



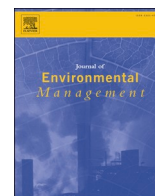
## **Environmental risk assessment of using antifouling paints on pleasure crafts in European Union waters**

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## Research article

## Environmental risk assessment of using antifouling paints on pleasure crafts in European Union waters

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## ABSTRACT

To ensure sustainable use of antifouling paints, the European Union have developed a new environmental risk assessment tool, which a product must pass prior to its placement on the market. In this new tool, environmental concentrations are predicted based on estimated release rates of biocides to the aquatic environment and risk characterization ratios are calculated in regional spreadsheets. There are currently two methods in use to predict release rates of biocides; a calculation method and a laboratory method. These methods have been believed to overestimate environmental release of biocides and therefore fixed correction factors to reduce the release rate can be applied. An alternative method, known as the XRF method, has recently been developed and used to derive field release rates from antifouling paints. The aim of this study was to review the new environmental risk assessment tool and assess how the choice of release rate method and application of correction factors impact the approval of antifouling paint products. Eight coatings were environmentally risk assessed for usage in four European marine regions; Baltic, Baltic Transition, Atlantic and Mediterranean; by applying release rates of copper and zinc determined with the different methods. The results showed none of the coatings to pass the environmental risk assessment in the Baltic, Baltic Transition and the Mediterranean if field release rates were used. In contrast, most of the coatings passed if the correction factors were applied on the release rates obtained with the calculation or laboratory method. The results demonstrate the importance of release rate method choice on the outcome of antifouling product approval in EU. To reduce the impact of antifouling paints on the marine environment it is recommended that no correction factors should be allowed in the environmental risk assessment or preferably that site-specific field release rates are used. If the regulation in the European Union (and elsewhere) continues to allow correction factors, the pressure of biocides to the environment from leisure boating will result in degradation of marine ecosystems.

## 1. Introduction

An unprotected surface area immersed in seawater will within minutes to days be fouled by different organisms (Bixler and Bhushan, 2012). This so-called biofouling can be of considerable concern, primarily for shipping and leisure boating, as it increases fuel consumption and maintenance costs as well as shortens dry-docking intervals (Davidson et al., 2020; Schultz et al., 2011). The most common strategy to prevent biofouling is to coat the hull with antifouling paint that contains and leaches biocides (Almeida et al., 2007; Amara et al., 2018). Historically, many unsustainable biocides have been used in antifouling paints including arsenic, lead, mercury and organotin compounds such as tributyltin (TBT) (Antizar-Ladislao, 2008; Yebra et al., 2004). These

biocides were all efficient in preventing biofouling, but they also impacted non-target organisms and created adverse effects on marine ecosystems and have therefore been phased out from the antifouling paint market (Miller et al., 2020; Thomas and Brooks, 2010).

Today, most antifouling paints are based on copper compounds, e.g. cuprous oxide (Amara et al., 2018). Copper is an essential element for all living organisms and play important roles in many metabolic processes (Ochoa-Herrera et al., 2011). However, copper may also be toxic to most species when concentrations exceed levels that are physiologically required (Strivens et al., 2020). Due to anthropogenic activities such as mining and smelting, municipal wastes, agricultural and industrial emissions the concentration of copper in the environment has increased (Morrone et al., 2019). In addition, emissions of copper from antifouling

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paints have shown to impact water quality negatively. For example, copper concentrations exceeding acute and chronic water quality have been found in recreational boat marinas located in the US (Schiff et al., 2004), Sweden (Kylin and Haglund, 2010), and Finland (Lagerström et al., 2020a). As a response, legislation has been passed in Washington State that will ban the use of certain copper based products in both freshwater and marine environments (Heine and Nestler, 2019). California is also considering similar regulations and local restrictions are in place in e.g. San Diego Bay (Miller et al., 2020).

In the European Union, a new harmonized product approval process has been developed under the Biocidal Product Regulation (BPR, Regulation (EU) 528/2012) requiring antifouling paints to pass an environmental risk assessment (ERA) prior to their placement on the EU market. In addition to the biocides, substances added into the paint formulation labelled as environmental Substances of Concern (SoCs), e. g. zinc oxide, shall be considered. In the ERA, environmental concentrations are modelled based on the estimated release rate of the biocides and SoCs from the paint surface to the water. The modelling is performed in a newly developed Excel calculation tool which automatically generates predicted environmental concentrations (PECs) of the biocides and SoCs in pleasure craft marinas within four European marine regions (Baltic, Baltic Transition, Atlantic and Mediterranean). The PEC values are subsequently divided with pre-defined predicted no effect concentrations (PNECs) to produce risk characterization ratios (RCRs) of the individual biocides/SoCs as well as cumulative RCRs if more than one biocide/SOC is included in the product. If the RCR is less than unity ( $<1$ ), the concentration in the environment is likely to be lower than the critical threshold value; the risk of adverse effects is considered low. If the ratio is higher than unity ( $>1$ ), risk for adverse effects exists and actions to reduce the risk are recommended. This includes higher tier refinements where correction factors (CFs) can be applied to the biocidal release rate (ECHA, 2017). For product approval, an efficacy assessment of the product is also required where the applicant must demonstrate that the product meet certain efficacy criteria in preventing biofouling. This antifouling paint product approval scheme under the EU BPR is shown in Fig. 1.

Since the outcome of the ERA depends on the release rates of the

biocides used in the paint formulation, it is important that the submitted release rates accurately predict environmental release in the relevant region of intended use. Currently, two standardized release rate methods are recommended for risk assessments in EU. The first is a laboratory “rotating cylinder” method where release rates are determined in artificial seawater (ASTM, 2005) and the second one is a calculation method (ISO, 2010). Besides these methods, a new method allowing for determination of field release rates using an X-Ray Fluorescence spectrometer (XRF) has been developed by Ytreberg et al. (2017) and further modified by Lagerström et al. (2018) and Lagerström and Ytreberg (2020).

The main aim of the current study was to review the new ERA calculation tools for antifouling paints developed under the EU BPR and assess how different release rate methods (the rotating cylinder method, the calculation method and the XRF method) impact the authorization of antifouling products in the EU. A second aim was to investigate differences in environmental sensitivity in terms of acceptable biocidal input between the different marine regions and how that will affect the product approval and impact the antifouling paint market. Finally, the highest acceptable release rates of copper and zinc from antifouling paints were determined in the different marine regions. Eight commercial coatings were used in the current study. The PECs of copper and zinc in different pleasure craft marinas located in the marine regions Baltic, Baltic Transition, Atlantic and Mediterranean were modelled using release rates obtained from the three release rate methods. Risk characterization ratios were subsequently calculated to determine if usage of the coatings would pose an unacceptable risk for the marine environment depending on the choice of release rate method.

## 2. Materials and methods

### 2.1. Study area and modelling tool

MAMPEC (Marine Antifouling Model to Predict Environmental Concentrations) is a widely accepted model used in regulatory purposes worldwide to predict environmental concentrations of biocides in the marine environment. The model is used by several EU member states to

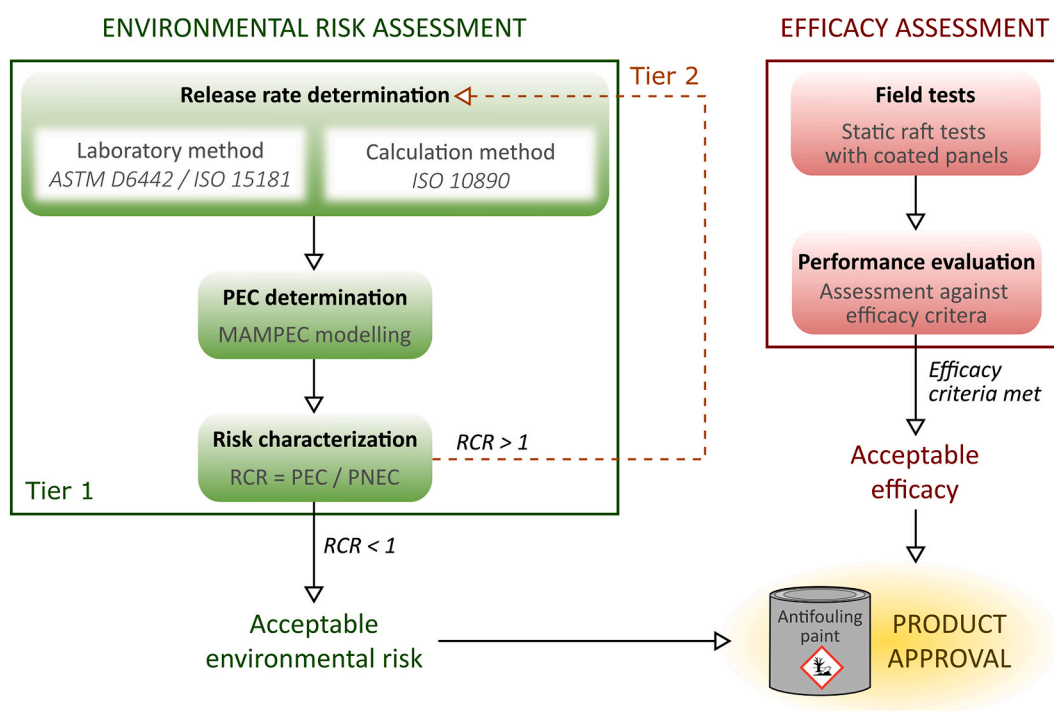
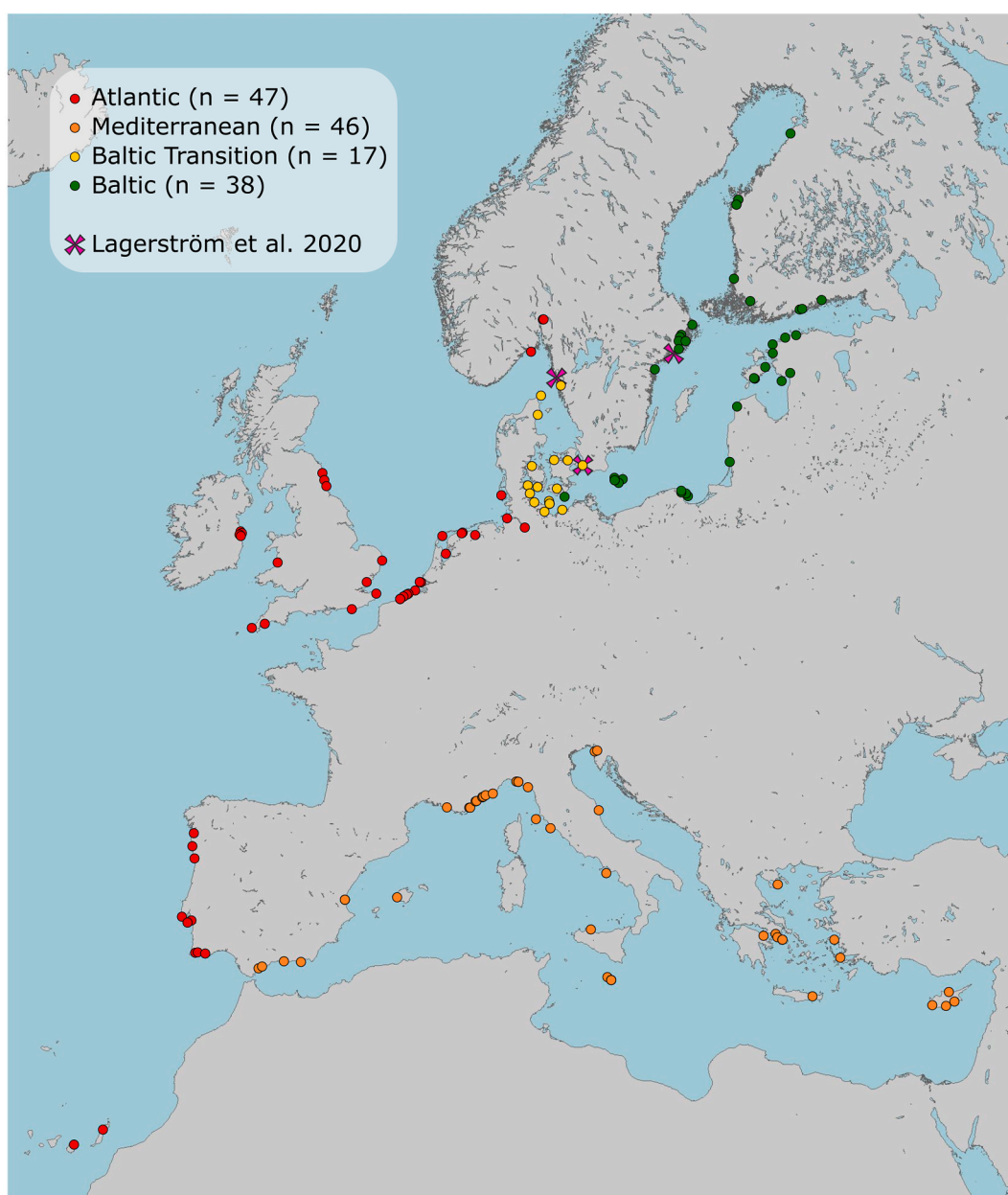


Fig. 1. Product approval process for antifouling paints under the European Union Biocidal Products Regulation (BPR, Regulation (EU) 528/2012).

determine PEC-values of biocides in different environments such as harbors, marinas and ship lanes. In MAMPEC, several scenarios exist, including two OECD scenarios representing a typical European commercial harbor and a pleasure craft marina, respectively. However, as these OECD scenarios do not reflect environments with low tidal differences, e.g. the Baltic Sea, several countries have developed their own scenarios to be used for the ERA of antifouling products, while other countries have approved all products irrespective of the biocidal release rate, as long as the biocides used are approved by the EU. Therefore, the antifouling paint market differs substantially between EU countries. In order to harmonize the regulation and to capture the wide range of conditions across EU waters, the Environment Working Group of the Biocidal Product Committee developed an agreed set of pleasure craft scenarios (ECHA, 2017) representative of the marine regions of the Atlantic (47 marinas), Mediterranean (46 marinas), Baltic Transition (17 marinas) and Baltic Sea (38 marinas) (Fig. 2). The purpose of this harmonization is to allow for Mutual Recognition under the BPR as the

ERA is performed by region, rather than by country. For the product approval applications, the applicant fills in the release rates of the different substances into newly developed Excel calculation tools which automatically generate PECs and RCRs for all individual marinas within a region. The Excel workbooks, one for each approved substance, can be accessed via the PT21 Emission Scenario Document pages on the ECHA website (ECHA, 2020). The PECs and RCRs are produced for the dissolved and particulate fractions of the substance, both inside the marina and its surrounding environment. For the risk characterization, the dissolved concentration inside the marina is proposed to be used. The 90th percentile concentration, based on the PECs determined for each individual pleasure craft marina, is used to calculate an overall RCR per region. Thereby, the applicant directly receives information about the risk characterization of the product in the different regions. If more than one biocide or SoCs is used in the product, a PEC/PNEC summation approach shall be used in a separately provided Excel calculation tool to determine the cumulative RCR (ECHA, 2017).



**Fig. 2.** Distribution of pleasure craft marinas, per marine region, included in the Excel calculation tool. The three exposure sites (Nynäshamn, Baltic Sea; Malmö, the Sound and Kristineberg, Kattegat) used by Lagerström et al. (2020b) to measure field release rates are also shown.

**Table 1**  
Tier 1 and Tier 2 release rates for Cu and Zn, as submitted to the Swedish Chemicals Agency for the risk assessment of the products, and site specifically field release rates using the XRF method obtained from Lagerström et al. (2020b).

Paint	Area of use	Product	Manufacturer	Cu <sub>2</sub> O (wt %, ww)	ZnO (wt %, ww)	Baltic Sea						Baltic Transition						Field, Kattegat					
						Tier 1		Tier 2		Field, Baltic Sea		Tier 1		Tier 2		Field, Oresund		Field, Kattegat					
						Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn
A	Baltic Sea and Baltic Transition	Lefant Nautica Copper <sup>(c)</sup>	Lefant	7.0	20 – 100	3.0 <sup>a</sup>	-	1.03 <sup>a</sup>	-	1.9	8.2	3.0 <sup>a</sup>	-	1.03 <sup>a</sup>	-	1.9	7.7	5.4	16.5				
B	Baltic Sea and Baltic Transition	Millie Light Copper <sup>(c)</sup>	International	6.1	10 – 25	3.0 <sup>a</sup>	4.0	0.97 <sup>a</sup>	1.32 <sup>a</sup>	2.2	5.8	3.0 <sup>a</sup>	4.0 <sup>a</sup>	0.97 <sup>a</sup>	1.32 <sup>a</sup>	3.9	5.4	7.1	6.3				
C	Baltic Sea and Baltic Transition	Cruiser One <sup>(c)</sup>	Jotun	8.5	2.5 – 25	2.5 <sup>a</sup>	5.8 <sup>a</sup>	0.9 <sup>a</sup>	2.0 <sup>a</sup>	4.4	6.4	2.5 <sup>a</sup>	5.8 <sup>a</sup>	0.9 <sup>a</sup>	2.0 <sup>a</sup>	3.6	5.9	4.4	10.1				
D	Baltic Transition	VC17m <sup>(c)</sup>	Hempel	17.96 <sup>c</sup>	-	N/A	N/A	N/A	N/A	8.0	0	15.11 <sup>a</sup>	-	5.21 <sup>a</sup>	-	7.9	0	13.3	0				
E	Baltic Transition	Racing VK <sup>(c)</sup>	Hempel	22.02	10 – 25	N/A	N/A	N/A	N/A	7.0	9.4	13.0 <sup>a</sup>	-	4.48 <sup>a</sup>	-	7.2	6.7	18.9	10.3				
F	Baltic Transition	Hard Racing Xtra <sup>(R)</sup>	International	33.1	10 – 25	N/A	N/A	N/A	N/A	8.1	6.2	32.0 <sup>b</sup>	-	6.2 <sup>b</sup>	-	6.9	4.4	16.7	4.2				
G	Baltic Transition	Biltema Antifouling <sup>(R)</sup>	Biltema	13	20 – 25	N/A	N/A	N/A	N/A	5.0	7.7	12.8 <sup>b</sup>	-	2.37 <sup>b</sup>	-	6.3	5.5	12.1	9.7				
H	Baltic Transition	Micron Superior <sup>(c)</sup>	International	31.93	2.5 – 25	N/A	N/A	N/A	N/A	7.8	2.7	12.44 <sup>a</sup>	3.66 <sup>a</sup>	4.29 <sup>a</sup>	1.26 <sup>a</sup>	12.0	2.9	27.5	4.1				

N/A denotes release rates not available in the product approval since the products were not risk assessed for usage in the Baltic Sea

- denotes Zn not to be included in the product approval and therefore release rates were not available

<sup>a</sup> denotes release rates calculated with the CEPE mass-balance method.

<sup>b</sup> denotes release rates obtained using the Rotating Cylinder method.

<sup>c</sup> denotes Cu powder

## 2.2. Methods for release rate estimation

Two methods are currently approved to be used for release rate determination in the EU, a mass-balance calculation method developed by CEPE (EU, 2006), from now on referred to as the “calculation method” and a rotating cylinder laboratory method, from now on referred to as the “rotating cylinder method”. In addition, two *in situ* release rate methods exists: the “Dome method” developed by the US Navy (Finnie, 2006; Valkirs et al., 2003), and the “XRF method” first developed by Ytreberg et al. (2017) and later modified by Lagerström et al. (2018) and Lagerström and Ytreberg (2020). A detailed description of the different release rate methods is shown in Supporting Material.

If the coating fails to pass the initial Tier 1 assessment, correction factors (CFs) of 2.9 (calculation method) and 5.4 (rotating cylinder method) can be used through simple division (EU, 2006). These corrected release rates can be used for a further Tier 2 assessment. The CFs were originally proposed by Finnie (2006) as the calculation and rotating cylinder method have shown to overestimate environmental release rates of copper determined in field. However, the proposed CFs are based on field data from one coating only and the applicability of a universal CF to predict environmental release rates of biocides for any type of coating has been questioned (Lagerström et al., 2018). In addition, neither the rotating cylinder nor the calculation method and the CFs consider changes in environmental parameters known to govern leaching of biocides such as salinity, pH and temperature (Lagerström et al., 2020a, 2020b; Ytreberg et al., 2017).

## 2.3. Antifouling coatings investigated and release rates of copper and zinc

The release rates of copper and zinc from eight antifouling coatings were used in the study. The coatings are authorized to be used on pleasure crafts in Sweden and were selected based on their Cu<sub>2</sub>O concentrations to achieve a wide range (6.1–31.9% Cu<sub>2</sub>O) (Table 1). In Sweden, regional restrictions exist for antifouling paints where stricter regulation is applied for coatings used in the Baltic Sea as compared to the Swedish west coast. Consequently, different antifouling paints exist for the Baltic Sea market as compared to the Swedish west coast. The release rates of copper and zinc, determined using the calculation method or the rotating cylinder method, were obtained from the product authorization reports submitted to the Swedish Chemicals Agency (Table 1). Five of the coatings were authorized to be used on the Swedish West coast only, i.e. in the Baltic transition region, while the other three were authorized to be used both in the Baltic Sea and on the Swedish West coast. Therefore, release rates are available for all eight coatings in the Baltic transition. As five of the coatings were environmentally risk assessed for exposure in the Baltic transition only, release rates for exposure in the Baltic Sea were not submitted during the product authorization.

Site-specific field release rates from the eight coatings determined with the XRF method were obtained from Lagerström et al. (2020b) and used for assessment against the release rates submitted in the product authorization reports (Table 1). These average daily *in situ* release rates between day 14 and 56 of exposure were determined during the summer season of 2018 at three sites along the Swedish coast: Nynäshamn, (Baltic Sea, salinity 6.4 PSU), Malmö (the Sound, Baltic transition, salinity 7.5 PSU) and Kristineberg Marine Research and Innovation Centre (Kattegat, Baltic transition, salinity 27 PSU) (Table 1). The exposure sites are shown in Fig. 2.

## 2.4. Exposure assessment and risk characterization

The release rates used in the application for product approval (Tier 1 and 2) as well as the site-specific field release rates determined with the XRF method were used in the new Excel calculation tool to environmentally risk assess the different coatings for use on pleasure crafts in the four marine regions. Noteworthy, zinc was not included in the



application for product approval for paints A, D, E, F and G. However, zinc was shown to be released from four of these coatings (A, E, F and G) when measured with XRF (Table 1). This will impact the outcome of the risk assessment, as the exposure assessment using the field derived release rates considers both Cu and Zn for all coatings (except paint D which did not contain zinc) while that using Tier 1 and Tier 2 release rates from product approval for paints A, D, E, F and G is based on copper only.

The release rates obtained with the rotating cylinder (Product F and G) are applicable to all marine waters and would hence be used in the ERA for all four marine regions. However, the release rates obtained with the calculation method (Product A–E and H) are derived based on the recommended paint film thickness and lifetime of the coating (see equation 1 and 2) and reflects emissions during usage recommendations for the regions Baltic Sea and Baltic Transition (Table 1). It is unknown if other dry paint film thicknesses would have been recommended for the Atlantic and Mediterranean market which would have increased or decreased the release rates and impacted the result of the ERA. Therefore, the assessment in the Atlantic and Mediterranean was performed to explore if the coatings theoretically would pass the ERA or not.

For the Baltic Sea region, field determined release rates obtained in Nynäshamn were used. For the Baltic transition region, release rates from both the Sound and Kattegat were used. For the Atlantic and Mediterranean, no site-specific field release rates were available. However, as the salinity at the Kattegat exposure site (27 PSU) is close to what is expected in most Atlantic and Mediterranean marinas (35 PSU), the field release rates obtained at Kattegat were also used for the ERA of the Atlantic and Mediterranean regions.

The application factors (the fraction of pleasure crafts assumed to use the product), PNEC-values and background concentrations are already fixed in the Excel calculation tool (Table 2) and the release rates of biocides are the only parameters required for the tool to derive PECs and RCRs. The 90th percentile concentration, based on the PECs determined for each individual pleasure craft marina, is used to calculate PEC/PNEC-ratios per region. When the coatings contain more than 1 biocide and/or SoC a PEC/PNEC summation approach was used to calculate the cumulative risk of the product ( $\Sigma$ RCR).

### 3. Results and discussion

#### 3.1. ERA in the marine regions

The three coatings approved for use on pleasure crafts in the Swedish Baltic Sea (Paint A, B and C) all failed the risk assessment when Tier 1 release rates of the standardized methods were used in the Baltic Sea region (Fig. 3). Tier 2 release rates showed  $\Sigma$ RCR < 1 for Paint A and Paint B, while Paint C had a  $\Sigma$ RCR of 1.05. Although not intended for use in the Baltic Sea, the remaining five coatings also fail the ERA using Tier 1 release rates. Only one coating (Paint G,  $\Sigma$ RCR = 0.99) would pass in a Tier 2 assessment, but as zinc was not included in the product application of this paint (as well as those of paints A, D, E and F) its exposure assessment is lacking given that the XRF method showed zinc to indeed be released from all coatings except paint D (Table 1). Only one coating (paint B) out of the three coatings passing the Tier 2 assessment (paints

A, B and G) included zinc in the product approval. When Baltic Sea field release rates were used, all eight coatings showed  $\Sigma$ RCRs above 2, thus failing the ERA. Notably, the PEC/PNEC ratios of zinc alone was > 1 for all paints except paint H (0.56) and paint D which did not contain any zinc (Supporting Material Table S1), highlighting the importance of including SoCs in the ERA of antifouling products.

In the Baltic Transition, only one coating (Paint A) had  $\Sigma$ RCR  $\leq$  1 when Tier 1 release rates were used whereas the Tier 2 assessment showed half the coatings (paints A, B, C and G) to obtain  $\Sigma$ RCRs  $\leq$  1 (Fig. 3 and Supporting Material Table S2). The remaining four coatings displayed  $\Sigma$ RCRs between 1.2 and 1.5. All coatings posed an unacceptable risk to the marine environment when field release rates obtained in the Sound were used ( $\Sigma$ RCR = 1.8–2.9). The  $\Sigma$ RCRs were even higher when the Kattegat-derived release rates were used ( $\Sigma$ RCR = 2.5–5.6). The higher  $\Sigma$ RCRs for Kattegat are a result of the higher release rates obtained in the more saline Kattegat (27 PSU) as compared to the Sound (7.5 PSU).

All coatings except coating F showed  $\Sigma$ RCRs < 1 when Tier 1 release rates were used in the ERA for the Atlantic. When Tier 2 release rates were used, all coatings showed  $\Sigma$ RCRs well below 1 (around 0.5) (Fig. 3 and Supporting Material Table S3). All coatings except paints E and H would pass the ERA if field derived release rates were used. In the Mediterranean, only the paints designed for the Swedish Baltic Sea (Paint A, B and C) pass the assessment using Tier 1 release rates, while all eight coatings showed  $\Sigma$ RCR < 1 when Tier 2 release rates were used (Fig. 3 and Supporting Material Table S4). The results showed no coatings to pass the ERA in the Mediterranean when field derived release rates were used.

#### 3.2. Acceptable environmental risk

The results from the ERA show all paints to pose an unacceptable risk to the environment in the Baltic, Baltic Transition and the Mediterranean when region-specific field release rates were used. As such, the antifouling paints assessed here would need to reduce the release rates of copper and/or zinc in order to be approved for these regions. The maximum allowable release rate combinations of copper and zinc to pass the ERA, defined as  $\Sigma$ RCR = 1, in the four marine regions were derived from the Excel calculation tool and are shown in Fig. 4. The differences in the extent of acceptable releases between marine regions, i.e. the positioning of the lines representing  $\Sigma$ RCR = 1, reflect differences in environmental sensitivity in terms of biocidal input. For example, acceptable risks for the Baltic marinas can only be obtained if the release rate of copper is  $\leq$  2.4  $\mu\text{g}/\text{cm}^2/\text{d}$  (assuming no zinc to be present in a coating). The corresponding maximum allowable release rates of copper in the Baltic Transition, Mediterranean and the Atlantic are; 3.4  $\mu\text{g}/\text{cm}^2/\text{d}$ , 9.0  $\mu\text{g}/\text{cm}^2/\text{d}$  and 21.4  $\mu\text{g}/\text{cm}^2/\text{d}$ , respectively. The differences can be explained by differences in water exchange capacity where the average tidal difference for the pleasure craft set used for the Baltic Sea is 0.2 m as compared to the higher average tidal differences in Mediterranean (0.53 m) and the Atlantic (2.97 m) (Shan-I et al., 2013).

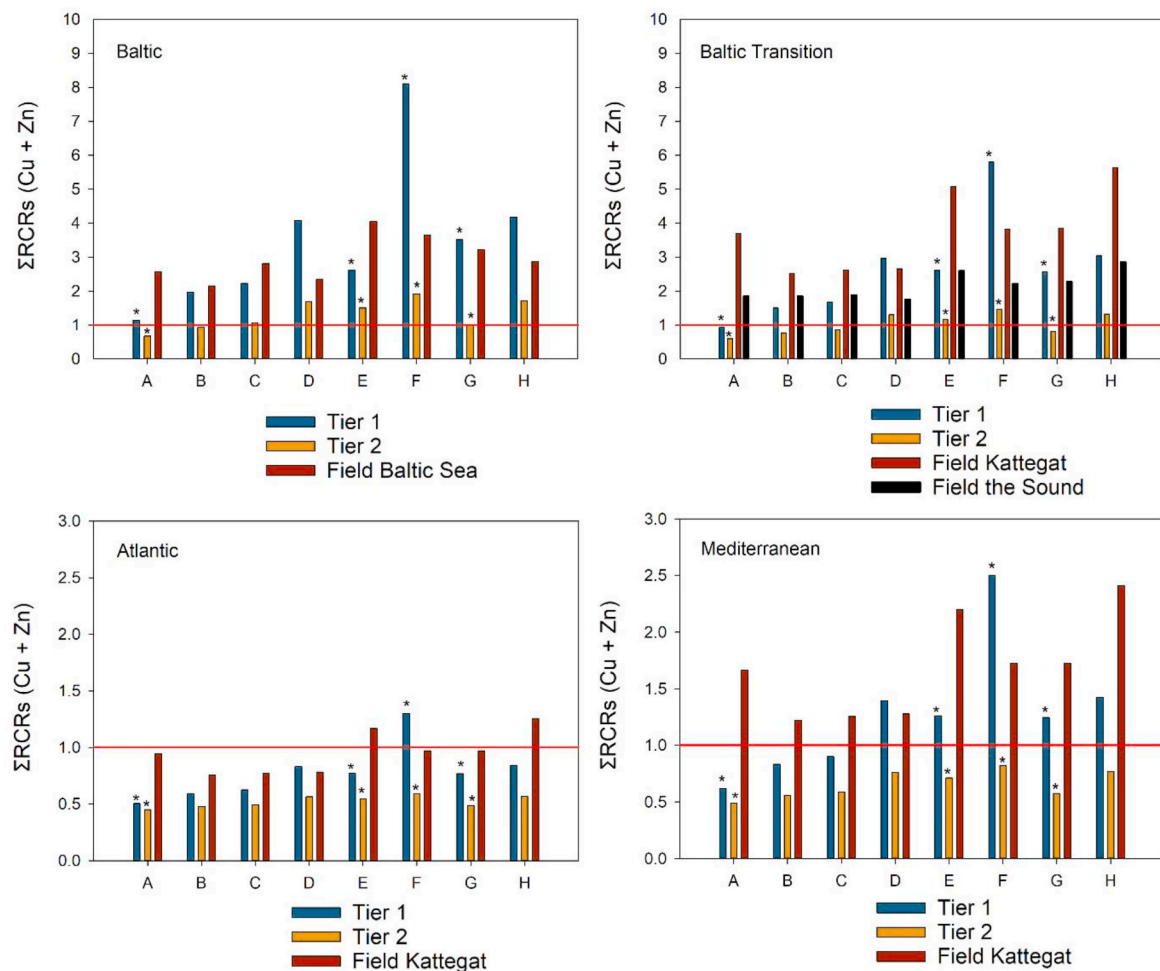
#### 3.3. Implications for the antifouling paint market and its regulation

Antifouling paints are considered effective if static field tests show a surface coverage of macrofouling < 25% (European Chemical Agency, 2018). However, as macrofouling on ship and pleasure boat hulls translates to severe economic costs due to fuel penalties (Schultz et al., 2011), the critical release rate of copper ( $\text{RR}_{\text{crit}}$ ) to prevent the attachment of macrofouling (defined as 0% surface coverage of macrofouling) is a more accurate efficacy parameter for the boating and the shipping sector. The antifouling efficacy and  $\text{RR}_{\text{crit}}$  of copper has been investigated by Lindgren et al. (2018) where a generic biocide and zinc free coating was spiked with cuprous oxide. The release rates of copper were determined with the XRF method during exposure time d14–d56. The  $\text{RR}_{\text{crit}}$  of copper was determined based on a number of experimental

**Table 2**

Predicted no-effect concentrations (PNEC), background concentrations and application factor used for the different regions in the Excel calculation tool.

Region	PNEC ( $\mu\text{g}/\text{L}$ )		Background concentrations		Application factor	
	Cu	Zn	Cu	Zn	Cu	Zn
Atlantic	2.6	3.4	1.1	0	0.95	0.9
Mediterranean	2.6	3.4	1.1	0	0.95	0.9
Baltic Transition	2.6	3.4	1.1	0	0.95	0.9
Baltic	2.6	3.4	1.1	0	0.95	0.9



**Fig. 3.** Sum of Risk Characterization Ratios ( $\Sigma$ RCRs) of copper (Cu) and zinc (Zn) for paint A – H depending on release rate method (Tier 1 and Tier 2 release rate submitted during product approval or region-specific field release rates determined with the XRF method) and in what marine region (Baltic, Baltic Transition, Atlantic and Mediterranean) the paints shall be used. For Paint D, that did not contain any Zn, the RCR is based on Cu release rate only for all release rate methods. The red line shows  $\Sigma$ RCR = 1. \* denotes RCRs are based on Cu release rates only as release rates of Zn was not included in ERA submitted for product approval. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

coatings containing increasing cuprous oxide concentrations that were exposed in a pleasure craft marina in Gothenburg, i.e. in the region Baltic Transition (Lindgren et al., 2018). All investigated copper coatings were efficient in preventing macrofouling and the  $RR_{crit}$  of copper was determined to be  $\leq 4.7 \mu\text{g}/\text{cm}^2/\text{d}$ , indicating that coatings with even lower release rates of copper may be sufficient in preventing macrofouling. Hence, low-leaching efficient coatings could pass the ERA in the Baltic Transition where the maximum allowable release rate is  $3.4 \mu\text{g}/\text{cm}^2/\text{d}$  (Fig. 4).

$RR_{crit}$  of copper in the Baltic Sea and Baltic Transition can also be estimated from the studies by Lagerström et al. (2020b) and Lagerström et al. (2018) where commercial coatings were used. The  $RR_{crit}$  of copper from these studies, determined using the XRF method, holds a higher uncertainty since all the coatings contained and released zinc which also may impact the coatings antifouling performance. Fig. 5 gives a geographical overview of the  $RR_{crit}$  estimates and the acceptable release rates in the two regions. The  $RR_{crit}$  of copper in the Baltic is lower than the acceptable release rate to pass the ERA indicating that antifouling coatings with a substantial lower release rate of copper, than what is currently available on the market, could be developed without compromising the requirements of antifouling efficacy. For the Baltic Transition, the situation is more complex where the  $RR_{crit}$  is lower than the acceptable release rate in the Sound, while the northern parts of the region show  $RR_{crit}$  to be higher than the acceptable release rate. Hence,

in order to pass the ERA in the Baltic Transition, coatings must have a lower release rate of copper than expected to be required to deter macrofouling in the northern part of the region.

The field determined release rates from the eight coatings studied here are not only typically above the maximum allowable but also sometimes greatly in excess of  $RR_{crit}$ . As shown in Lagerström et al. (2020b), copper emission could be reduced by as much as 80% for some paints without any loss in efficiency. The large discrepancy between the copper release rate of products currently on the market and release rates needed to deter fouling. This is likely because release rates derived using inappropriate methods and correction factors have, thus far, been admissible in the ERA. As demonstrated here, application of correction factors in the Tier 2 assessment nearly always results in a product passing the ERA. No longer permitting the use of correction factors would incentivize the development of lower-leaching, more sustainable coatings as well as promote the development of biocide free alternatives such as silicone coatings which currently only hold a small percentage of the market.

To ensure sustainable management of our coastal environments it is crucial that we understand the links between human activities, the pressure they pose on the environment and how that pressure may impact the environment and human welfare (Elliott et al., 2017; Elliott and O'Higgins, 2020). Thus, it is important that environmental release rates of biocides from antifouling paints are used to predict impacts on

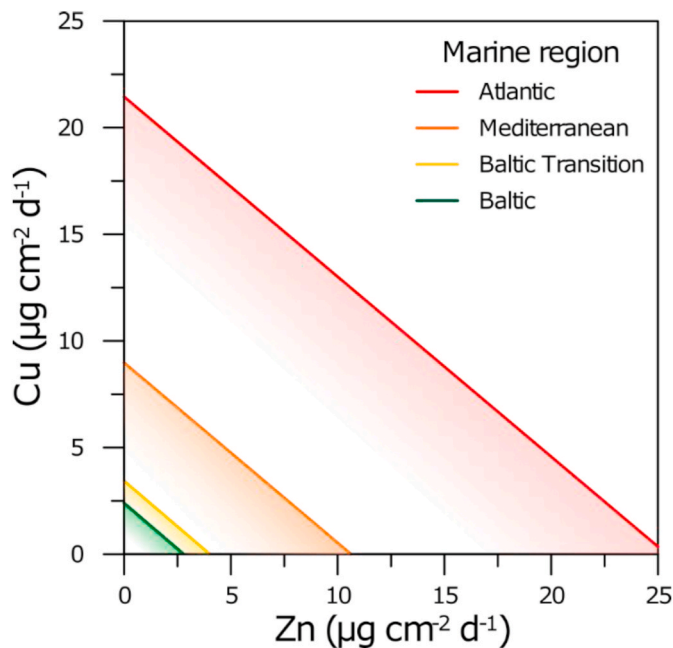


Fig. 4. Maximum allowable release rates of copper (Cu) and zinc (Zn) to obtain a Sum of Risk Characterization Ratios (ΣRCRs) equal 1 in the different marine regions. Areas above line denotes unacceptable risks, i.e. ΣRCRs > 1 while areas below the line denotes acceptable risks, i.e. ΣRCRs < 1.

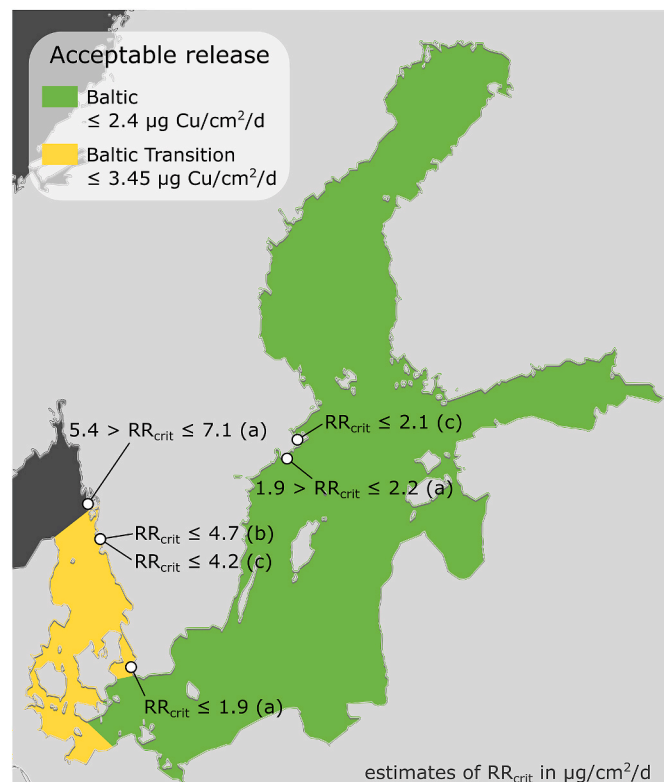


Fig. 5. Maximum acceptable release rates of copper in the Baltic Sea (green) and the Baltic Transition (yellow) for a product to pass the environmental risk assessment (ΣRCR ≤ 1) and critical Cu release rates (RRcrit) at five locations along the Swedish coast obtained from Lagerström et al. (2020b) (a), Lindgren et al. (2018) (b) and (Lagerström et al., 2018) (c). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

the environment. In a study by Lagerström et al. (2020a) a comparison between modelled (using MAMPEC) and measured environmental concentrations of copper inside a marine in Sweden was performed. The study used release rates submitted for product approval (determined with the calculation method and Tier 2) and site-specific field release rates determined with the XRF method. The study showed good agreement between measured concentrations and the modelled concentration when the field derived release rates were used. However, when the release rates from the products' authorization applications were used, the modelled concentration were 2-fold lower than the measured concentrations. This suggests the XRF method to accurately predict environmental concentrations while the calculation method (using Tier 2) underestimated the load of copper. Hence, if the regulation in EU (and elsewhere) continue to allow Tier 2 release rates, the pressure of copper to the environment from leisure boating will result in degradation of marine ecosystems. Reduction of copper emissions is also required to improve the environmental status of our oceans, seas and coasts. For example, the latest environmental status assessment of Swedish coastal water bodies showed 27% of the assessed water bodies not to fulfil the requirements for good ecological status with respect to copper, i.e. they displayed copper concentrations in surface seawater or in sediment exceeding the environmental quality standard (WISS, 2020). Similar patterns have been reported for other coastal areas, i.e. Burant et al. (2019) showed 51% of water samples from Californian saltwater marinas to exceed the chronic environmental quality standard. This further motivates the need to accurately determine the load of copper from different anthropogenic activities, including emissions from antifouling paints.

#### 4. Conclusions

This study shows that the release rate method chosen in the ERA of antifouling products will have a large impact on the estimated pressure of biocides as well as the outcome of the ERA. If site-specific release rates determined in the field with the XRF method are used, none of the eight products assessed in the current study would pass the ERA in the Baltic, the Baltic Transition or in the Mediterranean. However, most of the coatings would pass the ERA if Tier 2 release rates obtained using the calculation method or the laboratory method are used. Ideally, it is recommended that site-specific field release rates are used to estimate pressures of biocides in future environmental risk assessments of antifouling products, but as a first step, Tier 2 correction factors should no longer be permitted when using existing standardized methods. Increasing the accuracy of the predicted pressure of biocides, would act to ensure sustainable leisure boating in European coastal waters in the future. Since copper pollution is a worldwide problem, particularly in semi-enclosed pleasure craft marinas, the recommendation to use field specific release rates in environmental risk assessments also applies to other regions and countries outside of EU.

#### Credit author statement

Erik Ytreberg: Writing – original draft, Conceptualization, Formal analysis, Methodology, Maria Lagerström: Writing – review & editing, Conceptualization, Visualization, Formal analysis Sofia Nöu: Investigation, Writing – review & editing Ann-Kristin E. Wiklund: Writing – review & editing, Funding acquisition, Conceptualization.

#### 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.



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## Appendix A. Supplementary data

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## References

- Almeida, E., Diamantino, T.C., de Sousa, O., 2007. Marine paints: the particular case of antifouling paints. *Prog. Org. Coating* 59, 2–20.
- Amara, I., Miled, W., Slama, R.B., Ladhari, N., 2018. Antifouling processes and toxicity effects of antifouling paints on marine environment. A review. *Environmental Toxicology and Pharmacology* 57, 115–130.
- Antizar-Ladislao, B., 2008. Environmental levels, toxicity and human exposure to tributyltin (TBT)-contaminated marine environment. A review. *Environment International* 34, 292–308.
- ASTM, 2005. Standard Test Method for Determination of Copper Release Rate from Antifouling Coatings in Substitute Ocean Water. ASTM Method D 6442-05, p. 9.
- Bixler, G.D., Bhushan, B., 2012. Biofouling: lessons from nature. *Phil. Trans. Math. Phys. Eng. Sci.* 370, 2381–2417.
- Burant, A., Zhang, X., Singhasemanon, N., 2019. Antifouling Paint Biocide Monitoring and Modeling to Support Mitigation, Pesticides in Surface Water: Monitoring, Modeling, Risk Assessment, and Management. American Chemical Society, pp. 491–517.
- Davidson, I.C., Smith, G., Ashton, G.V., Ruiz, G.M., Scianni, C., 2020. An experimental test of stationary lay-up periods and simulated transit on biofouling accumulation and transfer on ships. *Biofouling* 36, 455–466.
- ECHA, 2017. PT 21 Product Authorisation Manual (Environmental Risk Assessment). Available at: <https://echa.europa.eu/sv/guidance-documents/guidance-on-biocides-legislation/emission-scenario-documents>.
- ECHA, 2020. Accessed 2020-09-14. <https://echa.europa.eu/sv/guidance-documents/guidance-on-biocides-legislation/emission-scenario-documents>.
- Elliott, M., Burdon, D., Atkins, J.P., Borja, A., Cormier, R., de Jonge, V.N., Turner, R.K., 2017. “And DPSIR begat DAPSI(WR(M))” - a unifying framework for marine environmental management. *Mar. Pollut. Bull.* 118, 27–40.
- Elliott, M., O'Higgins, T.G., 2020. From DPSIR the DAPSI(WR(M)) Emerges... a Butterfly – ‘protecting the natural stuff and delivering the human stuff’. In: O'Higgins, T.G., Lago, M., DeWitt, T.H. (Eds.), *Ecosystem-Based Management, Ecosystem Services and Aquatic Biodiversity: Theory, Tools and Applications*. Springer International Publishing, Cham, pp. 61–86.
- EU, 2006. HARMONISATION of LEACHING RATE DETERMINATION for ANTIFOULING PRODUCTS under the BIOCIDAL PRODUCTS DIRECTIVE. Workshop Report Endorsed at the 26th Meeting of Representatives of Members States Competent Authorities for the Implementation of Directive 98/8/EC Concerning the Placing of Biocidal Products on the Market (11-14 September 2007). Workshop Report Ispira, Italy, 12 December 2006.
- Finnie, A.A., 2006. Improved estimates of environmental copper release rates from antifouling products. *Biofouling* 22, 279–291.
- Heine, L., Nestler, A., 2019. Promising practices for alternatives assessment: lessons from a case study of copper-free antifouling coatings. *Integrated Environ. Assess. Manag.* 15, 867–879.
- ISO, 2010. Paints and Varnishes — Modelling of Biocide Release Rate from Antifouling Paints by Mass-Balance Calculation. International Standards ISO 10890, p. 2010.
- Kylin, H., Haglund, K., 2010. Screening of antifouling biocides around a pleasure boat marina in the Baltic Sea after legal restrictions. *Bull. Environ. Contam. Toxicol.* 85, 402–406.
- Lagerström, M., Ferreira, J., Ytreberg, E., Eriksson-Wiklund, A.-K., 2020a. Flawed risk assessment of antifouling paints leads to exceedance of guideline values in Baltic Sea marinas. *Environ. Sci. Pollut. Control Ser.* 27, 27674–27687.
- Lagerström, M., Lindgren, J.F., Holmqvist, A., Dahlström, M., Ytreberg, E., 2018. In situ release rates of Cu and Zn from commercial antifouling paints at different salinities. *Mar. Pollut. Bull.* 127, 289–296.
- Lagerström, M., Ytreberg, E., 2020. Quantification of Cu and Zn in Antifouling Paint Films by XRF. *Talanta*, p. 121820.
- Lagerström, M., Ytreberg, E., Wiklund, A.-K.E., Granhag, L., 2020b. Antifouling paints leach copper in excess – study of metal release rates and efficacy along a salinity gradient. *Water Res.* 186, 116383.
- Lindgren, J.F., Ytreberg, E., Holmqvist, A., Dahlström, M., Dahl, P., Berglin, M., Wrangé, A.-L., Dahlström, M., 2018. Copper release rate needed to inhibit fouling on the west coast of Sweden and control of copper release using zinc oxide. *Biofouling* 34, 453–463.
- Miller, R.J., Adeleye, A.S., Page, H.M., Kui, L., Lenihan, H.S., Keller, A.A., 2020. Nano and traditional copper and zinc antifouling coatings: metal release and impact on marine sessile invertebrate communities. *J. Nanoparticle Res.* 22, 129.
- Morroni, L., Sartori, D., Costantini, M., Genovesi, L., Magliocco, T., Ruocco, N., Buttino, I., 2019. First molecular evidence of the toxicogenetic effects of copper on sea urchin *Paracentrotus lividus* embryo development. *Water Res.* 160, 415–423.
- Ochoa-Herrera, V., León, G., Banihani, Q., Field, J.A., Sierra-Alvarez, R., 2011. Toxicity of copper(II) ions to microorganisms in biological wastewater treatment systems. *Sci. Total Environ.* 412–413, 380–385.
- Schiff, K., Diehl, D., Valkirs, A., 2004. Copper emissions from antifouling paint on recreational vessels. *Mar. Pollut. Bull.* 48, 371–377.
- Schultz, M.P., Bendick, J.A., Holm, E.R., Hertel, W.M., 2011. Economic impact of biofouling on a naval surface ship. *Biofouling* 27, 87–98.
- Shan-I, C., Thomason, J., Prowse, G., 2013. Defining Typical Regional Pleasure Craft Marinas in the EU for Use in Environmental Risk Assessment of Antifouling Products, Available at: [https://echa.europa.eu/documents/10162/16908203/pt21\\_regional\\_marina\\_scenario\\_study\\_en.pdf/be35dd48-86ca-4e30-b6ac-425ed5ffd5a4](https://echa.europa.eu/documents/10162/16908203/pt21_regional_marina_scenario_study_en.pdf/be35dd48-86ca-4e30-b6ac-425ed5ffd5a4).
- Strivens, J., Hayman, N., Rosen, G., Myers-Pigg, A., 2020. Toward validation of toxicological interpretation of diffusive gradients in thin films in marine waters impacted by copper. *Environ. Toxicol. Chem.* 39, 873–881.
- Thomas, K.V., Brooks, S., 2010. The environmental fate and effects of antifouling paint biocides. *Biofouling* 26, 73–88.
- Valkirs, A.O., Seligman, P.F., Haslbeck, E., Caso, J.S., 2003. Measurement of copper release rates from antifouling paint under laboratory and in situ conditions: implications for loading estimation to marine water bodies. *Mar. Pollut. Bull.* 46, 763–779.
- WISS, 2020. Water Information System Sweden, a Database Developed by the Competent Authorities of the Swedish Water Districts, the County Administrative Boards and the Swedish Agency for Marine and Water Management. Accessed: 2020-06-23. <https://viss.lansstyrelsen.se/>.
- Yebra, D.M., Kiil, S., Dam-Johansen, K., 2004. Antifouling technology—past, present and future steps towards efficient and environmentally friendly antifouling coatings. *Prog. Org. Coating* 50, 75–104.
- Ytreberg, E., Lagerström, M., Holmqvist, A., Eklund, B., Elwing, H., Dahlström, M., Dahl, P., Dahlström, M., 2017. A novel XRF method to measure environmental release of copper and zinc from antifouling paints. *Environ. Pollut.* 225, 490–496.