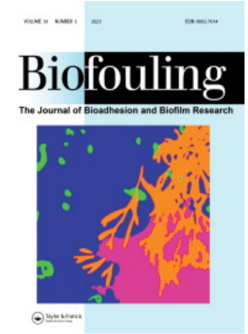

Biofouling

The Journal of Bioadhesion and Biofilm Research

ISSN: (Print) (Online) Journal homepage: https://www.tandfonline.com/loi/gbif20

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To cite this article: Florian Weber & Naser Esmaeili (2023): Marine biofouling and the role of biocidal coatings in balancing environmental impacts, Biofouling, DOI: 10.1080/08927014.2023.2246906

To link to this article: https://doi.org/10.1080/08927014.2023.2246906

BIOFOULING
https://doi.org/10.1080/08927014.2023.2246906

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REVIEW ARTICLE

Marine biofouling and the role of biocidal coatings in balancing environmental impacts

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ABSTRACT
Marine biofouling is a global problem affecting various industries, particularly the shipping industry due to long-distance voyages across various ecosystems. Therein fouled hulls cause increased fuel consumption, greenhouse gas emissions, and the spread of invasive aquatic species. To counteract these issues, biofouling management plans are employed using manual cleaning protocols and protective coatings. This review provides a comprehensive overview of adhesion strategies of marine organisms, and currently available mitigation methods. Further, recent developments and open challenges of antifouling (AF) and fouling release (FR) coatings are discussed with regards to the future regulatory environment. Finally, an overview of the environmental and economic impact of fouling is provided to point out why and when the use of biocidal solutions is beneficial in the overall perspective.

ARTICLE HISTORY
Received 19 May 2023
Accepted 7 August 2023

KEYWORDS
SPC; FRC; ROV; invasive aquatic species (IAS) antifouling; GHG emissions

Introduction

Marine biofouling describes the growth of marine organisms on submerged structures and is a global problem affecting various industries. It compromises the efficiency of vessels, offshore energy infrastructure, as well as the performance of technical equipment in contact with sea water, such as heat exchangers and sensor systems (de Carvalho 2018). Depending on the geographical location, temperature, salinity, nutrition, and light conditions, there are distinct fouling pressures (Antunes et al. 2019). This is a particular problem in the shipping industry, as vessels travel long distances crossing various ecosystems and environmental conditions that contributes to their fouling susceptibility. To comprehend the scale of biofouling in the marine industry the available surface area can be approximated. Between 1999 and 2013, about 120,000 vessels above 100 gross tons were registered as active. Their estimated surface area of 571 km^2, demonstrates that surfaces equivalent to the total land area of France require protection (Moser et al. 2016).

The problem is that fouling causes an increase in drag forces requiring up to 86% additional shaft power (Schultz 2007). Since the added fuel consumption is directly related to increased greenhouse gas (GHG) emissions, it is obvious that fouled ship hulls have a significant contribution to the environmental footprint of the shipping industry. In response to this problem the efficiency of vessels has been addressed with various regulations. With the newly introduced annual carbon intensity index (CII) ratings reported in the ship energy efficiency management plan (SEEMP), the industry continuously faces stricter regulations to maintain their fleet.

Besides the increase in fuel consumption and GHG emission, biofouling can also lead to the spread of invasive aquatic species (IAS). Fouling occurring on commercial and recreational vessels spread IAS, which endanger biodiversity and cause secondary biofouling problems (Williams et al. 2013; Havel et al. 2015). With the introduction of International Maritime Organisation's (IMO) ballast water management regulations in 2017, the first step towards environmental protection against invasive organisms was taken (Gollasch et al. 2007), however external surfaces still harbour IAS (BIMCO 2022).

To keep the external hull surfaces clean, manual cleaning protocols or protective coatings that prevent the attachment and growth of marine organisms on surfaces are commonly used. Since manual cleaning is often not a cost-effective option, most submerged surfaces employ a protective antifouling coating (Hopkins et al. 2021). Throughout history, efforts have been

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made to protect the hulls of ships. From the early use of copper sheets to modern polymer coatings, the most significant example of effective solutions were coatings containing tributyltin (TBT) developed in the 1960s (Omae 2003). However, due to the toxicity of TBT their use was banned in 2008. This caused the rise of tin-free cuprous oxide-based coatings, which currently dominate the market (Champ 2000; Ciriminna et al. 2015).

The issue with antifouling coatings is that they must provide protection against a broad diversity of fouling organisms in vastly different climates. Modern antifouling coatings are typically tailored to specific use cases, such as long idle periods, vessel speeds, and trade regions, taking water temperature and main fouling organisms into consideration. Upon deviation from the originally specified trade route, significant fouling or coating deterioration may occur. Therefore, new methods to manage biofouling, which withstand todays challenging requirements in the shipping industry, are highly sought after. Currently, there is a trend to implement proactive cleaning (grooming) with remotely operated vehicles (ROV) as cost-effective alternative to reactive cleaning during drydocking (Song C and Cui 2020). Further, the use of fouling release (FR) coatings rises as alternative to the established antifouling (AF) coatings (Lejars et al. 2012; Hu et al. 2020). Finally, with the continuous increase of digitalization and data collection of vessels and ports, the marine sector might be at the forefront of a digital revolution allowing accurate prediction of biofouling (Lee et al. 2008).

This review provides a comprehensive overview about important aspects of biofouling starting from the adhesion strategies of the most common biofouling organisms to a summary of today's commercially available antifouling coatings. Further, recent developments in the antifouling coating research and open challenges are presented in context with their potential to reduce emission and the spread of IAS. Finally, an overview of the regulatory landscape that directs the future requirements in the maritime industry is given.

Biofouling

Fouling of marine surfaces starts with the exposure to seawater. Its progress is often classified into soft or hard fouling with various degrees (Figure 1). Soft fouling describes the initial biofilm layer comprised of biomacromolecules, bacteria, diatoms, and microalgae (Antunes et al. 2019). With time, a transition to macrofouling occurs when higher organisms such as barnacles, mussels, tubeworms, and bryozoans conquer the surfaces (Cao et al. 2011). Although the fouling progress is often idealised in a sequential manner, it is rather dynamic with individual organisms exploring new surfaces in parallel until they find cues for their settlement (Richmond and Seed 1991).

Upon exposure to sea water, pristine surfaces face nonspecific adsorption of dissolved organic matter and ions as the initial conditioning film (Jain and Bhosle 2009; Wang Y et al. 2020). Simultaneously physically driven adsorption of bacteria, diatoms, and algae spores occurs. Once these organisms settle on the surface, they develop a biofilm matrix, which protects them against environmental influences (Callow and Callow 2011; de Carvalho 2018). Commonly, surface topography and physico-chemical properties

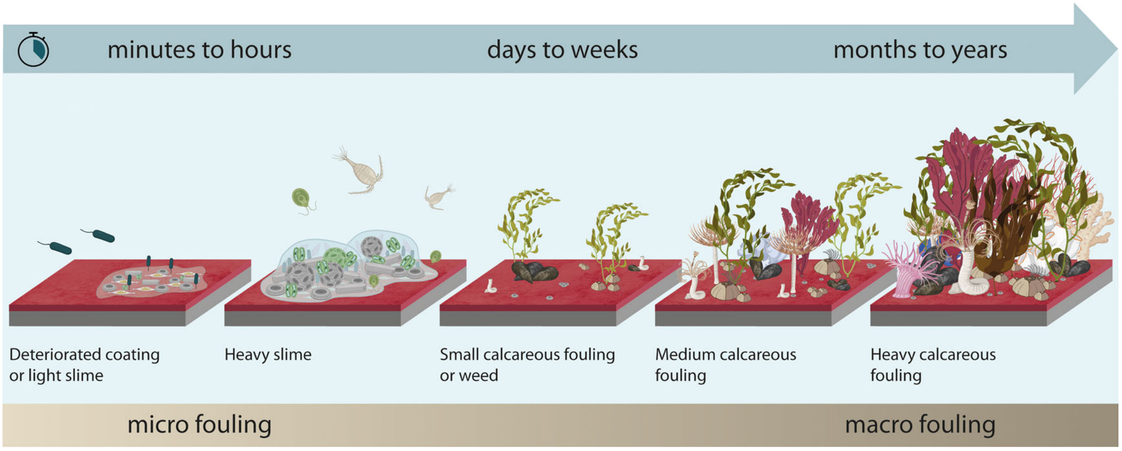

Figure 1. Biofouling is a temporal and highly dynamic process starting with the adsorption of small organic molecules and microorganisms onto the surface. It is followed by the settlement of spores and larvae of higher organisms and plants exploring new habitats. Depending on various parameters, such as location, temperature, salinity, nutrients, and properties of the surface, a unique fouling layer develops.

drive these interactions between surfaces, biomolecules, and organisms (Ozkan and Berberoglu 2013). Extensive research on biomolecule adsorption on surfaces with different energies by Baier et al. identified minimum fouling at a surface energy around 22 mN/m (Baier 2006). Often, this low fouling nature of such surfaces is also observed for micro and macrofouling organisms (Callow and Fletcher 1994). Their specific adhesion phenomena are thus discussed in the following.

Microfouling is a biofilm containing bacteria, diatoms, and microalgae and other Protista (Dobretsov and Rittschof 2020; Salta et al. 2013). One of the main species in marine biofilms are alpha-proteobacteria, for which *C. crescentus* has become a widely used model organism. This specialized holdfast adhesin is composed of glucose, mannose, xylose, and N-acetylglucosamine, and presents high adhesion strength to hydrophobic surfaces (Berne et al. 2013; Hershey et al. 2019).

Diatoms express an adhesive glue (mucilage) through their raphe that consists of diverse polysaccharides (80–84%), proteins (7–10%), and uronic acids (18–19%) (Dugdale et al. 2006; Lachnit et al. 2019). It has been shown that physical surface properties do not influence the initial settlement of diatoms, but their adhesion strength also peaks on hydrophobic surfaces (Thompson and Coates 2017).

With the vast diversity of species found in marine biofilms, there is a consensus that there can be positive and negative interactions between micro and macrofouling organisms (Clare et al. 1992). Biofilms are often cues for larval settlement of higher organisms in the first phase of ‘behavioural searching’, in which surfaces are probed for suitability (Thompson and Coates 2017; Dobretsov and Rittschof 2020). Biofilms can however also prevent settlement of bryozoans and barnacles (Maki et al. 1988; Wieczorek and Todd 1997). This interaction is driven by bacterial quorum sensing molecules that some marine organisms are able to recognise (Salta et al. 2013).

The settlement macrofouling organisms is often more complex. *Ulva* green algae zoospores repeatedly probe surfaces before they discharge self-aggregating glycoproteins forming an adhesive pad (Callow et al. 2005; Callow and Callow 2006). In contrast to diatoms and bacteria, *Ulva* zoospores preferentially adhere to hydrophilic surfaces (Callow et al. 2005; Thompson and Coates 2017). Their negatively charged cell wall further causes a noteworthy pseudo-settlement on positively charged surfaces that paralyses the spores (Ederth et al. 2008).

Barnacle cyprids explore suitable places for settlement using a temporary adhesive system before their permanent attachment (Okano et al. 1996). Their cement is made up of 90% proteins, which provides extraordinary strength through enzymatically cross-linking (Kamino 2006; Aldred and Clare 2008; He LS et al. 2013). The adhesion in aqueous environment is facilitated by the secretion of a lipidaceous phase-separating fluid that removes water from the surfaces to allow optimal surface coverage of the cement (Gohad et al. 2014; Fears et al. 2018).

The adhesion mechanism found in blue mussels (*Mytilus* spp.) inspired decades of research of underwater adhesives. Their adhesive byssal plaque contains mussel foot proteins (Mfp) containing 3, 4-dihydroxyphenylalanine (DOPA) (Bandara et al. 2013). The strong adhesive properties stem from the ability of DOPA to form strong hydrogen bonding, hydrophobic interactions, and covalent crosslinks (Saiz-Poseu et al. 2019). Like barnacles, mussels also use fatty acid secretions to displace surface bound water for improved wetting of their adhesive protein matrix (He Y et al. 2018).

While algae, barnacle and mussel adhesion is well documented, relatively little is known about detailed attachment mechanisms of tube worms, bryozoan, hydrozoan, and tunicates. One study of *Hydroides elegans*, indicated that their settlement is induced by *Pseudoalteromonas* biofilms (Hadfield et al. 2021). Yet their adhesive system is not well documented. Only for the larvae of the tunicate *Ciona intestinalis,* a glycoprotein rich adhesive containing DOPA residues has been reported (Zeng et al. 2019).

Evidently, there is a broad variety of organisms with different adhesion mechanisms, which have not been investigated thus far. Particularly studies of interactions between fouling organisms in communities are still lacking. This is in part due to the lack of tools to accurately describe such complicated interactions. However, metabolomic (metabolites), proteomic (proteins) and transcriptomic (RNA) approaches become progressively more used (Dobretsov and Rittschof 2023). Transcriptomics demonstrated the impact of biocides on the natural diversity of biofilms. A change from biofilms abundant in alphaproteobacteria to biofilms consisting of predominantly gammaproteobacteria upon exposure to copper pyrithione/Cu_2O was observed (Winfield et al. 2018; Kim et al. 2021). This change of biofilm structure becomes a crucial factor for the subsequent macrofouling, where it was observed that non-biocidal coatings allow high biodiversity and coverage during static field immersion tests (Papadatou et al. 2021; Bressy et al. 2022). This aligns with the observation that based on the geographical location, the initial composition of biofilms also determines the subsequent macrofouling composition (Swain et al. 2000).

Taking all these observations into consideration, fouling could in future be predicted depending on the type of coating and geographic location. To achieve this, there is a need for data-driven models as well as large datasets to train such models. While there are models available, they are limited to individual species or other factors (Vellwock et al. 2019). Further, there is a lack objective high throughput methods to produce data for large databases. On this front, automated detection tools are being developed to quantify fouling based on image data (Papadatou et al. 2022; Krause et al. 2023). This may foster future projects establishing open databases similarly to the OCEANIC project that collects data along European coastal regions (Vinagre et al. 2020). However, until global public databases are established, most expertise on biofouling prevention lies with companies providing and maintaining commercial antifouling solutions that are introduced in the next chapter.

Biofouling prevention technologies

Biofouling can be limited using coatings or mechanic cleaning systems. Amongst the coatings systems there are three categories, (i) biocidal antifouling (AF) coatings, (ii) fouling release (FR) coatings, and (iii) hard coatings (Figure 2). Hard coatings are mostly used in combination with mechanical cleaning systems and not described in full detail here. The following sections will focus on AF and FR technologies (Table 1), followed by an overview of proactive cleaning measures (grooming). Finally, the modern standard of performance monitoring based on ISO 19030 is presented, which improves drydock scheduling for cleaning maintenance.

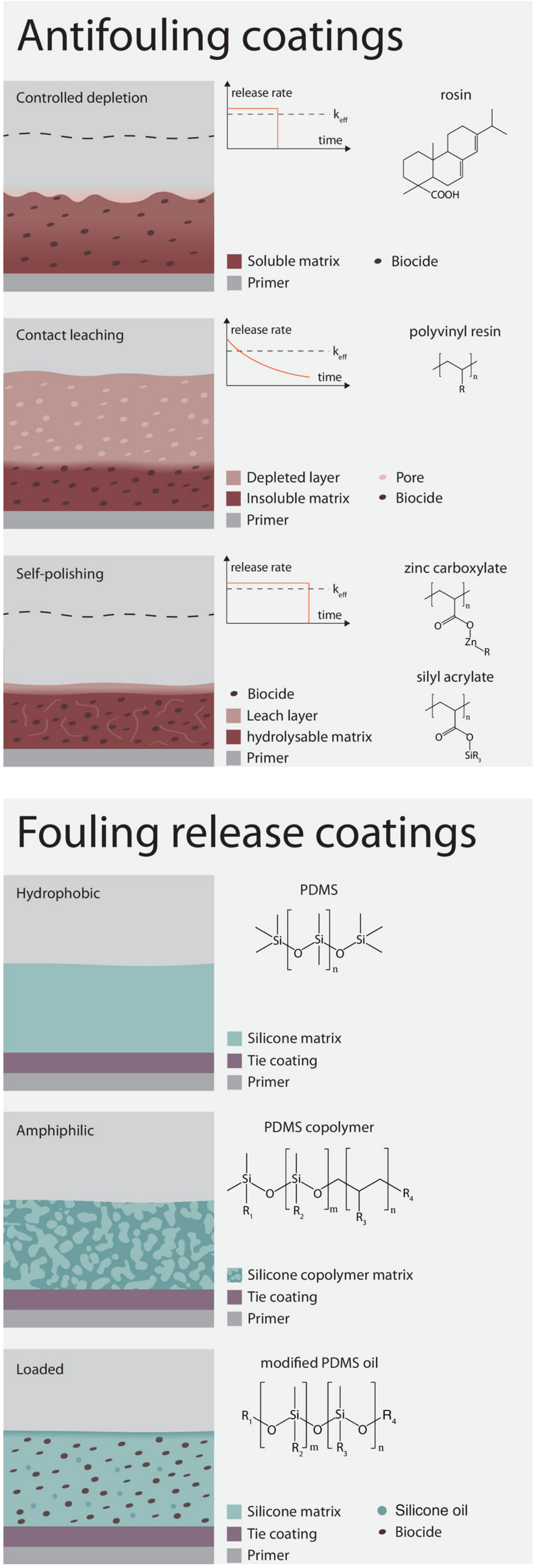

Figure 2. Most applied coatings to prevent biofouling in the marine industry are antifouling (AF) and fouling release (FR) coatings. Besides AF and FR coatings, hard coatings are used specifically with external cleaning systems.

Antifouling coatings

Today the commercial market for hull coating solutions is largely dominated by biocidal antifouling (AF) coatings. In 2011, approximately 94% of the applied coatings were reported to be biocidal AF coatings (Lindholdt et al. 2015). Three years later, their market share decreased to 90% indicating the shift in the industry towards the new technologies (Ciriminna et al. 2015). Despite that, AF coatings are still widespread products whose performance rely on the controlled release of biocidal molecules through diffusion and

Table 1. Overview of the main commercial coatings systems of the market leading manufacturers available in 2023.

Antifouling (AF) coatings		
Manufacturer	Name	Technology
AkzoNobel	Intersmooth	Cu/Silyl acrylate SPC
	Interspeed	CDP
	Interswift	Blend of Intersmooth and Interspeed
Chugoku	Seaflo Neo	CF Z and CF Premium: Cu-free Zn acrylate SPC SL M and SL Z: Silyl methacrylate SPC
	Sea Grandprix	500: Zn acrylate SPC 1000L: Silyl acrylate SPC
	Sea Premier	1000: Zn acrylate SPC 3000: Silyl methacrylate SPC
Hempel	Atlantic+	Acrylic SPC
	Dynamic	Silyl acrylate SPC
	Globic	Nano-acrylate SPC
	Olympic+	Acrylic SPC
	Oceanic+	Zn carboxylate SPC
Jotun	SeaForce	Ion exchange SPC
	SeaMate	Silyl acrylate SPC
	SeaQuantum	Silyl (meth)acrylate SPC
Nippon	A-LF Sea	100: Zn acrylate SPC 250, 400, and 600: Cu-silyl-acrylate SPC
	Aquaterras	Biocide-free amphiphilic micro-domain SPC
	Fastar	Amphiphilic nano-domain silyl-acrylate SPC
	Ecoloflex	Original: Cu acrylate SPC Hyb: Cu-silyl-acrylate SPC
PPG	ABC	SPC
	Amercoat	CDP
	Sigma Alphagen	SPC
	Sigma Ecofleet	SPC
	Sigma Nexeon	Cu-free Zn acrylate SPC
	Sigma Sailadvance	Zn methacrylate CSP
Fouling release (FR) coatings		
AkzoNobel	Intersleek	Biocide-free fluoropolymer
AST Inc	SLIPS® Dolphin™	Biocide-free
Chugoku	Bioclean (Bioclean+)	Biocide-free (Biocidal)
Hempel	Hempaguard	Biocidal silicone hydrogel (Actiguard)
	Hempasil	Biocide-free silicone enhanced hydrogel
Jotun	SeaQuest	Biocide-free
	SeaQuest Endura	Biocidal
PPG	Sigmaglide	Biocide-free pure PDMS

SPC: self-polishing copolymer; CDP: controlled depletion polymer; CSP: controlled surface-active polymer

matrix degradation. To improve these processes, various binder technologies have been developed (Figure 2).

The first antifouling coatings were either based on water-soluble, or on insoluble matrices (Almeida E et al. 2007). While controlled depletion coatings based on a soluble rosin matrix provide a constant release of biocides, they suffer from various problems, such as low biocide loading and short lifetime. Blending rosin with various synthetic polymers however allows the control of the degradation rate in modern systems. In contrast, contact leaching coatings are made from an insoluble matrix based on high molecular weight polymers, such as polyvinyl resins (Woods Hole Oceanographic Institution 1952), and contain a high biocide load. These properties lead to their drawback of a steady decline of released biocides throughout the lifetime of the coating and the build-up of a porous top layer depleted of active molecules which facilitates biofouling (Almeida E et al. 2007).

Due to these drawbacks the modern standard AF coating is based on self-polishing copolymers (SPC). This technology combines the stability of high-molecular weight polymers and a controlled chemical degradation of functional groups that allow a more precise control over the biocide release. The degradation mechanism is either based on ion-exchange or hydrolysis reactions. Ion-exchange binders contain Cu- or Zn-acrylates (carboxylate), which change solubility after sodium ions in seawater replace the metal ions. Hydrolysis based self-polishing technologies use a labile ester bond in silyl-acrylate polymers which breaks in aqueous environment releasing water-soluble siloxanes (Bressy et al. 2009). Today, most research focuses on tuning the degradation, water uptake, and mechanical properties of silyl-acrylate resins by modifying the molecular structure of the silyl sidegroup (Zhou et al. 2015; Xie et al. 2018). Thereby, control over the leach layer is obtained to maintain the thin layer of biocide depreciated matrix on the surface of SPC coatings as thin as possible.

Beyond these principal binder technologies, a broad variety of additional modifications address mechanical and rheological properties of AF coatings. Waxes, pigments, filler particles and the volatile organic content (VOC) make out the unique composition of each manufacturer's antifouling solution (Yebra et al. 2004). Today, the antifouling solutions for the low budget market are typically dominated by Zn/Cu acrylate based SPC products, while premium products utilize the more advanced silyl-acrylate technologies (Table 1). In addition, manufacturers have proprietary technologies that separate them from their competition. Nippon FASTAR and (A-)LF-SEA coatings are according to their website inspired by tuna fish skin and include hydrogel components to trap water at the surface creating a slippery hull-to-water interface (Nippon 2023). Hempel's Atlantic, Oceanic, and Olympic paints contain microfibers that enhance surface smoothness and mechanical stability (Hempl 2023). PPG

Sigma advertises their Sailadvance RX and GX series with controlled surface-active polymers (CSP) that function as a lubricant on the coating/water interface (PPG 2023). Jotun's SeaQuantum Ultra, Plus, and Classic III series feature microZone technology which extends the time between the activation of biocides and their dispersion (Jotun 2023). Thereby activated biocides retain closer to the hull surface and create a dense protective barrier against fouling according to the manufacturer. Another technology found in Jotun's Seaforce Active paint, is the incorporation of hydractive particles, which slow down water uptake and reduce the rate of biocide release. This claims to result in a longer lifetime of the antifouling coating (Jotun 2023).

Fouling release coatings

FR coatings were developed as eco-friendly biocide free coating alternatives. Their mechanism is based on minimizing the adhesion force between the coating surface and fouling organisms. The main component of FR coatings is PDMS due to the synergism of low surface tension and low elastic modulus of silicones (Brady and Singer 2000). This effectively reduces adhesion strengths of most micro and macro fouling organisms below 100 Pa and 0.5 MPa respectively (Oliveira and Granhag 2016). Thereby, shear forces detach adherent organisms while a vessel is moving at operating speed above 15 knots (Dafforn et al. 2011). Due to low erosion, FR coatings also excel in terms of their surface roughness yielding a low skin friction coefficient, which reduces fuel consumption and GHG emissions (Lindholdt et al. 2015). Although FR coatings currently have a low market share, their use shows a positive trend. A survey by BIMCO reports that 18% of ship owners and operators are actively exploring the use of FR coatings in their fleet (BIMCO 2019). This is reflected in the increasing number of products developed by the major coating suppliers.

Today there is a broad variety of FR coatings spanning from pure silicone coatings to modified PDMS binders and different additives in the silicone matrix (Table 1). Contrary to their environmentally benign origin, the current trend is to include biocides, due to the poor performance of silicone-based coatings under static conditions (Truby et al. 2009). FR coatings typically suffer from slime fouling, which is related to the strong adhesion of diatoms and bacteria to hydrophobic surfaces (Holland et al. 2004; Zhao et al. 2019). Therefore, much effort is put into advancing the technology and alter the surface chemistry to create self-lubricating surfaces with dynamic surface antifouling effects that can compete with AF coatings for applications with long idle periods (Xie et al. 2019).

Self-lubricating silicone surfaces started with the incorporation of hydrophobic silicone oils in PDMS (Truby et al. 2000). In these coatings, silicone oils, migrate towards the water interface to form a continuous oil film that prevents organisms to permanently attaching (Zhang et al. 2013). Yet, the introduction of hydrophobic silicone oils still did not perform satisfactorily against microfouling. To combat biofilms, the next generation used hydrophilic polyethylene glycol (PEG) functionalised oils. The increased hydrophilicity of the new formulation showed a promising reduction in the adhesion of proteins, bacteria, and diatoms (Hawkins and Grunlan 2012; Hawkins et al. 2014, 2017). Today, the hydrophobic properties of silicone coatings are balanced with a variety of oils with different modifications, such as different building blocks and branching, which give FR coatings tuneable amphiphilic characters.

For the next generation of FR coatings, advances from the medical industry, such as zwitterionic molecules and quaternary ammonium compounds, may be utilized (Liu et al. 2013; Yeh et al. 2014; Martinelli et al. 2018). However, an issue with mobile hydrophilic components is the potential depletion into the aqueous environment (Camós Noguer et al. 2017a). So far, released silicone oils are not considered as detrimental to the environment due to their inert properties. With extended use and chemical variation, their potential toxic effect at elevated concentrations in closed marinas should be kept in mind (Lagerström et al. 2022). However, this can be prevented through direct functionalisation of the backbone of the silicone matrix, changing the molecular weight of oils, or increasing the density of the silicone matrix (Mazan et al. 1995; Camós Noguer et al. 2017b).

Beyond academic research, there is also active research by manufacturers to boost the performance of their FR coatings. AkzoNobel incorporates biorenewable long chain waxy sterols from sheep's wool (Lanion technology) in their Intersleek 1001 to target slime fouling through their hydrophilic character (AkzoNobel 2023). Hempel developed the ActiGuard system, which traps the biocide in a hydrogel layer at the surface of their Hempaguard coatings (Sørensen et al. 2015). Likewise, Jotun recently released its biocide containing SeaQuest Endura line in which the diffusion and release of biocides and amphiphilic oils is controlled through their proprietary Elastotain cross-linking technology (Jotun 2023). In contrast, PPG is currently the last major manufacturer holding on to pure PDMS coatings (PPG 2023). Instead, their Sigmaglide coatings feature a proprietary technology that allows silicone molecules to reconnect and rearrange to regenerate the integrity of the coating.

One still unsolved issue with silicone-based coatings is the requirement of a tiecoat that links them to anticorrosive primers. This adds time and costs in the application process. Consequently, self-stratification may be utilized in future FR coatings. Pioneered by Webster et al. siloxane-polyurethane coatings were developed by combining the benefits of a silicone-based interface towards the aqueous environment with a primer-compatible polyurethane matrix (Majumdar and Webster 2005; Ekin and Webster 2007). In the group's further research, various oils with hydrophilic, amphiphilic, or zwitterionic character were investigated to adapt the fouling resistance in different conditions (Majumdar et al. 2009; Galhenage et al. 2016; Rahimi et al. 2020; Benda et al. 2021). Similar studies were later also conducted for epoxy-based coatings (Lemesle et al. 2021; Rahimi et al. 2022), demonstrating the applicability of phase separation for a continuous release of surface-active oils (Cui et al. 2015; Camós Noguer et al. 2018).

As an alternative to active biocide release, covalent attachment of organic biocides to the PDMS matrix is researched (Thomas J et al. 2004; Xie et al. 2015; Silva et al. 2019, 2021). This method presented improved fouling resistance compared to biocide free formulations while simultaneously limiting the release of toxic substances (Ferreira et al. 2020). If biocides in FR coatings are necessary to guarantee optimal performance, this may be a key technology to control their environmental impact.

Mechanical cleaning

Hauling out boats for seasonal cleaning is well known for many leisure boat owners. This is undoubtedly unfeasible for large vessels. Therefore, in water hull cleaning is one of the key aspects of maintaining the performance of commercial vessels without needing to dry-dock. Classically, divers inspect and clean the hull, but they are gradually replaced by fully autonomous robots, or remotely operated vehicles (ROV) that reduce labour costs and diving accidents. At present, several established companies and start-ups offer ROV cleaning at various scales targeting professional and recreational use (Table 2). Once this equipment is broadly available, proactive cleaning during (un-)loading can become a vital part of the antifouling management plan compared to the currently employed reactive cleaning protocols (Scianni and Georgiades 2019).

The implementation of mechanical cleaning for vessels with existing coating systems is however discussed. Although studies indicate that proactive cleaning reduces soft and hard biofouling on FR as well as SPC coatings without significant deterioration (Tribou and Swain 2015, 2017; Hunsucker et al. 2018), manufacturers are sceptical. Since AF coatings are not designed for repeated mechanical cleaning premature AF depletion may occur and paint manufacturers advise the use of hard coatings for the use with proactive ROV cleaning. ROV manufacturers answer the problem with offering different brush stiffness or waterjets pressures specific to fouling as well as coatings type.

Another drawback of in-water hull cleaning is the problem with waste disposal. Without the implementation of capture mechanisms, biologic waste containing IAS and paint residue accumulates around cleaning stations creating problems like ballast water disposal (Scianni and Georgiades 2019). To combat this issue, many grooming equipment employ filter systems. Depending on the implemented filtration level, the number of dispersed solids and heavy metal ions in the water after filtration can be reduced significantly (Tamburri et al. 2020). In this situation, appropriate infrastructure that manages waste for its save disposal is required at every port. This sparks the discussion about the global implementation of ROV

Table 2. In water hull cleaning systems using remotely operated vehicles (ROV).

ROV	Removal technology	Adhesion	Usage	Waste collection	Company
HullSkater	non-abrasive brushes	Permanent magnet wheels	PRO-1	no	Jotun AS
Keelcrab	nylon or stainless-steel brushes	Low pressure	PRO-2	optional	Keelcrab
ECOsubsea	Soft Jets	Low pressure	PRO-2	yes	ECOsubsea
Envirocart	Non-contact blades	Low pressure	PRO-2	yes	CleanSubSea
Fleet Cleaner Robot	High pressure jets	Magnetic attachment	PRO-2	yes	Fleet Cleaner
Hullbot	Microtextured Cleaning Heads	Octo-thrusters	PRO-2	no	Hullbot Pty Ltd
Armach	Brush heads	Low pressure	PRO-2	no	Armach Robotics
ITCH	Soft-brush system	Ship's propulsion energy	PRO-3	no	Shipshave
HullBug	soft brushes and water jet	Low pressure	PRO-2	yes	SeaRobotics
HullWiper	variable-pressure seawater jets	Low pressure	PRO-2	yes	HullWhiper
TAS Robot	8 different brushes	Slipping locomotive	PRO-2	yes	TAS Global
Mepus-UICR	Cavitation jets	Not specified adaptive adherent function	REA	no	ZhiZheng Ocean Technology Company
SeaBadger	Variable pressure water jets	Not specified	REA	yes	HydroHull Cleaning

PRO-1: Proactive during idling through crew and/or licensed operator – ROV vessel based; PRO-2: Proactive during idling through crew and/or licensed operator – ROV port based; PRO-3: Proactive in transit through crew; REA: Reactive during idling through licensed operator.

cleaning. Should ROVs stay at ports, giving the port most control over the cleaning process, or should ROVs stay with the ship to give the shipowners control over the integrity of their hull integrity? Currently, only Jotun and Shipshave offer ROVs that stay with the ship (Table 2).

Despite these drawbacks, the consensus is that proactive cleaning provides a cleaner hull than reactive cleaning. Detachment of early stage microfouling is far easier than detachment of macrofouling and a cleaner hull contributes to the overall performance and fuel consumption of a vessel (Swain et al. 2022). Nevertheless, under certain circumstances, such as damage to coatings or high fouling pressure, fouling may still occur. In these cases, performance monitoring introduced in the following section allows inferences on cleaning interventions.

Performance monitoring

Since the introduction of the automated identification system (AIS) based on terrestrial (2007) and satellite tracking (2013), data from a broad variety of vessels is accessible. Particularly trade routes, vessel speed, idle times, and water conditions are of interest to predict the fouling status. Additionally, ISO 19030 introduced in 2016, allows continuous monitoring of the performance of vessels to creates important insight on the fouling status.

With these tools on hand, the top paint companies started to expand their product line with online monitoring tools, which help to predict biofouling and guarantee the performance of their antifouling coatings. For example, AkzoNobel's Intertrac HullCare as well as Chugoku's monitoring & Analysis Program (MAP) harness the power of big data collected by their software. Combined with ISO19030 these tools provide guidance for maintenance advice. Jotun also offers a hull performance solution (HullKeeper), which claims optimised hull performance regardless of the applied coating. The use of the program enables ship operators to take full control of their operations with hull monitoring, fouling risk alerts, inspections and advisory services helping them to identify potential fouling problems. Further, 3rd party companies, such as WE4SEA, entered the market providing competitive online tools for ship monitoring.

Today, 34% of ships are still inspected based on calendar schedule, followed by 24% using semi-automatic calculations (BIMCO 2022). Online hull performance solutions are on 3rd place with an implementation rate of 15%. Clearly, there is a market for advanced analysis software which tailors the cleaning schedule to the individual need of the ship owner. Since the change to an on demand-based schedule has a high potential to reduce costs and time for drydocking, data driven approaches using digital twins to model and predict performance are likely to become a future standard (Coraddu et al. 2019).

Currently used biocides

As previously mentioned, biocides are key components of all AF and some FR coatings. After the ban of organotin compounds, such as tributyltin (TBT), their market share of up to 70% was taken by cuprous oxide (Cu_2O) (Evans et al. 2000; Chambers et al. 2006). Today, 75% of all paint formulations contain Cu in the form of Cu_2O, CuSCN, or metallic copper, whereas Cu free coatings use predominately Zn (Paz-Villarraga et al. 2022). Cu_2O is a broad acting biocide exerting its antifouling effect on soft and hard fouling organisms through a variety of interaction mechanisms affecting cell homeostasis and cell integrity (Table 3). Depending on the fouling pressure at intended geographical locations, the Cu content used in SPC coatings varies between 7 and 75 wt% (Lindgren et al. 2018). As an estimate, the average release rate to deter barnacle settlement is 4.5 µg cm^{-2} day^{-1} (Lindgren et al. 2018).

Since the toxicity of copper is lower than tin, some organisms, such as certain algae, tolerate high concentrations of copper, rendering it necessary to include additional biocides in the paint that target individual organisms (Voulvoulis et al. 1999; Manzo et al. 2008; Amara et al. 2018). These booster biocides (Table 3) are added to the formulations at concentrations between 0.1 and 10 wt% (Bressy et al. 2022; Cima and Varello 2023).

Most booster biocides that were introduced as alternative to TBT claim a fast degradation rate and low bioaccumulation. However, studies on the environmental fate are still ongoing and elevated concentrations is often found in marine sediments (de Campos et al. 2022). Additionally, marine biologists agree that more knowledge is needed on the mechanistic details of pharmacodynamics and pharmacokinetics, particularly in non-target species. Therefore, the European regulatory body (ECHA) requires the evaluation and approval of both biocides and antifouling coating systems. These proceedings led to the latest ban of Irgarol 1051 (cybutryne), which is a biocide that inhibits electron transfer during photosynthesis in algae (Amara et al. 2018; Soon et al. 2019).

Future developments

With regards to the further development of biocides, the current excessive costs and processing times for approvals slow down the market access of novel compounds. Although there are many experimental studies on eco-friendly and nature-derived products that deter biofouling without being toxic (Villa and Cappitelli 2013), they lack up-scaling for the translation into industry. An example of such a non-toxic approach is to tackle the establishment of biofilms through inhibiting quorum sensing. Thereby, the communication between bacteria and other organisms may interrupt the biofouling cascade (Dobretsov et al. 2011).

Table 3. Biocides approved by the European BPR (exception diuron and zinc pyrithione which are in the approval process) and their toxicity (Amara et al. 2018).

Name	Brand name	Target species	Mode of action
Cu_2O		Broad-spectrum antifouling agent	Competitive binding to metal ligands, creation of reactive oxygen species, interaction with nucleic acids (Borkow and Gabbay 2005)
DCOIT	Sea-Nine™ 211 N. Rocima™ 200	Broad-spectrum antifouling agent targeting soft fouling	Creation of radicals causing intracellular oxidative stress (Chapman and Diehl 1995)
Diuron		Diatoms and algae	Photosynthesis inhibitor (Jung et al. 2017)
Chlorothalonil		Hard fouling organism, crustaceans, and invertebrates	Inhibition of thiol-dependent proteins and enzymes (Tillman et al. 1973)
Dichlofluanid (DCF)	Preventol®	Macro algae, diatoms, and invertebrate fouling organisms	No information about toxic mechanism of DCF and its degradation products (Jung et al. 2017)
Zinc/Copper pyrithione	Zn/Cu Omadine	Broad-spectrum antifouling agent targeting soft fouling	Inhibiting several cellular processes, such as ATP balance, membrane transport, and protein synthesis (Dahllöf et al. 2005)
Tralopyril	Econea®	Broad-spectrum antifouling agent targeting hard fouling	Interfering with mitochondrial function (Vilas-Boas et al. 2022)
Zineb		broad-spectrum antifouling agent targeting soft and invertebrate fouling organisms	Inhibitor of metabolic pathways through thiol interactions within proteins and enzymes (Hunter and Evans 1990)
Medetomidine	Selektope	Barnacle larvae	Receptor stimulation, causing hyperactive swimming behaviour (Dahlström et al. 2000)

Alternatively, the protective biofilm matrix and adhesive patches could be weakened through enzymes that degrade the polysaccharides expressed by fouling organisms (Dobretsov et al. 2007). The adhesion to surfaces can also be suppressed through nano-structured surfaces topographies. This was achieved by incorporating various NP, such as carbon nanotubes, or β-MnO_2 nanorods, into FR coatings (Sun et al. 2018; Selim et al. 2019). In contrast, formulating copper and zinc-based AF coatings with nano-sized Cu_2O or ZnO particles did not boost the efficacy compared to micro sized particles, indicating that the effect is based on unique morphological properties (Miller et al. 2020).

Meanwhile, industry is focusing on established compounds and optimising their release profiles. This is primarily addressed through encapsulation techniques, such as developed for Sea-Nine Ultra (Andersson Trojer et al. 2015). An added benefit of encapsulating toxic compounds is a more secure handling during paint production. Another way to tune the distribution of active components are vascular like networks that aid the migration of active components (Howell et al. 2014).

With the rise of global sustainability goals, there is also a trend to move towards green chemistry and improve the environmental footprint of existing coating technologies. To fulfil these goals, chemicals can either be sourced from renewable and recycled sources (Lemesle et al. 2021) or substituted with less toxic compounds. Examples amongst many are the use of biopolymers or the replacement of tin-based catalysts used in FR coatings with titanium-based alternatives (Karak 2016; Wang D et al. 2017). Latter is particularly relevant since heavy metal catalysts as well as cyclic low molecular weight siloxane compounds are toxic to marine organisms (Rittschof et al. 2022).

Finally, there are also advances of antifouling technologies that do not require chemical components. There is noteworthy progress in development of UV-C panels with low power LEDs embedded in a silicone matrix for the application on exterior hull surfaces (Hijnen and Jongerius 2018). Until this technology is applicable for large structures, their use is at least practical for smaller submerged components and sensors. Alternatively, ultrasound-based technologies are effective in preventing biofouling by creating cavitation bubbles (Legg et al. 2015). While there are concerns about their application in open waters due to noise levels that impact other marine organisms, it can be an effective option for land-based operations. For small internal surfaces heat may be an option but it is energy intensive compared to the other solutions (Song C and Cui 2020).

Environmental and economic impacts

Greenhouse gas emissions

Despite the detrimental toxic effects of biocidal coating solutions, they are necessary to combat the increase in fossil fuel consumption associated with elevated drag. Even light soft fouling results in added 10 to 16% required shaft power to maintain a vessel's speed. Beyond soft fouling, the power loss due to hard calcareous fouling can reach up to 85% depending on fouling degree and vessel speed (Schultz 2007).

It is recognised that the emission of greenhouse gasses (GHG) in marine shipping significantly contributes to the global temperature rise (Poloczanska and Butler 2010). Despite that, shipping is still the most efficient method for global trade in contrast to other transport sectors, emitting about 7 gCO_2/t km (European Environment Agency 2022). The fourth IMO GHG study conducted in 2020 estimates that GHG emissions in the shipping sector have increased by 10% from 977 million tonnes in 2012 to 1076 million tonnes in 2018, representing 2.89% of the global anthropogenic CO_2 emissions (Figure 3). Nonetheless, between 2012 and 2018 the carbon intensity index for international shipping has improved by 29% compared to estimates in 2008 (International Maritime Organization 2020). This indicates that the global number of vessels increases with a simultaneous improvement in efficiency. This is due to stricter regulations such as the IMO's Annex VI to MARPOL 73/78 in 2011 and the enforcement of EEDI and EEOI standards in 2013 (Chen et al. 2019). These efficiency standards resulted in a significant drop in NO_2 and CH_4 emissions and led to reduced CO_2 emissions. Yet current predictions still anticipate a rise of CO_2 emissions totalling 1700 million tons by 2050, whereas the IMO target is set to 340 million tons.

This target may not be reached with improved hull performance only. Although up to 10% of fuel can be saved by an optimal hull surface, alternative fuel saving techniques must be taken into consideration (Bouman et al. 2017). Changes in hull design, alternative fuels, electric power and propulsion systems, and voyage optimizations have the potential to reduce fuel consumption and GHG emissions by up to 77% (Bouman et al. 2017).

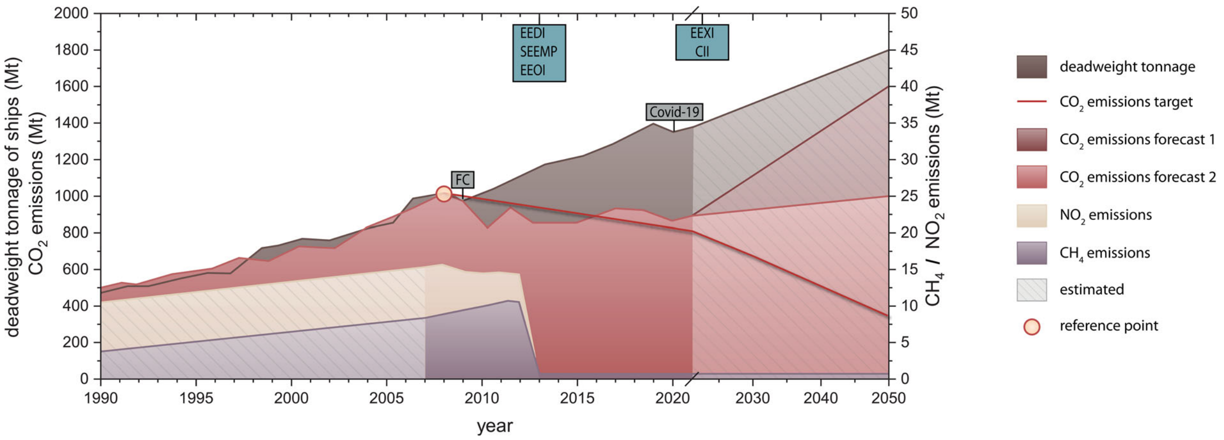

Figure 3. Development and prediction of the global shipping industry and their GHG emissions. Since 2013, the IMO enforces regulations to gradually reduce the CO_2 emissions as well as limit N_2O and CH_4 emissions compared to their reference point in 2008 (Chen et al. 2019). Predictive models based on pessimistic (1), and optimistic (2) scenarios forecast the development of GHG emissions up to 2050 (Bouman et al. 2017).

Invasive aquatic species

Biofouling not only affects the emissions of GHGs but also has a significant effect on biodiversity (Figure 4a,b). The main factors for translocation of species are ballast water and fouled hulls of vessels trading all around the globe (Gollasch et al. 2007; Georgiades et al. 2021). This caused the establishment of today's most common IAS: cholera, water flea, bay barnacles, Northern Pacific seastars, zebra mussels, Asian kelp, European green crabs, and various algae (International Maritime Organization 2023a, 2023b). Consequently, the control of harmful non-indigenous species causes a significant economic impact. In the United States alone 147 billion dollars are spent annually to cope with IAS (Lovell et al. 2006).

Although ballast water disposal is regulated, hull fouling remains an issue at large commercial harbours hosting IAS. Therefore, biologic monitoring protocols have to be implemented to obtain deeper knowledge about types of IAS and their spreading vectors. This would aid the development of predictive models of vulnerable sites for IAS to protect these environments in future (Lee et al. 2008; Vander Zanden and Olden 2008; Goldsmit et al. 2018). Particularly with the climate change, new locations, which are still free of IAS, may become endangered (Figure 4c,d) (Havel et al. 2015). To protect vulnerable sites more efficiently, the potential propagule pressure is estimated based on vessel surface area and ballast water (Ceballos-Osuna et al. 2021). Although this model is still relatively simple, it provides an indication for ports how to allocate their resources. Future adaptations based on vessel specific trade route histories may improve the estimates of cumulative IAS transmission potential.

Microplastics

The use of protective coatings often sparks the discussion about their impact on marine microplastic levels. It cannot be denied that paint particles found in sediments around harbours and marine industrial areas originate from coating application and maintenance activities (Takahashi et al. 2012; Song YK et al. 2014). While most studies generalise decorative and AF coatings, it should be distinguished between their intended use. Today's SPC-type coatings erode and dissolve over time in the ocean. These degradable polymers are not always considered as microplastics due to the lack of definition of the term microplastic (Verschoor et al. 2016; Shi et al. 2021). Despite that, there are still concerns about the impact of potential toxic additives (Gaylarde et al. 2021).

Typically, seawater sampling in coastal areas attest a variety of microplastics contamination (Figure 4e). Amongst the found polymeric materials, PP, PE, PS, PET, PVC, and Nylon are most common, representing about 90% of microplastics (Vianello et al. 2013; Phuong et al. 2018). Often, they originate from textiles and insufficient treatment of communal wastewater (Claessens et al. 2011; Haave et al. 2019; Nikki et al. 2021).

Hence, proper collection of paint residues and industrial waste would be the first method to prevent the further decrease microplastic levels. A report of the Dutch National Institute for Public Health and the Environment credits that with the establishment

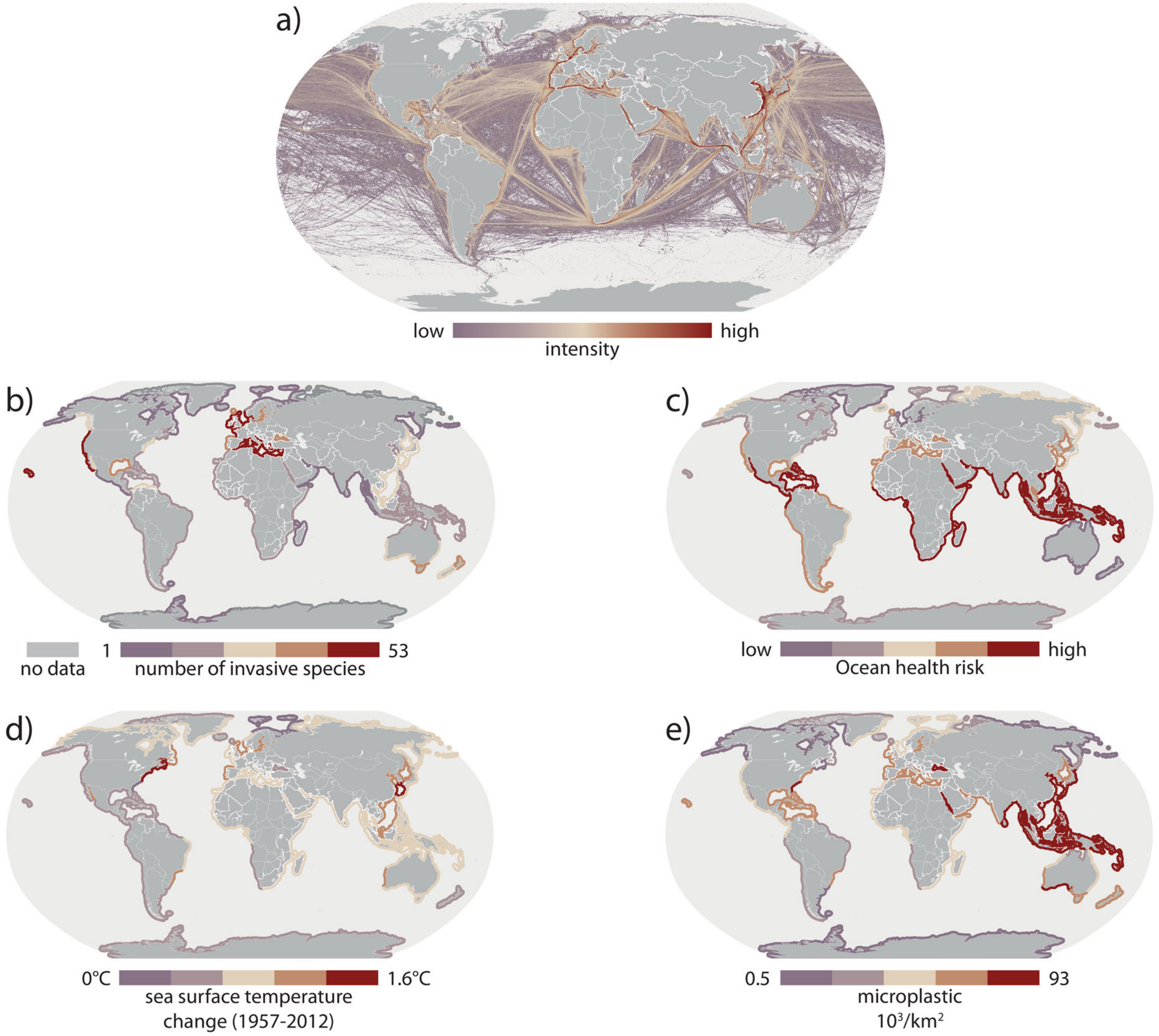

Figure 4. (a) Main shipping trade routes based on AIS data from 2015 to 2021 (Adam Symington 2023). (b) Number of alien invasive species reported in coastal areas in 2008 (Molnar et al. 2008). (c) health condition of the marine environment based on ten diverse socio-ecological indices (Halpern et al. 2012). (d) change in sea surface temperature between 1957 and 2012 (Belkin 2009). (e) amount of floating microplastics (particles < 4.75 mm) in coastal areas based on modelled data in 2012 (Lebreton et al. 2012). Images (b) to (e) were adapted from onesharedocean.org (Jay O'Reilly 2023).

of good environmental practice at their ports, the collection of paint material being removed during professional sandblasting and maintenance activities is 97.5% (Verschoor et al. 2016). Despite best practice, the Dutch authorities estimate about 200 t/y microplastic emission from their shipping sector.

Performance loss in marine infrastructure

Fouling not only affects hulls but also internal systems of vessels. In case that vital engine cooling components are affected, the impact of fouling on the operating costs is exponential compared to a linear behaviour of hull fouling (Davidson et al. 2023). Fouling is however not only an issue for vessels, but also for other marine infrastructure, such as marine energy production, aquaculture, and port infrastructure (Hopkins et al. 2021). The renewable energy sector mostly faces the problem to maintain an efficient power generation to compete with traditional methods of energy generation. Studies on static offshore installations in the northern Beibu gulf of China found fouling reaching up to $30\,kg/m^2$ (Yan et al. 2006). This causes an increase in hydrodynamic structural displacement volume, structural weight, and flow instability, which affect the integrity of marine equipment (Jusoh and Wolfram 1996). Other effects are the physical obstruction of sensors or in-/outlets that compromises essential functions of marine equipment. For static infrastructure, fouling that deteriorates protective coatings causing corrosion is most detrimental and significantly contributes to operation and maintenance costs of offshore equipment (Röckmann et al. 2017). Since maintenance costs account for 25–30% of the total lifecycle costs in windfarms, biofouling prevention has a high potential to save costs (Loxton et al. 2017).

The last industry worth mentioning being affected by biofouling is the aquaculture sector. Biofouling of nets can act as reservoir for fish pathogens that threaten their health. Therefore, biofouling management is also extremely important for eco-friendly fish farming. Currently, the direct biofouling management costs for a typical salmon farm are estimated to be US$420,000 to $493,600 per production cycle, equating to 2.2% of the production costs (Bloecher and Floerl 2021). Fouling resistant nets and equipment would improve the farming conditions of fish while simultaneously reducing costs and staff safety risk associated with cleaning.

Future regulatory and policy perspectives

With Cu based coatings becoming the most dominant biocidal product used today, elevated levels of Cu in water and marine soils are found at various test sites. Particularly enclosed areas and waters with high salinity seem to be most affected (Lagerström et al. 2020). At such sites, the Cu levels in sediment reaches up to 20-fold increase over natural Cu levels in seawater (Hobbs et al. 2022). This is often due to contamination with eroded paint particles which accumulate in the sediment (Soroldoni et al. 2017). Since laboratory tests typically dissolve all compounds in acid (Finnie 2006), it may be an overestimated value, but with the future climate change and ocean acidification this may become more problematic (Doney 2006).

Currently it is not foreseeable how long Cu containing products are still approved. In addition, booster biocides face the same problem since numerous studies have detected their potential bioaccumulation (Konstantinou and Albanis 2004; Thomas K 2009). With uncertainties of their environmental fate, future restrictions may be introduced. At the forefront of regulating the use of biocides in commercial coatings is South Korea. Their regulatory authorities already mandate the maximum concentration of certain biocides, such as Selektope and DCOIT, in coatings to be <1.0 wt.%, with further biocides likely to follow these thresholds (Kim 2021).

Currently the marine industry also faces the issue with a highly fragmented landscape of individual laws on the use of biocides. Within the EU, antifouling paints are regulated through the BPR which requires an environmental risk assessment stating the product's biocidal release rates according to standard test methods. These processes and standards differ around the world, which causes a high level of bureaucracy and time for approvals.

This fragmented regulatory landscape is also detrimental in terms of establishing alternative biofouling control technologies. With ROV cleaning becoming increasingly important, the industry needs to decide how the use is implemented regarding waste collection and treatment. This could be inspired by regulations on ballast water, where a ballast water treatment plan mandates the exchange of ballast water away from coastal waters and limits the maximum number of viable organisms discharged (Salleh et al. 2021). To accurately monitor these limits, cost-effective test and verification methods need to be defined. Particularly a quantification of organism viability would improve complying with regulations (Drake et al. 2014).

With regards to the efficiency of vessels and their GHG emissions the current EEDI and EEOI standards are being further developed. From 1st January 2023, it will be mandatory for all ships to calculate their attained Energy Efficiency Existing Ship Index (EEXI) and report their annual operational carbon intensity indicator (CII) rating. These metrics, defined by the SEEMP Part III, are basis for every vessel's mandatory performance documents, which are reported and stored starting from 2024. This paves the way to reduce the carbon intensity of all ships by 40% by 2030 compared to 2008 (Figure 3). The European Commission also proposed to include the shipping industry into their emissions trading system. In the period from 2025 to 2027, ships above 5000 t traveling to and from the EU must obtain allowances that cover 40% to 100% of their GHG emissions of previous years. This inclusion will further cap the allowed emissions of GHG and simultaneously fund future research and technologies through the EU's innovation and climate investment funds.

Compliance with the regulations also requires efficient auditing methods. Visionary ideas propose digital cleanliness passports which are hosted decentralized on the blockchain where the cleaning history is checked by local authorities (Abdallah et al. 2022).

Conclusions

Marine organisms have developed a broad variety of strategies to conquer surfaces under water causing loss of performance and increase of GHG emissions in the marine industry. Further, the spread of invasive aquatic species, host of pathogens, and loss of biodiversity are primary concerns of biofouling. To combat the growth of marine organisms on submerged surfaces, the use of biocidal coatings is widespread. However, their future use is not guaranteed due to bioaccumulation and toxicity concerns to non-target species.

Therefore, other preventive methods must be implemented and standardized for a unified global use. Amongst available technologies are proactive grooming and fouling release coatings, each coming with their own current drawbacks. Water cleaning requires appropriate waste treatment to avoid the spread of IAS and dispersion of microparticles. Biocide-free FR coatings still offer limited performance in terms of soft fouling, particularly during long idle periods which pressures manufacturers to add booster biocides.

Currently, there is still a lack of knowledge regarding mechanistic principles of biofouling and biologic adhesion cues for marine organisms. Additionally, the effect of voyage variation and idle periods in different environments on fouling are not well documented. The rise of data science tools, machine learning, and artificial intelligence could evolve to the future of fouling prevention. Through the collection of large data assets correlating vessel activity and the occurrence of fouling species, it may be possible to predict biofouling and suggest mitigation strategies.

Until environmentally friendly technologies perform at the level of classic antifouling coatings, their potential toxicity has to be weight against the reduction of GHG emissions and prevention of IAS. Ultimately, biocidal AF coatings should be phased out to avoid loss of biodiversity and costly wastewater treatment. However, currently, their performance and costs are unmatched. The most promising available alternative, which balances the environmental impact with the toxicity of biocides, are FR coatings containing low amount of booster biocides.

The future shift in the industry is also dependent on governmental guidelines. Since the industry currently faces a fragmented regulatory landscape, companies are hesitant to establish innovative technologies ahead of their approval. Therefore, representatives from all parties and countries must collaborate with the regulatory bodies and work towards a set of global rules.

Acknowledgements

The authors thank Jotun for their commercial insights and relevant discussions that contributed to this work. Figure 1 uses graphical elements from BioRender.com.

CRediT authorship contribution statement

F.W.: conceptualization, investigation, visualization, writing - original draft, writing - review & editing, N.E.: conceptualization, project administration, writing - review & editing. All authors have read and agreed to the published version of the manuscript.

Disclosure statement

No potential conflict of interest was reported by the author(s).

Funding

This work was supported by the Norwegian Research Council under Grant [number 327720].

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