn	0.09	156	13.6	15	141	82.5	5.9	73	66.7	7	123	54.27
V	1.03	0.82	0.71	0.64	0.82	0.07	0.77	0.8	0.43	0.81	0.81	0.4
Fe	0.08	104	0.08	0.12	0.86	0.21	0.04	0.24	0.21	0.08	35.03	0.17
Zn	9.31	1 310	171	12.6	180	71	24.4	125	194	15.44	153	145
Ni	0.38	3.08	11.1	1.05	0.91	4.1	0.59	1.81	137	0.67	1.93	50.73
As	1.34	3.89	1.24	1.62	2.07	0.92	1.95	2.28	1.08	1.64	2.75	1.08
Sn	1.43	1.93	6.18	1.38	1.67	10.3	1.31	1.59	2.38	1.37	1.73	6.29

of Cu, Mn, and Fe in the discharged water was lower than that in the contaminated water, so it could be removed through the filter.

For heavy metals in ship hull wash water, it would be beneficial to secure a diversity of samples from various types of ships in the future.

4. Conclusion

Based on the analysis results of general, inorganic, and organic constituents in the wastewater, it has been confirmed that the treatment of wash water is feasible. Currently, the International Maritime Organization (IMO) is in the process of developing agreements for treatment standards, making the development of this treatment system necessary. The treatment of ship hull wash water is deemed to require the application of a comprehensive treatment approach. Through this study, the need for coagulation, precipitation, and microbial disinfection systems has been identified. In future research, treatment methods will be applied, and the treatment outcomes will be analyzed.

Research on various attached-growth biological treatment methods is essential. In the future, we will secure diverse attached-growth biological treatment systems and continue research on biological and heavy metal treatment.

Acknowledgement

This work was partly supported by IITP (Institute of Information & Communications Technology Planning & Evaluation) grant funded by the Korea government (MSIT) (RS-2023-0022378, Development of industry-level technology bridging ICT for coal2biomethane) and KIMST (Korea Institute of Marine Science & Technology Promotion) funded by the Ministry of Oceans and Fisheries, Korea (No. 20210500, Development of treatment technology of marine bio-fouling on ship hull).

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