Sustainability ranking of antifouling coatings for leisure boats - Balancing efficacy and environmental impact

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Sustainability ranking of antifouling coatings for leisure boats – Balancing efficacy and environmental impact

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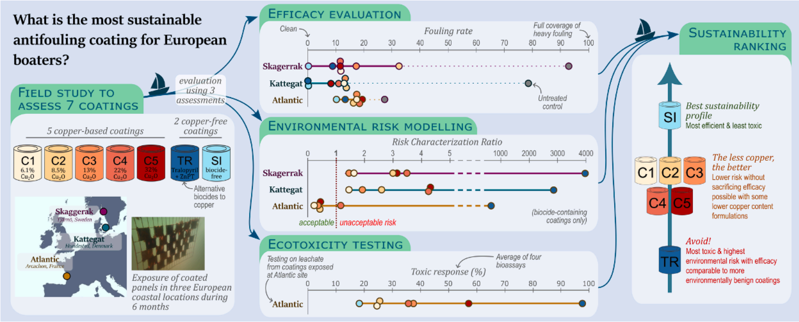
HIGHLIGHTS

- Sustainability assessment of seven antifouling coatings for leisure boats.
- Combined field efficacy, environmental risk modeling and ecotoxicity testing.
- Biocide-free silicone coating ranked most sustainable with low toxicity.
- Tralopyril-based coating exhibited high toxicity and environmental risk.
- Copper coatings met environmental risk criteria only at one of three study sites.

GRAPHICAL ABSTRACT



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ABSTRACT

Antifouling coatings are widely used on leisure boats to prevent biofouling, yet their environmental impacts vary significantly depending on formulation. This study systematically evaluated seven commercial antifouling products, including five copper-based coatings (with varying cuprous oxide content), one tralopyril-based coating and one biocide-free silicone foul-release coating. The evaluation included three complementary approaches: field-based efficacy testing, environmental risk assessment (ERA) modeling and ecotoxicological assays. Field trials were conducted over six months at three European coastal sites (Skagerrak, Kattegat and Atlantic) to assess the antifouling performance. Biocide release rates were quantified using X-ray Fluorescence and used to model environmental risks in accordance with EU guidelines. Acute toxicity of leachates was tested on four marine species (*Aliivibrio fischeri*, *Phaeodactylum tricornutum*, *Artemia salina* larvae and *Oryzias melastigma* larvae). The biocide-free silicone coating ranked highest in sustainability, combining strong antifouling efficacy

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with minimal acute toxicity. The copper-based coatings typically showed comparable efficacy at all sites despite differences in copper release but some only passed ERA thresholds in one of the sites (Atlantic). Lower copper content formulations generally exhibited reduced environmental impact. The tralopyril-based coating, despite being marketed as "environmentally safe", demonstrated the highest toxicity and environmental risk. These findings highlight the need for environmental assessment of antifouling products to validate environmental safety claims and to promote lower impact formulations. While ERA modeling is suitable for biocidal products, ecotoxicological testing remains essential for biocide-free alternatives. Improved and standardized leaching protocols are needed to better reflect real-world conditions and support sustainable product development.

1. Introduction

Antifouling coatings are widely applied to vessel hulls to prevent the settlement of biofouling organisms, which otherwise leads to increased hydrodynamic drag and fuel consumption [1]. Copper, particularly in the form of cuprous oxide (Cu_2O), is currently the most widely used biocide in antifouling paints globally [2]. Its effectiveness against a broad spectrum of marine organisms has made it a default choice for decades [3]. However, the widespread use of copper-based coatings has led to elevated concentrations of copper in marina sediments and waters, raising concerns about toxicity to non-target species and long-term ecological impacts [4–6].

To address these environmental concerns, three main strategies can be employed to reduce the impact of antifouling coatings. The first strategy is to reduce the release rate of copper from antifouling coatings. Studies have shown that many antifouling coatings for leisure boats release copper in excess of the necessary dose to inhibit fouling, suggesting that dose reduction without compromising on efficacy is possible [7, 8]. The effective dose may however differ between water bodies: while a copper release rate of 2 $\mu g/cm^2/day$ is enough to prevent macrofouling in the brackish Baltic Sea (salinity $\leq$ 8 PSU) [7], a release rate of 7 $\mu g/cm^2/day$ may be required in the more saline North Sea/Atlantic ($\geq$ 20 PSU) [8]. Hence, to reduce environmental copper loads, regional-specific products with tailored doses need to be developed. Even so, it is not certain that the required dose to prevent biofouling would be low enough to pass the environmental risk assessment in all waters [9].

The second strategy involves using so called "copper-free" coatings that incorporate more environmentally sustainable biocide(s). Among these, tralopyril has gained attention as a non-copper biocide and is often marketed in copper-free coatings as an environment-friendly option due to its rapid degradation in aquatic environments [2]. Similar to copper, tralopyril (4-bromo-2-(4-chlorophenyl)-5-(trifluoromethyl)-1H-pyrrole-3-carbonitrile) is a broad-spectrum antifouling agent, effective against a wide range of fouling organisms such as barnacles, hydroids, mussels, oysters and polychaetes [10]. Tralopyril is a pyrrole-based compound that acts by disrupting mitochondrial function in fouling organisms [11]. Although tralopyril itself degrades relatively quickly, one of its degradation products, BCCPCA (3-bromo-5-(4-chlorophenyl)-4-cyano-1 H-pyrrole-3-carboxylic acid), has been found to be persistent in sediments [12]. Moreover, tralopyril is often used in combination with zinc pyrithione (ZnPT), a booster biocide that enhances efficacy against soft fouling organisms [2]. ZnPT, while also known for rapid degradation, has been shown to be toxic to a wide range of marine species including algae, crustaceans and fish at low concentrations. Additionally, the stability and toxicity of ZnPT metabolites are mostly unknown [13].

The third strategy is to use biocide-free antifouling coatings, where silicone-based foul-release coatings currently seem to be the most promising alternative. These coatings rely on low surface energy and mechanical properties rather than chemical toxicity [14]. The combination of low surface energy and elastic modulus reduces the adhesion strength of fouling organisms, enabling a 'self-cleaning' effect through removal by water shear forces during navigation [15]. It has nonetheless been demonstrated that such biocide-free coatings can match or exceed the performance of copper-based paints in certain regions, even during static conditions [16, 17]. However, concerns remain regarding the potential emission of silicone oils from these coatings and ecotoxicological data on new formulations are still limited [17].

No comprehensive comparison of the efficacy and environmental risks of these three strategies has been made so far. The present study aims to address this gap by systematically evaluating the performance and potential ecological impact of five copper-based antifouling coatings (with different Cu_2O contents), one tralopyril-based coating and one biocide-free foul-release coating. Efficacy testing paired with biocide release rate measurements was conducted over a six-month field study in three European coastal locations. Using previously published results on copper release rates from the copper-based coatings [8] and new release rate data for the tralopyril-based coating, environmental risk assessments (ERA) on biocidal release rates were performed in accordance with EU guidelines. Ecotoxicity testing on leachates from field-exposed coatings was also conducted on all coatings using bioassays on four marine species. The study compares efficacy and environmental risk across three coastal locations with contrasting physical properties and biofouling intensities, with the aim of ranking the products with regards to their overall sustainability, i.e. their ability to balance efficacy with environmental toxicity.

2. Materials & methods

2.1. Field exposure

2.1.1. Studied products and market availability

Seven different market products (black color) were assessed in this study (Table 1) through field tests in coastal waters of three European countries: Sweden, Denmark and France. Five of these products were traditional copper-based coatings, containing cuprous oxide as the sole active substance, with concentrations ranging from 6% to 32% by weight. They were marketed as self-polishing (C1), polishing (C2, C5) or hard (C3, C4) and represent the spectrum of copper coatings available to boaters in the studied countries. However, while products with high concentrations (20–40%, such as C4 and C5) are commonly found in all three countries, those with lower concentrations (<15%, such as C1 – C3) are not readily available to boaters in France [18]. Additionally, one product with a different biocidal composition (tralopyril and ZnPT) and a biocide-free silicone coating were also tested. The tralopyril-based coating (TR) is currently available only on the Danish and French markets, whereas the silicone coating (SI) can be purchased in all three countries. A control coating, consisting of a biocide-free primer was also included as a reference for the efficacy assessment to characterize the site-specific natural fouling pressure (see 2.2.).

2.1.2. Study locations

All three immersion sites, already described in Lagerström et al. (2025) [8], were located in close proximity to the coast (Fig. 1A). In France, the panels were exposed in Port de la Vigne, a recreational marina with 310 berths located in Arcachon Bay, a marine nature park established in 2014 (44.6744°N, −1.2400°E). This marine protected area hosts rich biodiversity which is essential to maintain ecological function. Alongside, the bay supports several marine professional activities such as fishing, oyster farming, boat construction and

Table 1
Coatings assessed in this study. The content of the active substances was extracted from the Swedish (coatings C1-C5) and French (coating TR) authorities' public biocidal products registries [18, 19]. The zinc oxide content range was obtained from the coatings' safety data sheets. Further details about the coatings can be found in Table SI1.

Abbreviation	Coating type	Active substances (wt%, ww)			
		Cuprous oxide (Cu_2O)	Zinc pyrithione (ZnPT)	Tralopyril	Zinc oxide (ZnO)
C1	Copper (biocidal)	6.1	-	-	$\geq 10 - \leq 25$
C2	Copper (biocidal)	8.5	-	-	$\geq 10 - \leq 25$
C3	Copper (biocidal)	13	-	-	7–10
C4	Copper (biocidal)	22.02	-	-	$\geq 10 - \leq 25$
C5	Copper (biocidal)	31.93	-	-	$\geq 2.5 - < 25$
TR	Tralopyril and zinc pyrithione (biocidal)	-	7.0	4.0	15 – 20
SI	Silicone foul-release (biocide-free)	-	-	-	-
Control	Primer (biocide-free)	-	-	-	-

maintenance, as well as a wide range of marine recreational activities. Arcachon Bay is an attractive area with an increasing population density and tourism. It is also the site where the harmful effects of the antifouling biocide tributyltin (TBT) were first documented in the early 1980's, prompting a French ban on organotin-containing antifouling paints in 1982 [20]. This ultimately paved the way for global restrictions under the International Convention on the Control of Harmful Anti-fouling Systems on Ships (AFS Convention) adopted in 2001 [21]. For the Danish location, the testing platform of the CoaST Maritime Test Center was used (55.9661°N, 11.8427°E). The exposure platform is located in the Port of Hundested, which encompasses both recreational and commercial port areas. The port sits at the outlet of Isefjord, a designated Natura 2000 protected area (Natura 2000 nr. 153: *Havet og kysten mellem Hundested og Rørvig*). The panels in Sweden were exposed at Tjärnö Marine Laboratory (58.8818°N, 11.1340°E), located on the Swedish west coast, close to the Norwegian border. Tjärnö Marine Laboratory is located by Kosterhavet National Park, Sweden's first marine national park. Hence, all three sites are areas where on-going efforts to preserve high natural values co-exist with recreational and commercial maritime activities.

2.1.3. Panel preparation and immersion

All biocidal coatings (C1-C5 and TR) were applied to 2 mm thick PVC panels (Poly Vinyl Chloride, 10 ×10 cm) using an automatic, motorized film applicator (TQC AB3120, TQC Sheen B.V., Rotterdam, Netherlands). As described in Lagerström et al., 2025 [8], panels were first coated with a primer layer (the same as the Control) at a wet film thickness (WFT) of 100 µm. Subsequently, an 8 cm wide stripe of each biocidal coating was applied across the 10 cm width of the panels. A WFT of either 300 or 400 µm was applied, using identical application and curing conditions for all coatings, with the choice of WFT adjusted solely to ensure that the resulting dry film thicknesses (80–120 µm) matched or exceeded each respective products' recommendations for one boating season. All panels were left to air-dry for at least two days. Four parallel sets of panels, with three replicates per biocidal coating, were prepared for each of the three exposure sites to be able to monitor changes in biocide release rate over time by X-Ray Fluorescence (see 2.3). The panels were randomly mounted on metal frames and immersed at depths between 35 and 95 cm at each site.

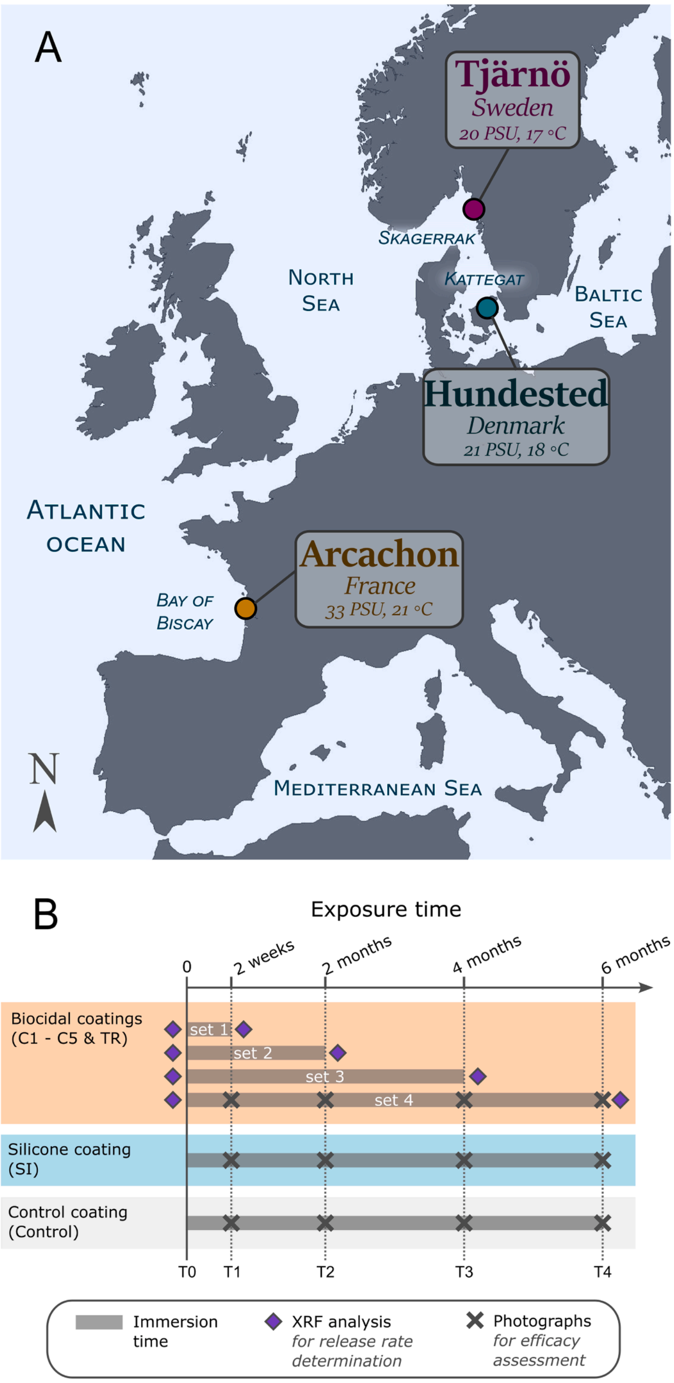

Fig. 1. Map showing exposure locations of panels coated with antifouling coatings in Europe and the average salinity and temperature for the sites during the field study (A) and an overview of the exposure scheme and assessments for the different products during field exposure (B).

Initial immersion at the three locations occurred within a week of each other in spring 2023 (end of April/beginning of May). Panels were subsequently retrieved at 4 different timepoints: 2 weeks (T1), 2 months (T2, 56 days), 4 months (T3, 119 days) and 6 months (T4, 182 days). For the biocide-free silicone coating (SI), the panels were prepared by the coating manufacturer, following the recommended application instructions, and immersed for the full 6 months of the field experiment (Fig. 1B). At the French location, blank (uncoated) PVC panels were also exposed for 6 months to be used as field controls for the ecotoxicological tests (see 2.4.). Salinity and temperature were monitored at all sites during the immersion period. The 6-month exposure period (May–October) was selected for the field experiment as it corresponds to the typical timing and duration of the recreational boating season in all three countries. For the French location, this peak period was later confirmed through a 2023 boat census based on aerial and satellite imagery [22].

2.2. Efficacy evaluation

To assess the efficacy of the different products against biofouling, photographs of the panels were taken at each of the designated timepoints (Fig. 1B). Biofouling coverage of the panels was evaluated using a modified version of the fouling rating system from the US Naval Ships' Technical Manual (NSTM) [23], summarized in Table 2. Fouling rate categories (FR) range from 0 (clean surface) to 100 (extensive fouling with multiple types of organisms), with soft fouling (FR10–FR30) including biofilms, macroalgae, tunicates and ascidians, and hard fouling (FR40–FR90) comprising of calcareous organisms like tubeworms, barnacles, mussels and oysters. Modifications were made to improve classification accuracy: juvenile barnacles (<2 mm) were assigned to FR20 and encrusting bryozoans were included under FR40, following refinements proposed by Oliveira and Granhag (2020). Additionally, FR70 was redefined to include the presence of either tubeworms or barnacles, rather than requiring both.

Panel photographs were analyzed with ImageJ to outline and quantify the surface coverage (in %) of each FR category, and Inkscape (v1.3.2) was used to estimate the area of juvenile barnacles (brightness cutoff of 0.3). Surface coverage was based on the entire organism, except for tunicates and algae, where only attachment points were considered, following NSTM guidelines. To avoid edge effects, organisms located within 15 mm of panel edges or not physically attached to the surface (e.g. overhanging growth) were excluded, following ASTM D 6990 (2011) [46]. A final weighted fouling rating (FR_w) was calculated to obtain an overall score for each panel [16], where the FR category values (0 – 100) were multiplied with their respective observed surface coverage (in %) and subsequently added together according to the following equation (where n is the total number of FR categories detected per panel):

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

Table 2
Fouling rate scale adapted from the Naval Ships' Technical Manual [23]. Modifications applied in this study are shown in italic.

Fouling Rate (FR)	Fouling Type	Description
0	-	Clean, foul-free surface
10	Soft	Incipient slime, painted surfaces visible beneath the fouling
20	Soft	Advanced slime, painted surfaces obscured by the fouling. *Juvenile barnacles ≤ 1 mm.*
30	Soft	Soft fouling up to 76 mm in length and up to 6.4 mm in height (e.g. filaments, sea cucumbers)
40	Hard	Tubeworms less than 6.4 mm in height or diameter. *Encrusting bryozoans.*
50	Hard	Barnacles less than 6.4 mm in height or diameter
60	Hard	Combination of tubeworms and barnacles less than 6.4 mm in height or diameter
70	Hard	Tubeworms and/*or* barnacles greater than 6.4 mm in height or diameter
80	Hard	Closely packed tubeworms or barnacles (less than 6.4 mm in height) growing on top of each other
90	Hard	Dense growth of tubeworms with barnacles, 6.4 mm or greater in height; *presence of mussels or oysters*
100	Composite	Soft and hard fouling present, with soft fouling growing over various forms of hard growth

The earliest timepoint with the highest observed FR_w on the Control was selected as the most suitable for the efficacy comparison between the antifouling coatings studied at the three sites. To assess differences in coating performance, one-way ANOVAs ($\alpha = 0.05$) were conducted for each site. Where significant differences were found, post hoc testing (Tukey's HSD) was performed (JMP v. 18.2.2).

2.3. Release rates and environmental risk modelling

2.3.1. Release rates of biocides and substances of concern

Field release rates (µg/cm^2/day) were determined using X-ray fluorescence (XRF) spectroscopy, one of few methods that enables quantification of in-situ biocide release from antifouling coatings [24–26]. All biocides in the tested coatings contain signature elements (i.e. Cu from Cu_2O, Zn from ZnO or ZnPT or Br from tralopyril) detectable by XRF. By performing non-destructive measurements of these elements at defined points on coated panels before and after exposure, biocide release rates can thus be calculated.

2.3.1.1. Copper coatings (C1-C5). The derivation of copper and zinc release rates for coatings C1 – C5 has already been described in detail in Lagerström et al., 2025 [8] and is therefore only briefly summarized here. An XRF instrument (Delta-50, Innov-X, Olympus) was calibrated to determine the area concentrations of Cu and Zn (in µg/cm^2) for each coating. To monitor changes in metal release over time, four parallel sets of panels were prepared to enable retrieval and subsequent XRF measurement at four different timepoints (Fig. 1B). XRF analysis was conducted in the same measurement points (four in total) on each panel both before and after exposure. Post-exposure, all XRF measurements were made on air-dried panels and after gentle removal of any obstructing fouling present on the panels. The difference in area concentration between timepoints, i.e. the metal release in µg/cm^2, was averaged for each panel (n = 4 points), resulting in three replicates per timepoint (n = 3 panels). The cumulative releases over time were then plotted and release rates (in µg cm^{-2} day^{-1}) derived from the linear regression slopes between consecutive timepoints. The uncertainties of the derived release rates are given as the 95% confidence interval of the slope of the fitted regressions.

2.3.1.2. Tralopyril and zinc pyrithione coating (TR). The release rates of tralopyril from the coating TR were inferred from the determined loss of bromine (Br). The Br peak was detected by XRF using the same acquisition parameters as for the copper coatings (beam energy of 40 kV, 20 s measurement time with triplicate spectra acquired for each analysis point). The characteristic X-Ray emission line of Br Kα peak has an energy of 11.9 kg electron volt (keV) and the Br peak was integrated between 11.59 – 12.30 keV and, just as for Cu and Zn, normalized against the background scatter signal integrated between 20.3–20.8 keV [8, 26].

Calibration curves were established for both Br and Zn for coating TR. Standards were prepared according to the same procedure as that of the paint-specific Cu and Zn calibration curves for C1 – C5. A detailed description of this work can be found in the Supporting Information Text SI1. The relationship between the area concentration of Br and Zn (in µg cm^{-2}) and their respective Kα peak areas were established. For all coatings, data points were fitted to polynomial curves of the second degree with r^2-values ≥ 0.998 (**Figure SI1**). The limits of quantification, defined as the mean blank value plus ten standard deviations, were determined to 4.4 µg cm^{-2} for Br and 65.1 µg cm^{-2} for Zn.

2.3.1.3. Statistical tests. One-way ANOVAs with post hoc testing (Tukey HSD) were used to assess statistical differences in cumulative Br and Zn releases between the three study sites (due to variations in environmental parameters such as salinity and temperature) for coating TR. To evaluate temporal differences in release rates, an analysis of means (ANOM) was carried out (JMP v. 18.2.2) for all coatings and XRF-determined elements (Cu, Zn and Br). In the ANOM analysis, the mean release rates from each of the four studied time intervals were compared to their grand mean at a significance level of 5%, following the methodology outlined by Nelson et al. (2005) [47]. Release rates that fall outside the calculated upper or lower decision limits are considered statistically higher or lower than the overall average.

2.3.2. Environmental risk modelling

Environmental risk for the biocide-containing coatings (C1–C5 and TR) was assessed using Risk Characterization Ratios (RCRs), in line with the environmental risk assessment (ERA) framework under the Biocidal Products Regulation (BPR) (EU) 528/2012 and ECHA guidelines for antifouling product approval [27]. Emission scenario calculation tools provided by ECHA, based on modelling in MAMPEC (Marine Antifouling Model to Predict Environmental Concentrations), were used to estimate predicted environmental concentrations (PEC) for the dissolved phase inside marinas. RCRs are calculated as the ratio of the predicted environmental concentration (PEC) to the predicted no-effect concentration in seawater (PNEC). RCR > 1 indicates an unacceptable risk to marine life, potentially preventing the product from being approved for use.

Following guidelines, PECs were modelled for all biocide(s) and substances of concern (SoC) (e.g., Zn) present in each coating, namely: Cu and Zn for coatings C1–C5, and tralopyril, Zn and ZnPT for coating TR. Tralopyril release rates were calculated under the assumption that all Br emissions were in the form of tralopyril. This assumption is supported by the close agreement between the Br concentration of the dry coating determined via standard addition in this study (14,747 mg/kg, see Supporting Information) and the Br concentration expected if the coating's stated content of tralopyril (4.0 wt%) was the sole source of Br (13,826 mg/kg). The release rates of ZnPT for coating TR were derived from those of Zn, under the assumption that ZnPT release followed the same dynamics as total Zn. Both ZnPT and ZnO contribute to the total Zn release, but the XRF release rate measurement cannot distinguish between the two Zn sources. Additionally, the ratio between the two substances in the coating was unknown as concentrations of substances not defined as biocides (e.g., ZnO) were not disclosed by manufacturers (Table 1). The ZnPT contribution to total Zn was therefore estimated (to 12%) by using the known ratio of ZnPT to tralopyril in the coating. Release rates of ZnPT alone could then be derived for the subsequent calculations.

Release rates were entered into the Excel calculation tools provided in the PT21 Emission Scenario Document by ECHA [28], which outputs the 90th percentile RCR for different marina scenarios for the marine regions of the Baltic (38 marinas), Baltic Transition (17 marinas), Atlantic (47 marinas) and Mediterranean (46 marinas). Release rates from the field study sites were only assessed for their geographically corresponding region, i.e. Baltic Transition (Tjärnö, Sweden and Hundested, Denmark) and Atlantic (Arcachon, France). The combined risk of multiple biocides and SoCs present in a coating was assessed using the additional mixture assessment tool, which first sums the individual RCR values of the substances before determining the 90th percentile $\sum$RCR for the product. As no emission scenario tool exists for ZnPT, all ZnPT release rates were converted to copper pyrithione (CuPT) equivalents and used as input in the CuPT emission scenario document. ZnPT is known to transchelate into CuPT in seawater [13], supporting the relevance of this approach. Furthermore, the marine PNEC value of CuPT (17.6 ng/L), although expressed as CuPT, was initially derived for total pyrithione [29].

Application factors of 95% for Cu and 90% for other substances were used, following ECHA guidelines [27]. The calculation was performed on the average release rates, as well as their lower and upper 95% confidence estimates allowing the propagation of 95% confidence intervals for the final $\sum$RCR. To evaluate whether the calculated $\sum$RCRs significantly exceeded the threshold value of 1, a one-sample *t*-test ($\alpha = 0.05$) was applied. The ERA was carried out for each time interval, but the interval with the most frequent significant exceedances of 1 was selected for a worst-case assessment, following the precautionary principle.

2.4. Ecotoxicity testing

2.4.1. Leachate preparation

Four different short-term toxicity assays with marine species (*Aliivibrio fischeri*, *Phaeodactylum tricornutum*, *Artemia salina* larvae and *Oryzias melastigma* larvae) were conducted using leachate solutions from coated panels exposed at the Arcachon Bay site. Leachates were produced in filtered (<0.45 µm) artificial seawater (ASW) (Instant Ocean® Sea Salt; salinity: 33 g/L and pH 8.0–8.5). The panels were immersed in glass jars (washed three times with 70% ethanol and three times with distilled water) at a ratio of 1 L of ASW to 100 cm^2 of paint surface for 24 h at 23°C, in the dark, with orbital agitation (IKA-KS 501 digital) at 115 rpm. After 24 h, the leachates were filtered at 0.45 µm to remove particles and then stored either refrigerated at 4°C to be used within two days or frozen at −20 °C for longer storage.

For a worst-case assessment, leachates were produced from panels retrieved at the timepoint with the highest expected releases of potentially harmful substances. Accordingly, leachates were generated from the triplicate panels exposed for two months (T2) for the biocidal coatings (C1-C5 and TR). Meanwhile, non-exposed panels (T0) were selected for coating SI as previous studies have shown that silicone coatings may exhibit higher toxicity when fresh [30, 31]. For comparison, three of the ecotoxicity assays were also performed on coating SI at the 6-month timepoint (T4). Triplicate uncoated and unexposed (T0) PVC panels were leached to serve as controls. To investigate the putative toxicity of the ageing of PVC panel itself and of any attached fouling, PVC panels exposed in the field for 6 months (T4) were also leached and tested using the marine bacteria assay.

Time-dependent release patterns were also explored with the bacterial assay through extended testing of coatings C3 and TR, using leachates from triplicate panels retrieved at all timepoints (T1-T4). As coating TR holds two active substances (tralopyril and ZnPT) that should rapidly degrade in the marine environment through hydrolysis and/or photodegradation [13, 32], a photodegradation test was carried out whereby additional bacterial assays were performed on T2 leachates after exposure to sunlight. To ensure optimal light conditions and penetration, leachates were placed in quartz beakers and exposed outdoors under clear skies (22th and 23th of July 2024 from 10:00 AM to 16:00 PM, temperature from 16°C in the morning to 27°C in the afternoon) for 6 ($n = 6$) and 12 ($n = 3$) hours. Results were compared to non-irradiated leachates ($n = 6$). An overview of the leached panels and their corresponding tests is provided in Table 3.

2.4.2. Ecotoxicity assays

2.4.2.1. Marine bacteria (Aliivibrio fischeri), bioluminescence inhibition. Microtox was carried out on the leachates using *Aliivibrio fischeri* according to ISO 11348–3:2007. Bioluminescence was measured (Luminometer BioLight® Toxy, R-Biopharm) after 30 min of bacteria exposure at 15°C. Potassium dichromate ($K_2Cr_2O_7$) solutions were used as positive and negative controls for the test at concentrations of 200, 100, 50, 25, 12.5 and 0 mg/L in NaCl at 20 g/L. For the paint leachate tests, the negative control was artificial seawater (ASW) at 33 g/L.

2.4.2.2. Marine microalgae (Phaeodactylum tricornutum), growth inhibition. Growth inhibition tests were performed on the marine diatom *Phaeodactylum tricornutum* using a microbiotest (Algaltoxkit M., MicroBioTests) in 96 wells micro-plates. Plates were placed in an orbital shaker at 120 rpm for 72 h at 22°C in a thermoregulated chamber under constant white LED illumination (RetroLED bright, 1560 Lm, SuperFish). The microalgae were counted in a Malassez counting chamber. The negative control was ASW and the positive control was $K_2Cr_2O_7$ at

Table 3
Overview of ecotoxicity assays performed on leachates from triplicate panels either unexposed (T0) or exposed in Arcachon Bay in France for 2 weeks (T1), 2 months (T2), 4 months (T4) or 6 months (T4). Leachates from the triplicate panels were tested in three sets (n = 9), except for certain coatings and assays where one (*n = 3) or two (**n = 6) sets were tested. A summary of the chemical analyses performed on the leachates are also displayed.

Treatment	Marine bacteria *Aliivibrio fischeri* Bioluminescence inhibition (30 min)	Marine microalgae *Phaeodactylum tricornutum* Growth inhibition (72 h)	Marine microcrustacean larvae *Artemia salina* Immobilization (72 h)	Marine fish larvae *Oryzias melastigma* Mortality (48 h)	Chemical analysis of leachate (a) Cu, Zn, As, Cd (b) Si (c) Tralopyril, BCCPCA, ZnPT, PT, diPT
C1	T2	T2	T2	T2	T2 (a)
C2	T2	T2	T2	T2*	T2 (a)
C3	T1, T2, T3, T4 + EC50 (T2)	T2	T2	T2	T2 (a)
C4	T2	T2	T2	T2*	T2 (a)
C5	T2	T2	T2	T2	T2 (a)
TR	T1, T2, T3, T4 + EC50 (T2) + photodegradation test (T2)	T2 + EC50 (T2)*	T2	T2 + EC50 (T2) *	T2 (c)
SI	T0, T4	T0, T4	T0**, T4**	T0	T0 (a,b), T4 (a,b)
PVC (panel blank)	T0, T4	T0	T0*	T0	T0 (a,b,c), T4 (a,b)
ASW (negative control)	Control	Control	Control	Control	Control (b,c)

concentrations from 1 to 32 mg/L in ASW. (33 g/L). The percentage inhibition of growth rate for each treatment was calculated according to ISO 8692:2012 at 72 h [33].

2.4.2.3. Marine microcrustacean (Artemia salina), larval immobilization. *Nauplii* of *Artemia salina* were produced after a 48h-culture of cysts in ASW at 24°C with continuous air bubbling. For each replicate, ten *nauplii* were incubated in 10 mL of leachate in clean glass jar for 72 h at 20°C. Each condition was tested in triplicate. The negative control was ASW and the positive control was a 5 mg/L $CuSO_4$ solution in ASW. (33 g/L). Dead *nauplii* were counted after 48 and 72 h of exposure. The immobilization rate was calculated as the ratio of cumulative immobile larvae over 72 h to the initial number of larvae in each treatment.

2.4.2.4. Marine fish (Oryzias melastigma), larval mortality. Embryos of the marine medaka at 3–4 days post-fertilization were obtained from the Marbec fish facility, Plateforme Experimentale Marine de Palavas (34250 Palavas, France). Between 14 and 16 fish larvae at hatching (9–10 days post fertilization at 28°C) were exposed for 48 h to 10 mL of 100% leachate in a glass beaker. Each condition was tested in three replicates. The beakers were placed on an orbital shaker at 30 rpm, under a 14 h/10 h light/dark cycle at 26°C in a thermoregulated chamber (Snijders Scientific). At 24 h, dead larvae were counted and removed from the beaker, and 50% of the medium was renewed. At 48 h, dead larvae were counted again and the remaining fish were euthanized. ASW (33 g/L) was used as a negative control. Mortality rate was calculated as the ratio of dead larvae within 72 h to the initial number of larvae in each treatment.

2.4.2.5. Statistical analysis and determination of toxic values. A one-way ANOVA followed by Dunnett's post-hoc test was used for all four assays to assess differences between treatments, using PVC (T0) as the control group (α=0.05, JMP v. 18.2.2).

Serial dilutions of T2 leachate solutions from 100% to 6.25% in ASW were tested to derive reference toxic values for the C3 (bacterial assay) and TR coatings (bacterial, microalgae and fish larvae assays) (Table 3). The Effective Concentration inducing 50% of the maximum effect (EC50 values) of the coating leachates were calculated based on dose–response curves fitted to the experimental data.

2.4.3. Chemical analysis of leachates and RCR calculation

To interpret the observed effects and compare laboratory leaching results with those for the field via XRF, leachates were analyzed for their biocides, transformation products and other elements of concern (Table 3). Silicon (Si) was also analyzed in selected leachates to investigate indications of potential release of silicone oils from coating SI. Details on the chemical analysis methods can be found in the Supporting information Text SI2.

Pairwise comparisons of release rates between lab and field conditions were conducted using two-tailed *t*-tests (α=0.05, JMP 18.2.2) to assess differences in leaching behavior. To evaluate the influence of biocides and substances of concern (Zn) on the observed toxic effects across the four bioassays, linear regression analysis was performed between the $\sum RCR$ of leachates and the corresponding effects. $\sum RCR_{leachate}$ was calculated for each leachate as the sum of the ratios between the average measured concentrations (MEC) of active substances and substances of concern (i) and their PNEC (dissolved, in marine or aquatic environment) using the following equation:

$$\sum_{i}^{n} RCR_{\ leachate} = \frac{MEC_i}{PNEC_i} \tag{2}$$

3. Results & discussion

3.1. Efficacy assessment

To support a conservative evaluation of antifouling performance, the studied coatings were assessed at a timepoint of high fouling pressure. The monitored fouling pressure on the Control (Fig. 2A) was therefore used to select a suitable evaluation timepoint for the efficacy assessment. For the two locations in the Baltic Transition (Tjärnö and Hundested), peak fouling was observed after 4 months of exposure (T3), after a steep increase in FR_w. At the Atlantic site (Arcachon), the fouling development was more gradual, reaching its peak at the 6-month observation point (T4). These findings could support the selection of T4 (6 months) for the efficacy evaluation. However, a decline in FR_w was observed at Tjärnö by the final retrieval (T4). This reduction may be attributed to seasonal factors (e.g., lower temperatures, reduced biological activity), detachment of overgrown organisms or the natural completion of fouling life cycles (**Figure SI2**). Additionally, as discussed with regards to coatings C1-C5 in Lagerström et al. (2025), prolonged static conditions past 4 months were observed to slow or even halt the release of copper. This was hypothesized to be due to a lack of coating surface polishing, a process which requires more dynamic exposure conditions. Hence, to not underestimate the coating performance, the efficacy evaluation was carried out based on the fouling evaluation at T3 (4 months, Fig. 2B).

The weighted fouling rating (FR_W) at timepoint T3 for the copper coatings generally ranged between 10 and 20 across all three locations (Fig. 2C), reflecting that the predominant fouling observed on the panels was either light (FR10) or heavy (FR20) slime (Table 2, Fig. 2B). Overall, the performance of the copper coatings did not differ significantly from

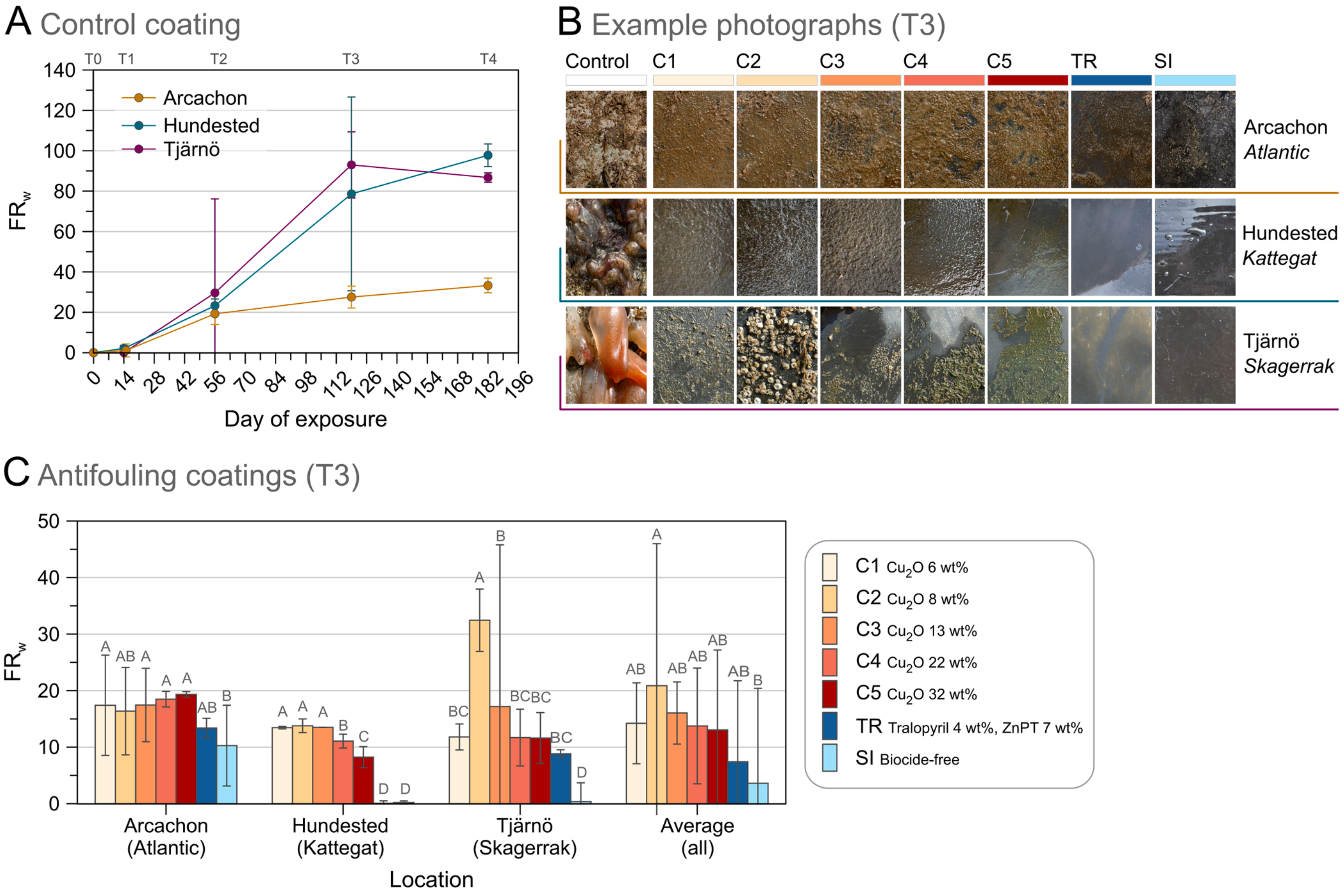

Fig. 2. Change in average weighted fouling rate (FR_W) on the Control coating over time at the three locations (A), example photos of panels retrieved after four months exposure T3 (B) and average weighted fouling rates (FR_W) for the studied antifouling coatings (T3) at the three locations (C). Error bars show the 95% confidence interval (n = 3). The results from the ANOVA with Tukey's HSD post-hoc testing for each site and the grand average are shown by the letter above the bars, where treatment not connected by the same letter are significantly different (α= 0.05).

one another, with two exceptions. At the Skagerrak site (Tjärnö), coating C2 exhibited substantial barnacle coverage (~ 40%), resulting in an average FR_W exceeding 30. Meanwhile, at the Kattegat site (Hundested), coatings with the highest copper content (C4 and C5) demonstrated a slightly improved ability to prevent slime accumulation. Although significant, this difference in average FR_W was relatively modest. The alternative, non-copper coatings TR and SI were either not significantly different from the copper coatings or outperformed them at one (TR) or two (SI) of the locations.

A comparison of the average FR_W values for each coating across test sites reveals that the coatings' performances did not generally differ significantly from one another. The only significant difference was between coatings C2 and SI. The elevated FR_W of coating C2 was primarily driven by its poor performance at the Skagerrak site, whereas the lower FR_W of coating SI reflected its strong performance at both the Kattegat and Skagerrak sites.

Although FR_W values within sites were relatively consistent, differences in fouling type and severity were evident across sites. In particular, the copper-based coatings accumulated considerably heavier slime at the Atlantic site compared with Kattegat and Skagerrak. While differences in biocide release could partly explain this, the Atlantic site also exhibited the highest release rates of copper and zinc from these coatings (Section 3.2), suggesting that site-specific characteristics of the slime fouling community and its tolerance to copper were more influential. This interpretation is further supported by the silicone coating, which also showed the highest slime coverage at Arcachon. At Kattegat, coatings C4 and C5 resisted slime more effectively, consistent with their higher copper release rates. The sensitivity of microalgae to copper varies between species, with some fouling diatoms known to be markedly more tolerant than others, which likely contributes to the observed variation in slime accumulation between sites [34].

The failure of coating C2 to prevent barnacle (*Amphibalanus improvisus*) settlement at Skagerrak (but not Kattegat) also suggests spatial differences in barnacle settlement pressure. Previous work showed that this coating exhibited copper release rates below the threshold needed to fully deter settlement [8]. Although the release rates of coatings C1 and C3 were also below this threshold, they displayed much lower barnacle coverage (single and small individuals only), indicating that the poorer performance of C2 cannot be explained by release rates alone. Additional coating properties such as surface smoothness and/or hardness may have contributed to its reduced performance. Furthermore, barnacles were more abundant at the Skagerrak site than at the Kattegat site (Figure SI2C), which may explain why C2 performed adequately at one site but not the other. These findings underline the importance of multi-site field testing to evaluate antifouling coatings under differing biological and environmental conditions to accurately assess their suitability for use across regions.

3.2. *Environmental risk modelling*

3.2.1. *Release rates*

The emissions of Cu, Zn and Br and their variation over time for the six biocidal coatings are presented in Fig. 3. As discussed in Lagerström et al. (2025) [8], the copper-based coatings can roughly be divided into lower (C1, C2 and C3) and higher (C4 and C5) leaching products, with the higher Cu_2O contents in the latter category likely contributing to the

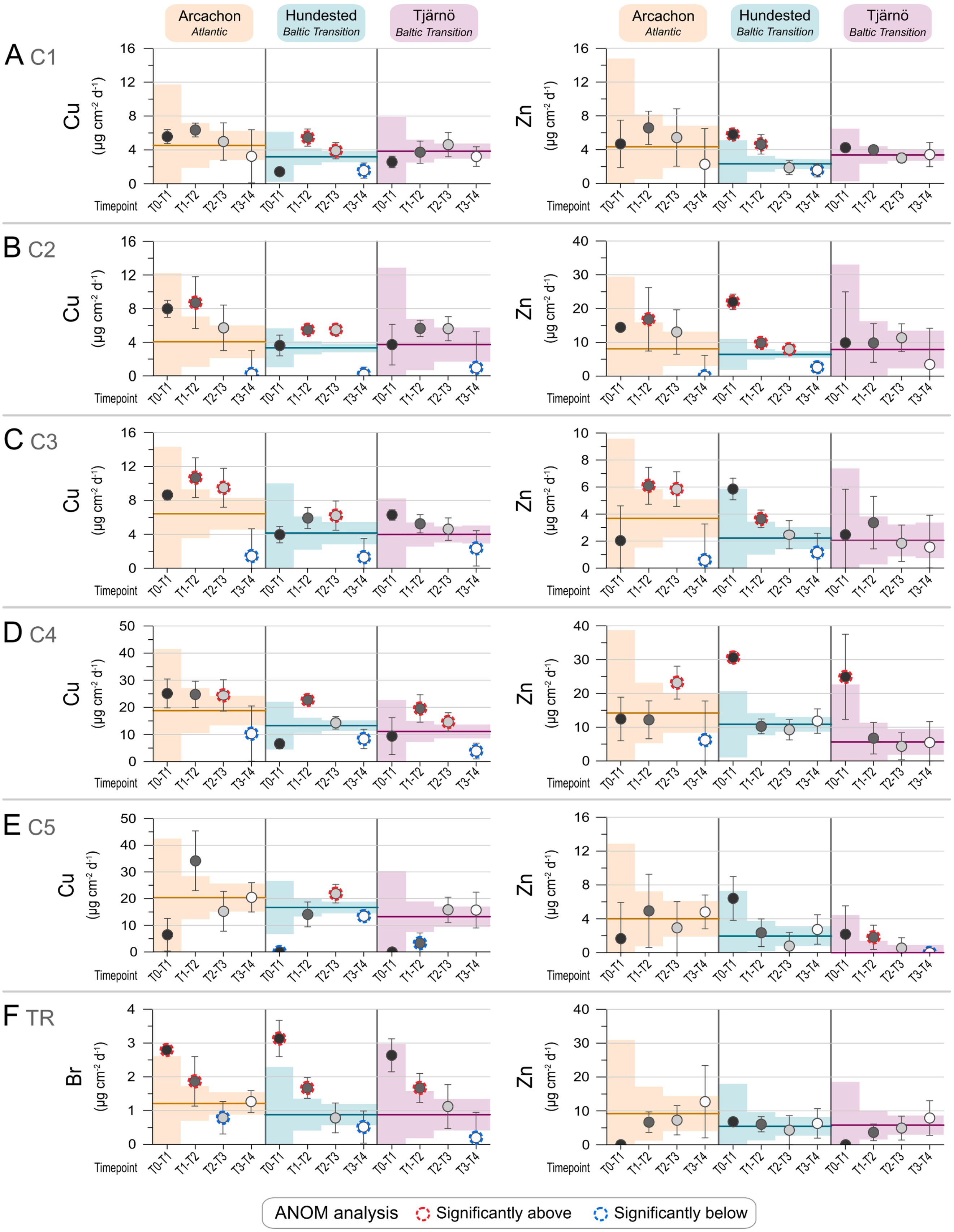

Fig. 3. Release rates of Cu, Zn and Br for the five biocidal coatings (C1-C5 and TR) during the different time intervals of field exposure at the three studied coastal locations (labels on x-axis of bottom graph). Error bars represent the 95% confidence interval of the release rate determination. The colored fields show the 95% confidence interval of the ANOM test, while the colored lines represent the overall average. Red and blue outlines highlight data points significantly above or below the overall average, respectively. Note that the scaling of the y-axes have been adjusted to fit the data and therefore differ between graphs.

more rapid releases (Fig. 3A-E). The previously published work also proposed that the higher Cu release rates observed at Arcachon compared to Hundested and Tjärnö to be linked to the higher salinity and/or temperature at this site, as it can enhance the solubility of Cu_2O particles as well as that of the paint binder [35–37]. The parameters affecting the release rates of Zn from AF coatings are not as well understood. While studies have shown that ZnO content and salinity appear to play a role, their effects have been observed only for certain formulations, suggesting other parameters may be of higher important [7, 25]. Nonetheless, the results of the studied coatings here suggest that higher salinity and/or temperature of the Arcachon waters had a promotional effect also on the release rates of both Zn and Cu, as demonstrated by the higher mean release rates.

For coating TR, the cumulative loss of Br (in µg cm^{-2}) was found to not be significantly different between the three locations for the first 4 months, as indicated by the ANOVA performed for timepoints T1 ($p = 0.4352$), T2 ($p = 0.5298$) and T3 ($p = 0.6098$). This is reflected in the Br release rates, whose magnitude and general decreasing trend over this time period are similar for all three locations (Fig. 3F). At the end of the field study, the total Br loss from the coating at the Arcachon site (on average 248 µg cm^{-2}) was however found to be significantly higher ($p = 0.0026$) compared to Hundested (196 µg cm^{-2}) and Tjärnö (190 µg cm^{-2}). As seen in Fig. 3F, this is due to a drop in Br release rates at the two latter sites in the final (T3-T4) time interval. Similar decreases in release rate during T3-T4, typically affecting both Cu and Zn, were also observed for the other biocidal coatings. According to the ANOM analysis, the time period with the most release rates significantly below the overall average was T3-T4 (20 instances). As previously mentioned in 3.1., this may be attributable to the long-term static exposure conditions. Disregarding this final time interval, the similar Br release patterns across the three sites during the first 4 months of exposure indicate that differences in temperature, salinity or small-scale flow do not seem to impact the leaching behavior of tralopyril from the studied product.

Contrary to Br, no significant deviations from the overall average were observed for Zn (to which both ZnO and ZnPT contribute) during any time interval or at any of the sites for coating TR. While the cumulative release of Zn was initially significantly higher at Hundested as compared to the other two sites (illustrated in Fig. 3F by their low releases at T0-T1), no such differences could be detected by the last two timepoints (4 and 6 months). Hence, coating TR generally behaved similarly at all three sites when it comes to both Br and Zn releases. This is in stark contrast to the copper-based coatings C1-C5 whose release rates were significantly influenced by site-specific conditions. Another difference lies in the release rate trend, as evidenced by the ANOM results. While coating TR typically exhibited the highest Br releases during the initial two weeks of exposure, Cu and Zn emissions of coatings C1-C5 were instead significantly elevated at later stages, specifically the T1-T2 (1–2 months) and T2-T3 (2–4 months) intervals.

3.2.2. *Risk characterization ratios*

The ERA modelling results showed that the risk ratios ($\sum$RCR) significantly exceeding the threshold of 1 were most frequently detected during time intervals T1-T2 and T2-T3, with 14 instances of exceedance each (**Table SI3**). The results for T2-T3 were selected for the worst-case assessment as this timepoint coincides better in time with the efficacy evaluation (carried out at T3).

The ERA results indicated that acceptable environmental risks were exclusively achieved in the Atlantic marine region and only for a subset of the biocidal products. Here, all copper-based coatings except C4 attained an $\sum$RCR < 1 (Fig. 4). None of the biocidal coatings demonstrated an acceptable environmental risk in the Baltic Transition region, regardless of whether release rates from Hundested or Tjärnö were applied. These results are consistent with previous findings, indicating that higher release rates of biocides and substances of concern are permitted in the Atlantic marine region compared to the Baltic Transition, primarily attributed to the greater water exchange capacity in the modelled marinas, driven by larger tidal differences [9].

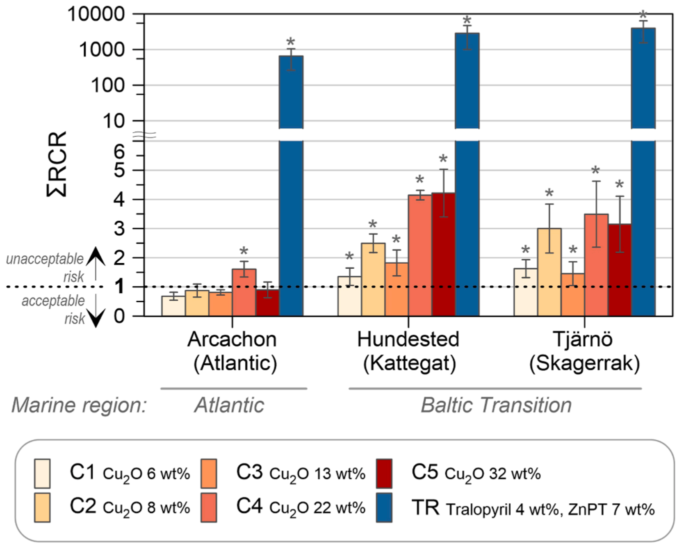

Fig. 4. Summed risk characterization ratios ($\sum$RCR) for the biocide-containing coatings, modelled for either the Atlantic (Arcachon) or Baltic Transition (Hundested and Tjärnö) marine regions based on the XRF average release rates of biocides and substances of concern for the time interval T2-T3. Error bars represent the RCRs calculated based on only the lower or only the upper 95% confidence estimates of the release rates. Asterisks show RCRs significantly above 1. Note that the scale on the y-axis is logarithmic after the break at 6. No $\sum$RCR was calculated for coating SI as it does not contain any biocides or substances of concern.

Exceptionally elevated risk ratios were determined for all marine regions for coating TR. The highest values were observed for the Baltic Transition region, with $\sum$RCR reaching 2850 and 3958 when using release rates from Hundested and Tjärnö, respectively. The lowest $\sum$RCR, albeit still elevated at 654, was determined for the Atlantic marine region. The $\sum$RCRs for coating TR were derived based on the release rates of three substances: Tralopyril, Zn and ZnPT (expressed in CuPT equivalents). Since the calculation is conducted in a stepwise manner, beginning with individual RCR calculations for each substance, it is possible to evaluate the contribution of each to the final $\sum$RCR of the modelled marinas and identify the primary driver of the overall risk. In the Baltic Transition region, tralopyril accounted for an average of 84% and 87% of the $\sum$RCR across the 17 modelled marinas, based on Hundested and Tjärnö release rates, respectively. Similarly, for the 47 Atlantic marinas, the contribution from tralopyril was 85% on average. The remaining portion of the $\sum$RCR was mainly attributed to ZnPT, as Zn contributed $\leq 0.02\%$ in all regions. Despite tralopyril being the major contributor to the $\sum$RCR, it is important to note that ZnPT emissions alone resulted in unacceptable risks, with RCRs of 100 (Atlantic), 611 (Baltic Transition, Hundested) and 681 (Baltic Transition, Tjärnö).

The PNEC values for the two organic biocides used in the ECHA emission scenario documents were derived using high assessment factors (100) to account for uncertainties in the ecotoxicity data [11]. However, even when the ERA was repeated using PNEC values without assessment factors, the resulting $\sum$RCRs still exceeded the threshold of 1. Theoretically, to meet ERA requirements in the least stringent region (Atlantic), a product emitting only tralopyril would need a release rate ≤ 0.0062 µg cm^{-2} d^{-1}. This is nearly 600 times lower than the release rate entered into the emission scenario for the Atlantic region here (3.5 µg cm^{-2} d^{-1}). For comparison, the release rate of tralopyril for the studied product was also estimated using a mass-balance calculation method (ISO 10890) [38], which is approved by ECHA to derive release rates for the ERA [27]. This method calculates the average release rate over the coating's lifetime based on parameters such as biocidal content, recommended dry film thickness and specified lifetime. Using manufacturer-provided data and assuming a 12-month lifetime, the calculated release rate was 2.8 $\mu g\ cm^{-2}\ d^{-1}$, closely aligning with the field-based value of this study. These findings suggest that the product would not pass the ERA regardless of release rate method used. Coating TR was introduced to the Danish and French markets prior to the implementation of the ERA procedure applied here. As a result, it has not yet undergone a formal ERA for market placement. Nevertheless, it currently remains available for purchase while its approval status is pending.

3.3. Ecotoxicological testing

3.3.1. Comparison of release rates between field and laboratory

Laboratory release rates of Cu and Zn from coatings C1–C5 were determined through chemical analysis of leachates obtained during 24-hour leaching experiments (**Table SI4**). The panels had been previously exposed for two months at the Arcachon Bay site and leachate results can thus be compared to the average release rates during their field deployment (T0–T2) (Table 4). Copper release rates under laboratory conditions were, on average, 5.4 times lower than those observed in the field, with statistically significant differences for all five coatings. In contrast, zinc release rates were only 1.7 times lower on average, and significant differences were observed for fewer coatings (C2, C3, and C5). Notably, for coating C5, the zinc release rate in the laboratory exceeded that observed in the field.

The discrepancy between Cu and Zn may be attributed to the differing solubility of their paint pigments: ZnO has lower solubility than Cu_2O in seawater, which typically results in preferential Cu release during exposure [25]. This trend was confirmed for the T2 panels, where a greater proportion of Cu was released from C1-C5 relative to the initial content in these coatings during field exposure. However, under laboratory conditions, the opposite was observed: Zn was released preferentially over Cu. One possible explanation is the influence of artificial seawater properties on dissolution behavior, potentially favoring Zn over Cu. Another hypothesis is that field exposure led to the formation of distinct pigment dissolution fronts, with a ZnO-rich layer forming above the Cu_2O layer [39]. During laboratory leaching, this ZnO front may have been more readily accessible compared to the underlying Cu_2O layer for the short duration (24 h) of the leaching step.

The overall lower release rates during laboratory leaching may lead to an underestimation of ecotoxicity in the conducted bioassays compared to real-world conditions. Oppositely, the use of artificial seawater devoid of dissolved organic carbon could result in an overestimation of toxicity, as organic matter in the environment can bind metals such as copper and reduce their freely available concentrations [40]. Moreover, the distinct differences in copper release rates observed among coatings in the field were not reproduced under laboratory conditions (Table 4). The short-term leaching in the laboratory therefore seems to mask formulation-specific differences that may only emerge over longer exposure periods. Taken together, these factors underscore the need for cautious interpretation of the ecotoxicity results presented in this study.

Table 4
Average laboratory and field release rates (± 1 standard deviation, n = 3 panels) of copper and zinc for coatings C1-C5 exposed in Arcachon Bay for the panel set retrieved at T2.

	Copper release rates ($\mu g/cm^2/day$)		Zinc release rates ($\mu g/cm^2/day$)	
Coating	Field (Arcachon Bay, T0-T2)	Laboratory (24 h leaching in ASW)	Field (Arcachon Bay, T0-T2)	Laboratory (24 h leaching in ASW)
C1	6.1 (± 0.3)	2.4 (± 0.4)	6.1 (± 0.8)	5.5 (± 1.7)
C2	8.5 (± 1.4)	2.6 (± 1.4)	16.2 (± 4.3)	4.8 (± 2.5)
C3	10.1 (± 1.1)	2.1 (± 0.9)	5.0 (± 0.4)	1.9 (± 0.8)
C4	24.9 (± 2.0)	2.1 (± 0.3)	12.2 (± 2.3)	10.8 (± 1.3)
C5	26.7 (± 5.0)	6.1 (± 1.6)	4.1 (± 1.8)	12.0 (± 0.6)

3.3.2. Ecotoxicological assay results

Although leachates may not be fully representative of field conditions, as discussed previously, they can nonetheless inform on the comparative toxicity between products. This is particularly relevant for coating SI whose potential environmental risk cannot be assessed using the emission scenarios of the ERA for antifouling products due to its lack of biocides and substances of concern.

Four acute bioassays were performed for the seven different coatings (Fig. 5**, Table SI5**). For each test, significant differences were evaluated against an unexposed (T0) PVC panel. The two coatings exhibiting the highest ecotoxicity, with significant differences compared to PVC (T0) in all four tests, were coatings C5 and TR, with the latter demonstrating particularly high acute toxicity with responses at or close to 100% across all tests. Exempting coating C5, the other four copper-based coatings C1-C4 generally tested similarly against the PVC (T0) for all tests except the fish larvae assay. The lack of differing results between C1-C4 could be due to their similar leachate concentrations of copper which were found to poorly reflect the distinct differences in copper release rates observed in the field (Table 4).

Meanwhile, the coatings with the lowest apparent ecotoxicity, showing significant differences compared to PVC (T0) in only one of the bioassays, were: C2 (bacterial assay), C4 (bacterial assay) and the unexposed SI (T0) coating (microcrustacean assay). Notably, leachates from SI panels exposed in the field for six months (T4) showed no significant differences compared to PVC (T0) in any of the tests, indicating minimal ecotoxicity after prolonged immersion. While this study evaluated a six month pre-immersion period, previous studies have shown that 1–2 months of immersion may be sufficient to reduce or eliminate ecotoxicological effects of leachates from silicone-based coatings [30, 31], suggesting that the release of harmful substances may be largely limited to the initial immersion phase.

Chemical analysis of the leachates revealed no significant difference in dissolved concentrations of Cu, Zn, As or Cr for SI (T0) or SI (T6) leachates in comparison with blanks (ASW) or PVC (T0) leachates (**Table SI4**). However, silicon (Si) concentrations were significantly elevated in SI (T0) leachates (1.3 mg/L) compared to ASW (0.45 mg/L) and PVC (T0) leachates (0.54 mg/L). The higher Si concentration could be due to the release of silicone oils, known to be release from silicone-based foul-release coatings [17]. Elevated Si concentrations were also observed in T4 leachates from both the SI coating (1.4 mg/L) and PVC panels (1.2 mg/L). While the PVC panel at T4 was heavily colonized by fouling organisms (similarly to the control coating, Fig. 2) which likely contributed to the Si enrichment of the leachate, the SI coating was virtually free of fouling at T4 (Fig. 2), suggesting that Si emissions were still occurring from the coating itself after six months exposure. The similar Si concentrations observed in leachates from SI panels at both T0 and T4 suggest a consistent release of silicone substances from the coating matrix which thus cannot account for the observed reduction in toxicity between the two timepoints. Further ecotoxicological testing and chemical characterization of initially released substances would be necessary to identify less toxic alternatives. Despite these uncertainties, the ecotoxicological results here suggest that the SI coating was the least acutely toxic of the evaluated products, especially after extended field exposure. This aligns with previous studies indicating that silicone-based coatings generally exhibit lower toxicity compared to copper-based formulations [17].

3.3.3. RCR of leachates

The lowest derived $\sum RCR_{leachate}$ was obtained for coating SI with values of 3.8 and 4.0 for the T0 and T4 panel leachates, respectively. Oppositely, the highest derived $\sum RCR_{leachate}$ was obtained for coating TR at 10,349 (**Table SI4**). For this coating, concentrations of organic transformation compounds in the leachates exceeded those of parent compounds. The hydrolysis of ZnPT (into PT and PT_2) and tralopyril

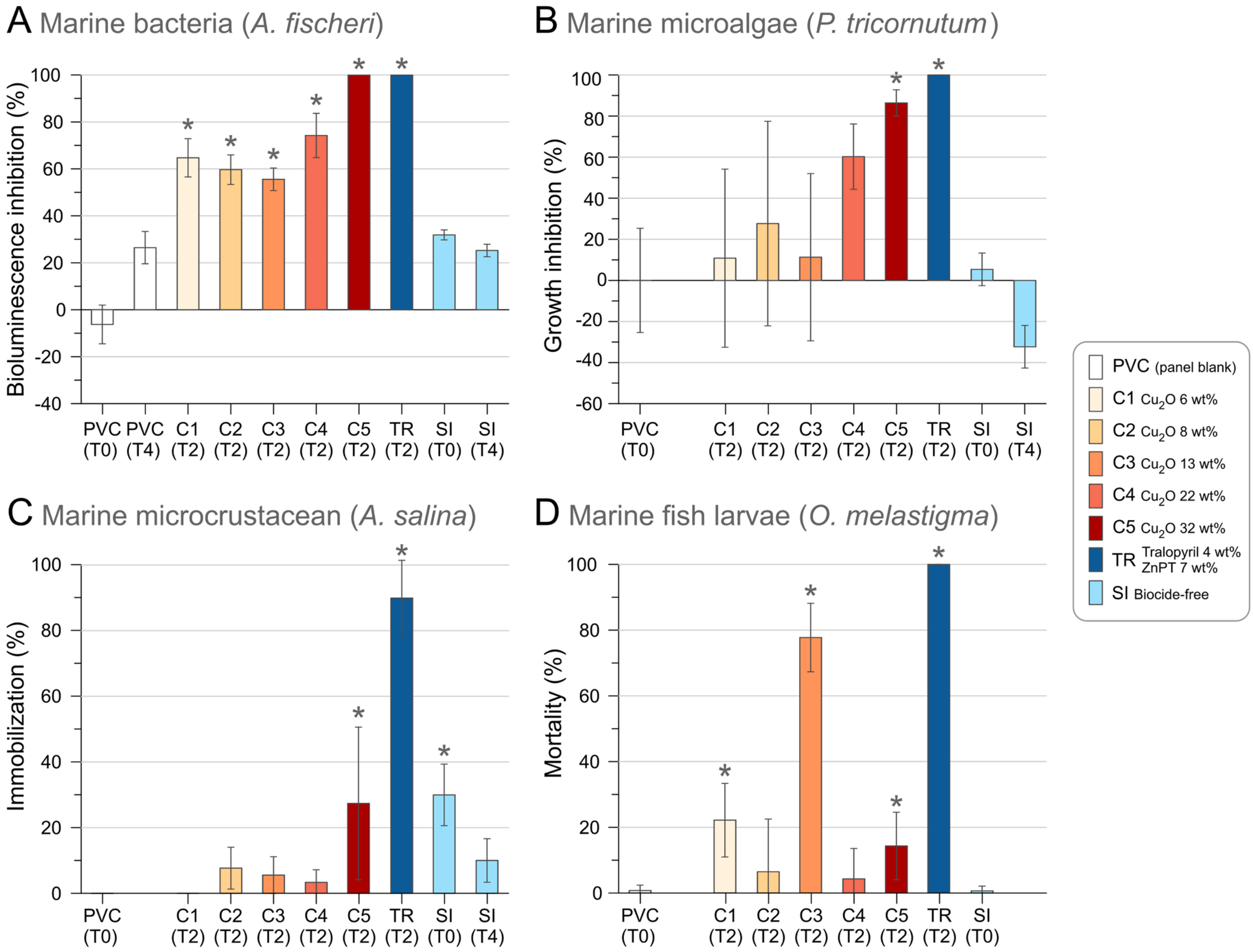

Fig. 5. Average ecotoxicity assay responses for marine bacteria (A), marine microalgae (B), marine microcrustacean (C) and marine fish larvae (D). Error bars show the 95% confidence interval. Asterisks show leachates resulting in significantly different responses compared to PVC (T0), i.e. an uncoated and unexposed PVC panel.

(into BCCPCA) is rapid, typically on the order of minutes to hours and can occur also during sample analysis [13, 32]. For pyrithiones, the total risk is accounted for by the $\sum RCR_{leachate}$ calculated from the total pyrithione concentration (PT_{Total}, **Table SI4**). The contribution of the bromine-based compounds is based on the parent compound tralopyril and the degradation product BCCPCA. Analytical results showed that ≥ 99.9% of the total bromine in the leachate was present as the more benign degradation compound BCCPCA, indicating a high rate of transformation. Consequently, the contribution of tralopyril and BCCPCA to the $\sum RCR_{leachate}$ was only 20%, compared to 80% from PT_{Total}. As it unclear whether similar degradation occurred during ecotoxicological testing and as the contribution of Zn could not be evaluated for this leachate due to insufficient sample volumes, it is difficult to conclude on the contribution of each substance to the toxicity of TR leachates with any certainty. Nevertheless, this coating exhibited the highest $\sum RCR_{leachate}$ and consistently demonstrated the strongest toxic responses across the four bioassays.

Significant relationships between $\sum RCR_{leachate}$ and the observed toxic effects of coating leachates were observed for the bacteria (r^2=0.84, p = 0.0013) and microalgae (r^2=0.73, p = 0.0068) assays, but not for the microcrustacean (r^2=0.20, p = 0.2665) and fish larvae assays (r^2=0.31, p = 0.1913) (**Figure SI4**). The latter two assays were found to be the least sensitive, exhibiting the lowest overall toxic responses which may explain the lack of statistical significance. Nonetheless, when the average toxic response across all four bioassays was considered, a significant correlation with $\sum RCR_{leachate}$ was observed (r^2=0.72, p = 0.0079) (Fig. 6). These findings suggest that the concentrations of active substances, their transformation products and zinc are important explanatory factors for the observed toxicity.

3.3.4. Additional test results for coating TR

The elevated toxicity observed for coating TR prompted additional testing of this product, alongside coating C3 for comparison. Leachates from panels retrieved at all timepoints were evaluated using the bacterial assay and all were found to be significantly different compared to PVC (T0) for both coatings. However, while the toxicity of C3 varied over time, likely reflecting changes in copper and zinc releases, the ecotoxicological response for coating TR was consistently higher, with ≥ 98% bioluminescence inhibition across all tested timepoints (Fig. 7). The difference in toxicity between the two coatings is further emphasized by the EC50-values derived for the bacterial assay on T2 leachates, where the EC50 of TR (23% leachate concentration) was nearly three times lower than that of C3 (68%). The EC50-values derived for coating TR for the other ecotoxicological tests were similarly low at 23% for the microalgae assay and 16% for the fish larvae assay (**Figure SI4**).

To further investigate the persistence of toxicity, bacterial assays were performed on TR (T2) leachates following 6 and 12 h of solar irradiation during a clear day in July. After 6 h of exposure, toxicity decreased but remained significantly higher than the PVC control (T0) (Fig. 7). Extending irradiation to 12 h did not result in any further

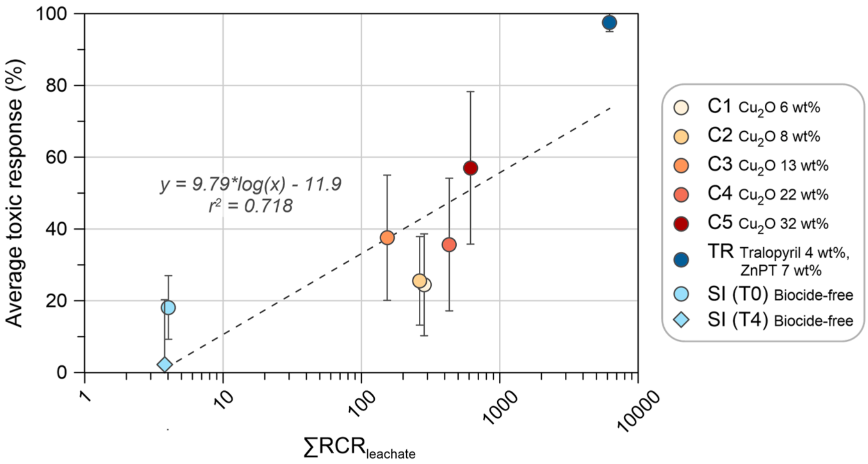

Fig. 6. Linear regression analysis between $\sum RCR_{leachate}$ and the average toxic effects (in %) of coating leachates across the four bioassays. Note that $\sum RCR_{leachate}$ was log-transformed. Error bars show the standard error (n = 4 assays, except for SI (T4) where n = 3 assays).

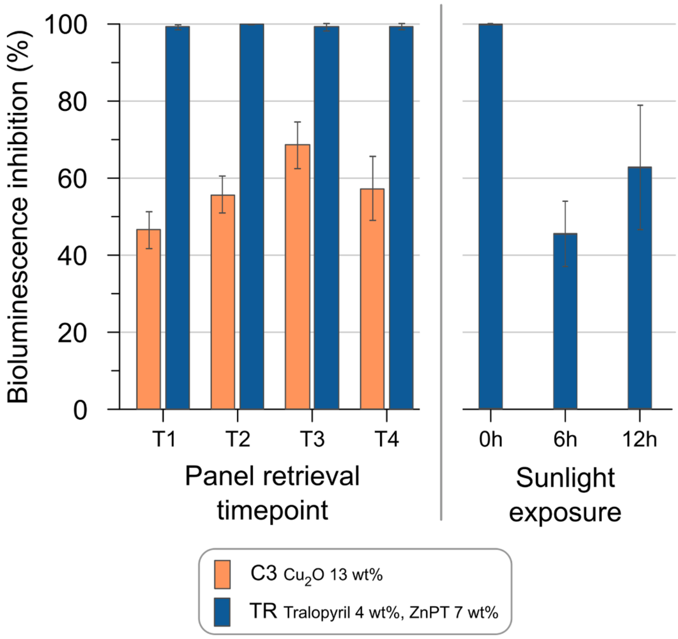

Fig. 7. Average ecotoxicological response of marine bacteria (*Aliivibrio fischeri*) for the extended testing of coatings C3 and TR. Leachates were obtained from panels retrieved after different field exposure durations: T1 (2 weeks), T2 (2 months), T3 (4 months), and T4 (6 months), with n = 9 bacterial assays for each timepoint. For coating TR, the effect of sunlight exposure on toxicity was evaluated using T2 leachates subjected to 0 h (n = 6 assays), 6 h (n = 6), and 12 h (n = 3) of solar irradiation. Error bars show the 95% confidence interval.

reduction, with inhibition stabilizing around 50–60%, a level comparable to that observed for the copper-based coatings C1–C4 (Fig. 5A). These results suggest that, despite favorable conditions for photodegradation, which are unlikely to occur consistently during actual use, particularly in shaded areas beneath submerged hulls, emissions from coating TR may still pose an environmental risk. This persistent toxicity could be attributed to the presence of non-degraded parent compounds, degradation products holding harmful properties, the presence of other additives in the formulation or a synergic combination of these factors. To more clearly identify the underlying cause of the observed toxicity, future studies would require a chemical characterization of the leachates, ideally using targeted and non-targeted analytical approaches, to identify degradation products and additive-related compounds, coupled with toxicity identification evaluations (TIE) to link specific chemical species to observed toxic effects [41]. Regardless of the underlying mechanism, the consistently high toxicity observed throughout this study indicates that coating TR is not an environmentally friendly alternative to copper-based coatings.

3.4. *Sustainability ranking*

The results from the three evaluation approaches applied to the seven coatings (Table 5) were qualitatively assessed to determine how effectively each product balances antifouling efficacy with environmental impact. Environmental sustainability was evaluated using two approaches, each with inherent limitations. The ERA does not capture mixture toxicity as it focuses only on emissions of biocides and zinc, but it is able to reflect in-use conditions through the incorporation of site-specific field-measured release rates. In contrast, the ecotoxicity testing reflects the toxicity of the complete leachate mixture but was conducted using leachates from only one site and under laboratory conditions that were not fully representative of in-use exposure. Consequently, the two approaches were applied in a complementary manner: ecotoxicity results were used primarily to support the ranking of the biocide-free coating SI, whereas the ERA was considered more appropriate for evaluating the environmental performance of the biocidal coatings. It is important to note that the sustainability assessment presented here is constrained to the end-points included in this work and does not encompass all possible environmental concerns (e.g. chronic toxicity or species-specific sensitivity). The results should therefore be interpreted within the scope of the parameters assessed, rather than as an absolute determination of overall environmental sustainability. The efficacy evaluation showed that, when averaged across the three study sites, the antifouling performance of most coatings was comparable (Fig. 2B). Site-specific results revealed nonetheless that the copper-free coatings SI and TR significantly outperformed the copper-based coatings in certain locations. However, the environmental assessments revealed stark differences between the two: the biocide-free silicone coating SI exhibited minimal toxicity, with significant acute effects observed for only one of the four assays. Given that these effects were no longer observed at the six months timepoint (T4), coating SI thus had the lowest average toxicity of all studied products. It should be acknowledged, however, that this study evaluated only one commercial silicone foul-release coating, and both antifouling performance and ecological impact may vary among silicone coatings depending on their specific formulation; the conclusions drawn here should therefore not be generalized to all silicone-based coatings. In addition, the extent to which silicone oils leach from these coatings and how long they persist

Table 5
Summary of results from the three evaluations performed on the antifouling coatings. Note that SI T4 ecotoxicity results (*) are based on three (and not four) bioassays.

Coating	Coating type	Efficacy assessment		Environmental sustainability assessment			
				Environmental risk modelling		Ecotoxicity testing	
		Assessed timepoint	Average FR_w (%), n = 3 sites	Assessed interval	Average RCR, n = 3 sites	Assessed timepoint	Average toxic response (%), n = 4 bioassays
C1	Copper (6.1 wt% Cu_2O)	T3	14.2	T2-T3	1.2	T2	24.4
C2	Copper (8.5 wt% Cu_2O)	T3	20.9	T2-T3	2.1	T2	25.5
C3	Copper (13 wt% Cu_2O)	T3	16.0	T2-T3	1.4	T2	37.6
C4	Copper (22.0 wt% Cu_2O)	T3	13.8	T2-T3	3.1	T2	35.7
C5	Copper (31.9 wt% Cu_2O)	T3	13.1	T2-T3	2.8	T2	57.0
TR	Tralopyril (4.0 wt%) and ZnPT (7.0 wt%)	T3	7.4	T2-T3	2487	T2	97.5
SI	Silicone (biocide-free)	T3	3.6	-	-	T0 T4*	18.1 2.2*

in marine environments is still largely unknown, highlighting a key knowledge gap for future sustainability assessments.

In contrast to coating SI, coating TR demonstrated consistently high toxicity in the bioassays and a high environmental risk in the ERA across all timepoints, indicating continuous harmful emissions over the whole product lifetime. Coating TR is therefore identified as the least sustainable option even though it is marketed on the manufacturer's website as "formulated to be safe for the environment, adhering to modern environmental standards and regulations" [42]. Although this product is currently only available in Denmark and France pending formal approval, its existence is surprising given that the ECHA assessment report for its main active substance, tralopyril, states that it would not be used in products intended for boats < 25 m [11]. Even though the MAMPEC model used in the ERA accounts for environmental degradation, tralopyril would need to be released at exceedingly low rates (e.g. $\leq 0.0062\ \mu g\ cm^{-2}\ d^{-1}$ in the Atlantic region and $\leq 0.0014\ \mu g\ cm^{-2}\ d^{-1}$ in the Baltic Transition) to meet regulatory thresholds. At such minimal releases, it is questionable whether a tralopyril-based coating would retain any meaningful antifouling efficacy.

Finally, for the copper-based coatings, four out of five products passed the ERA modeling of the Atlantic site (Arcachon Bay, France). However, the use of copper-containing coatings needs to be reduced in Arcachon Bay as copper concentrations in port sediments have significantly increased over the last decade, surpassing ecological effect thresholds [43]. Copper levels in oysters in the bay have also been rising over the past 40 years, with isotopic analyses identifying antifouling coatings as the primary source [44]. Meanwhile, it has been demonstrated that high copper loadings from antifouling coatings are not required to achieve acceptable antifouling performance in this location [8]. None of the copper-based coating passed the environmental risk assessment for the Baltic Transition sites (Hundested, Denmark and Tjärnö, Sweden), indicating that they cannot be considered environmentally sustainable across all the studied regions. Comparison of environmental risk ($\sum$RCR) and average toxicity response in the ecotoxicity tests showed that coatings with lower copper content generally present a lower environmental impact (Table 5). In terms of efficacy, all copper coatings (except for C2 at Hundested), performed similarly. This further confirms that increasing the copper release does not provide additional benefits and that optimizing formulations toward reduced copper content represents a more sustainable pathway to reduce environmental impact while maintaining effective protection against fouling.

The three studied locations represent temperate coastal systems with comparable seasonal patterns. As such, the conclusions may not be directly transferable to regions with substantially different environmental conditions, such as tropical or subtropical waters with year-round fouling pressure, areas with markedly different hydrodynamics or locations dominated by distinct biofouling communities. Nevertheless, the broadly consistent results obtained across the Kattegat, Skagerrak and Atlantic sites with respect to both coating efficacy and environmental impact suggest that the general patterns observed here are likely to be transferable across similar temperate coastal regions. Additionally, a previous study in which coating SI was exposed in Tjärnö (Skagerrak), Malmö (Kattegat) and Askö (Baltic Sea) for up to one year demonstrated high efficacy for this coating, comparable or better to that of Cu_2O-based coatings for ships which also contained cobiocides [17]. This suggests that the conclusions presented here may also extend to lower-salinity environments such as the Baltic Sea, and that the performance of coating SI appears to be robust across multiple years and environmental settings.

At present, no single antifouling solution can be considered universally optimal. Although biocide-free silicone foul-release coatings constitute a promising alternative to conventional biocidal systems, their adoption among leisure boat owners remains limited. Lagerström et al. (2022) identified several key barriers to uptake, including end-user perceptions and attitudes, with boat owners express uncertainty regarding the long-term performance and durability of silicone systems compared to traditional biocidal paints [17]. Practical constraints further hinder adoption, such as the need for more careful surface preparation and concerns about performance on low-speed or infrequently used vessels. Moreover, silicone foul-release coatings are only suitable under specific operational conditions, notably for vessels that do not undergo beaching and are not moored in drying harbors given their low mechanical strength and susceptibility to mechanical damage [45]. In addition, existing regulatory and market structures continue to favor familiar copper-based products. While the new ERA procedure under the BPR may, in some cases, lead to the removal of high-leaching copper products from the market in certain marine regions (as illustrated here for coatings assessed under Baltic Transition conditions), it offers little regulatory leverage for sensitive Atlantic sites. Most copper-based products in this study meet the ERA criteria when evaluated using the environmental parameters of the Atlantic region, highlighting the limitation of the ERA procedure and indicating that the protection of areas such as the marine nature park of Arcachon Bay requires additional measures. In such areas, local management measures could promote a progressive transition toward more sustainable biocide-free coatings where appropriate, while restricting the use of biocidal products to moderate biocide concentrations and to cases where biocide-free alternatives are demonstrably inappropriate. The effectiveness of regulatory local instruments is contingent upon their support by coherent national and international regulatory frameworks that ensure their legal robustness and enforceability.

4. Conclusion

Drawing on the three complementary evaluation approaches (efficacy testing, environmental risk modeling and ecotoxicological assays), the biocide-free silicone coating (SI) emerged as the most sustainable option. It demonstrated strong antifouling performance across all study sites with minimal acute toxicity, particularly after extended field exposure. The copper-based coatings followed in the ranking, with formulations containing lower concentrations of cuprous oxide generally exhibiting reduced environmental impact while maintaining comparable efficacy to higher-copper products. However, copper-based coatings were only able to meet environmental risk thresholds in one of the sites (Arcachon Bay). The tralopyril-based coating (TR) ranked lowest due to its consistently high toxicity across all bioassays and extremely elevated risk characterization ratios in the ERA, which exceeded acceptable limits by several orders of magnitude in all modeled marine regions. These findings challenge the perception of such 'copper-free' products as inherently safer and highlight the importance of environmental impact assessments to guide sustainable antifouling product selection. While environmental risk assessment models such as MAMPEC may be used to evaluate biocide-containing products, ecotoxicological assays remain the only method for assessing biocide-free coatings. However, this study revealed discrepancies between laboratory and field leaching behavior, indicating that further research is needed to develop leaching protocols that can adequately reflect in-use conditions. Finally, while the ecotoxicity tests in this study focused on acute toxicity, future work should also investigate potential long-term exposure effects across a wider range of species.

Environmental implication

Most antifouling coatings for leisure boats contain biocides that are hazardous due to their toxicity to non-target marine organisms and their persistence in sediments. Their use poses ecological risks in marinas, threatening biodiversity and contaminating food webs. This study ranks the sustainability of seven commercial coatings by integrating antifouling efficacy, environmental risk modeling and ecotoxicological testing. By identifying biocide-free silicone coatings as the most sustainable option and revealing the severe risks of tralopyril-containing products, the work provides evidence to guide informed decision-making and regulatory development aimed at reducing harmful emissions and promoting environmentally safer antifouling strategies.

CRediT authorship contribution statement

Rémy Boisserie-Gimenez: Writing – review & editing, Investigation. **Laura Veensalu:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis. **Ann I. Larsson:** Writing – review & editing, Methodology, Investigation, Formal analysis. **Lola Racofier:** Writing – review & editing, Investigation. **Jérôme Cachot:** Writing – review & editing, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Kai Bester:** Writing – review & editing, Methodology. **Florane Le Bihanic:** Writing – review & editing, Resources, Methodology, Investigation. **Maria Lagerström:** Writing – original draft, Visualization, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Xavier Cousin:** Writing – review & editing, Resources. **Gwendolina Limon:** Writing – review & editing, Methodology, Investigation. **Marcel Butschle:** Writing – review & editing, Methodology, Investigation.

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. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2026.141600.

Data availability

All data is shared in the supplementary material.

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