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Recovery from TBT pollution in English Channel environments: A problem solved?

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ABSTRACT

Following recognition of effects in the 1980s, tributyltin (TBT) has been monitored at sites in the English Channel to evaluate the prognosis for biota – spanning the introduction of restrictions on TBT use on small boats and the recent phase-out on the global fleet. We describe how persistence and impact of TBT in clams *Scrobicularia plana* has changed during this period in Southampton Water and Poole Harbour. TBT contamination (and loss) in water, sediment and clams reflects the abundance and type of vessel activity: half-times in sediment (up to 8y in Poole, 33y in Southampton) are longest near commercial shipping. Recovery of clam populations – slowest in TBT-contaminated deposits – provides a useful biological measure of legislative efficacy in estuaries. On rocky shores, recovery from imposex in *Nucella lapillus* is evident at many sites but, near ports, is prolonged by shipping impacts, including sediment legacy, for example, in the Fal.

1. Introduction

The rise of the organotin (OT) 'problem', stemming from the use of tributyltin (TBT) and, to a lesser extent, triphenyltin (TPT), as biocides in antifouling (AF) preparations, has been extensively reviewed, as have the regulatory measures invoked to halt damage to vulnerable species (nb oysters, stenoglossan gastropods) and coastal ecosystems (Champ and Seligman, 1996; de Mora, 1996; Bray, 2006; Arai et al., 2009). Despite the large amount of research and statutory monitoring that has been conducted (Alzieu, 2000; Hawkins et al., 2002; Waldock et al., 1999), there are still significant gaps in our knowledge of the long-term behaviour and impact of TBT – notably, the recovery of ecosystems. Indeed although improvement is ultimately expected, globally, controversy exists as to whether, in the short-term, conditions may worsen in some parts of the world, particularly areas in Asia (Bray, 2006).

Assessing TBT pollution in sediments is particularly difficult, as it may be present in different forms, adsorbed or as paint chippings (Turner, 2010). Furthermore, unlike water – for which there is an Environmental Quality Standard (EQS) of 2 ng L^{-1} (0.8 ng L^{-1} as Sn) – there are no agreed quality criteria for sediments. The outlook for recovery of TBT – damaged ecosystems subjected to heavy boating/shipping activity (typified by the English Channel) therefore remains unclear, particularly in locations with persistent sedimentary sinks, and is exacerbated by ambiguity over the longevity of TBT residues. Published estimates of TBT half-times in sediment are highly variable – from weeks to years – depending on the nature of the benthic deposits. Persistence appears to be greatest in anoxic sediments: half-times in excess of ten years have been predicted where biological degradation is suppressed by poor mixing and lack of oxygenation (Maguire, 2000). However, there is a lack of published field evidence to confirm these predictions.

Studying such aspects of recovery presents policy makers and regulators with a unique opportunity to understand the efficacy of their actions on the TBT legacy. In this context, two crucial pieces of legislation are likely to have influenced recuperation from TBT pollution in the Channel (with similar measures being adopted in many countries worldwide). The first of these, in 1987 in the UK (1982 in France), banned the use of TBT on vessels <25 m (the leisure market), but allowed larger vessels (commercial and naval fleets) to continue usage, based on the premise that biocide leachates on large ships would be diluted, harmlessly, on the open seas. However, doubts over this assumption led to the second legislative initiative – based on IMO's proposal to remove OT-based AFs from the global fleet (beginning in 2003 with the recommendation to halt new application of OT-containing coatings, and the final adoption and ratification of the antifouling (AF) treaty in September 2008, which effectively enforced removal, or sealing, of all tin-based coatings http://www.imo.org/OurWork/Environment/AntifoulingSystems/Pages/Default.aspx). The latter was pre-empted by EU legislation in January 2008; elsewhere, sealing or removal was permissible until 2013 (to allow for the projected five-year life of a TBT coating newly applied in 2008).

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UK involvement in monitoring post-legislation change in TBT status has included statutory observations (Cefas, 1998, 2000; OSPAR, 2004, 2008; Gubbins et al., 2008) and *ad hoc* studies, such as this. Our aims were to compare TBT trends and impacts in estuarine and coastal areas in the English Channel, including ports and harbours hosting different quantities and types of vessel (leisure craft and commercial shipping). Given the partitioning behaviour of TBT in favour of particulates (Langston and Pope, 1995) and the anticipated persistence of TBT in this phase, we have examined long-term impacts on populations of the sediment-dwelling bivalve *Scrobicularia plana* to determine the efficacy of regulatory measures in achieving chemical and ecological improvements. This clam typically accounts for a significant proportion of the infaunal biomass and energy-flow in unpolluted estuaries bordering the north-eastern Atlantic (Casagranda and Boudouresque, 2005). However, as a deposit-feeder, *S. plana* is vulnerable to a number of sediment-associated contaminants (Ruiz et al., 1994): at heavily TBT-contaminated sites, typified by the Solent area of the English Channel, clam numbers appeared to decline considerably during the 1980s, as TBT concentrations peaked (Langston et al., 1990, 1994a; Langston and Burt, 1991). Intuitively, clam abundance should recover as TBT levels fall, following pollution control measures. The question is: to what extent and over what timescales are improvements accomplished?

Two areas on the Channel coast of the UK were singled out to address these issues – Poole Harbour and Southampton Water. Poole Harbour was originally selected for study in the 1980s as a possible worst-case situation with regard to TBT contamination (Langston et al., 1987). This large natural lagoon is utilised by several thousand leisure vessels (and a relatively small number of commercial ships). Because of the predominance of small craft, it was ideal for judging the effectiveness of the 1987 legislation prohibiting the use of TBT paints on boats <25 m. Monitoring of water, sediment, and *S. plana*, has taken place here at intervals over the last two decades. Comparable monitoring has been undertaken over similar timescales in Southampton Water (hosting commercial and naval ships, in addition to yachts).

To supplement our understanding of improvements in TBT status in estuarine habitats in the Channel area we also present data on imposex in dogwhelks *Nucella lapillus* from rocky shores, including time-series at previously-sampled sites in south-west England subjected to varying shipping and boating intensities. Imposex, the imposition of male characteristics (penis and vas deferens development) on females is initiated in *N. lapillus* by TBT concentrations as low as 0.5 ng L^{-1} (Bryan et al., 1986, 1987). Severe expression prevents egg-laying and, prior to TBT legislation, led to localised extinction of many UK south coast (Channel) populations. TBT-induced imposex in these marine snails remains the most specific and sensitive example of the threat of endocrine disrupting chemicals (EDC) on Channel ecosystems and helped inform the development of TBT legislation. The effect of these regulatory measures on reducing the threat of TBT should, by now, have taken effect in the Channel. Indeed, surprisingly rapid (10y) recovery of populations (for a species lacking a planktonic larval stage) has been described in *N. lapillus* at three UK locations, including SW England, by Colson and Hughes (2004). These authors suggest, from observations of genetic diversity in recolonizing individuals, that the migrant pool (crawling larvae and rafting individuals from adjacent populations) could be larger and more mobile than anticipated in organisms with such limited dispersal characteristics, thereby promoting revival of populations. To confirm the hypothesis that dogwhelk recovery is well underway and widespread in the region, we have examined trends in imposex in *N. lapillus* over the last quarter of a century at a number of sites in SW England, in the context of TBT contamination.

2. Materials and methods

2.1. Sampling sites: Poole and Southampton

2.1.1. Poole Harbour

The narrow harbour entrance and limited tidal range result in relatively restricted flushing of the primary basin (Fig. 1A). Similar features produce an even less energetic tidal regime in the secondary basins of Holes Bay and Lytchett Bay. As a consequence of the high density of leisure vessels and restricted water exchange, elevated TBT residues were encountered in various parts of the Harbour during the 1980s (Langston et al., 1987). Since boat-traffic is dominated by small craft, Poole is an ideal location to judge the effectiveness of the 1987 legislation. Water, surface sediments and, where available, sediment-dwelling clams *S. plana,* were sampled for TBT on various occasions between 1987 and 2009 at sites shown in Fig. 1A. Marinas and moorings are predominantly situated along the northern shoreline, between Lytchett Bay and the Harbour mouth. In contrast, the southern shoreline is relatively undeveloped. Numbers of 'adult' bivalves m^{-2} were estimated by counting their characteristic siphon marks on the mud surface. Quantitative biological sampling (30 cores, totalling 0.1 $m^2 \times 10$ cm deep, sieved at 0.5 mm) was also employed on occasions as a measure of juvenile bivalve recruitment success.

2.1.2. Southampton Water

Southampton lies at the confluence of the Test, Hamble and Itchen Estuaries (Fig. 1B) and harbours commercial shipping and warships (mainly in the Test Estuary and Southampton Water), together with thousands of yachts and other small craft (notably in the Hamble and, to a lesser extent, the Itchen Estuary). TBT sampling in water, sediments and *S. plana* from nine sites in these estuaries (Fig. 1B) was started in 1986, prior to TBT restrictions, when the waterway became one of the most heavily TBT-polluted areas of the UK. *S. plana* were sampled semi-quantitatively in timed searches, to estimate clam densities and size cohorts. An earlier baseline study of total clam numbers in timed searches was undertaken at some of these sites in 1978. Quantitative sampling of juvenile clams in sediment cores, described above for Poole Harbour, was also undertaken on most occasions.

Analytical protocols for OT in Poole and Southampton samples were comparable. Water (0.5 m sub-surface) was collected in 1L glass-stoppered, acid-washed glass bottles and acidified with 5 mL L^{-1} high purity HCl. Intertidal sediment samples were skimmed from the surface and returned to the laboratory on ice, sieved through 100 μm polypropylene mesh with 50% seawater and allowed to settle overnight before decanting off excess water. The resulting sediment slurry was well mixed and aliquots dispensed for OT analysis, determination of water and organic content (based on the weight loss of dried sediment after ashing for 6 h at 400 °C). *S. plana,* dug by hand or garden fork, were returned live to the laboratory and cleaned for 3 days (50% offshore seawater) prior to analysis. Shell length, and tissue wet and dry weights were recorded. OT analyses were performed on homogenised batches of six pooled *S. plana.*

Tributyltin (TBT) and dibutyltin (DBT) were determined by analysis of hexane-extractable forms of Sn, using graphite furnace atomic absorption (Langston et al., 1994b). Hexane extracts were shaken with 1 M NaOH to separate DBT from TBT. Reagent blanks were put through the same procedure. Detection limits for TBT and DBT in tissues and sediment, expressed as Sn on a dry weight basis, were 5–10 ng g^{-1} (as Sn) and for seawater, 0.2 ng L^{-1} (as Sn). The accuracy of analytical methods was established using reference materials and internal quality control checks, and corrected by the use of standard additions on all samples. Recovery of TBT from

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