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# Use of Antifouling Paints on Ship Hulls over Past Four Decades and Consequent Imposex: A Review

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**Abstract:** ***Review provides the historic stance, general overview of gastropod populations being served as biosensors and developments in imposex detection. Typically muricoid species belonging to genus Nucella and Thais have been found to be good bioindicators globally. Although up to seven (7) imposex developmental stages have been described based on organotin accumulation by an organism and resultant morphological expression (penis and vas deferens development in females) due to endocrine disruption and steroidal imbalance. From Pakistan phenomenon of imposex has been described in nine (9) species of meso and neogasrtropods. Imposex stages 1-4 and 4+ have been found in examined muricids, bursid and buccinid species which revealed the moderate contamination effects on gastropod populations found along the Pakistan coast. Some archaeogastropods from Japan and Pakistan have also been tested respectively for reproductive fitness due to possible contamination effects.***

**Keywords:** shipping traffic, antifouling paints, gastropods, endocrine disruption

## 1. Introduction

The phenomenon of imposex in gastropod species is globally recognized as a cheap, easily applied biological indicator test and marine gastropod species being served as biosensors have provided a guideline in assigning priorities for more rigorous chemical analysis and ecological hazards caused by these compounds in the areas subjected to boat trafficking [1-3].

Imposex disarray first observed by Blaber [4] in *Nucella lapillus* and later on Smith [5] first ever used the term imposex for superimposition of male sex characters on female gonochoristic gastropods, which in extreme cases results in the sterilization and premature death of the female gastropod. Imposex in gastropods is induced mainly by an organotin compound tributlytin (TBT) and also by its sister product triphenyltin (TPhT), which are, used as biocidal agents, catalysts and stabilizers for polyvinyl chloride polymers. TBT based antifouling paints are widely used to prevent the organisms such as barnacles, seaweeds and others from clinging to ships and other submerged structures such as buoys, boat hulls and mariculture cages etc. Whereas, TPhT is mainly used as broad-spectrum fungicide and also in antifouling paints usually in combination with TBT [6-7].

TBT paints have been marketed since 1960's but their use become widespread in the 1970's with the development of self polishing copolymer paints [8]. The compounds of TBT and TPhT can be degraded by solar radiation, bacterial biodegradation or biological decomposition to their metabolites, such as, dibutyltin (DBT), monobutyltin (MBT), diphenyltin (DPT), and monophenyltin (MPT), respectively [9-10]. The chemicals release from the paint into the water column and are finally accumulated in sediments present in and around harbours and along shipping lanes. This chemical in the water column or sediments are taken up by marine organisms and finally enters the food chain [11-14].

The application of TBT based antifouling paints was banned in the late 1980s in many countries. Legislation have been implemented to restrict the use of TBT in anti fouling paints [15-16] then finally a global ban on the use of TBT from 2003 to 2008 was imposed by the International Maritime Organization (IMO) to restrict and to remove all existing coatings from ship hulls [17].

## 2. Global Imposex Induction Reports and Principal Biosensor Species Used In Biomonitoring

Imposex/intersex induction reports from different regions have been documented during past four decades and marine gastropod species served for organotin contamination biomonitoring. TBT is a well known endocrine disruptor (EDC) and its uptake results in musculanization by upsetting the hormonal system in gastropods at very low concentrations [18-19]. Although in some populations of gastropods affected with TBT contamination, aphally in males (lack of penis and incomplete development of vas deferens) has been reported in some individuals [20]. Due to the extensive use of TBT containing antifouling paints the incidences of imposex have been recorded in more than 260 species of gastropods world wide excluding Antarctic region [21]. Some information pertaining global imposex induction reports and principal species has been tabulated as given in Table 1. However, from Pakistan phenomenon of imposex has been described in nine (9) species of meso and neogasrtropods. Imposex stages 1-4 and 4+ have been found in examined muricids, bursid, turrid and buccinid species; namely *Thais carinifera, T. rudolphi, T. bufo, T. hippocastanu,, T. tissoti, Morula granulata, Bursa granularis, Turricula javana* and *Babylonia spirata* which revealed the moderate contamination effects on gastropod populations found along the Pakistan coast [22-23].

### 2.1. Organotin Toxicity and Chemical Testing Practices

Later on the basis of toxicity chemical testing in tissue samples it was proven that imposex is related to TBT accumulation. Smith [24-25] found strong evidence that imposex is induced by leaching of TBT from antifouling paints used on boats and fish farm-fields. Bryan *et al.* [26] provided strong evidences that imposex in the Atlantic dog whelk, *N. lapillus* is associated with TBT contamination. These evidences were further strengthened by the laboratory experiments carried out by Bryan *et al.* [26] and Gibbs *et al.* [27]. They exposed the adult animals collected from unpolluted sites exposed to different concentration of TBT. TBT and TPhT are both considered as equally potent inducer of imposex and are accepted as the most toxic biocide ever introduced into the sea [15, 28-30]. In these experiments the bioaccumulation of tin by adult females was accompanied by increased development of imposex. The TBT has been primarily reported to cause imposex in prosobranch gastropods at concentration as low as 0.5-1 ng Sn/ $l^{-1}$ and can induce reproductive failure at high concentrations [26, 31]. In severe conditions due to accumulation of these compounds the reproductive capacity of the populations of the gastropod species is sometimes lowered to the point of no egg capsule formation and consequently leads to the absence of juveniles from the population [26]. At high concentrations typically above 5 ng/ l the females have a fully grown penis and a vas deferens comparable in size with those of mature males [26, 32]. In some species the vas deferens obstruct the vulva, causing a blockage of the oviduct [32]. This prevents the females from laying eggs, often resulting in the accumulation of eggs in the oviduct and the eventual rupture of the capsule gland and causes female sterility [31, 33-34]. The presence of other male sex organs such as the prostate, seminal vesicles and testes have been noted in some females affected by high TBT concentrations (Gibbs *et al.*, 1988; Tan, 1997) [27, 32].

### 2.2. Archaeogastropods Being Served As Biosensors

The development of ovo-testis or spermatogenesis in female gonads have been observed in muricid and buccinid neogastropods [27, 35-37]. In archaeogastropods the sexes are not identified by any superficial characteristics so morphological expression of TBT contamination is not likely. Only information on the effect of TBT on reproductive behavior of archaeogastropod is that on abalone species, *Haliotis madaka* and *H. gigantea.* TBT has so far been known to induce spermatogenesis (ovo-testis) and contraction of oocytes in females of archaeogastropod *H. gigantea* and *H. madaka* (abalone) at the TBT contaminated site and also in female exposed to different concentration of TBT in laboratory experiments in Japan. Beside that in Japan decline in the population of *H. madaka* was also observed at sites contaminated with organotin compounds [38-40]. Similarly histological studies were carried out from Pakistan to examine the possible correlation between reproductive fitness of two archaeogastropod species, namely *Turbo coronatus* and *Monodonta canalifera.* In both species ovo-testis development or ovarian spermatogenesis was not evident. Spawning individuals of both sexes were found throughout the year at shipping vicinity area and the reference site but spawning appeared to be better synchronized at cleaner site [22-23].

Use of antifouling paints on ship hulls was started in 1960 and remains widespread over four decades. Consequent global problem of "imposex" in marine gastropods was stay behind a matter of concern worldwide. This article reviews the part of biosensor marine gastropod species in detection of organotins (OTC) contamination by means of serving as a cheap and easy bio-tool world over from more than four decades and to date for screening and reliable testing of presence of OTC in ports, marinas and adjoining natural environment.

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**Table 1:** Global imposex/intersex induction reports from different regions and principal marine gastropod species served for organotin contamination biomonitoring

| *Country/ Region* | *Species* | *Reports provided by* |
|---|---|---|
| U. K | *Nucella lapillus Urosalpinx cineraria* | Blaber (1970), Smith (1971, 1980, 1981) Bryan *et al.* (1986) and Gibbs *et al.* (1988, 1991, 1999) |
| Germany | *N. lapillus*<br>*Littorina littoria*<br>*Trivia arctica*<br>*T. monacha*<br>*Ocinebrina aciculata*<br>*O. erinacea*<br>*Hinia reticulata* | Stroben *et al.*, (1992); Fioroni *et al.*, (1992); Bauer *et al.* (1995); Oehlmann *et al.* (1991, 1996, 2000) |
| Canada | *Nucella emarginata*<br>*N. lamellosa*<br>*N. canaliculata*<br>*N. lima*<br>*Searlesia dira*<br>*Amphissa columbiana Neptunea phoenecia*<br>*Colus halli* | Bright & Ellis, (1990). |
| Spain | *N. lapillus* | Barrerio *et al.* (1999) |
| Malta | *Hexaplex trunculus* | Axiak *et al.* (2003) |
| | | |
| | | |
| Australia | *Thais orbita*<br>*Morula granulata* | Foale, (1993); Wilson *et al.* (1993); Reitsema & Spickett, (1999); Gibson & Wilson, (2003). |
| South East Asia (Singapore, Malaysia and Indonesia) | *Thais luteostoma*<br>*T. bitubercularis*<br>*T. clavigera*<br>*T. jubilaea*<br>*T. gradata*<br>*Morula musiva*<br>*Cronia margariticola Drupella rugosa*<br>*Naquetia capucina Chicoreus capucinus* | Ellis & Pattisina, (1990); Tan, (1997; 1999). |
| India | *Cronia konkanensis*<br>*B. spirata*<br>*Thais bufo*<br>*T. rudolphi,*<br>*T. tissoti*<br>*Ocenebra bombayana Gyrineum natator* | Vishwakiran and Anil (1999); Vishwakiran *et al.* (2006); Murugan *et al.* (2006). |
| Israel | *Stramonita haemastoma*<br>*H. trunculus* | Rilov *et al.* (2000) |
| Hong Kong and Taiwan | *T. clavigera*<br>*T. rufotincta* | (Hung *et al.*, 2001; Cheng & Liu, 2004) |
| Japan | *T. clavigera*<br>*Babylonia japonica* | Horiguchi *et al.* (2001, 2006). |
| Thailand | *Chicoreus capucinus*<br>*T. distinguenda*<br>*T. bitubercularis*<br>*T. rufotincta*<br>*Morula musiva*<br>*M. granulata*<br>*M. margariticola* | Bech (2002 a, b) |
| Brazil | *Thais haemastoma*<br>*T. deltoidae*<br>*T. rustica* | Fernandez *et al.* (2002); Camillo *et al.* (2004); Queiroz *et al.* (2007); Da Costa *et al.* (2008). |
| China | *T. clavigera* | Shi *et al.* (2005) |
| Pakistan | *Thais carinifera*<br>*T. rudolphi*<br>*T. bufo*<br>*T. hippocastanum*<br>*T. tissoti*<br>*Morula granulate*<br>*Turricula javana*<br>*Bursa granularis*<br>*Babylonia spirata* | Afsar (2009); Afsar *et al.* (2010, 2012, 2013, 2014). |
| Portugal | *N. lapillus* | Oliveira *et al.* (2011) |