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Sediments indicate the continued use of banned antifouling compounds

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ABSTRACT

Antifouling paints are widely used to avoid organisms settling on boat hulls. The active ingredients in the paints have differed over the years where lead, TBT, irgarol and diuron have been deemed too harmful to non-target organisms and subsequently been banned within the EU. Most of these compounds however are persistent in the environment and can cause problems long after they are deposited.

We have examined if present-day and banned substances used in antifouling paints can be found in sediments in a national park on the Swedish west coast. Sampled locations include waterways, natural harbours and small marinas for leisure crafts to investigate if number of visiting boats affect the concentration of antifouling compounds in sediments.

Few significant differences were found when comparing the different locations types, suggesting that overall boat presence is more important than specific mooring sites, however, several banned antifouling compounds were found in the surface sediments.

1. Introduction

Biofouling is the growth of unwanted organisms on submerged structures. On ships biofouling adds friction, which reduces speed and thereby increase fuel consumption and subsequently also emissions (Abbott et al., 2000). Effective antifouling methods are therefore needed, not just for the pure financial and environmental benefits of decreased fuel use but also to prevent introduction of non-indigenous species that might otherwise be transported on ship hulls (Dafforn et al., 2011).

Antifouling paints are currently the most commonly used technique to combat fouling organisms, and most of the commercially available paints are self-polishing and function by slowly releasing the active ingredient to create an unsuitable environment for settling (Buskens et al., 2013). The active ingredients in paints differ in type and concentration, although Cu is the most common (Thomas and Brooks, 2010; KemI, 2017). Zinc in the form of ZnO, is added for its polishing properties and is therefore not classified as an active ingredient. The use of active ingredients differs between countries where in Sweden Cu is the only allowed substance for leisure boats. However, former active ingredients like TBT (tributyltin) and the herbicide irgarol can still be found in the environment due to their relative persistence. In Sweden, both Cu and Zn have also been used as active ingredients in the form of Cu- and Zn pyrithiones.

1.1. TBT

TBT was commonly used in antifouling paints from the 1960s, but by the mid-1970s it was found to be responsible for malformations and population decline of the pacific oyster, *Crassostrea gigas*, in Arcachon, France (Alzieu et al., 1986). Further studies have shown that it has adverse effects on many different groups of organisms (Sousa et al., 2014). Due to its toxicity towards non-target organisms TBT was banned in the late 1980s on vessels smaller than 25 m (EC Directive 89/677/EEC, 1989), but it took until 2008 for the IMO (International Maritime Organization) member states to ratify a global ban on all ships.

TBT degrades slowly in the environment and can be stored in anoxic sediments for decades (Fent, 2006). The degradation products, DBT and MDT (di- and monobutyltin) are also persistent in sediments (Sarradin et al., 1995) and although less toxic than the parent compound, they are still a cause for environmental concern (Hoch, 2001).

1.2. Irgarol 1051/diuron

Herbicides have been added to antifouling paints to boost their efficacy, since some algae are tolerant to heavy metals such as Cu (Reed and Moffat, 1983). Irgarol and diuron both function by disrupting the electron transport by binding to photosystem II thereby inhibiting photosynthesis. Consequently, they affect all organisms that perform

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Fig. 1. Map of sampled locations. WW = waterway, NH = natural harbour.

photosynthesis and therefore also have negative effects on non-target organisms. A study on periphyton communities (Dahl and Blanck, 1996) showed effects on community level when subjected to irgarol concentrations occurring in Swedish coastal waters. Irgarol was later also found to have induced selection in periphyton resulting in more tolerant communities (Blanck et al., 2009). Additional studies have shown that irgarol decreases nematode abundance and shifts the community composition to more tolerant species (Hannachi et al., 2016) and lethal effects was shown by Gallucci et al. (2015) when exposing nematode assemblages to irgarol and diuron.

As of January 2016, the EU Commission decided not to continue the authorization of irgarol as an antifouling product (Commission Implementing Decision (EU) 2016/107) and there has not been any new permissions to use irgarol as an active ingredient in antifouling paint for leisure crafts in Sweden since the old permit expired in 2010 (KemI, 2017). Irgarol degrades slowly with a half-life of around 200 days (Zhang et al., 2008) and can have a long residence time in marine systems (Ranke, 2002).

Diuron on the other hand is short-lived with a half-life of 14 days in anaerobic sediments. However, diuron associated with paint particles showed no signs of degradation over 42 days in marine sediments (Thomas et al., 2003). Diuron was phased out during 2008 (Commission Decision, 2007/565/EC) and is a priority substance under the water framework directive (WFD). It has never been permitted as an active ingredient in antifouling paints for leisure boats in Sweden (KemI, 2017).

1.3. Metals

Heavy metals like Pb, Hg and Cu have been used in antifouling paints as they have a negative effect on growth rate and reproduction on fouling organisms but they can also be lethal in high concentrations. On a global scale, Cu is the most common heavy metal used as a biocide (Thomas and Brooks, 2010). Zinc is used mainly for its polishing properties, especially in paints with a low copper content, but also in sacrificial anodes. These anodes are often made up of alloys containing small amounts of Cd, which can also exist as an impurity in recycled Cu used in antifouling paint (Annex XV Restriction Report, 2015). Lead and mercury were previously used as active ingredients in antifouling paints but are no longer allowed, as is the case for other heavy metals like Cr and Co that were used as pigments.

An abundance of data exists describing the concentrations of antifouling compounds in commercial harbours and marinas (Boxall et al., 2000; Thomas et al., 2002; Di Landa et al., 2009), and many studies have been dedicated to describing the effects these compounds may have when released into the environment (Gammon et al., 2009; Gallucci et al., 2015; Eklund et al., 2016). Natural harbours and waterways for leisure boats are not as well studied with regard to the emissions from boating activities. Several natural harbours along the Swedish coast are subjected to a high degree of visiting boats during the summer months and could be viewed as seasonal marinas. In the Kosterhavet National Park, an archipelago on the Swedish west coast, boat tourism have been frequent since the 1970s (personal communication, Prof. Kerstin Johannesson) and it is still a very popular destination during summer with over 500,000 visitors annually, mainly from Sweden and Norway. The two main islands in the archipelago have a permanent population of just above 300 people, but largely the area is uninhabited.

This study aims to test if leisure boat activity has given rise to increased sediment concentrations of antifouling associated compounds compared to pre-industrial levels (NV Rapport 4914, 1999) and if concentrations in natural harbours can be related to number of visiting boats.

2. Method

Sediment cores were taken at ten locations, including four popular natural harbours (NH) and two small marinas (Fig. 1). None of the sampled sites are completely devoid of boat presence, but four additional locations were chosen as they had no known moorings. These locations could be considered as leisure craft waterways (WW).

Samples were collected during three occasions in July, August and September 2014. Cores were taken with a kayak-sampler and the top 2 cm was collected for analysis. Five cores were taken at each location to ensure sufficient material for analysis. Sediment from two cores was pooled for metal and organotin analysis and two other cores was pooled