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Citation for the original published paper (version of record):
Lagerström, M., Wrange, A., Soares Reis de Oliveira, D. et al (2022). Are silicone foul-release coatings a viable and environmentally sustainable alternative to biocidal antifouling coatings in the Baltic Sea region?. Marine Pollution Bulletin, 184.
http://dx.doi.org/10.1016/j.marpolbul.2022.114102

(article starts on next page)

Marine Pollution Bulletin 184 (2022) 114102

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Are silicone foul-release coatings a viable and environmentally sustainable alternative to biocidal antifouling coatings in the Baltic Sea region?

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ARTICLE INFO

Keywords:
Foul-release coating
Antifouling coating
Antifouling performance
Ecotoxicology
Sustainable products

ABSTRACT

To combat unwanted fouling on immersed hulls, biocidal antifouling coatings are commonly applied to vessels trafficking the Baltic Sea. Here, the efficacy, environmental sustainability and market barriers of silicone foul-release coatings (FRCs) was assessed for this region to evaluate their viability as replacements for biocidal coatings. Coated panels were exposed statically over a 1 year period at three locations in the Baltic Sea region to assess the long-term performance of a biocide-free FRC and two copper coatings. The FRC was found to perform equally well or significantly better than the copper coatings. Even though most silicone FRCs on the market are biocide-free, a review of the literature regarding toxic effects and the identity and environmental fate of leachables shows that they may not be completely environmentally benign, simply for the lack of biocides. Nonetheless, FRCs are substantially less toxic compared to biocidal antifouling coatings and their use should be promoted.

1. Introduction

Biofouling is the unwanted colonization of man-made structures by marine organisms and constitute a particular challenge for ships and leisure boats as a fouled hull increases the hydrodynamic drag of vessels, affecting both speed, fuel consumption and maneuverability (Yebra et al., 2004; Jin et al., 2022). The increased fuel consumption results not only in an economic loss for the operator, but also in increased gaseous (greenhouse gases, sulphur and nitrogen oxides) and particulate air emissions to the environment (Cullinane and Cullinane, 2013). Additionally, as hull fouling communities may harbor non-indigenous species, fouled ship and leisure boat hulls can act as vectors for the unintentional introduction and spread of invasive species (Hewitt et al., 2009; Fernandes et al., 2016). Unfortunately, even though it is both financially and environmentally beneficial to prevent hull fouling, the most common antifouling technique today does so to the detriment of the marine environment. Present-day ships and leisure boat hulls are typically coated with biocide-containing antifouling paints, formulated to release one or more active (toxic) substances to repel or poison settling organisms (Finnie and Williams, 2010). Most coatings contain an inorganic copper-based primary biocide (e.g. cuprous oxide or cuprous thiocyanate) together with one or more organic booster biocides (e.g. copper pyrithione, zineb, DCOIT) (Paz-Villarraga et al., 2022). As these compounds are biologically active, their direct release and dispersion into the marine environment can lead to adverse effects on non-target organisms (Dafforn et al., 2011; Amara et al., 2018). Additionally, as copper does not degrade in the environment, the water, sediment and/or soil compartments in and around areas and facilities where antifouling paints are either applied, maintained or used (e.g. ports, harbors, marinas, shipping lanes, boatyards, shipyards, recycling facilities, etc.) tend to be subject to copper accumulation and pollution (Dafforn et al., 2011; Thomas and Brooks, 2010; Turner, 2010; Eklund and Eklund, 2014; Costa et al., 2016; Boyle et al., 2016; Daehne et al., 2017; Lagerström, 2019; Soon et al., 2021).

For the Baltic Sea, the use of biocidal antifouling paints is of particular concern as it is a heavily trafficked marine region, by ships and recreational boats alike. It has been estimated that at least 2000 ships navigate within its borders at any given moment, and shipping is

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https://doi.org/10.1016/j.marpolbul.2022.114102
Received 1 February 2022; Received in revised form 13 June 2022; Accepted 1 September 2022

projected to increase as a result of the modal shift of transport from road to sea that will take place in Europe (Matzcak et al., 2018). Leisure boats are also ubiquitous in the countries bordering its shores, with nearly two million vessels in the Swedish and Finnish boat parks alone (ICOMIA, 2018). A recent study on the loads of copper to the Baltic Sea shows that the use of antifouling paints on ships and leisure boats is the single largest anthropogenic source of copper, accounting for a third of the total load (Ytreberg et al., 2021). The Baltic Sea is a sensitive sea with brackish water, low biodiversity and high anthropogenic impact. The large number of vessels combined with a slow water exchange rate of approximately 30 years makes it environmentally relevant to minimize the use of biocidal antifouling paints on both recreational and commercial vessels in this semi-enclosed sea (Korpinen et al., 2012).

The leading market alternative to traditional biocidal coatings are so called foul-release coatings (FRCs), which act to prevent the attachment of fouling organism through physical, rather than chemical, action. These coatings can have either silicone- or fluoropolymer based binders, but all commercially available systems are silicone-based (Lejars et al., 2012). Today, all major coating companies with paint products for marine application, market at least one silicone-based product (Kim, 2021). Whereas FRCs held <1 % of the ship market in 2009, this share had risen to nearly 10 % in 2014 (Ciriminna et al., 2015). Silicone FRCs are composed of crosslinked silicone elastomers, most commonly poly (dimethylsiloxane) (PDMS), and prevent the attachment of biofouling by means of their non-stick properties. Their low surface energies and elastic modulus act to reduce the adhesion strength of fouling organisms which can then be readily removed by the water shear force during cleaning or navigation, the latter enabling the coating to "self-clean" (Brady and Singer, 2000). In addition, silicone FRCs have low surface roughness and are hydrodynamically smoother than traditional biocidal coatings leading to reduced drag, if unfouled (Townsin and Anderson, 2009). The optimization of silicone FRCs is a field of on-going and active research, with investigated modifications targeting the main drawbacks of this coating system, namely fouling on idle periods and poor mechanical strength (Hu et al., 2020). As a result, the composition of commercial silicone FRCs has changed considerably over the past few decades. There is a great number of published studies describing various chemical alterations and additions to silicone-based matrices and their effect on coating properties. However, only a few of these strategies have been implemented in commercially available products (Barletta et al., 2018) and are described next.

While the first generation FRCs launched in the 1990's and early 2000's consisted mainly of hydrophobic PDMS-based formulations, most second and third generation products hold amphiphilic surface properties (Camós Noguer, 2016; Gevaux, 2019) (Fig. 1). First generation coatings had difficulties in preventing slime adhesion as diatoms could adhere more strongly to their hydrophobic surfaces, impeding their release from PDMS-coated surfaces even at high speeds (> 30 knots) (Holland et al., 2004). Such diatom slimes can significantly increase the frictional resistance of the vessel and also influence the subsequent settlement of macrofouling organisms (Lejars et al., 2012; Schultz, 2007). The more recently launched products are therefore amphiphilic with coating surfaces that combine domains of hydrophilic and hydrophobic character in order to confuse organisms during settlement and adhesion (Galli and Martinelli, 2017). As such, these coatings are not only foul-releasing, but also foul-resistant, especially during static conditions (Hu et al., 2020). To produce these surface-active coatings, amphiphilic additives of varying chemical composition, are employed. These substances, most commonly amphiphilic block copolymers, segregate at the coating surface where the hydrophobic block (e.g. PDMS-based) acts as a tether to the surface, while the hydrophilic block (e.g. polyethylene glycol(PEG)-based) is usually extended to seawater and provide the non-fouling properties (Camós Noguer et al., 2017). In Hempel's biocide-free coatings for ships (Hempasil X3+) and leisure boats (Silic One), these additives consist of hydrophilic-modified silicone oils that migrate to the surface of the coating to enable the formation of a hydrogel, i.e. a complex polymer network with the ability to bind large amounts of water. At the aqueous interface, the siloxane chain of the substance acts to anchor it in the coating, while the hydrophilic moieties are hydrated upon contact with water to form the hydrogel layer over the hull (Ciriminna et al., 2015; Camós Noguer, 2016; Noguer, 2021). The hydrogel has been shown to delay diatomic settlement compared with a traditional foul-release silicone coating (Thorlaksen et al., 2010). The incorporation of fluoropolymers is another approach that has been adopted for some products (e.g. Intersleek 900 and its successor Intersleek 1100SR) to generate a coating surface with

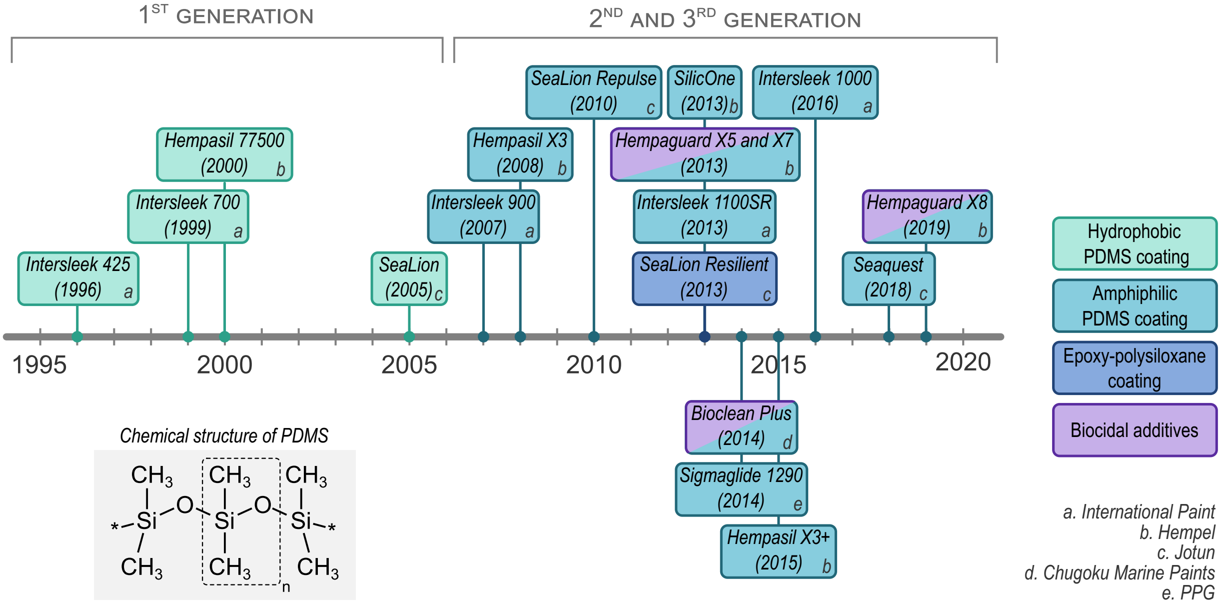

Fig. 1. Examples of commercial silicone foul-release coatings (launch year in parenthesis) and their change in characteristic over time (Lejars et al., 2012; Fathom, 2013; Ciriminna et al., 2015; Camós Noguer, 2016; Gevaux, 2019). Note that all products listed here are intended for use on ships, except SilicOne which is intended for recreational vessels. Also shown is the typical chemical structure of the PDMS elastomer backbone in foul-release coatings, with methyl side groups and chain-ends (*) that can be hydroxy-, amino- or alkoxy-terminated.

amphiphilic properties (Hu et al., 2020).

Other types of additives to silicone FRCs include hydrophobic oils and biocides. The addition of silicone oils consisting of differently substituted, non-crosslinked PDMSs has been reported for some first generation commercial coatings, with concerns raised regarding the environmental repercussions of releasing these persistent compounds into the marine environment (Nendza, 2007). Additions of other types of oils such as paraffin and mineral oils have also been reported and the latest launched product by International paints (Intersleek 1000) contains lanolin oil (Gevaux, 2019; Watermann et al., 1997). Although the first generation of FRCs were biocide-free, biocidal additives can now also be found in some market products (e.g. Hempaguard and Bioclean Plus). Finally, the modification of silicone-based FRCs with epoxy-segments to improve upon the mechanical strength has been adopted in, for example, Jotun's SeaLion Resilient. The epoxy-polysiloxane resin is meant to reduce mechanical damage, while still providing a glossy, smooth surface to prevent both drag and the settlement of fouling organisms (Jotun, 2013).

Even though silicone coatings have been readily available on the market for many years, scientific studies comparing the efficacy of foul-release and traditional copper coatings are lacking for the Baltic Sea region. To date, only one such study has been performed at a location in Kattegat, near Gothenburg on the Swedish west coast, by Oliveira and Granhag (2020). In their study, the efficacies of a biocide-free FRC (Sigmaglide 1290 by PPG), as well as a self-polishing copper-containing ship paint were evaluated over a one year period through static panel testing. The results showed the FRC to be significantly less fouled compared to the biocidal coating. The overall aim of this study was to determine whether biocide-free FRCs indeed constitute a viable option to traditional copper coatings for the wider Baltic Sea region, both from an efficacy and environmental sustainability standpoint. This work has therefore been divided into three sections. Firstly, the long-term performance of two commercial copper coatings and one silicone FRC were compared. Coated panels were exposed in the field at three locations in the Baltic Sea region holding low to high fouling pressures over a one year period to assess the efficacy of the coatings during worst-case (static) conditions. The hydrogel-based FRC, Silic One by Hempel, was used as it is currently the only FRC widely available to recreational boaters around the Baltic Sea. This coating is also based on the same hydrogel technology as Hempasil X3+, intended for ships. The implication of the obtained results can thus be discussed in relation to both ships and leisure boats operating in the Baltic Sea region. As described, the composition of commercial silicone FRCs can vary and reports of different types of leachables from these products prompted the second part of this work. Here, the aim was to assess whether commercial silicone FRCs constitute a viable alternative to biocidal coatings from an environmental point of view. Their environmental sustainability was therefore investigated through a review of the published literature with regards to two aspects, namely, their toxicity to aquatic organisms, particularly in comparison to copper coatings, and the chemical nature and potential environmental hazard of possible leachables. In the third and final part, the barriers that may currently be hindering the establishment of silicone FRCs on both the professional and recreational markets were analyzed.

2. Materials & methods

2.1. Efficacy study

2.1.1. Panel preparation and exposure sites

Four coatings were included in the study: a biocide-free paint (Control), two copper-containing antifouling paints (Copper 1 and Copper 2) and a silicone FRC (Foul-release) (Table 1). While the copper paints are intended to be used on commercial ships, antifouling paints of similar composition are available to leisure boat owners in most countries around the Baltic Sea. The silicone FRC used in this study (Silic One) is marketed for recreational vessels, but Hempel also has a ship coating (HempaSil X3+) based on the same hydrogel technology. The results from these coatings are thus relevant for both ships and leisure boats. All investigated coatings were applied to 15 × 15 cm PVC (Poly Vinyl Chloride) panels. Panels were sanded and then painted with a roller, with a first layer of primer (Hempel's Underwater Primer 26030), followed by a top coating. For the silicone coating, a layer of tie coat (Hempel's Silic One Tiecoat 27450) was also applied before the top coat. Four replicates of each treatment were prepared for each exposure site.

The panels were attached to frames which were deployed in July 2020 at a depth of 1.5 m (exposure depth of 1.5 ± 0.5 m for the panels) at three different research stations along the Swedish coast: Askö Laboratory (Station 1), Kristineberg Marine Research Station (Station 2) and Tjärnö Marine Laboratory (Station 3). Salinity and temperature measurements at 1 m depth were obtained from monitoring sites at the research stations themselves (Stations 2 and 3), or from nearby buoys (Station 1) (Kristineberg Marine Research Station, 2021; SMHI, 2021; Tjärnö Marine Laboratory, 2021). Station 1 is located in the Baltic Proper and held an average salinity of 6 PSU, whereas stations 2 and 3 with respective average salinities of 24 and 26 PSU, are located in Skagerrak (Fig. 2). Panels were hauled and photographed approximately every month over a one year period, for later visual assessment of type and surface coverage of fouling organisms. Unfortunately, the panels at Station 3 were disturbed after 8 months of exposure, likely by eider ducks feeding on blue mussels that dominated the panels. The experiment at Station 3 therefore had to be terminated early in March 2021.

2.1.2. Fouling assessment

The fouling on the panels was rated on a scale from 0 to 100 according to a classification system from the Naval Ships' Technical Manual (NSTM) of the US Navy (2006). A fouling rate (FR) of 0 represents a clean and foul-free surface, while FR10 and above represent different types of fouling organisms, with increased fouling rate indicating increased fouling severity (Table 2). Fouling rates of 10–30 represent soft fouling, e.g. various types of algae, whereas FR40 and above represent hard fouling, i.e. calcareous fouling. The NSTM scale does not include a recommended rating for the classification of bryozoans. When present, a fouling rate of 40 was used for bryozoans as it was deemed most representative. The surface coverage of each identified rating type was estimated according to ASTM D 6990 (2011). In

Table 1

Coatings used in the study. Information regarding the content of active substances (cuprous oxide and DCOIT) were obtained from the Swedish Chemicals Agency public pesticide register (Swedish Chemicals Agency, 2021). The zinc oxide content range was collected from the products' safety data sheets.

Coating	Product name	Manufacturer	Color	Cuprous oxide (wt%, ww)	Copper pyrithione (wt%, ww)	DCOIT[a] (wt%, ww)	Zinc oxide (wt%, ww)
Control	Underwater Primer 26030	Hempel	Aluminium grey	–	–	–	–
Copper 1	Sigmarine 530	PPG	Redbrown	39.02	–	2.53	10–25
Copper 2	SeaForce 60 (spl)	Jotun	Red	31.6	1.5	–	10–25
Foul-release	Silic One	Hempel	Black	–	–	–	–

[a] 4,5-Dichloro-2-octyl-isothiazolone.

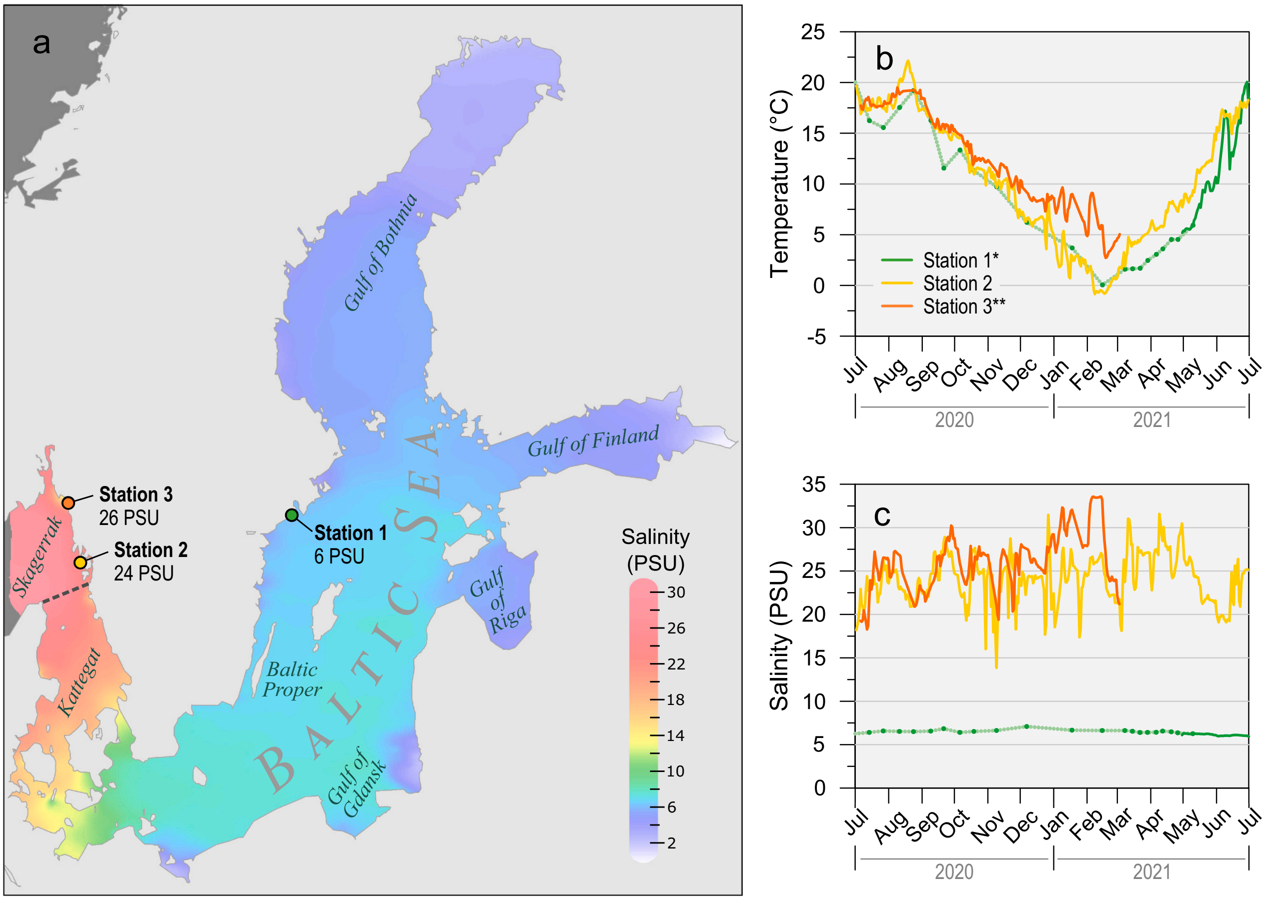

Fig. 2. Map showing the location of the three stations (a). The average salinity during the study is indicated below each station. The average daily temperature (b) and salinity (c) at 1 m depth for the whole study period are also shown. Daily averages of salinity and temperature were only available for the last few months of exposure at Station 1 (*) due to maintenance work on the buoy at Askö B1. The dotted green lines are instead showing ~ monthly data (dots show the exact sampling point) at 0 m depth at the same location for the period July 2020–April 2021, as measured by the national environmental monitoring program at station B1. The exposure at Station 3 (**) had to be terminated ahead of time, after roughly 8 months, which is also reflected in the temperature and salinity graphs. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

accordance with this method, biofouling attachment within 13 mm from all edges of the panel was disregarded in the analysis. The assessed surface coverages of fouling rates were used to characterize the fouling pressure at the three stations (through evaluation of the fouling on the control paint) and to compare the efficacy of the studied antifouling coatings in their ability to deter different types of fouling organisms.

To make an overall comparison between treatments, a single weighted fouling rate, FR_w, was also determined for each replicate panel (Oliveira and Granhag, 2020). The values (0–100) of each identified fouling rating category were multiplied with their corresponding observed surface coverage (in %) and summed according to the following equation (where $n = 100$):

$$FR_w = \sum_{i=0}^{n} \frac{FR_i \times Surface\ coverage_i}{100} \tag{1}$$

The calculated FR_w were used to test for significant differences between the copper and foul-release coatings at each observation point in time by performing one-way ANOVAs with post-hoc testing (Tukey's HSD). All statistical tests were performed in JMP® Pro 15 with a significance level of 5 % ($\alpha = 0.05$).

2.2. Assessment of environmental impact of silicone foul-release coatings

As silicone FRCs are either biocide-free or contain lower amounts of biocides compared to conventional antifouling coatings, they are generally considered more environmentally friendly (Ciriminna et al., 2015). However, concern has been raised over the fact that they may contain environmentally persistent leachables (e.g. silicone oils) that could in part be responsible for coating performance (Nendza, 2007; Buskens et al., 2013). To assess the overall environmental impact of silicone FRCs, two aspects were investigated through a review of the published literature, namely 1) their toxicity to aquatic organisms, and 2) the chemical nature and potential environmental hazard of possible leachables. For the former, a literature search was performed in order to identify ecotoxicological studies including commercial FRCs. For the latter, substances that have been reported in the literature which may be contained and leached from commercial silicone coatings and may be of environmental concern were reviewed.

2.3. Assessment of market barriers for silicone foul-release coatings

To date, there is only one company, Hempel, that markets biocide-free silicone coatings for both ships (Hempasil X3+) and recreational vessels (Silic One, tested in this study), with the latter coating readily available to boaters in most countries around the Baltic Sea. To gain insights with regards to the obstacles that may currently hinder the end-users of each of these markets from switching to a biocide-free silicone coating, an interview with Hempel representatives was carried out, in addition to reviewing the existing literature.

Table 2
Fouling rate scale used in the study to assess the type of fouling present on the coatings. Adapted from the Naval Ships' Technical Manual (NSTM) (US Navy, 2006). The corresponding change in required shaft power for an Oliver Hazard Perry class frigate (FFG-7) at a speed of 15 knots, as derived by Schultz (2007), is also shown for reference.

Fouling type	Fouling rate (FR)	Description	Change in required shaft power for an FFG-7
–	0	Clean, foul-free surface	–
Soft	10	Incipient slime, painted surfaces visible beneath the fouling	11 %
	20	Advanced slime, painted surfaces obscured by the fouling	
	30	Soft fouling up to 76 mm in length and up to 6.4 mm in height (e.g. filaments, sea cucumbers)	21 %
Hard	40	Tubeworms <6.4 mm in height or diameter	35 %
	50	Barnacles <6.4 mm in height or diameter	
	60	Combination of tubeworms and barnacles <6.4 mm in height or diameter	
	70	Combination of tubeworms and barnacles >6.4 mm in height or diameter	54 %
	80	Closely packed tubeworms or barnacles (<6.4 mm in height) growing on top of each other	
	90	Dense growth of tubeworms with barnacles, 6.4 mm or greater in height	86 %
Composite	100	Soft and hard fouling present, with soft fouling growing over various forms of hard growth.	

3. Results & discussion

3.1. Efficacy of a silicone coating relative to copper coatings in the Baltic Sea

3.1.1. Differences in fouling pressure between stations

The fouling pressure, i.e. the intensity and types of fouling organisms, that ships and leisure boat hulls may face in the Baltic Sea region can vary, both in time and space (Wrange et al., 2020). Hence, to evaluate how the investigated coatings perform under different fouling conditions, they were exposed at three different stations. Here, the fouling pressure of each station, as recorded on the control panels, is described.

At station 1 located in the Baltic Sea, the fouling pressure was relatively low with mainly soft fouling consisting of slime (FR10–20) and filamentous algae (FR30). However, barnacles (*Amphibalanus improvisus*) at low densities were also observed after two months of exposure (Fig. 3, Table S1). The weighted FR, FR_w, at station 1 never exceeded 30 during the entire study reflecting the soft fouling and low number of barnacles observed (Fig. 4). This supports previous observations that barnacle settlement occurs 1–3 times per year in the Askö region where the panels were exposed (Wrange et al., 2020).

At station 2 the fouling was more intense, including a higher diversity of organisms and more calcareous and crust-forming species, but also filamentous algae (Fig. 3). Already during the first month of exposure, the panels became colonized by tubeworms (FR40) and crust-forming bryozoans that rapidly became the dominant fouling types on the panels. Although a few barnacles appeared on the panels during the winter months, the major settlement of barnacles occurred from April, resulting in multiple layers of fouling forming. This pattern is reflected in the calculated FR_w, which gradually increased during the first 5 months and then remained constant during winter (at around 40), after which it increased rapidly to above 80 after the barnacle settlement in April (Fig. 4, Table S1). After one year of exposure, the panels displayed multiple layers of fouling including tubeworms, bryozoans, barnacles, oysters, tunicates and filamentous algae (Figs. 4 and 5b).

At Station 3, the fouling pressure was the highest, with fouling rapidly increasing already after the first month of exposure, starting with settlement of blue mussels (*Mytilus edulis*), barnacles and filamentous algae (Figs. 3 and 5c). An $FR_w > 90$ was obtained already after 4 months of exposure (Fig. 4). The fouling was strongly dominated by blue mussels with an underlying layer of barnacles, but tunicates (*Ciona intestinalis*) were also present on the panels. The species diversity was lower compared to station 2 which is likely explained by the strong dominance of the three mentioned species. After 8 months, a sudden reduction of fouling was observed on the panels, indicated that the heavily fouled panels had been disturbed, most likely by eider ducks tearing off the mussels from the panels. The main feeding season of eider ducks occurs during the late winter months and eider ducks are common in the area during winter. However, the loss of mussels may also be partly a result of wave action causing large heavy mussel aggregations to fall of the panels. Station 3 was a more exposed site, compared to the other two stations since the panels were hanging from a floating jetty in the middle of a semi-enclosed bay. In contrast, the panels at station 1 and 2 were hanging from jetties closer to the shoreline. The loss of fouling on the panels resulted in early termination of the panel monitoring at station 3 (Figs. 4 and 5a).

3.1.2. Performance of tested coatings

The average amount and type of fouling, as classified according to the fouling rate scale of the US Navy (2006), present on all copper and foul-release coatings over the course of the one year study period are shown in Fig. 5. For comparison of background fouling pressure, the classified fouling is also shown for the control paint. The average fouling rating and replicate variability for all coatings can be found in the supplementary information (Tables S1–S3).

3.1.2.1. Copper coatings. The two copper paints (Fig. 5d–f and g–i) showed nearly parallel results at all three exposure sites, both regarding the severity (i.e. fouling rating) of the fouling community and its temporal changes. At station 1, both copper coatings were foul-free for the first 2–3 months, after which mainly incipient slime (FR10) and/or, to a small extent, advanced slime (FR20) were observed during the remainder of the one year exposure period (Fig. 5d and g, Table S1). Even though the average portion of the surface covered by fouling ($FR > 0$) was somewhat higher for Copper coating 2 during September–May, this difference is not significant compared to Copper coating 1 (see Table S1 of the supplementary information).

At station 2, where the fouling pressure was more intense, panels coated with either of the two copper paints nonetheless remained foul-free for the first 2–3 months of exposure (Fig. 5e and h, Table S2). The fouling during the subsequent 3 months consists of variable surface coverages (10–80 %) of incipient slime (FR10). After 6 months, i.e. from January 2021 and onward, the fouling coverage steadily increases to gradually reach 100 % in March after which no parts of the panels were foul-free for the remaining part of the experiment. During this time period, incipient slime remains the dominant type of fouling on the panels with average surface coverages of around 80 % for Copper coating 1 and 60 % for Copper coating 2. The rest of the fouled surface was principally covered by advanced slime (FR20). Just as for station 2, incipient (FR10) and advanced (FR20) slime are the dominant types of fouling present on the copper coatings exposed at station 3 (Fig. 5f and i, Table S3). However, their establishment occurs much earlier in time, with slime observed on both copper paints already at the very first reading after only 1 month of exposure. Whereas it took 8 months for various types of slime to completely colonize the surface of the coated

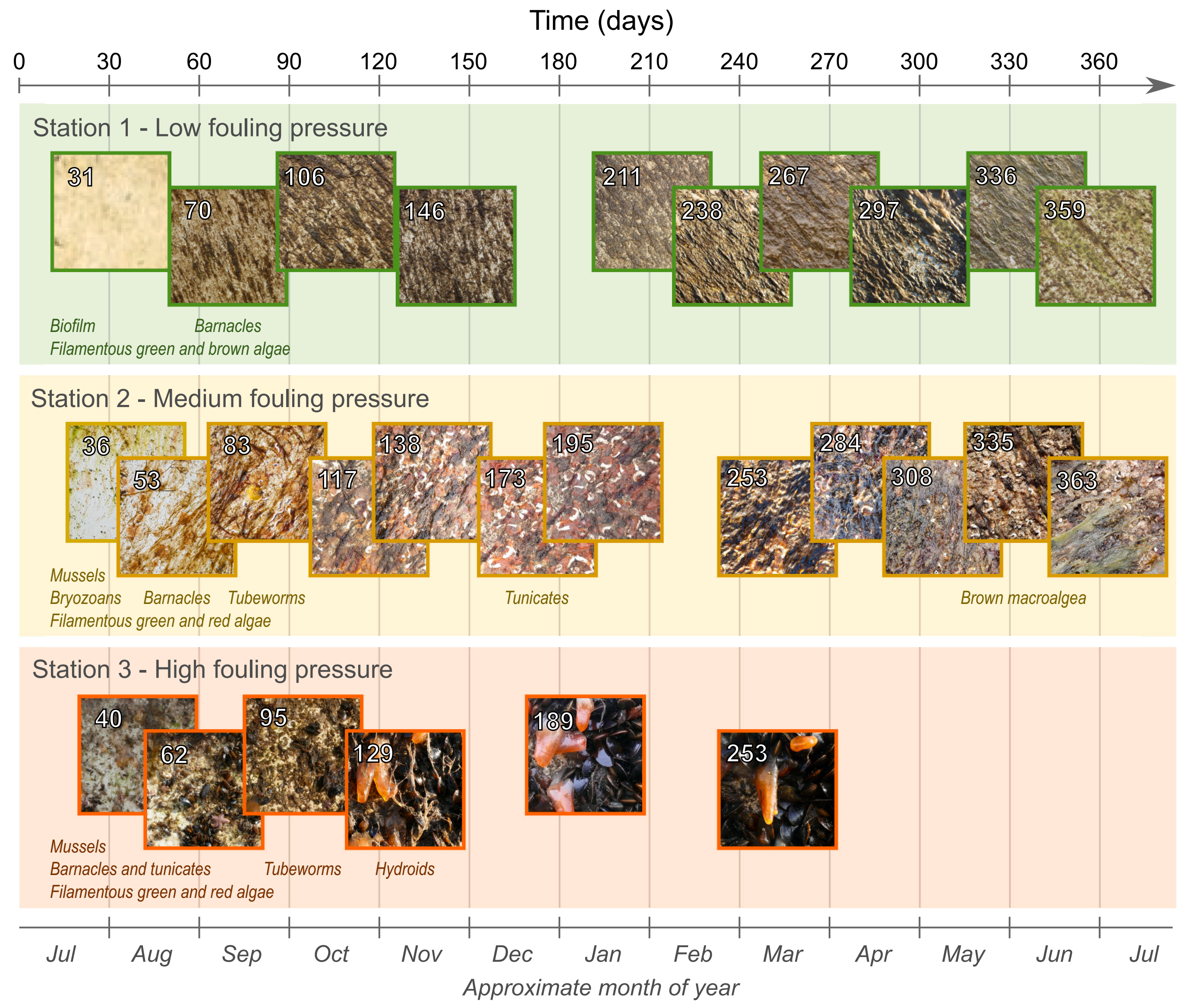

Fig. 3. Example photographs of control panels and main fouling organisms at the three study sites. Numbers on photographs indicate the number of days the panel had been immersed when the picture was taken.

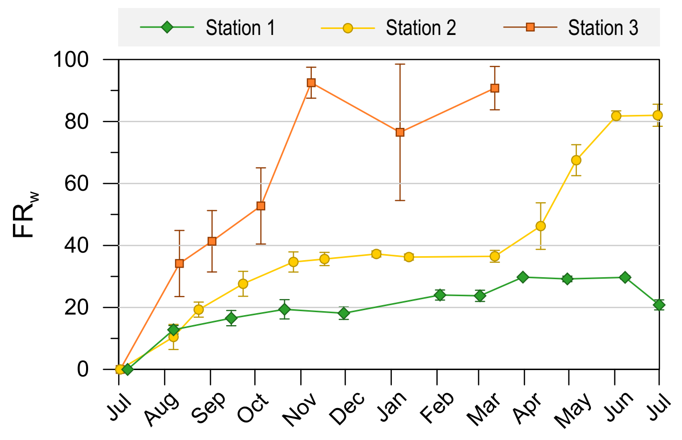

Fig. 4. Calculated weighted fouling rate (FR$_W$) for the control coating at the three stations. Error bars show the standard deviation (n = 4 panels). The exposure at Station 3 was terminated ahead of time, after roughly 8 months (in March 2021).

panels at Station 2, this process only took 3 months at Station 3. At Station 1, with the lowest fouling pressure and salinity, this only occurred for one of the copper coatings (Copper coating 1) after 11 months of exposure.

3.1.2.2. Silicone foul-release coating. For the biocide-free FRC, small occurrences (≤10 %) of slime and algae were present on the coating surface within the first month of exposure at Station 1 (Fig. 5 j, Table S1). The surface coverage of soft fouling (FR10–30) then gradually increased from 10 to 20 % coverage over the first four months (July–October), to 60 % the subsequent 4 months (November–February) and 80 % during the 9th month of exposure (March). Complete surface coverage (100 %) was observed in June after 11 months of exposure. Even though occurrences of advanced slime (FR20) and filamentous algae (FR30) were observed, incipient slime (FR10) was the dominant type of soft fouling across the whole study period. At the last observation after 12 months of exposure, the surface coverage of fouling was markedly lower, dropping from an average of 100 % the previous month down to 13 %, likely due to the self-cleaning properties of the FRC. The fouling accumulated on the panel up until that point may have gotten

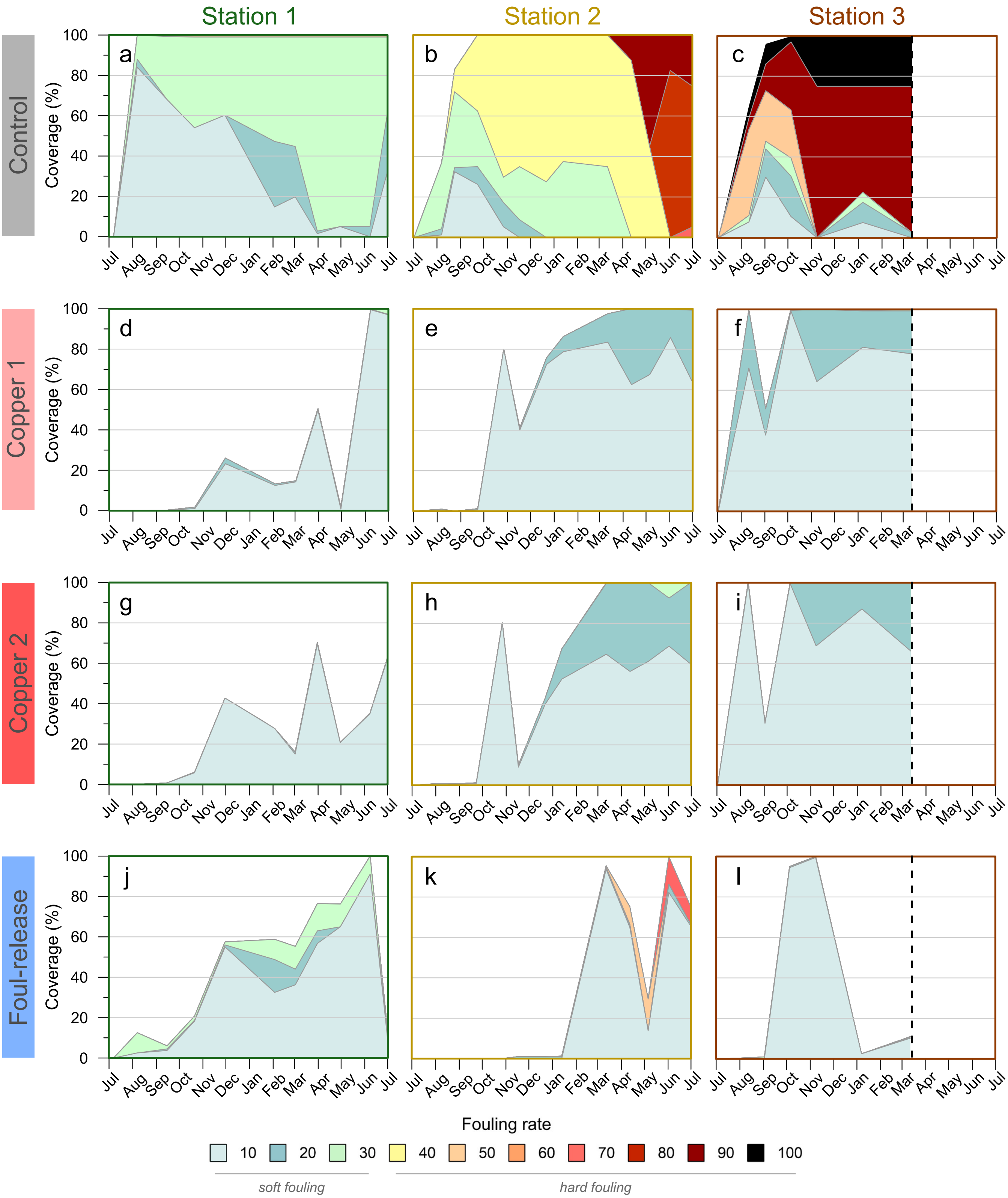

Fig. 5. Average coverage and type of fouling (shown here as fouling rate) on static controls (a–c) and painted panels with copper (d–f and g–i) and silicone foul-release (j–l) coatings during 1 year of continuous exposure at each of the three stations. The exposure at Station 3 was terminated ahead of time, after roughly 8 months (in March 2021).

too heavy to be able to remain adhered to the slippery surface of the coating. It is also possible that the fouling was removed by the action of increased local currents.

At station 2, the FRC remained essentially completely foul-free during the first 6 months of exposure (Fig. 5k, Table S2). From January and onward, incipient slime (varying between ~20–90 % surface coverage) and smaller occurrences of barnacles (1–16 % surface coverage) were observed on the panels. Surprisingly, no barnacles or other hard fouling was observed on the FRC at station 3 (Fig. 5l, Table S3), even though the control panels can testify to an intense fouling pressure with colonization by hard fouling already within the first month of exposure (Fig. 5c). This can be compared to Station 2 where fouling rates >40 were only observed on the control panels 6 months into the study (Table S2). It is nonetheless possible that hard fouling would have eventually established on the FRC at Station 3, if the study had not been terminated early at this site due to the disturbance of the panels. Although not measured in this study, it is also possible that differences in local flow conditions could have impacted the results. If local currents were markedly stronger by one station as compared to another, this could in theory benefit the performance of the FRC at that station by enabling it to self-clean to a greater extent. The large decrease in incipient slime (FR10) coverage between December 2020 and January 2021 at Station 3, could, for instance, have been the result of the self-cleaning mechanism of the coating. It should however be noted that due to light reflection caused by the glossy nature of the FRC surface it was particularly challenging to discriminate incipient slime (FR10) from a foul-free surface (FR0). The FR10 category corresponds to soft fouling resulting in light shades of red or green on the surface of the coating with the painted surface still visible underneath the fouling (US Navy, 2006). The shiny surface of the FRC may have contributed to increased uncertainties in the assessment of surface coverage for this particular type of light fouling.

3.1.3. *Comparison between copper and foul-release coatings*

While the two copper coatings performed similarly to each other at all three sites, it is clear from Fig. 5 that they performed differently compared to the FRC. To make an overall comparison between coatings, weighted fouling rates (FR_w) were calculated (see Eq. (1)) and tested for significant differences using ANOVAs.

At the location with the lowest fouling pressure, Station 1, the extent and type of fouling found on the copper coatings were similar. This is also reflected in the calculated FR_w, where the statistical comparison using ANOVA confirms that the performances of the two copper coatings was not significantly different at any point in time over the 1 year study (Fig. 6a). At Station 1, the presence of incipient slime (FR10) as the main fouling type was common to all coatings. However, advanced slime and algae (FR20 and 30) were present to a larger extent on the FRC (2–29 % of panel area, depending on time point) as compared to Copper paints 1 (0–3 %) and 2 (0–1 %) (Fig. 5). This consequently resulted in a higher average FR_w for the FRC (Fig. 6a). However, the statistical analysis shows that the FR_w of the FRC was only significantly different compared to either of the two copper coatings at two points in time (August 2020 and May 2021). Hence, the copper coatings did not generally perform significantly better than the FRC at this site.

At Station 2, holding an intermediate fouling pressure, both coating types were able to effectively deter fouling during the first three months of the experiment (July–September), but a clear difference between coatings can be observed during the subsequent fall/winter months (October–January). At this time, the FRC was commonly found to have a significantly lower FR_w compared to both copper coatings: while the FR_w of the FRC was constant and nearly zero, the two copper coatings show an increase in their FR_w compared to previous months (July–September) (Fig. 6b). This is particularly interesting given that the FR_w of the control paint does not testify to any increase in fouling pressure at this station during this time. Quite oppositely, the fouling growth appears to have more or less stagnated between October and March (Fig. 4). The reduced performance of the copper coatings may thus instead be coupled to lowered release rates of biocides from the paints. The water temperature at Station 2 dropped rapidly after the month of September (Fig. 2b). This decrease in temperature could have impacted the release rate of biocides in the coating, through reduced biocide and/or paint binder dissolution rates (Ferry and Carritt, 1946; Rascio et al., 1988), enabling the attachment and growth of slime and filamentous algae. During the last 4 months of the study, both the copper coatings and the FRC were found to perform similarly again. This is due to the fact that some calcareous fouling, mainly barnacles, was able to attach to the FRC, resulting in an FR_w comparable to those of the copper coatings. However, the extent of the attachment of the calcareous fouling was characterized by large variations between replicates, as reflected by the error bars. Some individual panels thus performed better compared to the copper coatings. The large variation between replicate FRC panels may partly be explained by the fact that the fouling was loosely attached to the FRC panels and easily got detached if touched, e.g. when carefully removing large floating filamentous algae that sometimes got stuck on the panel frame used, despite gentle handling during sampling. Upon retrieving the panels in October 2021, it was also noted that the previously observed barnacles on the FRC panels were gone, indicating self-cleaning properties. Thus, for Station 2, the results show that the FRC had a similarly good or even, at certain time points, better performance than the copper coatings.

At Station 3, the calculated FR_w of the FRC was generally not significantly different compared to those of the copper coatings during the first 6 months of exposure (July–December) (Fig. 6c). At the following assessments (in January and March 2021) the FR_w of the FRC was however significantly lower in comparison. At this time, the incipient biofilm (FR10) observed on the coating previously was no longer visible on the photographs of the panels leading to a reduction of the average FR_w from 10 to ≤ 2. As discussed previously, this could have been the result of self-cleaning by the coating and/or related to uncertainties in the coverage assessment of fouling classified as FR10 due to light reflection caused by the coating surface.

In summary, with the exception of only two time points at Station 1, the FRC was found to performed equally well or significantly better than the studied copper coatings regardless of exposure site or time. Even though the copper coatings differ both in the amount of added cuprous oxide and the type of booster biocide (Table 1), they generally performed equally well, with statistical difference only observed at a few time points at Station 2 and 3 (Fig. 6).

3.1.4. *Implications for shipowners and boaters in the Baltic Sea region*

To be placed on the European market, biocide-containing antifouling paints must be approved by the competent authority in the individual member state. The approval can only be granted if the product both passes an environmental risk assessment and an efficacy assessment where the product is demonstrated to be efficient against its target organisms (European Parliament and Council, 2012). For the latter assessment, the European Chemicals Agency (ECHA) has provided guidelines for the efficacy testing of antifouling paints (ECHA, 2018). The guidelines are the same whether the paint is intended for recreational boats or ships. According to these, a static raft test with coated panels should be performed for a period of at least 6 months covering the period of peak fouling activity. The assessment of the efficacy is then based on the surface coverage of macrofouling, defined as large, distinct multicellular organisms visible to the human eye such as barnacles, tubeworms, or fronds of algae >5 mm. Translated to the NSTM fouling rate scale used in this study, this definition of macrofouling organisms roughly corresponds to FR $\geq$ 40. According to the guidelines, the result from a product is deemed acceptable if the macrofouling coverage is below 25 % (ECHA, 2018). All the tested coatings here, including the biocide-free silicone coating, were thus able to meet the efficacy requirement outlined by ECHA in a more than satisfactory way, even after a whole year's exposure.

Static testing of antifouling paints is typically considered a worst-case scenario as the stagnant conditions combined with the high fouling intensity of near-shore coastal waters act to strongly favor the settlement of fouling organisms (ECHA, 2018; Cassé and Swain, 2006). Indeed, for a commercial ship with little idle time, static tests such as those conducted in this study may therefore represent a worst-case scenario. For a recreational boat, on the other hand, the test may rather reflect the actual conditions of use. National surveys in Sweden

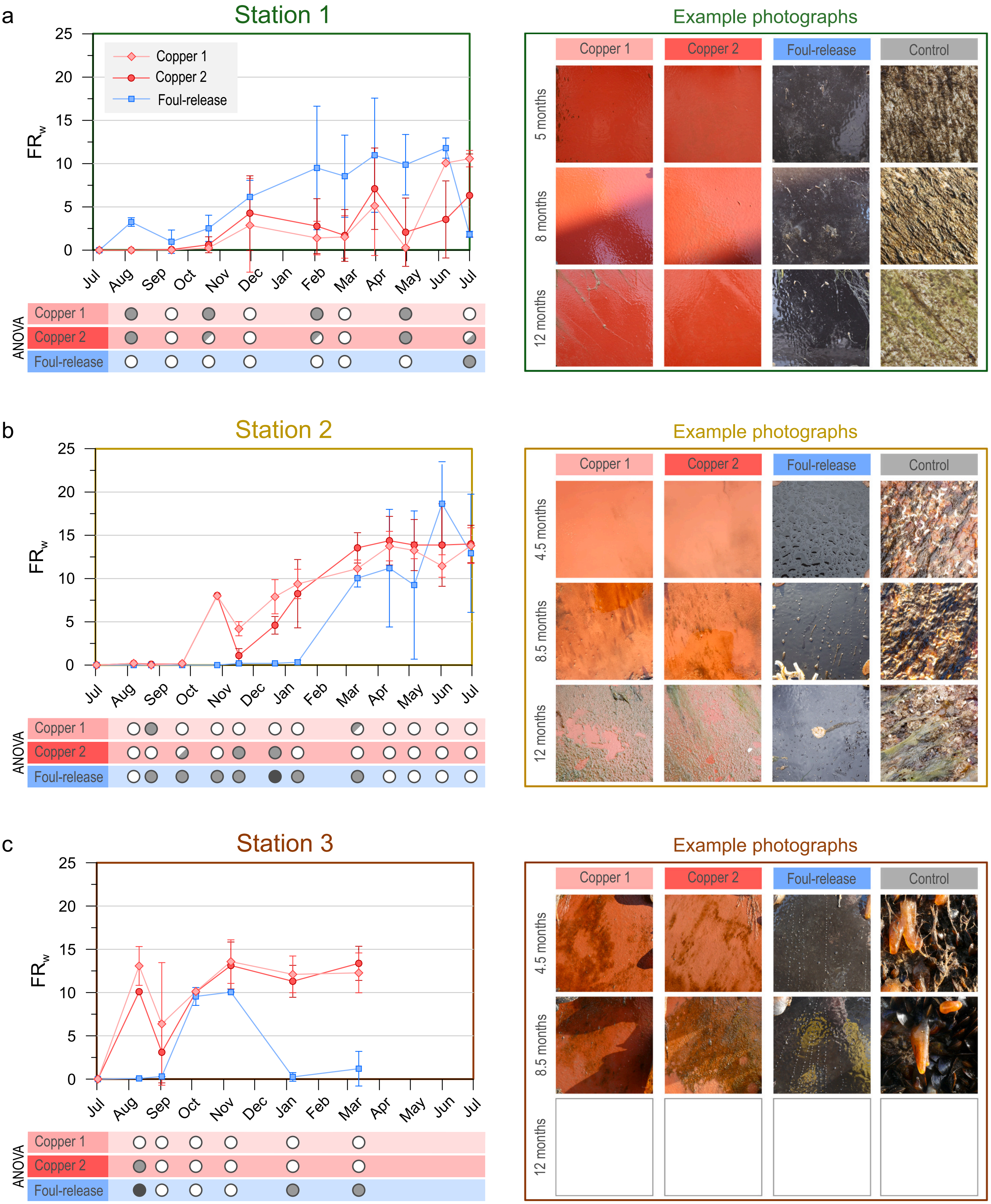

Fig. 6. Calculated weighted fouling rate (FR_w) for the copper and foul-release coatings at Station 1 (a), 2 (b) and 3 (c). Error bars show the standard deviation ($n = 4$ panels). The results from the ANOVAs with Tukey's HSD post-hoc testing are shown underneath each respective graph where dots not containing connecting colors correspond to FR_w-values found to be significantly different. Some example photographs of the coated panels and the control from three different time points are shown to the right of each graph. Note that fouling growing within 13 mm of panel edges was not considered in the evaluation (see Materials and Methods). The exposure at Station 3 was terminated ahead of time, after roughly 8 months (in March 2021).

have, for example, shown that leisure boats are idle during 85–90 % of the boating season (The Swedish Transport Agency, 2015; Swedish Transport Agency, 2021). The length of the boating season in the Baltic Sea is typically around 5 months, stretching from May to October. Boats are taken up in the fall and stored on land over the winter to avoid storm and sea ice damage. In this study, the coatings were not exposed for the full duration of the boating season of 2020 as panels were deployed in July, i.e. 2 months into the season. The control panels show nonetheless that the coatings were subjected to a substantial fouling pressure. In Fig. 6, photos of one of the replicate panels are shown after a time period of about 5 months, i.e. the approximate length of a boating season. At this time, the statistical tests showed no significant difference between the performance of the FRC and copper coatings at any of the locations. Even at the end of the exposure test after nearly twice the length of a boating season or more, i.e. after 8.5 (Station 3) or 12 months (Stations 1 and 2), the silicone FRC was found to be equally efficient or even superior to the copper-based paints. This result demonstrates that the tested biocide-free coating is indeed as suitable as traditional copper coatings for leisure boats.

Even though the amount of idle time can differ between ships depending on their activity level, the conditions of static panel tests generally represent a worst-case scenario for this type of vessel. However, as opposed to leisure boats which mostly tend to be idle in the same mooring location, ships may face more variable types of fouling organisms and pressures depending on their specific route. Nevertheless, even at the site with the highest fouling pressure (Station 3), the biocide-

Table 3

Design and toxicity end-points of ecotoxicological studies carried out with commercial silicone foul-release coatings. Studies are listed in chronological order. Test design details and key results from the studies are summarized in Table SI4 of the supplementary information.

Reference and type of study	Media	Tested silicone coatings	Negative control	Positive control	Coating condition	Exposed organism	Exposure time	Ecotoxicological endpoint
1. Truby et al. (2000) Exposure to immersed coating	Water of unspecified nature	RTV11 (a PDMS elastomer from GE Silicones) with and without added silicone oil (10 wt% PDMPDPS[a])	No	Yes An ablative copper coating	Fresh	Mysid shrimp (*Mysidopsis bahia*)	4 days	Lethality, LC50
						Silverside fish (*Menidia beryllina*)	4 days	Lethality, LC50
2. Karlsson and Eklund (2004) Exposure to leachate solution	Natural seawater (7 or 20 PSU)	Intersleek 700 (International paint)	No	Yes A copper coating with irgarol (Cruiser Superior, International paint)	Pre-immersed for 1 h	Macroalgae (*Ceramium tenuicorne*)	14 days	Growth inhibition, EC50
						Macroalgae (*Ceramium strictum*)	14 days	Growth inhibition, EC50
						Copepod (*Nitocra spinipes*)	14 days	Lethality, LC50
3. Watermann et al. (2005) Exposure to leachate solution or immersed coating	Natural seawater	10 anonymous commercial silicone foul-release coatings (top-coats, tie-coats and/or combined coating system)	Yes	No	Fresh	Marine bacteria (*Vibrio fischeri*)	30 min	Luminescence inhibition
					Pre-immersed for 6–8 weeks	Crustacean larvae (*Balanus amphitrite*)	5–7 days	Mortality rate
4. Feng et al. (2012) Exposure to immersed coating	Seawater (sea urchin) or embryo rearing medium (fish)	Intersleek 425 Intersleek 757 Intersleek 970 (all from International paint)	Yes	No	Fresh or pre-immersed for 1 month	Sea urchin (*Arbacia punctulata*)	48 h	Embryonic development (frequency of different developmental stages)
						Fish, Japanese medaka (*Oryzias latipes*)	72 h	Embryonic development (hatching success, larval mortality and swim bladder inflation)
5. Okamura et al. (2012) Exposure to leachate solution	Artificial seawater	1 anonymous commercial silicone foul-release coating (supplied by International Paint Japan K.K.)	No	Yes A copper coating with zinc pyrithione (International Paint Japan K.K.)	Pre-leached for different amounts of time (up to 45 days)	Marine bacteria (*Photobacterium leiognathi*)	1 h	Luminescence inhibition, EC50
						Marine diatom (*Skeletonema costatum*)	72 h	Growth rate inhibition, EC50
						Crustacean (*Artemia salina*)	48 h	Lethality, LC50
6. Piazza et al. (2018) Exposure to leachate solution	Natural seawater (37 PSU)	5 anonymous commercial silicone foul-release coatings	Yes	No	Pre-immersed for 24 h, 7 days, 14 days, 30 days, 2 months and 3 months	Crustacean larvae (*Amphibalanus Amphitrite*)	24 h	Acute toxicity (immobility percentage of larvae) and swimming speed alteration
7. Muller-Karanassos et al. (2021) Exposure to paint particles in sediment	Estuarine sediment	Particles from an anonymous commercial silicone foul-release coating	Yes	Yes	Pre-immersed for 5–10 days	Ragworm (*Hediste diversicolor*)	18 days	Feeding rate, weight change, burrowing, metallothionein-like protein
						Cockle (*Cerastoderma edule*)	18 days	Feeding rate, weight change, burrowing, metallothionein-like protein

[a] PDMPDPS = polydimethyldiphenylsilicone.

free silicone coating performed well. This is particularly interesting given that the static exposure represents a worst-case scenario for the FRC in particular as removal by the water shear force would have taken place on a navigating ship. Likely, the performance of this coating compared to the traditional copper ones would have been even more superior, had the test involved more dynamic conditions.

The Baltic Sea region stretches across a broad salinity gradient that affects the species richness and fouling intensity, with lower fouling pressure in the inner parts of the Baltic Sea and increasing towards to North Sea (Wrange et al., 2020). Location 3 is found in a high salinity region, with a generally high fouling pressure, also in comparison to other European waters (Canning-Clode, 2008). Hence, although this study was aimed at evaluating the suitability of biocide-free FRCs in the Baltic Sea, its results suggest that current commercial FRCs may also perform well in other seas.

3.2. Environmental impact of silicone foul-release coatings

3.2.1. Toxicity to aquatic organisms

Seven ecotoxicological studies (numbered from 1 to 7 in Table 3) carried out with commercial silicone FRCs were identified and reviewed (Truby et al., 2000; Karlsson and Eklund, 2004; Watermann et al., 2005; Feng et al., 2012; Okamura et al., 2012; Piazza et al., 2018; Muller-Karanassos et al., 2021). In these ecotoxicological assessments, test organisms were exposed either to the immersed coatings (studies 1, 3 and 4), to coating leachate solutions (studies 2, 3, 5 and 6) or to sediment amended with paint particles (study 7). However, even for the same type of exposure, the selection of organism, ecotoxicological endpoints, silicone coating types and coating pre-treatments (i.e. pre-immersion or not) differed. In particular, the choice of ratio between coating surface (in cm^2) and solution volume (in mL) in the studies using coating leachate solutions varied substantially from 1:0.44 down to 1:100. Test design details and key results from the studies are summarized in Table SI4 of the supplementary information.

Three of the studies (studies 1, 2 and 5) used a positive control (copper coating, typically with a booster biocide), which makes it possible to compare the ecotoxicological response between silicone FRCs and traditional copper-based coatings. The results show copper coatings to have a substantially higher toxicity compared to silicone FRCs towards all tested organisms which included bacteria, diatoms, macroalgae, copepods, shrimp and fish (Table SI4). For many organisms, the tested leachate concentrations from the silicone FRCs were not high enough to even enable the determination of an EC50 or LC50 value whereas effects from exposure to copper coatings were clear at quite low leachate concentrations (e.g. studies 1 and 2). Nonetheless, that does not necessarily mean that silicone FRCs are non-toxic to marine ecosystems. In the studies with a negative control (studies 3, 4 and 6), some ecotoxicological effects from exposure to silicone coatings or their leachates were observed in comparison to the control (no coating). In study 3, Watermann et al. (2005), found that, 3 out of 10 tested silicone top-coats were slightly toxic to marine bacteria (Watermann et al., 2005). Additionally, 100 % mortality of cypris larvae were observed during direct exposure to one of the tested coatings, as the larvae got stuck in silicone oil exuding from the coating, rendering them immobile. In study 4, Feng et al. (2012) compared the effects of fresh versus pre-immersed coatings on the embryonic development of both sea urchin and fish for two hydrophobic silicone coatings (Intersleek 425 and 757, where the latter is the top-coat of the Intersleek 700 system) and one fluoropolymer silicone coating (Intersleek 970, top-coat of the Intersleek 900 system) (Feng et al., 2012). When fresh coatings were used, all exposed sea urchin embryos failed to reach later developmental stages (pluteus or full pluteus) for 2 out of 3 coatings (Intersleek 425 and 970) as they stopped developing at the earliest, pre-prism stage. Similarly, a significantly negative effect for these same two coatings was seen on the hatching success of the Japanese medaka (*Oryzias latipes*) fish eggs. Additionally, inability of swim bladder inflation in all embryos was observed for all three coatings. These toxic effects were reduced when coatings were pre-immersed for 1 month under running seawater. Nonetheless, Intersleek 970 still displayed a clear negative impact on sea urchin embryonic development, and swim bladder inflation of the embryonic fish eggs was still completely lacking for both Intersleek 425 and 970 despite pre-immersion. Even though it is uncertain how representative the conditions of these static, low volume embryotoxicity tests (ratio of coating to volume of 1:1) are compared to environmental conditions, the study highlights that commercial FRCs may release active compounds, especially during their initial immersion. This was also confirmed in study 6, where Piazza et al. (2018) assessed the toxicity of 5 commercial coatings after different times of immersion (24 h–3 months), also using a low volume test (ratio of coating to volume of 1:0.44) (Piazza et al., 2018). Here, the ecotoxicological endpoints involved immobility and swimming speed alteration of barnacle cypris larvae. The authors found that negative effects were reduced with increased pre-immersion time, with no significant effects relative to the control for any of the coatings after 2 months. Prior to that, some differences between coatings were observed: while three of the coatings only exhibited acute (immobility) effects in the larvae for the shortest pre-immersion time (24 h), the other two still showed significant toxic effects after 2 weeks and 1 month of immersion, respectively.

Study 7, investigated the ecotoxicological impact of silicone paint particles on sediment-dwelling organisms (a worm and a cockle) (Muller-Karanassos et al., 2021). Here, both negative and positive controls were included, consisting of either unamended sediments (negative) or sediments with additions of historic (collected from abandoned boats) or modern biocidal paint particles (positive). The amounts of added paint particles were based on mean observed concentrations in the field and different sub-lethal effects were evaluated after 18 days (Table 3). The modern biocidal paint particles, which held higher concentrations of copper compared to the historic ones, were the most toxic. The test with modern biocidal paint particles resulted in adverse health effects in the ragworm after 18 days (reduced feeding rate and weight loss) and was acutely toxic to cockles with mortality of all replicates within the first 10 days of exposure. For the test with silicone paint particles, no significant differences were observed compared to the control except for weight change in cockles. This effect was however not substantial, and the authors generally conclude that the silicone paint particles did not cause adverse health effects on cockles or ragworms.

In most of the examined studies, silicone coating identities were anonymous, which is problematic as coating formulations will likely differ not only between manufacturers but also over time, as previously discussed (Fig. 1). Some general conclusions can nonetheless be drawn. In summary, even though a few studies point to possible adverse effects from silicone coatings on marine organisms, copper-containing coatings are considerably more toxic. Additionally, any potential release of active compounds from silicone coatings seem to principally occur within the first 2 months of immersion. This could perhaps explain why immobility of cypris larvae was not generally observed in study 3 where coatings were pre-immersed for 6–8 weeks, but readily observed in study 6. The ability of ecotoxicological studies to detect effects of any initially released active compounds will thus depend on whether any pre-immersion of the tested coating(s) was carried out. Additionally, the same toxic effects were not generally observed for all silicone coatings in studies where several products were tested in parallel (studies 4 and 6). Hence, the specific formulation of the coating (i.e. the identity of the specific product) is important. The results from one silicone coating are therefore not representative of all silicone coatings in general. This also suggests that additives, rather than the PDMS-elastomer present in all the coatings, are responsible for any observed effects. Finally, the exposure concentration matters. Intersleek 700, was tested in both studies 2 and 4 which had widely different approaches and, as a result, different outcomes. In study 2, the ratio of coating to volume was the lowest of any of the studies and 100 times lower compared to that of study 4. Hence, even though the leaching time was longer in study 2 (14 days) compared to study 4 (2 or 3 days), it is perhaps not surprising that the leachate concentration did not allow for a determination of EC50 or LC50 values for the endpoints of study 2. On the other hand, the environmental relevance of using high coating to volume ratios can be debated. In study 4, Feng et al. (2012) state that the aim was to model the specific effects on embryos near or on the coatings as they argue that embryos may easily enter the non-slip layer over the hull of a docked ship (Feng et al., 2012). While a high coating to volume ratio may thus be relevant for certain situations and organisms, Piazza et al. (2018) (study 6) argue that the results from their exposure scenario, which had the highest ratio (1:0.44), should not be used as evidence of the coatings' actual toxicity or potential environmental hazard, but merely to compare any potential release of active substances (Piazza et al., 2018). The results from the studies with concentrated leachate exposure nonetheless raise the issue of whether the fouling prevention mechanism of all commercial silicone coatings is indeed purely physical. More studies, where known commercial coating compositions are tested, are thus warranted. Additionally, the production of coating solution leachate should be standardized to enable direct comparisons between studies on different coatings.

3.2.2. *Leachables and their potential environmental hazard*

As discussed in the previous section, the toxicity of silicone FRCs in comparison to copper coatings appears to be very low. A few studies indicated nonetheless that some coatings may leach active substances during the first 1–2 months of immersion (Feng et al., 2012; Piazza et al., 2018). The chemical nature of these substances is not known and could be anything from catalysts, unreacted components that migrate to the surface of the polymer, solvents and/or low levels of toxic compounds in pigments and other additives (Feng et al., 2012). Substances that have been reported in the literature which may be contained and leached, intentionally or no, from commercial silicone coatings and may be of environmental concern have been summarized here.

The addition and leaching of hydrophobic silicone oils is widely mentioned in the literature (Lejars et al., 2012; Hu et al., 2020; Truby et al., 2000; Watermann et al., 2005). Such oils, which can be present in the range of 1–10 wt% of FRCs, are added with the aim to act as softeners but also to improve the antifouling performance. The oils migrate to form an oily film at the surface of the coating, thereby increasing its slippery nature and taking early stages of fouling with it when released to the sea (Lejars et al., 2012). The extent to which silicone oils may or may not be present in currently available coatings is not known as this information is not disclosed by manufacturers. A review by Nendza (2007), has expressed concern over the potential large-scale physical effects in the environments from this type of leachables. Nendza (2007) argues that the leaching of silicone oils, i.e. PDMS, could lead to the build-up of an oil film on sediments and, at high exposures, to the potential trapping and suffocation of marine organisms. Even though the scientific evidence for the latter appears to be somewhat weak, and the toxicity and bioaccumulation potential of pure PDMS is reportedly low (Stevens et al., 2001; Wang et al., 2021), relying on the leaching of persistent silicone-based fluids may not be the most environmentally sustainable way of providing antifouling protection. The recently researched degradable silicone foul release coatings which would rely on the erosion of the coating through hydrolysis, thereby intentionally releasing polysiloxanes into the environment is questionable for the very same reason, although this technology is not known to exist (yet) as a commercial product (Gevaux et al., 2019; Hu et al., 2021).

The addition of fluorinated substances constitutes another environmentally dubious approach to improve the performance of silicone FRCs. Even though the use of silicone-based materials together with fluorinated compounds has been argued to combine the best of “two worlds” in terms of antifouling performance, it has also been suggested that the use of *per*- and polyfluoroalkyl substances (PFAS) should generally be limited due to their high persistence and potential environmental hazard (Nurioglu et al., 2015; Glüge et al., 2020). Some researchers have thus called for the voluntary removal of fluorinated substances from silicone coatings, as fluorine-free replacements have been demonstrated to yield similarly good results (Vesco et al., 2019). When it comes to commercial silicone FRCs, Intersleek 1100SR (launched in 2013) has, for example, been reported to contain PFAS (Wang et al., 2020). Even though Intersleek 1100SR is still currently on the market, the latest launch by International paint (Intersleek 1000 in 2016) appears to be fluorine-free which could perhaps be a sign of the company's intention to phase out fluorinated substances. In the EU, several PFAS will be restricted over the coming years and a number of other PFAS are on the REACH Candidate List of substances of very high concern (ECHA, 2021). This may aid to limit the use of fluorinated compounds in FRCs in the future.

While the addition of hydrophobic silicone oils seems to have been implemented to improve the performance of PDMS-based coatings through their intentional release, 2nd and 3rd generation products (see introduction) may instead suffer from the unintentional release of amphiphilic oils. Even though these block copolymers are not bonded in the coating matrix, they are meant to remain “anchored” at the coating surface to provide the surface with the desired amphiphilic qualities able to deter fouling. The stability of these surface-active additives will depend on their chemistry, but a study on experimental coatings with concentrations of up to 7 wt% PDMS-PEG-based copolymers, also show coating formulation (e.g. the presence of biocides) and environmental parameters (e.g. seawater temperature) to impact their release rates (Camós Noguer et al., 2017). When loss or degradation of the added block copolymers occurs, a re-generation process can take place where copolymer molecules in the bulk of the coating diffuse to the surface to replace lost ones (Inutsuka et al., 2013). This process is however only effective so long as there is no shortage of copolymer molecules in the bulk of the coating. Hempel's hydrogel coatings (e.g. Silic One and its ship counterpart Hempasil X3+) utilize this technology. Hydrogel precursors are stated to be present in the PDMS matrix, able to regenerate the hydrogel surface of the coating in case of damage (Thorlaksen et al., 2010). A study involving the assessment of mass loss in silicone coatings when immersed in distilled water at room temperature, found that Hempasil X3 exhibited a loss of 8 wt% within roughly 8 months. This loss was hypothesized to be due to the leaching of non-bonded copolymers (Gevaux, 2019). It is not known whether the current version of the product, Hempasil X3+, may also exhibit such a mass loss. PPG's Sigmaglide 1290 is also claimed to have a dynamic surface regeneration which inhibits loss in performance throughout the product's lifetime (PPG, 2021). Oliveira and Granhag (2020) exposed panels coated with Sigmaglide 1290 and noted that “[…] *during the first three months from deployment a thin film of oil could be observed on the sea surface on retrieving the panels for monthly inspections*” (Oliveira and Granhag, 2020). These non-bonded oils could have been of either hydrophobic or amphiphilic nature (Gevaux, 2019). Given that paint formulations are considered proprietary information, the identities of additives in commercial products are unknown and, as a result, it is difficult to speculate about their potential toxicity. As for the environmental fate of amphiphilic additives, the anchoring part is typically PDMS-based, meaning that these substances are likely to be of a persistent nature. Efforts should therefore be made to avoid their release.

The occurrence and leaching of organotin compounds from silicone coatings has also been studied as the most common curing reaction to produce the PDMS elastomers in commercial silicone foul-release products involves the use of an organotin compound as a catalyst, most typically dibutyltin (DBT) or dioctyltin compounds (Lejars et al., 2012). Even though organotins have been banned in antifouling paints by the International Maritime Organization since 2008, small additions are permissible under the AFS convention (IMO, 2005). In the cases where DBT has been used, tributyltin (TBT) and monobutyltin (MBT) can also be present as impurities in the coating (Watermann et al., 2005). However, leaching tests of six commercial coatings demonstrated either undetectable or very low releases of MBT (≤ 0.0006 μg/cm^2/day), DBT (≤0.007 μg/cm^2/day) and TBT (≤0.0002 μg/cm^2/day) (Watermann et al., 1997). Even so, it has been suggested that it would be prudent to use other catalysts as such are available (Lejars et al., 2012; Watermann et al., 2005).

From a toxicity point of view, the intentional release of biocides likely constitutes the biggest threat to the marine environment when it comes to silicone FRCs. Biocidal silicone coatings have only been introduced to the market in recent years due to technical challenges (Fig. 1). The successful incorporation of active substances to improve the antifouling performance was made difficult by the fact that the silicone matrix can only support small additions of biocides without compromising the surface smoothness of the coating (Ciriminna et al., 2015). Added biocides may also become trapped in the highly crosslinked PDMS matrix and therefore not able to easily migrate to the surface (Thomas et al., 2004). To overcome these issues, the Hempaguard product series from Hempel, launched in 2013, contains microencapsulated biocides, enabling a controlled diffusion and release of the active substance, copper pyrithione (Ciriminna et al., 2015; Hempel, 2021). Even so, it should be noted that the amount of added biocides to Hempaguard is considerably less compared to conventional copper coatings (Radenovic et al., 2014). Chugoku's Bioclean Plus is also reported to contain copper pyrithione (Lloyd's Register, 2019). Although the nature of its incorporation in this coating is not known, the product is marketed to have a controlled linear release of active agents by the manufacturer (CMP, 2014). A more sustainable approach to gain the benefits of including an active substance in the formulation, while avoiding its release into the marine environment has been the target of some recent studies (Silva et al., 2019; Ferreira et al., 2020; Silva et al., 2021). Here, non-releasing biocidal coatings have been obtained through the grafting of a biocide (Econea) in the coating matrix of Hempasil X3+. The fixation of the biocidal agent was found to reduce the environmental impact, even during simulated wear scenarios (Silva et al., 2021). The utilization of this technique in commercial products would certainly aid to further mitigate any environmental risks associated with biocidal silicone FRCs.

3.3. *Barriers for the transition to biocide-free foul-release coatings*

Even though silicone FRCs seem to have similar or, in some cases, superior antifouling properties compared to traditional copper coatings, as demonstrated here and in a previous study in the Baltic Sea region (Oliveira and Granhag, 2020), and a substantially lower environmental impact, as discussed above, a majority of ship and leisure boat owners and operators still opt for the use of biocide-containing coatings on their vessel hulls (Ciriminna et al., 2015). Obstacles to their establishment on the market, as perceived by end-users and coating manufacturers, are discussed next in relation to both the recreational and professional markets.

Both the commercial and recreational markets are traditional when it comes to choice of antifouling system, their end-users tend to be skeptical towards non-biocidal solutions (Kim, 2021; Sandgren, 2021). For the recreational market, the biggest obstacle for the establishment of biocide-free silicone coatings, according to the coating manufacturer Hempel, is therefore related to regulation or, more precisely, the continued authorization of biocidal paints (Sandgren, 2021). Regulating the products placed on the market rather than expecting boaters to make informed and environmentally-friendly choices may thus be a more effective way towards sustainable leisure boating (Lepoša, 2017). Another perceived hurdle for this sector may lie in the initial application of the coating on the hull. As recreational boaters in the Baltic Sea region typically do not hire professionals but coat the hull themselves, the switch to a silicone coating system can be laborious. Previous coating layers with biocide-containing paints typically need to be removed, and a tie-coat applied before the application of the silicone top-coat. A tie-coat that can be applied directly on top of existing biocide-based coatings has however recently been placed on the market and could help ease the transition (Sandgren, 2021). The application instructions for e.g. Silic One are nonetheless stricter than for copper coatings, with regards to e.g. application conditions and drying times, to ensure proper adhesion of the coating (Noguer, 2021). Even so, although the initial application of the coating may involve more work, silicone coatings require less work than copper-based paints when the entire coating lifetime is considered. The guaranteed lifetime of Silic One, for example, is 2 years, with some customers reporting the coating to last up to 5 years (Sandgren, 2021; Watermann and Thomsen, 2018). This can be compared to the copper-based paints on the recreational market in the Baltic Sea which typically have service life of only 1 year. At the end of the recommended service life, the recommendation for Silic One (and Hempasil, the ship equivalent) is simply to clean the surface and reapply one new layer of top-coat (Noguer, 2021; Sandgren, 2021).

For the professional market, the barriers are somewhat different. According to Hempel, their biocide-free FRC (Hempasil X3+) is currently out-competed by their biocide-containing FRC (Hempaguard) (Noguer, 2021). This likely relates to the previously mentioned skepticism towards biocide-free solutions, as Hempel representatives state that shipowners are reported to want to be “on the safe side” and therefore opt for the silicone coating with biocides when given the option between the two (Noguer, 2021; Sandgren, 2021). Nonetheless, as the amount of biocide in Hempaguard represents a fraction of that present in conventional biocidal antifouling coatings, the gain for the marine environment is still considerable, even with this coating (Ciriminna et al., 2015). Hempaguard also has a better performance than Hempasil X3+, especially in warmer waters, but for shipowners planning to dry-dock every 2–3 years and therefore not requiring the 5 years of performance provided by Hempaguard, the biocide-free Hempasil X3+ constitutes a suitable and also less costly alternative (Noguer, 2021). Hempel representatives further state that demonstration of improved performance, i.e. reduced fuel consumption, rather than sustainability arguments tend to convince shipowners to make the transition to a silicone coating (Noguer, 2021; Sandgren, 2021). For shipowners, increased sustainability thus seems to be a bonus and not a primary motivator to switch coating system. A high initial investment cost has been reported as another barrier for these end-users (Kim, 2021). However, despite a higher cost of application, silicone FRCs yield a better return on investment compared to copper coatings when the whole lifetime of the coating is considered (Lejars et al., 2012). Not only is the performance typically better over time, resulting in fuel savings, but the service lifetime is also longer as silicone coatings do not rely on polishing or depletion mechanisms. This results in extended dry-docking intervals. For shipowners, a service lifetime of 3–5 years (depending on vessel activity and trade route) is recommended for Hempel's biocide-free FRC, but service lifetimes of as much as 7 years for the biocide-containing FRC has been reported by some (Noguer, 2021). In comparison, the typical service life of a conventional copper coating is 2–3 years (Lejars et al., 2012). Finally, even though vulnerability to damage is typically mentioned in the scientific literature as a drawback of FRCs, it is not perceived as a barrier for customers to change to a silicone coating (Noguer, 2021). Even so, research on self-healing silicone coatings is ongoing (Hu et al., 2020) and commercial alternatives based on more durable epoxy-polysiloxane, such as Jotun's Sealion Resilient, are already commercially available.

4. Conclusions

This study has demonstrated good performance of a biocide-free silicone FRC in the Baltic Sea region during static worst-case conditions, even at locations of intense fouling pressure. The efficacy of the tested FRC over the course of a full year was found to be equal or even superior in comparison to two copper coatings with booster biocides. These results show that silicone FRCs constitute both a viable and commercially available biocide-free option for both shipowners and leisure boat owners in this region.

Except for a few products, most silicone FRCs on the market are biocide-free. As such, they are subject to less legal scrutiny than biocidal coatings. However, this study can conclude that there is a large variability in the formulation of commercial silicone FRCs and that they may not be completely environmentally benign, simply for their lack of biocides. Some coatings have displayed toxicity to marine organisms, especially during the first months of immersion. The identity of the substance(s) responsible for the observed effects is however unknown. Several potential leachables from commercial silicone FRCs have also been identified here and include hydrophobic and amphiphilic silicone fluids, PFAS and organotin compounds. Although some substances, e.g. silicone fluids, may not constitute a threat to the environment from a toxicity point of view, many are highly persistent. Efforts to limit or avoid their release should therefore be made by producers. Ultimately, more transparent studies investigating the potential toxic effects of commercial products as well as the identity of leachables and their environmental fate are needed. Nonetheless, biocide-free silicone FRCs undeniably hold a substantially lower toxicity in comparison to biocidal copper coatings. Their use should therefore be promoted over that of conventional antifouling paints.

To encourage the transition to silicone FRCs going forward, different measures are required for the professional and recreational markets. While demonstration of cost saving seems to be the main argument to sway shipowners who are driven by profit, other approaches are required for the recreational market. With traditional end-users that tend to be skeptical towards biocide-free solutions, stronger regulation of biocidal coatings for leisure boats may be necessary. This could not only encourage end-users to switch coating system, but also incite more coating companies to launch silicone-based coatings for recreational vessels.

CRediT authorship contribution statement

Maria Lagerström: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization. **Anna-Lisa Wrange:** Resources, Investigation, Writing – original draft, Writing – review & editing. **Dinis Reis Oliveira:** Conceptualization, Methodology, Formal analysis, Investigation. **Lena Granhag:** Conceptualization, Methodology, Investigation, Resources, Writing – review & editing, Funding acquisition. **Ann I. Larsson:** Investigation, Writing – review & editing. **Erik Ytreberg:** Conceptualization, Methodology, Investigation, Resources, Writing – review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All data can be found in the Supplementary Information

Acknowledgements

This study was financed by the project HÅLL – Sustainable ship hull maintenance through development of decision support to the maritime industry and authorities (2019–2021), funded by the Swedish Transport Administration (Trafikverket) via Lighthouse Swedish Maritime Competence Centre under the “Hållbar sjöfart” program (Reference TRV 2019/27023, Grant number 5190054846). Erik Ytreberg and Lena Granhag were partly funded by BalticSea2020. Lena Granhag has also been supported by the project COMPLETE PLUS (Practical implementation of the COMPLETE project outputs and tools, #X023), co-financed by the European Union's funding Programme Interreg Baltic Sea Region in 2017–2020 (European Regional Development Fund) and Chalmers Area of Advance Transport. The authors would like to thank Albert Camós Noguer and Joakim Sandgren at Hempel for sharing their insights on market barriers for silicone foul-release coatings.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2022.114102.

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