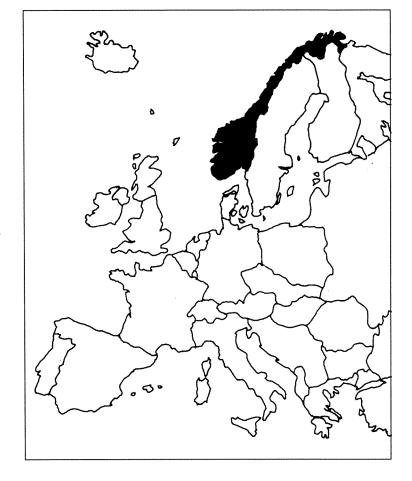
# **Report 693/97**

Client Norwegian State Pollution Control Authority

Contractor NIVA

Levels and environmental effects of TBT in marine organisms and sediments from the Norwegian coast.

A summary report





## Norwegian Institute for Water Research

# REPORT

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Abstract . Concentrations of TBT in sediments, blue mussels (Mytilus edulis) and dogwhelks (Nucella lapillus) from the Norwegian coast is summarised. The occurrence of imposex in dogwhelks along the coast is given. Most of the sediments contained concentrations below 100 ng/g d.w.. Sediment from 16 sites contained more than 1000 ng/g d.w. TBT whereas sediment from 17 sites contained between 100 and 1000 ng/g d.w of TBT. Maximum concentration of TBT was found in sediments from a navy harbour near the city of Bergen (103800 ng/g d.w.). Traces of TBT were, however, also found in sediments from remote, sparsely populated, areas along the coast. The concentrations of TBT in mussels from the Norwegian coast varied from less than 10 µg/kg wet weight to about 3000 µg/kg wet weight. The highest concentrations were found in and around harbours. Some degree of imposex occurred in all populations of dogwhelks except four stations in northern Norway (Finnmark). At about half of the total number of sites, a part of the female dogwhelk population was sterile. The concentration of TBT in the dogwhelks from the unaffected populations in Finnmark was below the detection limit (17 ng/g d.w.). The concentration of TBT in dogwhelks from affected populations was in the range 50-2676 ng/g d.w. It was observed a positive relation between concentration of TBT in dogwhelks and the degree of imposex. The results demonstrate that organotin compounds are present in most anthrophogenic influenced areas along the Norwegian coast. The major use of TBT in Norway is in antifouling agents on boats larger than 25 m. Total amount of TBT sold in Norway in 1994 was 36.7 t (23.4 t in antifouling paints for large boats). It is expected that a general reduction in the occurrence of organotin compounds along the Norwegian coast can only be achieved by enforcing restrictions on the use of such compounds on large vessels.

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Levels and environmental effects of TBT in marine organisms and sediments from the Norwegian coast.

A summary report

# **Preface**

The present report has been financed by the Norwegian State Pollution Control Authority (SFT). The report summarises available data on organotin (mainly TBT) along the Norwegian coast. The main purpose of the report is to summarise relevant national data as part of the International Maritime Organization's (IMO) effort to get a global overview of possible effects of antifouling agents in the marine environment. This will be part of the backgound information for an evaluation of the need to enforce further restrictions on the use of TBT in antifouling agents on ships. The report is also for use within the framework of OSPARCOM [Oslo and Paris Commissions and SIME (Working group on Concentrations, Ttrends and Effects of Substances in the Marine Environment]. Parts of the present report were submitted to SIME in 1996.

Most of the data presented are from reports within the State Pollution Monitoring Program of Norway, sponsored by SFT. The data on the concentration of organotin in dogwhelks are from work done by Norunn Følsvik at NIVA as part of her cand. scient. thesis. at the University of Oslo (Supervisors: Tyge Greibrokk, Einar M. Brevik, John Arthur Berge)

Oslo, April 10th, 1997

John Arthur Berge

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# **Summary**

Tributyltin (TBT) is one of the most toxic substances known. In the marine environment the main focus has been on the antifouling agent TBT although other organotin compounds may also be present. Unintentional effects of TBT have been discovered in populations of certain snail species (neogastropods), and have greatly reduced their densities. Effects of TBT has been observed primarely as the induction of male sex characters in females, known as "imposex". Imposex have been found in about one hundred species of gastropods worldwide. Imposex occurs because exposure to TBT results in the accumulation of a male hormone (testosterone) in female gastropods. Species particularly sensitive to TBT may show signs of effects at water concentrations of TBT as low as 1 ng/l. Many countries have therefore introduced restrictions on the use of TBT. In Norway, its use on boats less than 25 m in length has been prohibited since 1989 and in aquaculture a total ban of the use of TBT on fish cages was introduced in 1989/90.

In Norway the amount of TBT in antifouling paints, sold for use on ships larger than 25 m, has been in the range 11.7(1985)-28.5 (1991) ton/yr during the last 11 years. Approximately 90 % of the TBT used on the hull of ships and 10 % of the TBT used as wood preservatives are expected to end up in the environment. Total discharges of TBT from these two sources to the aquatic environment in Norway during the period 1985-1995 has been estimated to be in the range 18.7 ton/yr in 1989 to 57.3 ton/yr in 1985 (approximately 1-1.5 ton/yr of this was derived from wood preservatives). However, other sources may also contribute.

In order to evaluate the current status of organotin pollution along the Norwegian coast, sediments (100 stations) and blue mussels (*Mytilus edulis*) (63 stations) has been analysed for TBT (GC/MSD). The occurrence of imposex in dogwhelks (*Nucella lapillus*) were investigated in 41 populations sampled in 1993-1995 along the coast from Finnmark to the Skagerrak in the south. The degree of imposex was classified by the Relative Penis Size Index (RPSI) and the Vas Deferens Sequence Index (VDSI). The concentration of TBT was analysed in dogwhelks from 16 of the 41 populations characterised with respect to imposex. The quantification was done by gaschromatography equipped with an atomic emission detector (GC-AED).

The sediment concentrations of TBT in harbours in Northern Norway were highly variable. Most of the sediments contained concentrations below 100 ng/g d.w.. Sediment from 16 sites contained more than 1000 ng/g d.w. TBT, whereas sediment from 17 sites contained between 100 and 1000 ng/g d.w of TBT. The maximum concentration of TBT was found in sediments from a navy harbour near the city of Bergen (103800 ng/g d.w. TBT). The analysis show that many harbour sediments are contaminated with TBT. Analysis of sediment from more remote, sparsely populated, areas along the coast were few, but traces of TBT were also found in sediments form such sites.

The concentration of TBT in mussels from the Norwegian coast varied from less than 10  $\mu$ g/kg wet weight to about 3000  $\mu$ g/kg wet weight. The highest concentrations were found in and around harbours. Even in open waters, however, concentrations of up to 150  $\mu$ g/kg wet weight were found in mussels, which corresponds to concentrations in water exceeding the threshold for injury to sensitive organisms.

Some degree of imposex occurred in all investigated populations of dogwhelks except four stations in Northern Norway (Finnmark). At about half of the sites, a proportion of the female population was sterile (VDSI>4). The concentration of TBT in the gastropods from the unaffected populations in Northern Norway was below the detection limit (17 ng/g d.w.). The concentration of TBT in

dogwhelks from affected populations was in the range 50-2676 ng/g d.w. There seems to be a positive relation between concentration of TBT in dogwhelks and the degree of imposex .

The results demonstrate that organotin compounds are present in the environment in most anthropogenic influenced areas along the Norwegian coast. Biological effects on dogwhelks are widespread in most Norwegian coastal areas apart from some remote northern regions. Populations of *Nucella* from six stations investigated both in 1991 and in 1993 showed signs of reduction in VDSI and RPSI and thus indicate some imposex recovery.

It is expected that a general reduction in the occurrence of organotin compounds in the environment along the Norwegian coast best can be achieved by enforcing restrictions on the use of such compounds also on larger vessels.

# 1. Introduction

The use of organotin compounds as antifouling agents, fungicides, insecticides and bactericides has increased dramatically over the last 30 years (Huggett *et al.*, 1992 with references) and are considered among the most toxic compounds introduced to the marine environment by man (Goldberg, 1986). In the marine environment the main focus has been on the extremely effective antifouling agent tributyltin (TBT) although other organotin compounds may be present in water and sediment (Rosales *et al.*, 1992). Unintended environmental effects from TBT in antifouling paints was first found in the Arcachon Bay in France in the late 1970s on the Pacific oyster (*Crassostrea gigas*) (Alzieu *et al.*, 1986 with references, see also Ruiz et al. 1996). Effects of organotin compounds have since then been reported for a variety of other marine species (fish, gastropods, crustaceans, echinoderms, micro algae) at concentrations that may be found in anthropogenically influenced areas (Beaumont & Newman, 1986 a and b; Walsh *et al.*, 1986; Hall, 1988; Bryan *et al.*, 1986, 1989; Johansen & Møhlenberg, 1987; Bushong et al., 1990; Fent & Meier, 1994). In the last decade much attention has been paid to induction of male sex characters in female snails, known as "imposex", observed mainly in neogastropods (Bryan *et al.*, 1986; Gibbs & Bryan, 1986; Gibbs *et al.*, 1987; Horiguchi *et al.*, 1994). The first description of imposex was given by Blaber (1970).

Most investigations points to TBT as the agent responsible for imposex, however, there are some indications that other compounds/situations also may induce this condition in gastropods (Nias et al., 1993; Evans *et al.*, 1995). TBT-induced imposex is suggested to be caused by an inhibition of the cytochrome P-450 dependant aromatase system leading to an increase in the androgen (testosteron) level in the gastropods (Bettin *et al.* 1996).

Imposex in gastropods is believed to be a wide spread phenomenon (Ellis & Pattisina, 1990) and has in the open North Sea been related to shipping traffic intensities (Ten Hallers-Tjabbes *et al.*, 1994). In some cases local populations of dogwhelks (*Nucella lapillus*) were reduced or disappeared (Bryan *et al.*, 1986). Common non-neogastropods like *Littorina littorea* may, however, also be affected by TBT (Minchin *et al.* 1996). *Littorina* recruit by means of pelagic larvae, and consequently local populations do not so easily become extinct as *Nucella*, which lay their eggs directly on the substrate.

Unintended effects of TBT in the marine environment has led to restrictions on its use as antifouling agent on small boats in a number of countries. Monitoring in areas where such restrictions has been enforced indicate a reduction in environmental concentrations (IMO, 1994; Shiraishi & Soma, 1992; Evans *et al.*, 1995) and also some recovery in populations suffering from imposex (Evans *et al.*, 1991; Bryan *et al.*, 1993; Evans *et al.*, 1995), indicating that environmental problems caused by organotin has been reduced in areas which previously were heavily affected by small boats. In Norway restrictions on the use of TBT in antifouling agents for boats smaller than 25 m was enforced in 1989. The use of TBT on small aluminium boats were however allowed until 1993. A considerable amount of TBT was previously used in the aquaculture industry (table 1). A total ban on the use of TBT in the aquaculture industry (on fish cages) was enforced in 1989.

Investigations concerning the concentration of TBT in the marine environment of Norway started as late as in 1993-1994. Consequently, little is known about temporal trends of organotin contamination in Norway. Only limited data are also available on spatial distribution of organotin contamination in sediments (mostly lacking from areas outside harbours) and water. Investigations on possible effects on organisms apart form dogwhelks has yet not been performed in Norway. The Norwegian State Pollution Control Autority have, however; from 1997, in their monitoring of the Norwegian coast, included analysis of TBT in dogwhelks and and blue mussels in their monitoring. Investigations on imposex in dogwhelks and intersex in *Littorina littorea* have also been neluded.



### 2. Methods

### 2.1 Sampling

Sediments were generally sampled with a gravity corer (Niemistö, 1974) except for seven samples were an Ekman grab were used (Fagerhaug, 1997). The top 0-2 cm were analysed.

Composite samples containing 20-50 mussels (*Mytilus edulis*) from shallow water (0-2 m) were analysed from each station.

Composite samples of dogwhelks (*Nucella lapillus*) collected in the intertidal zone were analysed. The samples analysed from each station containing 30-60 female and male dogwhelks respectively.

Dogwhelks were analysed for imposex using the method of Gibbs *et al.* (1987). Both the Relative Penis Size Index (RPSI) and the Vas Deferens Sequence Index (VDSI) were calculated/recorded.

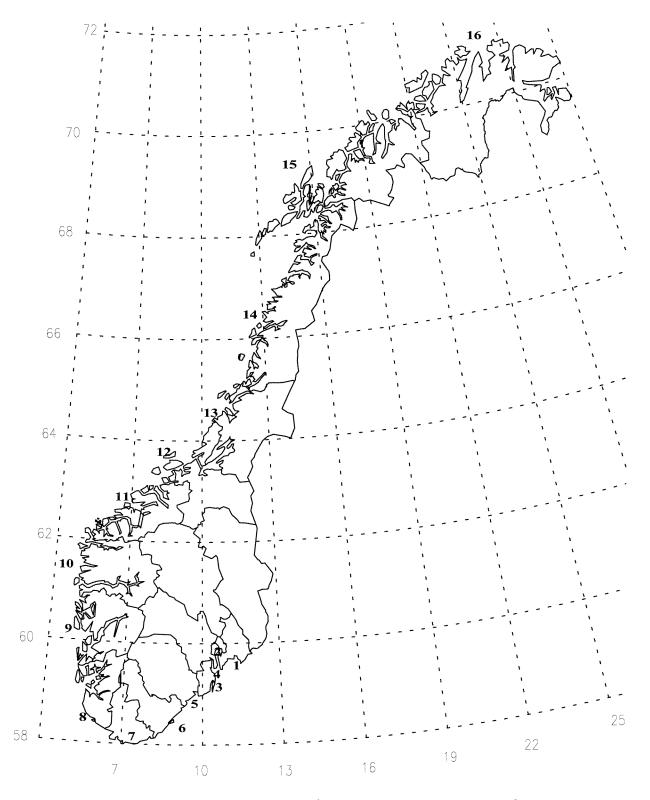
RPSI was calculated as follows:

RPSI=[(mean female penis length)<sup>3</sup> x (mean male penis length)<sup>-3</sup>] x 100%

Vas Deferens Sequence (VDS) recognises the development of a vas deferens in a female in 7 stages. Stage 0= Normal female, Stage 6= Genital papilla overgrown, aborted egg capsules can be seen. Individual snails with a VDS of 5 or 6 are prevented from releasing eggcapsules from their genital papilla and are sterile. VDSI represents the mean VDS for all the anlysed females from a station.

VDSI gives an indication of the reproductive ability of a population. Populations with a VDSI >4 indicate that a certain part of the female population are sterile.

A map of Norway is shown in figure 1.



**Figure 1.** Map of Norway showing latidute (58-72°N, left side) and longitude (7-25°E, bottom). The different counties along the coast are indicated. 1:Østfold, 2:Akershus/Oslo, 3:Vestfold, 4:Buskerud, 5:Telemark, 6:Aust-Agder, 7:Vest-Agder, 8:Rogaland, 9:Hordaland, 10:Sogn og fjordane, 11:Møre og Romsdal, 12:Sør-Trøndelag, 13 Nord-Trøndelag, 14:Nordland, 15 Troms, 16:Finnmark.

### 2.2 Determination of organotins:

#### 2.2.1 Blue mussels and sediments

Levels of organotin compounds in blue mussel (*Mytilus edulis*) and sediments were determined using acid dissolution of the samples, derivatisation with a Grignard-reagent (methylmagnesiumbromide) and analysis by GC-MSD. 1-2 g of the samples was dissolved in HCl, ascorbic acid and tropolone in methanol was added before extraction with n-hexane. The extract was dried with CaCl<sub>2</sub> to remove any traces of water. After removal of air by flushing N<sub>2</sub> into the sample-glasses, the extracts were derivatised by addition of methylmagnesiumbromide. Excess derivatizing reagent was removed by adding HCl and the final extract was concentrated before analysis by GC-MS.

Seven of the sediment samples from the county of Møre and Romsdal were analysed with a modification of the above method (Fagerhaug 1997).

#### 2.2.2 Dogwhelks

A different method, involving dervatization with sodiumtetraethylborate was adopted for all the analysis of dogwhelks (*Nucella lapillus*). Implementation of the method was part of a cand. scient. thesis (Norunn Følsvik). The method was developed because the Grignard-derivatization used for analysing sediments and mussels is both time-consuming and very sensitive to the effective removal of water and air from the samples. Direct ethylation with sodiumtetraethylborate has also other advantages compared to the Grignard-derivatization as it can be performed directly on wet samples and is far less time-consuming.

The samples (1-2 g) were dissolved in a 10 % solution of a tissue solubelizer (tetraethylamoniumhydroxid) in methanol before direct ethylation with NaBEt<sub>4</sub>. The organotins are present as "free" chlorides and hydroxides. The efficiency of the derivatisation is very pH-dependent and it is therefore urgent to adjust the pH before the derivatisation. Samples were shaken thoroughly and left for 10 minutes to complete the derivatisation. The tetraalkylated species were then extracted with n-hexane. The whole process was repeated once. Before analysis the samples were concentrated with nitrogen to approximately  $100~\mu L$ .

Quantification: The organotin compounds were identified and quantified by GC-AED. A Hewlett Packard-5890 gas chromatograph with a HP-5921A microwave induced plasma atomic emission detector was used. Injections were made by a HP-7673 automatic injector. Samples were injected onto a 30 m HP-5 column which mainly separates the compounds after increasing boiling point. Thus the lighter organotins reach the detector first and there the sample is atomised and exited and the intensity of characteristic lines is measured by a photodiodearray spectrophotometer.

<u>Calibration</u> was carried out by running a five point calibration curve for every series analysed. An internal standard was also used to compensate for losses during sample preparation. The different organotin species were identified by analysis of pure standard solutions.

The quality of the analysis was assured by analysing a standard for every 10 samples. In addition, the sample preparation was controlled by analysis of a standard reference material (NIES No. 11) and blank-samples. A home made biological "reference" material was also analysed to verify the long term reproducibility of the method. An analytical precision better than  $10\,\%$  ( RSD < 10%) was achieved when analysing parallel samples.

Comparison of the methods used for analysing M. edulis (section 2.2.1) and N. lapillus (section 2.2.2), indicate that the TBT concentrations in M. edulis probably are considerably underestimated (2-3 times) when using acid dissolution of the samples and derivatization with a Grignard-reagent

#### 2.2.3 Conversion factor

In this document the concentrations are usually given on a TBT basis. Other investigations may report results on a tin (Sn) basis. The following conversion can be used:

Concentration on TBT basis =2.44 x concentration on Sn basis.

# 3. TBT sources in Norway

Organotin compounds are generally of anthropogenic origin apart from methyltins, which may be produced by biological methylation. The total amount of TBT used in Norway after 1988 is reported to be dominated by the use of TBT in antifouling agents on boats larger than 25 m (table 1, see also Fjelldal, 1994) and as wood preservative (table 1, see also Fjelldal, 1994). World wide a large part of the consumption of TBT lies in the use as preservative for wood (Fent, 1996). A relatively large amount of TBT was previously used in the aquaculture industry (table 1). A total ban of such use was introduced in 1989/90.

The total annual amount of TBT in products sold in Norway during the last ten years has been in the range 28.7-73.2 ton (table 1). The total consumption of TBT in Sweden in 1989 is reported to be 400-500 t. Of this, close to 150 t was used in antifouling agents (Anonymous, 1989). The Norwegian consumption of TBT (table 1) seems to be relatively low compared to what has been reported for Sweden.

The amount of TBT used on small boats is assumed to be low since the ban was introduced in 1989 (table 1). The annual amount of TBT in antifouling paints sold for use on ships larger than 25 m has been more stable and are reported to be in the range 11.7-28.5 tons (table 1).

Approximately 10 % of the TBT used as wood perservatives and 90 % of the TBT used as antifouling agent on ships are expected to end up in the environment (Fjelldal, 1994) (see table 1 and table 2). Estimated total discharges of TBT to the aquatic environment from these two sources during the period 1985-1995 has been in the range 17.8-57.3 ton/yr (table 2).

It is claimed that the only/most important Norwegian source for the introduction of TBT to water is through antifouling agents (Fjelldal, 1994). Recent investigations have, however, shown that municipal wastewater and sludge may be an additional source for TBT and other organotins (Fent, 1996). Much of the TBT in the raw waste water is retained in the sludge. The amount retained (40-98%) depend on the performance of the treatment plant (Fent, 1996). The remaining TBT is discharged to aquatic systems. The concentration of TBT in waste water and sludge are so high that ecotoxicological consequences should be regarded. No information is available on the concentrations of TBT in wastewater from Norwegian sewage treatment plants.

It is assumed that the main source for the present introduction of TBT to the aquatic environment in Norway is through the use of antifouling paints on ships larger than 25 m. Sand blasting of the underwater hull of ships in order to remove old paint also be an important secondary source for TBT in harbours with dry docks. Other supplementary sources and routes for introduction of TBT to Norwegian coastal waters can not be excluded.

**Table 1.** Amount of TBT (ton) in products sold in Norway. Source: Norwegian State Pollution Control Authority. n.d.=no data, u.z.=usage assumed to be zero

| Year | Antifouling paints small boats | Antifouling paints | Antifouling paints all boats | Wood<br>preservatives |      | Total use in antifouling and as preservative |
|------|--------------------------------|--------------------|------------------------------|-----------------------|------|--|
| 1985 | 13.1                           | 11.7               | 24.8                         | 15                    | 33.4 | 73.2   |
| 1988 | 9.7                            | 16.2               | 25.9                         | 15                    | n.d. | 40.9   |
| 1989 | 1.2                            | 17.9               | 19.1                         | 15                    | n.d. | 34.1   |
| 1990 | 0.5                            | 23.0               | 23.5                         | 15                    | u.z. | 38.5   |
| 1991 | 1.5                            | 28.5               | 30                           | 12                    | u.z. | 42   |
| 1992 | 2.7                            | 26.6               | 29.3                         | 12                    | u.z. | 41.3   |
| 1993 | 0.5                            | 18.2               | 18.7                         | 10                    | u.z. | 28.7   |
| 1994 | 0.5                            | 23.4               | 23.9                         | 12.8                  | u.z. | 36.7   |
| 1995 | u.z.                           | 27.8               | 27.8                         | 12                    | u.z. | 39.8   |

**Table 2.** Estimated discharges of TBT from wood preservatives and antifouling paints. The estimated discharges from antifoulingpaints includes approximately 5 % triphenyltin. Source: Norwegian State Pollution Control Authority.

|      | Estimated discharges of TBT (ton) |                    |  |  |  |  |  |  |
|------|-----------------------------------|--------------------|--|--|--|--|--|--|
| Year | Wood preservatives                | Antifouling paints |  |  |  |  |  |  |
| 1985 | 1.5                               | 55.8               |  |  |  |  |  |  |
| 1988 | 1.5                               | 23.1               |  |  |  |  |  |  |
| 1989 | 1.5                               | 17.2               |  |  |  |  |  |  |
| 1990 | 1.5                               | 21.2               |  |  |  |  |  |  |
| 1991 | 1.2                               | 27.0               |  |  |  |  |  |  |
| 1992 | 1.2                               | 26.4               |  |  |  |  |  |  |
| 1993 | 1                                 | 16.8               |  |  |  |  |  |  |
| 1994 | 1.3                               | 21.5               |  |  |  |  |  |  |
| 1995 | 1.2                               | 25.0               |  |  |  |  |  |  |

### 4. TBT in the environment

Few if any studies on organotin compounds in the aquatic environment have been performed in Norway before 1990. Since then investigations have mainly focused on concentrations of TBT in coastal sediments, mussels and effects of TBT in dogwhelks (*Nucella lapillus*). Recently an investigation on organotin compounds (TBT, DBT and MBT) in dogwhelks has been started. Only a few analyses of TBT in fish from Norwegian coastal areas have been performed.

#### 4.1 TBT in sediments

Most of the sediments contained concentrations below 100 ng/g d.w. (appendix A, figure 2). Sediment from 16 sites contained more than 1000 ng/g d.w. TBT whereas sediment from 17 sites contained between 100 and 1000 ng/g d.w of TBT. The concentrations in the harbours were highly variable. Maximum and minimum concentration were 103800 ng/g d.w. (navy harbour) and <1ng/g d.w. (Stavern inner harbour), respectively (Appendix A). TBT is a manmade substance and is not reported to occur naturally in the environment.

Concentrations above the detection limit (1-5  $\mu$ g/kg d.w., Appendix A) thus indicate that TBT is discharged to the environment. The results show that some harbour sediments are heavily contaminated with TBT whereas the concentration of TBT in other harbours sediments are below the detection limit.

Resuspention of contaminated sediments may be a secondary source for the uptake of TBT in marine organisms.

Generally, there seems to be no obvious single factor responsible for the varying concentrations of TBT in the different harbour sediments. Distance to shipyards and quays, shipping traffic intensities, water renewal, bottom current regime and sedimentation are together responsible for the highly variable concentrations. Patchiness resulting from such factors probably is high, but has not been investigated in these introductory studies

Highly variable TBT concentrations in sediments has also been found in other areas (cf. table 3)

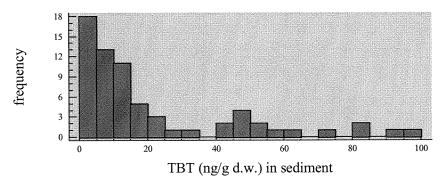
Analyses of sediment from more remote locations along the Norwegian coast are few and analysis of sediments from and near harbours are dominating in our data (Appendix A).

**Table 3.**Concenetrations of TBT (µg Sn/kg d.w. sediment) found in sediment in Canadian coastal harbours (selected values from Maguire et al. 1986).

| Area               | Concentration (µg Sn/ kg d.w.) | Concentration (µg TBT/ kg d.w.) |  |  |
|--------------------|--------------------------------|---------------------------------|--|--|
| British Columbia   |                                |                                 |  |  |
| Nanaimo Harbour    | <31)                           | <7.7                            |  |  |
| Tsehun Harbour     | 180                            | 439                             |  |  |
| Deep cove          | <31)                           | <7.7                            |  |  |
| Patricia Bay       | <31)                           | <7.7                            |  |  |
| Esquimalt Harbour  | 140-1400                       | 342-3416                        |  |  |
| Vancouver Harbour  | <3 <sup>1</sup> )-10780        | <7.7-26303                      |  |  |
| Nova Scotia        |                                |                                 |  |  |
| Pictou Harbour     | <10 <sup>2)</sup>              | <24.4                           |  |  |
| Sydney Harbour     | 10                             | 24.4                            |  |  |
| Halifax Harbour    | <10 <sup>2)</sup>              | <24.4                           |  |  |
| Newfoundland       |                                |                                 |  |  |
| Conception Bay     | 30                             | 73                              |  |  |
| Argentia Harbour   | <10 <sup>2)</sup>              | <24.4                           |  |  |
| St. John's Harbour | <31                            | <7.7                            |  |  |

<sup>&</sup>lt;sup>1</sup>Below limit of detection (approx. 3 μg Sn/kg d.w.)

<sup>2)</sup>TBT detected but concentration below quantification limit (approx 10 µg Sn/kg d.w.) but larger than detection limit.



**Figure 2.** Frequency distribution of the concentration of TBT in sediments collected from harbours along the Norwegian coast. Only sediments with concentrations below 100 ng/g d.w. are shown. (67 % of the data). All data are found in appendix A.

#### 4.2 TBT in blue mussels

Contamination with TBT in Norwegian harbours has been investigated in 1993-1994 using the common mussel (*Mytilus edulis*) as indicator of TBT-level. Mussels have been sampled from 63 harbours along the Norwegian coast from the county of Østfold in Skagerrak to Finnmark on the border to the Barents Sea (figure 1, Appendix B)

Median concentration of TBT in the mussels were 700  $\mu g/g$  d.w. Maximum and minimum concentration was 16649 and 35  $\mu g/g$  d.w., respectively (Appendix B). Mussels fromapproximately 50 % of the sites contained concentrations below 1000  $\mu g/g$  d.w. (figure 3). Examples of very high levels were found in several of the harbour localities (Table 1), where shipyards, drydocks and boating activities probably are the most dominant sources. The results also indicate TBT contamination even in mussels from some more remote reference areas (Table 1). Investigations from

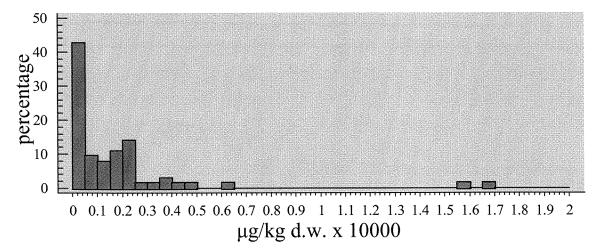
other areas indicate that TBT are associated to marinas or areas of high boat traffic (Short & Sharp, 1989; Short et al., 1989).

The concentrations of TBT in mussels from the Norwegian coast generally falls within the range found in other coastal areas around the world (table 4). It is noteworthy, however, that high concentrations still were found in most areas 3-4 years after the restrictions on use of TBT on small boats and aquaculture was enforced in Norway. TBT in antifauling agents used on large ships and from resuspention of TBT contaminated sediments are the most likely sources for this.

Time series on TBT concentrations in mussels are not available from the Norwegian coast. Results from other areas indicate that restrictions on the use of TBT on small boats have resulted in decreased concentrations in mussels (table 4). Investigations on transplanted mussels indicate that depression of TBT is a biphasic two-component process involving a rapid component (half life: 2.2-5.3 days) followed by a slower loss (half life: 28-69 days) (Page *et al.* 1995).

An assumed bioconcentration factor (BCF) of 10000 (Knutzen *et al.*, 1995 with references) gives TBT concentrations in ambient harbour waters mostly in the order of 10-50 ng TBT/l, in extreme cases up to about 300 ng/l. Similar calculations for localities far from land based sources gave values from <1 to 5-15 ng/l, which must be regarded as serious when compared to water quality criteria of 1-2 ng/l in Canada and UK (c.f Zabel *et al.*1988, Waite *et al.* 1991). It should be emphasized, however, that literature data on BCF varies considerably, - from < 5000 to >50000 (Knutzen et al., 1995 with references) - and that it is observations which indicate that BCF increase with reduced exposure. Consequently, the general use of a BCF as low as 10000 probably imply a safety factor when TBT exposure is relatively low.

# TBT in Mytilus



**Figure 3.** Frequency distribution of the concentration of TBT in mussels collected from harbours along the Norwegian coast.

Table 4. Data for TBT in mussels (mainly Mytilus edulis) from various countries.

| Area, year                            | TBT                   | TBT                       | Ref./comment                 |  |  |
|---------------------------------------|-----------------------|---------------------------|------------------------------|--|--|
|                                       | (μg /kg w.w.)         | (μg /kg d.w.)             |                              |  |  |
| SWEDEN                                |                       |                           |                              |  |  |
| West coast, stations 1984             |                       | ~700-92001)               | Bjørklund, 1987              |  |  |
| Stockholm archipelago, 1986           |                       | ~3000-40001)              | Bjørklund, 1987              |  |  |
| Stockholm archipelago (outer part),   |                       | <230 <sup>1)</sup>        | Bjørklund, 1987              |  |  |
| 1986,                                 |                       |                           |                              |  |  |
| West coast, stations, 1987-88         |                       | ~350-6300 <sup>1)</sup>   | Bjørklund, 1988,             |  |  |
| Baltic sea stations., 1987            |                       | ~350-1700 <sup>1)</sup>   | Bjørklund, 1988,             |  |  |
| Gothenberg area, 1992                 | 60-256                | ~360-1500 <sup>2)</sup>   | Granmo and Eklund, 1993      |  |  |
| West coast stations. 1993             | 20-91                 | ~120-540 <sup>2)</sup>    | Granmo and Eklund, 1993      |  |  |
| DENMARK                               |                       |                           |                              |  |  |
| Lyø south of Fyn, 1986                | $\sim 160^{2)}$       | ~1000 <sup>1)</sup>       | Zuolian og Jensen, 1989.     |  |  |
|                                       |                       |                           | Assumed "unpollute locality" |  |  |
| BRITISH ISLES                         |                       |                           |                              |  |  |
| Several estuaries, (1986)             | 330-2570              | ~2000-16000 <sup>2)</sup> | Waite et al., 1991           |  |  |
| Same estuaries as above. (1989)       | 70-810                | ~420-4900 <sup>2)</sup>   | Waite et al., 1991           |  |  |
| Several estuaries 1986                | 170-2570              | ~1000-15500 <sup>2)</sup> | MAFF, 1993                   |  |  |
| Same estuaries as above (1991)        | 20-250                | ~120-1500 <sup>2)</sup>   | MAFF, 1993                   |  |  |
| NETHERLANDS                           |                       |                           |                              |  |  |
| Eastern Scheldt, Grevelingen, 1988    | ~20-400 <sup>2)</sup> | 130-2300                  | Ritsema et al., 1991         |  |  |
| Waddeen Sea, 1988 (mussel farm)       | $<0.2^{2)}$           | <1                        | Ritsema et al., 1991         |  |  |
| PORTUGAL                              |                       |                           |                              |  |  |
| Sado estuary, 1986                    | $<7-50^{2}$           | <45-280                   | Quevauviller et al., 1989    |  |  |
| CANADA                                |                       |                           |                              |  |  |
| Several stations, 1986-89             | <12-10500             | ~70-63000                 | Thompson og Stewart, 1994    |  |  |
|                                       |                       |                           |                              |  |  |
| Georgia-stredet-Frazer River,         | ~20-130 <sup>2)</sup> | ~130-7801)                | Stewart og Thompson, 1994;   |  |  |
| 1992-93 (after TBT ban.)              |                       |                           |                              |  |  |
| USA                                   |                       |                           | TBT-begr. i USA 1988         |  |  |
| West coas stations., 1986-87          | 20-1000               | ~120-6000 <sup>2)</sup>   | Short og Sharp, 1989         |  |  |
| Auke Bay, Alaska, 1987-80             | <5-350 <sup>2)</sup>  | <25-2000 <sup>1)</sup>    | Short et al., 1989           |  |  |
| San Diego Bay                         | 68-1067               | ~400-6300 <sup>2)</sup>   | Stallard et al., 1989        |  |  |
| San Diego Bay, 1986-1988,             | 100-2100              | "600-12500 <sup>2)</sup>  | Valkirs et al., 1991         |  |  |
| San Diego Bay 1989-1990 (same         | 30-1700               | $\sim 180 - 10000^{2}$    | Valkirs et al., 1991         |  |  |
| stations as above)                    |                       |                           |                              |  |  |
| East- and West coast stations 1987-88 | ~17-540 <sup>2)</sup> | 105-3256                  | Uhler et al., 1989           |  |  |
| 45 East coast stations., 1989-90      | ~2-660 <sup>2)</sup>  | 10-4030                   | Uhler et al., 1993           |  |  |
| 31 West coast stations "              | ~2-230 <sup>2)</sup>  | 10-1380                   | Uhler et al., 1993           |  |  |

<sup>1)</sup> Recalculated fom Sn-based values to TBT (f=2.43).

<sup>2)</sup> Recalculated from wet weight values assuming: concentration (d.w.)/concentration (w.w.)=6/1.

### 4.3 TBT and imposex in dogwhelks

Imposex in the dogwhelk (*N. lapillus*) have been documented in Southern and Western Norway in 1991 (Harding *et al.*, 1992) and again in 1994 (Evans et al., 1996). A more comprehensive investigation of imposex in *N. lapillus* were carried out in 1993-1995 on 41 stations along most of the Norwegian coast. Preliminary results from this investigation (figure 4, table 5) indicate widespread effects of TBT along the Norwegian coast. Only a few stations (approximately 10%) showed no signs of imposex. These stations were located in rather remote sparsely populated areas mostly in, northern Norway (Finnmark).

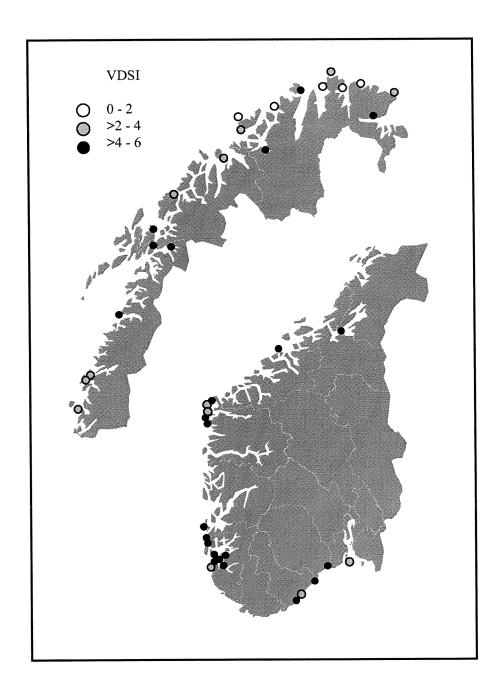
The results imply that populations from a majority of the stations contained sterile females (figure 4, VDSI>4, Gibbs & Bryan, 1986; 1987).

On Iceland imposex in *N. lapillus* has been found at 38 out of 45 localities studied and was most frequent near harbours (Svavarsson & Skarphédinsdóttir 1995). On Iceland imposex were found out to a distance of approximately 20 km from the nearest harbour. A preliminary evaluation indicate similar effects in Norway as found on Iceland. The absence of imposex at some remote locations in Iceland and Norway are noteworthy, as some indication of imposex usually are found at virtually all locations studied in more densly populated areas.

Dogwhelks from some of the stations investigated for imposex have also been analysed for TBT (table 5). The concentration levels of TBT in females and males were in the same range (figure 5). The results also show that the degree of imposex is related to the concentration of TBT in dogwhelks. (figure 6, figure 7).

Dogwhelks in populations with VDSI larger than 4 all contained more than 100 ng/g d.w. TBT. In contrast no clear sign of imposex was seen in gastropodes in population containing less than approximately 50-100 ng/g d.w. (figure 6, figure 7).

The main source for new TBT to the aquatic environments is probably through the continued use of TBT on large ships (see chapter 3). A link between shipping traffic intensity and imposex is also indicated since *N. lapillus* from sites near large harbours (shipping large amounts of goods) seems to be more heavily affected than those in areas with less traffic (figure 9).



**Figure 4.**Vas deferens sequence index (VDSI) in dogwhelks (*Nucella lapillus*) along the Norwegian coast, 1993-1995.

**Table 5.** Mean relative penis size index (RPSI) and vas deferens sequence index (VDSI) found in *Nucella lapillus* sampled along the Norwegian coast 1993-95. Preliminary data generated at NIVA. RPSI is a measure of the mean size of the female penis in relation to that of the male. VDSI recognises the development of a vas deferens in the female in 7 stages. Stage 0= Normal female, Stage 6= Genital papilla overgrown, aborted egg capsules can be seen. For further explanation see Gibbs *et al.*, 1987. Station numbers (no.) are referred on map (figure 4). TBT analysis were performed as part of an Cand scient. thesis (Norunn Følsvik).

| no. | Station                              | Date of collection | Pos. N<br>(NN° NN.NN') | Pos.E<br>(NNº NN.NN') | Count | VDSI | VDSI<br>St.dev. | RPSI (%) | TBT in females (ng/g d.w.) | TBT in males (ng/g d.w.) |
|-----|--------------------------------------|--------------------|------------------------|-----------------------|-------|------|-----------------|----------|----------------------------|--------------------------|
|     | AKERSHUS/OSLO                        |                    |                        |                       |       |      |                 |          |                            |                          |
| 1   | Færder                               | 13/09-93           | 59 01.55               | 10 31.70              | 20    | 3.95 | 0.22            | 10.34    | 138                        | 109                      |
|     | TELEMARK                             |                    |                        |                       |       |      |                 |          |                            |                          |
| 2   | Langesund (Fuglø)                    | 13/09-93           | 58 58.95               | 09 48.50              | 20    | 4.1  | 0.31            | 28.94    | 150                        | 145                      |
|     | AUST-AGDER                           |                    |                        |                       |       |      |                 |          |                            |                          |
| 3   | Risør                                | 13/09-93           | 58 43.80               | 09 17.50              | 20    | 4.2  | 0.41            | 8.38     | 193                        | 138                      |
| 4a  | Tromøy,<br>Alvekilen/Timmerstøberget | 18/06-95           | 58 27.92               | 08 53.75              | 52    | 3.46 |                 | 3.06     |                            |                          |
| 4b  | Tromøysund, northeast                | 04/10-94           | 58 30.77               | 08 56.79              | 20    | 4.3  | 0.66            | 8.14     | 138                        | 143                      |
|     | ROGALAND                             |                    |                        |                       |       |      |                 |          |                            |                          |
| 1   | Stavanger                            | 08/09-93           | 59 01.55               | 05 40.10              | 20    | 4.35 |                 | 55.5     | 293                        | 215                      |
| l   | G1-Fjoløy, Stavanger                 | 20/09-95           | 59 05.40               | 05 33.95              | 20    | 4.25 | I               | 24.9     |                            |                          |
| 5c  | G2-Alstein, Stavanger                | 18/09-95           | 59 05.40               | 05 31.68              | 20    | 4.6  | 0.5             | 45.38    |                            |                          |
| 5d  | G3-Grønningen, Stavanger             | 19/09-95           | 58 59.05               | 05 30.06              | 20    | 4.15 | 1               | 16.62    |                            |                          |
| 5e  | G4-Tungeneset, Stavanger             | 20/09-95           | 59 02.25               | 05 35.00              | 20    | 4.05 | l               | 33.81    |                            |                          |
| 5f  | G5-Vistnes, Stavanger                | 19/09-95           | 58 59.05               | 05 33.88              | 20    | 3.8  | 0.52            | 6.61     |                            |                          |
| 6   | Kopervik                             | 08/09-93           | 59 16.95               | 05 19.40              | 20    | 4.4  | 0.5             | 44.31    | 433                        | 325                      |
| 7   | Haugesund                            | 08/09-93           | 59 25.00               | 05 14.00              | 20    | 4.65 | 0.49            | 37.11    |                            |                          |
|     | HORDALAND                            |                    |                        |                       |       |      |                 |          |                            |                          |
| 8   | Espevær (Marholmen)                  | 08/09-93           | 59 34.75               | 05 08.90              | 20    | 4.15 | 0.67            | 6.02     |                            |                          |
|     | SOGN OG FJORDANE                     |                    |                        |                       |       |      |                 |          |                            |                          |
| 9a  | Florø, north                         | 28/08-93           | 61 36.41               | 05 03.33              | 20    | 4.72 | 0.55            | 91.37    | 470                        | 567                      |
| 9b  | Florø, south                         | 04/09-93           | 61 35.57               | 05 03.62              | 20    | 4.95 | 0.39            | 107.01   | 2673                       | 1178                     |
| 10  | Bremanger                            | 04/09-93           | 61 45.95               | 04 56.00              | 20    | 3.75 | 0.44            | 4.68     |                            | 411                      |
| 11a | Måløy, north                         | 04/09-93           | 61 58.43               | 05 08.50              | 20    | 4.9  | 0.31            | 71.22    |                            |                          |
| 11b | Måløy, south                         | 04/09-94           | 61 55.75               | 05 07.40              | 20    | 4    | 0               | 47.41    |                            |                          |
|     | MØRE ROMSDAL                         |                    |                        |                       |       |      |                 |          |                            |                          |
| 12  | Kristiansund                         | 02/09-93           | 63 06.40               | 07 45.15              | 20    | 4.25 | 0.44            | 30.89    |                            |                          |
|     | SØR TRONDELAG                        |                    |                        |                       |       |      |                 |          |                            |                          |
| 13  | Trondheim                            | 01/09-95           | 63 27.02               | 10 23.32              | 20    | 4.25 | 0.44            | 22.11    | 194                        | 166                      |

Table 6.(cont.)

| no. | Station             | Date of collection | Pos. N<br>(NN° NN.NN') | Pos.E<br>(NN° NN.NN') | count | VDSI | VDSI<br>St.dev | RPSI<br>(%) | TBT in<br>females<br>(ng/g<br>d.w.) | TBT in males (ng/g d.w.) |
|-----|---------------------|--------------------|------------------------|-----------------------|-------|------|----------------|-------------|-------------------------------------|--------------------------|
|     | NORDLAND            |                    |                        |                       |       |      |                |             |                                     |                          |
| 14  | Brønnøysund         | 29/08-93           | 65 28.35               | 12 11.93              | 20    | 3.55 | 1.15           | 2.23        |                                     |                          |
| 1   | Sandnessjø,. indre  | 28/08-93           | 66 01.55               | 12 38.20              | 23    | 4    | 0              | 26.57       | 220                                 | 344                      |
| l   | Sandnessjø,, ytre   | 28/08-94           | 66 02.95               | 12 40.60              | 20    | 2.65 | 1.09           | 0.27        | 111                                 | 158                      |
| l   | Bodø                | 27/08-93           | 67 17.25               | 14 21.80              | 30    | 4.5  | 0.51           | 37.04       | 152                                 | 183                      |
| 17  | Narvik              | 27/08-93           | 68 25.45               | 17 20.00              | 20    | 4.15 | 0.37           | 18.53       |                                     |                          |
| 18  | Ramsund             | 16/08-94           | 68 29.90               | 16 29.80              | 20    | 4.4  | 0.5            | 52.72       |                                     |                          |
|     | TROMS               |                    |                        |                       |       |      |                |             |                                     |                          |
| 19  | Harstad             | 16/08-94           | 68 49.30               | 16 33.92              | 20    | 4.75 | 0.64           | 45.36       |                                     |                          |
| 20  | Finnsnes            | 17/08-94           | 69 13.55               | 17 58.50              | 34    | 4.15 | 0.44           | 13.23       | 117                                 | 128                      |
| 21  | Skjervøy            | 19/08-94           | 70 02.15               | 21 00.30              | 27    | 3.89 | 0.32           | 8.71        |                                     |                          |
|     | FINNMARK            |                    |                        |                       |       |      |                |             |                                     |                          |
| 22  | Alta                | 20/08-94           | 69 59.20               | 23 18.80              | 20    | 4.1  | 0.72           | 14.5        | 156                                 | 167                      |
| 23  | Elenheim, st44      | 04/09-95           | 70 31.10               | 22 15.40              | 20    | 2.35 | 1.53           | 0.41        |                                     |                          |
| 24  | Sørøya, Åfjorden    | 05/09-95           | 70 37.89               | 22 22.99              | 20    | 0    | 0              | 0           | <18                                 | <18                      |
| 25  | Saughamnneset, ytre | 03/09-93           | 70 45.80               | 24 19.80              | 20    | 0    | 0              | 0           |                                     |                          |
| 26  | Honningsvåg         | 21/08-94           | 70 59.12               | 25 57.77              | 20    | 4.3  | 0.47           | 53.69       |                                     |                          |
| 27  | Kifjordneset        | 02/09-95           | 70 52.86               | 27 22.20              | 20    | 0    | 0              | 0           | <18                                 | <18                      |
| 28  | Mehamn              | 22/08-94           | 71 02.55               | 27 50.35              | 27    | 3.96 | 0.34           | 20.29       | 210                                 | 206                      |
| 29  | Tanafjorden         | 01/09-95           | 70 41.58               | 28 33.27              | 21    | 0    | 0.22           | 0.05        | 21                                  | <18                      |
| 30  | Båtsfjord           | 23/08-94           | 70 38,67               | 29 45.00              | 27    | 1.67 | 1.11           | 0.36        |                                     |                          |
| 31  | Vardø               | 24/08-94           | 70 22.65               | 31 06.50              | 23    | 3.52 | 0.99           | 0.75        | 48                                  | 46                       |
| 32  | Vadsø               | 24/08-95           | 70 04.48               | 29 42.90              | 20    | 4.3  | 0.57           | 6.11        |                                     |                          |

#### **TBT in Nucella**

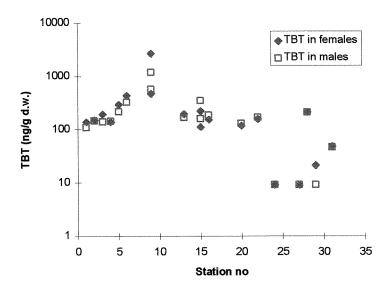


Figure 5. Concentation of TBT in male and female dogwhelks (*Nucella lapillus*). Concentration of TBT below the detection limit (<18 ng/g d,w,) is shown as 9 ng/g d.w. i the graph.

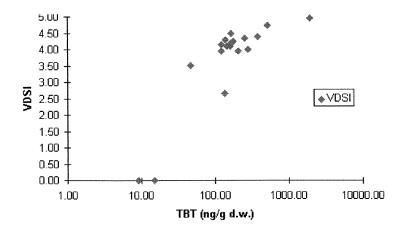
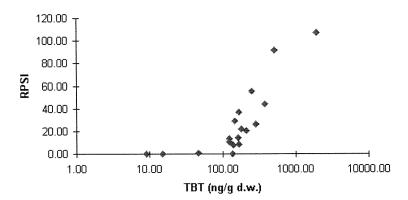


Figure 6. Vas deferens sequence index (VDSI) in dogwhelks (*Nucella lapillus*) as a function of the mean concentration in males and females. Concentration of TBT below the detection limit (<18 ng/g d,w,) is presented as 9 ng/g d,w in the graph.



**Figure 7.** Relative penis size index (RPSI) in dogwhelks (*Nucella lapillus*) as a function of the mean concentration in males and females. Concentration of TBT below the detection limit (<18 ng/g d.w.) is presented as 9 ng/g d.w. in the graph.

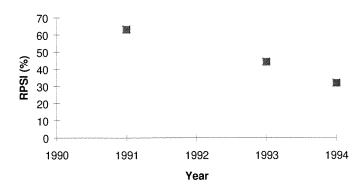
Some recent studies show some signs of recovery in populations suffering from imposex (Evans *et al.*, 1991; Bryan *et al.*, 1993; Evans *et al.*, 1995, Evans *et al.* 1996), indicating that environmental problems caused by organotin has been reduced in areas which previously were heavily affected by small boats. The first investigation recording imposex in Norway was conducted in 1991 (Harding *et al.*, 1992). All revisited Norwegian populations now show some recovery (table 7). At the station near Koppervik (only station visited 3 times) a consistant decrease in RPSI was indicated (figure 8).

**Table 7.** Relative penis size index (RPSI) and vas deferens sequence index (VDSI) found in *Nucella lapillus* sampled on the same stations along the Norwegian coast in 1991 (Harding *et al.* 1992), 1993 (Evans et al. 1996) and 1993-95 (data generated at NIVA, see table 5).

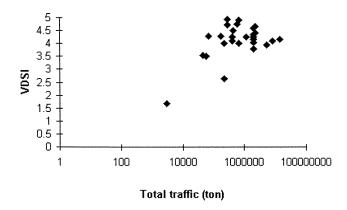
A:Adults, SA=subadults (15-21 mm), J=juvenlies (<15 mm)

| Station                              | Date of collection | Pos. N<br>(NN° NN.NN') | Pos.E<br>(NN° NN.NN')  | VDSI                         | RPSI<br>(%)                    |                        |
|--------------------------------------|--------------------|------------------------|------------------------|------------------------------|--------------------------------|------------------------|
| Færder (st. 15)                      | Sept-Oct. 1991     | 59 01.40               | 10 31.65               | A:4.5<br>J:4.0<br>SA: 4.0    | A:23<br>J:32,34<br>SA: 28,38   | Harding et<br>al. 1992 |
| Færder                               | 13/09-93           | 59 01.55               | 10 31.70               | 3.95                         | 10.34                          | NIVA                   |
| Langesund (Fuglø) (st. 14)           | Sept-Oct. 1991     | 58 58.95               | 09 48.50               | A:4.60                       | A:36.08                        | Harding et<br>al. 1992 |
| Langesund (Fuglø)                    | 13/09-93           | 58 58.95               | 09 48.50               | 4.1                          | 28.94                          | NIVA                   |
| Tromøy, most easterly tip (st. 13)   | Sept-Oct. 1991     | 58 30.64               | 08 57.40               | A:4.48                       | A:33.1                         | Harding et<br>al. 1992 |
| Tromøy,<br>Alvekilen/Timmerstøberget | 18/06-95           | 58 27.92               | 08 53.75               | 3.46                         | 3.06                           | NIVA                   |
| Koppervik (st.11)                    | Sept-Oct. 1991     | 59 17.00               | 05 19.35               | A:5.25                       | A:63.21                        | Harding et<br>al. 1992 |
| Kopervik                             | 08/09-93           | 59 16.95               | 05 19.40               | 4.4                          | 44.31                          | NIVA                   |
| Koppervik                            | 1994               | 59 16.95 <sup>1)</sup> | 05 19.40 <sup>1)</sup> |                              | 32                             | Evans et<br>al. 1996   |
| Haugesund (East of Storøy(St 4)      | Sept-Oct. 1991     | 59 25.00               | ?                      | A:5.36<br>J: 4.47<br>SA:4.39 | A:54.89<br>J:60.19<br>SA:63.06 | Harding et<br>al. 1992 |
| Haugesund                            | 08/09-93           | 59 24.99               | 05 14.02               | 4.65                         | 37.11                          | NIVA                   |
| Espevær (St 1)                       | Sept-Oct. 1991     | 59.34.93               | 05 08.36               | A:4.28<br>SA:4.18            | A:24.42<br>SA:29.25            | Harding et<br>al. 1992 |
| Espevær (Marholmen)                  | 08/09-93           | 58 34.75               | 05 08.90               | 4.15                         | 6.02                           | NIVA                   |

<sup>&</sup>lt;sup>1)</sup>Map in Evans et al. (1996) indicate that the station sampled is the same as used by NIVA in 1993.



**Figure 8.** RPSI in dogwhelks from Koppervik (Data from Harding *et al.* 1992, Evans *et al.* 1996 and NIVA.



**Figure 9.**VDSI as a function of total annual amount of goods traffic (1993) through the nearest large harbour (.

### 5. Conclusions

The main source for the introduction of TBT to the marine environment today is through the use of antifouling paints on ships larger than 25 m. Resuspention of contaminated sediments may also be a secondary source for TBT contamination and result in biological effects. However, other supplementary sources (wood preservatives, possible illegal use and sewage treatment plants) and routes for introduction of TBT to Norwegian coastal waters should also be considered. Approximately 90 % of the TBT used on the hull of ships and 10 % of the TBT used as wood preservatives are expected to end up in the environment.

Mussel and sediment data from Norwegian coastal areas clearly demonstrate that organotin compounds are present in most areas, with especially high concentrations found near harbours. Imposex in dogwhelks caused by TBT has also been recorded in almost all such areas. At about half of the total number of sites, part of the female gastropod population was sterile (VDSI>4). The concentration of TBT in the dogwhelks from unaffected populations in Northern Norway (Finmark) was below the detection limit (17 ng/g d.w:). The concentration of TBT in dogwhelks from affected populations was in the range 50-2676 ng/g d.w. The results also show that the degree of imposex is related to the concentration of TBT indogwhelks. (figure 6, figure 7).

The concentration levels found in sediment from harbours and in mussels and biological effects in dogwhelks clearly gives cause for further concern about other effects from TBT pollution and possible more widespread damage to small and less conspicuous species.

Restrictions on the use of TBT as an antifouling agent on small boats have been enforced in several countries (France, England, USA, Sweden, Norway and Japan among others). These restrictions have caused indications of improvements (reduced concentration in sediment, biota and reduction in frequency of imposex). Investigations on imposex in dogwhelks in Norway indicate indirectly, that the occurrence of TBT may have been somewhat reduced along parts of the Norwegian coast.

Little is known about temporal trends of organotin contamination in Norway. Only limited data are also available on spatial distribution of organotin contamination in sediments (mostly lacking from areas outside harbours) and water. Investigations on possible effects on organisms apart form dogwhelks has yet not been performed in Norway. The Norwegian State Pollution Control Autority have from 1997, included analysis of TBT in dogwhelks and in blue mussels in their monitoring. Investigations on imposex in dogwhelks and intersex in *Littorina littorea* are also included.

Our results is in accordance with other studies which clearly show that TBT still is a widespread problem and posing a threat to sensitive species, at least in coastal areas where shipping traffic intensities are above a certain level and in the vicinity of harbours. Consequently it must be assumed that significant reduction in the all over occurrence and unintended effects of organotin compounds only can be achieved by enforcing regulations on the use of such compounds also on large vessels (>25 m).

## 6. References

- Anonymous, 1989. Miljøfarliga ämnen. Exempelliste och vetenskapelig dokumentasjon. Rapport 19/89 fra Kemikalieinspektionen. Stockholm. ISBN 0284-1188.
- Alzieu, C., J. Sanjuan, J.P. Deltriel and M. Borel, 1986. Tin Contamintion in Arcachon Bay: Effects on Oyster Shell Anomalies. Mar. Pollut. Bull. 17 (11):494-498.
- Beaumont, A.R. and P.B. Newman, 1986a. Low level of tributyl tin reduce growth of marine micro-algae. Mar. Pollut. Bull. 17:457-461.
- Beaumont, A.R. and P.B. Newman, 1986b. Some effects of low concentrations of tributyl tin on planctonic organisms. Wat. Sci. Tech., 18: 299.
- Bettin, C., Oehlemann, J. and Stroben, E., 1996. TBT-induced imposex in marine neogastropods is mediated by an increasing androgen level. Helgoänder Meeresunters. 50, 299-317.
- Bjørklund, I., 1987. Skeppsbottnfärgernas miljöeffekter (in Swedish). Report 7/87 from Kemikalieinspektionen, 15 p. ISSN: 0284-1185.
- Bjørklund, I., 1988. Miljøeffekter av tennbaserade skeppsbottenfärger (in Swedish).. Report 6/88 from Kemikalieinspektionen. ISSN 0284-1185.
- Blaber, S.J.M., 1970. The occurrence of a penis-like outgrowth behind the right tentacle in spent females of *Nucella lapillus* (L.). Proc. malac. Soc. Lond. 39, 231-233.
- Bryan, G.W., Gibbs, P.E., L.G. Hummerstone and G.R. Burt, 1986. The decline of the Gastropod *Nucella lapillus* around south-west England: Evidence for the effect of tributyltin from antifouling paints. J. mar. biol. Ass. U.K., 66:611-640.
- Bryan, G.W., P.E. Gibbs, R.J. Huggett, L.A. Curtis, D.S. Bailey and D.M. Dauer, 1989. Effects of Tributyltin Pollution on the Mud Snail, *Ilyanasa obsoleta*, from the York River and Sarah Creek, Chesapeake Bay. Mar. Pollut. Bull., 20:458-462.
- Bryan, G.W., G.R. Burt, P.E. Gibbs and P.L. Pascoe, 1993. *Nassarius reticulatus* (Nassariidae: Gastropoda) as an indicator of tributyltin pollution before and after TBT restrictions, J. mar. biol. Ass. U.K., 73:913-929.
- Bushong, S.J., Ziegenfuss, M.C. and Unger, M.A., 1990. Chronic tributyltin toxicity experiments with the Chesapeake Bay copepod, *Acartia tonsa*. Environmental Toxicology and Chemistry, 9: 359-366.
- Ellis, D.V. and A. Pattisina, 1990. Widespread Neogastropod Imposex: A Biological Indicator of Global TBT Contamination. Mar. Pollut. Bull., 21:248-253.
- Evans, S.M., A. Hutton, Kendall, M.A. and A.M. Samosir, 1991. Recovery in Populations of Dogwhelks *Nucella lapillus* (L.) Suffering from Imposex. Mar. Pollut. Bull., 22:331-333.
- Evans, S.M., T. Leksono and P.D. McKinnell, 1995. Tributyltin Pollution: A Diminishing Problem Following Legislation Limiting the Use of TBT-Based Anti-fouling Paints, 1995. Mar. Pollut. Bull. 30, 14-21.
- Evans, S. M., Evans, P.M. and Leksono, T., 1996. Widespread Recovery of Dogwhelks, *Nucella lapillus* (L.), from Tributyltin Contamination in the North Sea and Clyde Sea. Mar. Pollut. Bull. 32, 263-269.

- Fagerhaug, 1997. Kartlegging av miljøgifter i marine sediment i Møre og Romsdal. Report no. 4/97 from the County Governor of Møre and Romsdal, 26 p + appendix (ISBN 82-7430-104-8).
- Fent, K. and W. Meier, 1994. Effects of Triphenyltin on Fish Early Life Stages. Arch. Environ. Contam. Toxicol. 27:224-231.
- Fent, K., 1996. Organotin compounds in municipal wastewater and sewage sludge: contamination, fate in treatment process and exotoxicological consequences. The Science of the Total Environment 185, 151-159.
- Fjelldal, J. C., 1994. Materialstrømsanalyse av tinnorganiske forbindelser (Material flow analysis of organotin compounds), SFT-report no. 94:07, 43pp.
- Gibbs, P.E. and G.W. Bryan, 1986. Reproductive failure in populations of the dog-whelk, *Nucella lapillus*, caused by imposex induced by tributyltin from antifouling paints. J.mar. biol. Ass. U.K., 66:767-777.
- Gibbs, P.E, G.W. Bryan, P.L. Pascoe and G.R. Burt, 1987. The use of the dog-whelk, *Nucella lapillus*, as an indicator of tributyltin /TBT) contamination. J. mar. biol. Ass. U.K., 67:507-523.
- Goldberg, E.D., 1986. TBT: an environmental dilemma. Environment, 28:17-44.
- Granmo, Å. og R. Eklund, 1993. Undersökning av miljögifter längs Bohuskusten 1993. Göteborgs Bohus Läns Vattenvårdsförbund, Uddevalla 30/9 1993. (Unpublished report). 16 p. + appendix.
- Hall, L.W. Jr., 1988. Tributyltin environmental studies in ChesapeakeBay, Mar. Pollut. Bull. 19, 431-438.
- Harding, M.J.C., S.K. Bailey and I.M. Davies, 1992. TBT imposex survey of the North Sea. Annex 7:Norway. Scottish Fisheries working paper No 10/92.
- Horiguchi, T., H. Shiraishi, M. Shimizu and M. Morita, 1994. Imposex and organotin compounds in *Thais clavigera* and *T. bronni* in Japan. J. mar. biol. Ass. U.K., 74:651-669.
- Huggett, R.J., M. A. Unger, P.F. Seligman and A.O. Valkirs, 1992. The marine biocide tributyltin. Assessing and managing the environmental risks. Environ. Sci. Technol. 26 (2):232-237.
- IMO (International Maritime Organization) 1994. Use of triorganotin compounds in anti-fouling paints. Results of TBT monitoring studies. Marine Environment Protection Committee 35 session Agenda item 17, 7p.
- Johansen, K. and F. Møhlenberg, 1987. Impairment of egg production in *Acartia tonsa* exposed to tributyltin oxide. Ophelia, 27:137-141.
- Knutzen, J., L. Berglind and E. Brevik, 1995. Introductory studies in Norwegian harbours. Organochlorines and tributyltin in the common mussel 1993-1994 (In Norwegian with English summary). NIVA-report no. 3296 (ISBN-85-577-2786-5), 79p
- Konieczny, R., 1994. Sedimentundersøkelser og tiltaksvurdering i forbindelse med kaiutbygging og mineryddingsfartøy (P-6084), Haakonsvern 1994 . NIVA-rapport nr. 3145, 56p.
- Konieczny, R. 1996. Sonderende undersøkelser i norske havner og utvalgte kystområder. Fase 3: Miljøgifter i sedimenter på strekningen Ramsund-Kirkenes . NIVA-rapport nr. 3423, 117 s.

- Konieczny, R. and A. Juliussen, 1995a. Sonderende undersøkelser i norske havner og utvalgte kystområder. Fase 1: Miljøgifter i sedimenter på strekningen Narvik-Kragerø . NIVA-rapport nr. 3275, 185 s.
- Konieczny, R. and A. Juliussen, 1995b. Sonderende undersøkelser i norske havner og utvalgte kystområder. Fase 2: Miljøgifter i sedimenter på strekningen Stavern-Hvitsten . NIVA-rapport nr. 3365, 109 s.
- MAFF, 1993. Monitoring and surveillance of non-radioactive contaminants in the aquatic environment and activities regulating the disposal of waste at sea, 1991. Aquatic environment monitoring report No 36. Directorate of Fisheries Research, Lowestoft.
- Maguire, R.J., R.J. Tkacz, Y.K. Chau, G.A. Bengert and P.T.S. Wong, 1986. Occurrence of organotin compounds in water and sediment in Canada. Chemosphere, 15:253-274.
- Minchin, D., Stroben, E., Oehlmann, J., Bauer, B., Duggan C., B. and Keating, M., 1996. Biological indicators used to map organotin contaminants in Cork Harbour, Ireland. Mar. Pollut. Bull. 32:188-195.
- Nias, D. J., S. McKillup and K. S. Edyvane, 1993. Imposex in *Lepsiella vinosa* from Southern Australia. Mar. Pollut. Bull., 26:380-384.
- Niemistø, L., 1974. A gravity corer for studies of soft sediments. Havforskningsinst. Skr. Helsinki, 238:33-38
- Page, D.S., Dassanayake, T.D. and Gilfillan, E., 1995. Tissue Distribution and Depuration of Field-Exposed *Mytilus edulis*. Mar. Environ. Res. 40:409-421.
- Quevauviller, P., R. Lavigne, R. Pinel og M. Astruc, 1989. Organotins in sediments and mussels from the Sado estuarine system (Portugal). Environ. Pollut. 57:149-166.
- Ritsema, R., R.W.P.M. Laane og O.F.X. Donard, 1991. Butyltins in marine waters of the Netherlands in 1988 and 1989; concentrations and effects. Mar. Environ. Res. 32:243-260.
- Ruiz, J.M., Bachelet, G., Caumette, P. and Donard, O.F.X., 1996. Three decades of tributyltin in the coastal environment with emphasis on the Arcachon bay, France. Environ. Pollut. 93, 195-203.
- Rosales, D., F. Pablos and I.L. Marr, 1992, Speciation and GC retention indices of some organotin compounds in water. Appl. Organomet. Chem. 6:27-38.
- Shiraishi, H. and M. Soma, 1992. Triphenyltin compounds in mussels in Tokyo Bay after restriction of use in Japan. Chemosphere, 24:1103-1109.
- Short, J.W. og J.L. Sharp, 1989. Tributyltin in bay mussels (<u>Mytilus edulis</u>) of the Pacific coast of the United States. Environ. Sci. Technol. 23: 740-743.
- Short, J.W., S.D. Rice, C.C. Brodersen and W.B. Stickle, 1989. Occurrence of tri-n-butyltin-caused imposex in the North Pacific marine snail *Nucella lima* in Auke Bay, Alaska. Mar. Biol., 102:291-297.
- Stallard, M.O., S.Y. Cola og C.A. Dooley, 1989. Optimization of butyltin measurements for seawater, tissue, and marine sediment samples. Appl. Organomet. Chem. 3:105-114.
- Svavarsson, J and H. Skarphédinsdóttir, 1995. Imposex in the dogwhelk *Nucella lapillus* (L.) in Iceland, Sarsia, 80: 35-40.
- Thompson, J.A.J. og C. Stewart, 1994. Organotin compounds in the coastal biota of British Columbia an overview. Can. Tech. Rep. Hydrogr. Ocean Sci. no. 155. 11 p.
- Ten Hallers-Tjabbes, C.C, J.F. Kemp and J.P. Boon, 1994. Imposex in Whelks (*Buccinum undatum*) from the Open North Sea: Relation to Shipping Traffic Intensities. Mar. Pollut. Bull., 28:311-313.

- Uhler, A.D., T.H. Coogan, K.S. Davis, G.S. Durell, W.G. Steinhauer, S.Y. Freitas og P.D. Boehm, 1989. Findings of tributyltin, dibutyltin and monobutylin in bivalves from selected U.S. coastal waters. Environ. Toxicol. Chem. 8: 971-979.
- Uhler. A.D., G.S. Durell, W.G. Steinhauer og A.M. Spellacy, 1993. Tributyltin levels in bivalve molluscs from the east and west coast of the United States: Results from the 1988-1990 national status and trends mussel watch project. Environ. Toxicol. Chem. 12:139-153.
- Waite, M.E., M.J. Waldock, J.E. Thain, D.J. Smith og S.M. Milton. 1991. Reductions in TBT concentrations in UK estuaries following legislation in 1986 and 1987. Mar. Environ. Res. 32: 89-111.
- Valkirs, A.O., B. Davidson, L.L. Kear, R.L. Fransham, J.G. Grovhoug og P.F. Seligman, 1991. Long-term monitoring of tributyltin in San Diego Bay California. Mar. Environ. Res. 12: 151-167.
- Walsh, G.E., L. L. McLaughlin, M.K. Louise, C. H. Deans and E.M. Lores, 1986. Inhibition of Arm Regeneration by *Ophioderma brevispina* (Echinodermata, Ophiuroidea) by Tributyltin Oxide and Triphenyltin Oxide. Ecotoxicology and Environmental Safety 12:95-199.
- Zabel, T.F., Seager, J and Oakly, S.D., 1988. Proposed environmental quality standards for list II substances in Water. Organotins. Water Research center (UK), report TR255, 73pp.
- Zuolian, C. og A. Jensen, 1989. Accumulation of organic and inorganic tin in blue mussel. *Mytilus edulis*, under natural conditions. Mar. Pollut. Bull. 20: 281-286.

# Appendix A.

Concentrations of TBT in surface sediment (0-2 cm) in and near harbours along the Norwegian coast.

SK= Coastal map, HS = harbour map.

- **A1**. Sediments collected in the northernmost part of Norway (Kirkenes-Ramsund) in August 1994 (data from Konieczny, 1996).
- **A2**. Sediments collected within and near harbours along the Norwegian coast from Narvik to Kragerø (data from Konieczny and Juliussen 1995a, Konieczny 1994, Fagerhaug 1997).
- A3. Sediments collected within and near harbours along the Norwegian coast from Stavern to Hvitsten in March 1994 (data from Konieczny and Juliussen, 1995b)

**A1.** 

| Station | Sample | Station name        | Station                   | GPS       | GPS       | Depth | Мар       | TBT         |  |
|---------|--------|---------------------|---------------------------|-----------|-----------|-------|-----------|-------------|--|
| code    | no.    |                     | description               | Pos. N    | Pos. E    | (m)   | referance | (ng/g d.w.) |  |
|         |        |                     | (NN° NN.NN') (NN° NN.NN') |           |           |       |           |             |  |
| KIR     | 02A    | Kirkenes harbour    | Outer harbour             | 69 43.380 | 30 06.490 | 12    | SK116     | 6           |  |
| VAS     | 02A    | Vadsø harbour       | Inner harbour Ø           | 70 04.450 | 29 44.420 | 6     | SK114     | 17          |  |
| VAS     | 01A    | Vadsø harbour       | Inner harbour V           | 70 04.370 | 29 43.170 | 8.5   | SK114     | 12          |  |
| VAR     | 02A    | Vardø harbour       | Inner harbour             | 70 22.610 | 31 06.490 | 18    | SK114     | 110         |  |
| VAR     | 01A    | Vardø harbour       | Outer harbour             | 70 23.080 | 31 06.553 | 47    | SK114     | 12          |  |
| BÅT     | 02A    | Båtsfjord harbour   | Inner harbour             | 70 37.325 | 29 42.377 | 20    | SK294     | 45          |  |
| BÅT     | 01A    | Båtsfjord harbour   | Outer harbour             | 70 38.184 | 29 43.952 | 23    | SK294     | 61          |  |
| BER     | 02A    | Berlevåg harbour    | Inner harbour             | 70 51.553 | 29 05.776 | 7     | SK294     | 85          |  |
| HON     | 03A    | Honningsvåg harbour | Outer harbour             | 70 58.950 | 25 57.670 | 66    | SK103     | 6           |  |
| HON     | 01A    | Honningsvåg harbour | Storbukta                 | 70 59.600 | 25 58.200 | 30    | SK103     | 18          |  |
| HAM     | 02A    | Hammerfest harbour  | Inner harbour             | 70 40.020 | 23 41.170 | 30    | SK98      | 12          |  |
| ALT     | 02A    | Alta harbour        | Bossekop                  | 69 57.970 | 23 14.200 | 21    | SK96      | 12          |  |
| ALT     | 01A    | Alta harbour        | Elvebakken                | 69 58.750 | 23 19.300 | 25    | SK96      | 9           |  |
| SKJ     | 02A    | Skjervøy harbour    | Inner harbour             | 70 01.902 | 20 59.072 | 30    | SK94      | 15          |  |
| SKJ     | 01A    | Skjervøy harbour    | Outer harbour             | 70 02.113 | 21 01.346 | 80    | SK94      | 12          |  |
| TRØ     | 04A    | Tromsø harbour      | Inner harbour/ship        | 69 39.085 | 18 57.865 | 4     | SK87      | 190         |  |
|         |        |                     | yard                      |           |           |       |           |             |  |
| TRØ     | 03A    | Tromsø harbour      | Tromsøysund N             | 69 40.480 | 18 59.850 | 50    | SK87      | 10          |  |
| TRØ     | 02A    | Tromsø harbour      | Near fuel station         | 69 39.603 | 18 58.480 | 8     | SK87      | 51          |  |
| TRØ     | 01A    | Tromsø harbour      | Tromsøysund S             | 69 38.430 | 18 57.050 | 8     | SK87      | 21          |  |
| FIN     | 02A    | Finnsnes harbour    | Outer harbour             | 69 13.810 | 17 57.670 | 36    | SK83      | 49          |  |
| FIN     | 01A    | Finnsnes harbour    | Finnfjorden               | 69 13.470 | 18 03.320 | 65    | SK83      | 34          |  |
| HAR     | 04A    | Harstad harbour     | Outer harbour             | 68 47.976 | 16 33.594 | 74    | SK487     | 190         |  |
| HAR     | 03A    | Harstad harbour     | v/ Kaarbø M.V.            | 68 47.785 | 16 32.749 | 18    | SK487     | 2400        |  |
| HAR     | 02A    | Harstad harbour     | Mathiassen M.V.           | 68 47.389 | 16 32.875 | 12    | SK487     | 1060        |  |
| HAR     | 01A    | Harstad harbour     | Gangsåsbotn               | 68 47.192 | 16 33.063 | 34    | SK487     | 1550        |  |
| RAM     | 02A    | Ramsund harbour     | Main quay North           | 68 29.499 | 16 30.084 | 45    | SK230     | 11          |  |
| RAM     | 01A    | Ramsund harbour     | Main quay South           | 68 29.356 | 16 30.385 | 54    | SK230     | 26          |  |

### A2 [appendix A(continued)].

| Sation | Sample | Station name                    | Station  | GPS                    | GPS                    | Depth |           | ТВТ                  |
|--------|--------|---------------------------------|--|------------------------|------------------------|-------|-----------|----------------------|
| code   | no.    |                                 | description  | Pos. N<br>(NN° NN.NN') | Pos. E                 | (m)   | referance | (ng/g d.w.)          |
| NAR    | 01A    | Narvik harbour                  | NV inlet to harbour  | 68 25.655              | 17 22.693              | 29    | HS461     | <5                   |
| BOD    | 02A    | Bodø harbour                    | Midle harbour  | 67 17.126              | 14 22.299              | 25    | HS476     | 5                    |
| BOD    | 03A    | Bodø harbour                    | Inner harbour  | 67 17.561              | 14 23.330              | 20    | HS476     | 5                    |
| SAS    | 01A    | Sandnessjøen harbour            | Høvding as.  | 66 01.250              | 12 38.700              | 30    | SK57      | 8                    |
| SAS    | 02A    | Sandnessjøen harbour            | Outer harbour  | 66 01.461              | 12 38.243              | 18    | SK57      | 92                   |
| SAS    | 03A    | Sandnessjøen harbour            | Inner harbour  | 66 01.365              | 12 37.554              | 10    | SK57      | 2570                 |
| BRØ    | 02A    | Brønnøysund harbour             | Inner harbour  | 65 28.469              | 12 12.428              | 35    | HS488     | 10                   |
| NAM    | 04A    | Namsos harbour                  | Inner harbour  | 64 27.944              | 11 29.234              | 23    | SK47      | <5                   |
| TRO    | 02A    | Trondheim harbour               | Ila basin  | 63 25.957              | 10 22.590              | 10    | HS458     | 15                   |
| TRO    | 04A    | Trondheim harbour               | Nyhavna Dora   | 63 26.477              | 10 25.740              | 8     | HS458     | 60                   |
| KRI    | 01A    | Kristiansund harbour            | Vågen  | 63 07.068              | 07 44.180              | 27    | HS454     | 75                   |
| KRI    | 03B    | Kristiansund harbour            | Dalasundet   | 63 07.280              | 07 46.200              | 53    | HS454     | 17                   |
| MOL    | 02B    | Molde harbour                   | Outer harbour  | 62 43.868              | 07 12.829              | 55    | SK33      | <5                   |
| 13     |        | Tomrefjorden, indre             | Ship yard,<br>mechanical                                       | 62 35.15 <sup>3)</sup> | 06 55.60 <sup>3)</sup> | 20-35 | SK53      | 16000                |
| 29     | -      | Lyngsvika, v/Kværner<br>kleven  | workshop<br>Ship yard,<br>mechanical<br>workshop               | 62 19.50 <sup>3)</sup> | 05 50.70 <sup>3)</sup> | 15-45 | SK30      | 5600                 |
| 28     | -      | Dalasundet øst                  | Ship yard,<br>industrial<br>dumpsite                           | 63 07.19 <sup>3)</sup> | 07 46.30 <sup>3)</sup> | 10-50 | SK454     | 2500                 |
| 5      | -      | Dalasundet vest                 | Ship yard,<br>industrial<br>dumpsite, sewage<br>treatmentplant | 63 06.98 <sup>3)</sup> | 07 45.60 <sup>3)</sup> | 10-30 | SK454     | 1700                 |
| 18     | -      | Fiskerstranda-Kvalsundet        | *  | 62 26.54 <sup>3)</sup> | 06 16.85 <sup>3)</sup> | 15-35 | SK456     | 1500                 |
| 10     | -      | Nerbøvågen                      | Ship yard  | 62 48.90 <sup>3)</sup> | 06 54.10 <sup>3)</sup> | 5-15  | SK33      | 700                  |
| 20     | -      | Lyngnevika, near<br>Ulsteineast | Ship yard  | 62 20.50 <sup>3)</sup> | 05 50.25 <sup>3)</sup> | 12-24 | SK30      | 700                  |
| MÅL    | 01A    | Måløy harbour                   | Ulvesund N   | 61 58.189              | 05 08.867              | 46    | HS490     | 8                    |
| St.3   | -      | Haakonsvern                     | Navy harbour   | 60 20.55 <sup>1)</sup> | 05 14.3 <sup>1)</sup>  | 7     | SK21      | 103800 <sup>2)</sup> |
| St.8   | -      | Haakonsvern                     | Navy harbour   | 60 20.55 <sup>1)</sup> | 05 14.3 <sup>1)</sup>  | 11    | SK21      | 1040                 |
| St.9   | -      | Haakonsvern                     | Navy harbour   | 60 20.55 <sup>1)</sup> | 05 14.3 <sup>1)</sup>  | 14    | SK21      | 2650                 |
| St.12  | -      | Haakonsvern                     | Navy harbour   | 60 20.55 <sup>1)</sup> | 05 14.3 <sup>1)</sup>  | 4.6   | SK21      | 1150                 |
| St.13  | -      | Haakonsvern                     | Navy harbour   | 60 20.55 <sup>1)</sup> | 05 14.3 <sup>1)</sup>  | 7     | SK21      | 490                  |
| St.16  | -      | Haakonsvern                     | Navy harbour   | 60 20.55 <sup>1)</sup> | 05 14.3 <sup>1)</sup>  | 9     | SK21      | 100                  |
| FLO    | 03A    | Florø harbour                   | Inner harbour N  | 61 36.320              | 05 03.390              | 52    | HS479     | 12                   |
| HAU    | 02A    | Haugesund harbour               | HMV  | 59 24.350              | 05 16.150              | 30    | HS469     | 1266                 |
| SAN    | 01A    | Sandnes harbour                 | Inner harbour  | 58 51.660              | 05 45.203              | 26    | HS485     | 23                   |
| STA    | 02B    | Stavanger harbour               | Ulnesgr. SW  | 58 59.069              | 05 42.777              | 57    | HS455     | 55                   |
| STA    | 03A    | Stavanger harbour               | Bangarvågen  | 58 59.584              | 05 43.383              | 12    | HS455     | 83                   |
| EGE    | 03A    | Egersund harbour                | Inner harbour  | 58 27.125              | 05 59.900              | 6     | HS467     | 42                   |
| FAR    | 02A    | Farsund harbour                 | Outer harbour  | 58 05.480              | 06 48.530              | 16    | HS477     | 10                   |
| MAN    | 02B    | Mandal harbour                  | Middle harbour   | 58 00.590              | 07 26.365              | 18    | HS457     | 103                  |
| LIL    | 03A    | Lillesand harbour               | Inner harbour  | 58 14.800              | 08 22.809              | 19    | SK8       | 8                    |
| GRI    | 03B    | Grimstad harbour                | Outer harbour  | 58 20.161              | 08 36.450              | 40    | SK8       | 12                   |
| ARE    | 03B    | Arendal harbour                 | East harbour   | 58 27.596              | 08 46.825              | 32    | HS453     | 22                   |
| KRA    | 02A    | Kragerø harbour                 | Tangen ship yard   | 58 52.105              | 09 26.000              | 62    | SK6       | 285                  |

<sup>1)</sup> Approximate locations, sediment from several stations in the same area analysed (see Konieczny, 1994 for more details)
2) Approximate value (instrument response exceeded calibration curve)
3) Approximate position (5-6 stations in the same area, see Fagerhaug 1997)

A3 [appendix A(continued)].

| Sation | Sample | Station name       | Station                | GPS       | GPS            | Depth | Map       | TBT         |
|--------|--------|--------------------|------------------------|-----------|----------------|-------|-----------|-------------|
| code   | no.    |                    | description            | Pos. N    | Pos. E         | (m)   | referance | (ng/g d.w.) |
|        |        |                    |                        |           | N') (NN° NN.NN |       |           |             |
| STV    | 01A    | Stavern harbour    | Main quay              | 58 59.795 | 10 02.753      | 12 m  | SK2       | 19          |
| STV    | 03A    | Stavern harbour    | Inner harbour          | 59 00.473 | 10 02.602      | 6 m   | SK2       | <1          |
| VIK    | 01A    | Viksfjorden        | Outer fjord            | 59 01.214 | 10 05.900      | 32 m  | SK2       | 3           |
| SAF    | 01A    | Sandefjordsfjorden | Near discharge         | 59 04.959 | 10 14.775      | 56 m  | HS480     | 46          |
| SAF    | 02A    | Sandefjordsfjorden | Volvo/Tranga           | 59 06.144 | 10 14.051      | 35 m  | HS480     | 227         |
| SAF    | 03A    | Sandefjordsfjorden | Hystastranda           | 59 06.333 | 10 13.945      | 28 m  | HS480     | 152         |
| SAF    | 04A    | Sandefjordsfjorden | Pronova/Jotun          | 59 06.492 | 10 13.762      | 26 m  | HS480     | 269         |
| SAF    | 05A    | Sandefjordsfjorden | Kilen                  | 59 07.580 | 10 14.260      | 5 m   | HS480     | 284         |
| SAF    | 06A    | Sandefjordsfjorden | Sentralhavna           | 59 07.460 | 10 13.830      | 8 m   | HS480     | 7           |
| SAF    | 07A    | Sandefjordsfjorden | Mechanical<br>workshop | 59 06.850 | 10 13.850      | 12 m  | HS480     | 1374        |
| SAF    | 08A    | Sandefjordsfjorden | Slipway SW             | 59 07.130 | 10 13.800      | 16 m  | HS480     | 979         |
| SAF    | 09A    | Sandefjordsfjorden | Tranga N               | 59 06.760 | 10 13.720      | 22 m  | HS480     | 253         |
| SAF    | 10A    | Sandefjordsfjorden | Outer fjord            | 59 05.360 | 10 14.560      | 53 m  | HS480     | 50          |
| SAF    | 11A    | Sandefjordsfjorden | Dumping site           | 59 04.390 | 10 14.950      | 70 m  | HS480     | 166         |
| SAF    | 12A    | Sandefjordsfjorden | Reference station      | 59 04.060 | 10 14.950      | 85 m  | HS480     | 19          |
| MEF    | 01A    | Mefjorden          | Outer fjord            | 59 03.677 | 10 17.300      | 26 m  | SK2       | 1           |
| VRE    | 01A    | Vrengensundet      | Chemical factory.      | 59 10.150 | 10 23.670      | 30 m  | SK2       | 15          |
| BOL    | 01A    | Bolærne            | North                  | 59 13.475 | 10 28.875      | 31 m  | SK2       | 4           |
| TØN    | 01A    | Tønsbergfjorden    | Træla area             | 59 14.900 | 10 26.700      | 11 m  | HS468     | 48          |
| VAL    | 02A    | Valløy-area        | Valløy- area           | 59 16.286 | 10 31.176      | 73 m  | HS468     | <1          |
| ÅSG    | 01A    | Åsgårdstrand       | Outer harbour          | 59 21.100 | 10 28.622      | 13 m  | HS481     | <1          |
| HOL    | 02A    | Holmestrand-area   | Hydro                  | 59 29.380 | 10 19.190      | 45 m  | SK3       | 1           |
| HOL    | 03A    | Holmestrand-area   | Inner harbour          | 59 29.410 | 10 19.180      | 15 m  | SK3       | 131         |
| HOR    | 01A    | Horten harbour     | Channel outlet         | 59 24.927 | 10 30.200      | 12 m  | HS486     | 1           |
| HOR    | 03A    | Horten harbour     | Inlet N                | 59 26.662 | 10 29.204      | 10 m  | HS486     | <5          |
| HOR    | 04A    | Horten harbour     | Central harbour        | 59 25.998 | 10 28.547      | 19 m  | HS486     | <5          |
| HOR    | 05A    | Horten harbour     | Inner harbour          | 59 25.696 | 10 28.615      | 11 m  | HS486     | 540         |
| HOR    | 06A    | Horten harbour     | Mellomøya              | 59 27.030 | 10 27.673      | 76 m  | HS486     | 7           |
| HOR    | 07A    | Horten harbour     | Løvøya                 | 59 26.870 | 10 25.475      | 50 m  | HS486     | 5           |
| HOR    | 08A    | Horten harbour     | North West             | 59 26.877 | 10 23.650      | 88 m  | HS486     | <5          |
| MOS    | 01A    | Moss harbour       | Channel South          | 59 24.970 | 10 38.910      | 24 m  | HS482     | 1977        |
| MOS    | 02A    | Moss harbour       | Inner harbour          | 59 26.250 | 10 39.230      | 34 m  | HS482     | <5          |
| MOS    | 03A    | Mossesundet        | Kambo NSO              | 59 28.970 | 10 40.830      | 83 m  | HS482     | 10          |
| MOS    | 04A    | Mossesundet        | Son harbour            | 59 31.450 | 10 41.000      | 24 m  | SK4       | <5          |
| HVI    | 01A    | Hvitsten           | Katten                 | 59 35.800 | 10 38.510      | 20 m  | SK4       | 6           |

# Appendix B.

TBT in the blue mussel (*Mytilus edulis*) from Norwegian harbours (\*) and other localities from the Norwegian coast 1993-1994. The provisional background concentrations are regarded to be near 3 μg/kg d.w.. Y: Outer, I: Inner, M: Central, N: North; S: South. (From Knutzen *et al.*, 1995). Please note that the TBT concentrations presented in this table probably are considerably underestimated (2-3 times). For further information see chapter 2.

|                           |          | Location               | Blue mus              | sel           |               |
|---------------------------|----------|------------------------|-----------------------|---------------|---------------|
| County/locality/date      |          | Pos. N<br>(NN° NN.NN') | Pos.E<br>(NNº NN.NN') | μg/kg<br>w.w. | μg/kg<br>d.w. |
| ØSTFOLD                   |          |                        |                       |               |               |
| Sponvikskansen            | 18/10-94 | 59 05.27               | 11 12.90              | 138           | 1290          |
| Singløykalven             | 18/10-94 | 59 05.63               | 11 08.50              | 145           | 1593          |
| N. Asmaløy                | 18/10-94 | 59 04.70               | 10 57.70              | 131           | -             |
| *Papper                   | 18/10-94 | 59 06.82               | 10 51,42              | 206           | 1731          |
| AKERSHUS/OSLO             |          |                        | ĺ                     |               |               |
| Near Rambergøya           | 30/10-94 | 59 52.70               | 10 42.85              | 99            | ~600          |
| Solbergstrand             | 29/10-94 | 59 37.35?              | 10 38.95?             | 6             | ~35           |
| VESTFOLD                  |          |                        |                       |               |               |
| Færder                    | 29/10-94 | 59 01.55               | 10 31.70              | 17            | ~100          |
| BUSKERUD                  |          |                        |                       |               |               |
| Mølen                     | 29/10-94 | 59 29.25               | 10 30.15              | 42            | ~250          |
| TELEMARK                  |          |                        |                       |               |               |
| Arøy/Langesundsbukta      | 13/09-94 | 58 59.90               | 09.47.80              | 59            | 386           |
| *Kragerø                  | 13/09-94 | 58 51.81               | 09 25.20              | 47            | 255           |
| Bjørkøy/Breviksfj.        | 28/10-94 | 59 01.35               | 09 45.40              | 174           | ~1050         |
| AUST-AGDER                |          |                        |                       |               |               |
| *Risør                    | 13/09-93 | 58 43.30               | 09 15.20              | 57            | 259           |
| *Arendal I                | 12/09-94 | 58 27.45               | 08 46.40              | 39            | 188           |
| *Arendal II               | 03/10-94 | 58 27.46               | 08 46.50              | 411           | 2258          |
| *Grimstad                 | 12/09-93 | 58 20.28               | 08 36.18              | 84            | 393           |
| *Lillesand                | 12/09-93 | 58 14.70               | 08 23,45              | 75            | 296           |
| VEST-AGDER                |          |                        |                       |               |               |
| *Mandal                   | 11/09-93 | 58 00,58               | 07 27,45              | 20            | 87            |
| *Farsund                  | 10/09-93 | 58 05,80               | 06 48,90              | 84            | 462           |
| ROGALAND                  |          |                        |                       |               |               |
| *Egersund                 | 09/09-93 | 58 26,82               | 05 59,12              | 103           | 440           |
| *Sandnes                  | 08/09-93 | 58 53,12               | 05 44,48              | 138           | 762           |
| *Stavanger Y              | 08/09-93 | 58 59,50               | 05 42,90              | 229           | 1279          |
| *Stavanger M              | 08/09-93 | 58 59,40               | 05 43,78              | 387           | 2331          |
| *Stavanger I              | 08/09-93 | 58 58,35               | 05 43,80              | 418           | 2211          |
| *Kopervik                 | 08/09-93 | 59 16,95               | 05 19,40              | 311           | 1620          |
| *Haugesund Y              | 08/09-93 | 59 24,80               | 05 14,10              | 29            | 125           |
| *Haugesund I              | 08/09-93 | 59 24,55               | 05 16,40              | 361           | 2162          |
| HORDALAND                 |          |                        |                       |               |               |
| Espevær                   |          | 59 35.10               | 05 08,65              | 25            | 133           |
| *Bergen I (Nordnes)       | 23/10-94 | 60 24,03               | 05 18,15              | 719           | 4730          |
| *Bergen II (Laksevåg)     | 23/10-94 | 60 22,87               | 05 19,75              | 921           | 6099          |
| *Bergen III (Damsgård)    | 29/11-94 | 60 23,03               | 05 19,05              | 2564          | 16649         |
| *Bergen IV (Solheimviken) | 29/11-94 | 60 23,62               | 05 17,05              | 2641          | 15720         |

### Appendix B (continued).

|                           |                      | Location               |                       | Blue mus      | Blue mussel   |  |
|---------------------------|----------------------|------------------------|-----------------------|---------------|---------------|--|
| County/locality/date      |                      | Pos. N<br>(NNº NN.NN') | Pos.E<br>(NNº NN.NN') | μg/kg<br>w.w. | μg/kg<br>d.w. |  |
| SOGN AND FJORDANE         |                      |                        |                       |               |               |  |
| *Florø N                  | 04/09-93             | 61 36,12               | 05 03,45              | 390           | 2020          |  |
| *Bremanger                | 04/09-93             | 61 46,33               | 04 56,30              | 54            | 286           |  |
| *Måløy S                  | 04/09-93             | 61 56,07               | 05 06,90              | 252           | 1537          |  |
| *Måløy N                  | 04/09-93             | 61 58.41               | 05 05,60              | 208           | 1051          |  |
| MØRE AND ROMSDAL          | 0 11 05 55           | 01 50.11               | 05 05.00              |               | 1,00,1        |  |
| *Ålesund S                | 03/09-93             | 62 28,15               | 06 09,10              | 324           | 1976          |  |
| *Ålesund N                | 03/09-93             | 62 28,53               | 06 09,10              | 420           | 2560          |  |
| *Molde                    | 02/09-93             | 62 44,20               | 07 10,00              | 40            | 227           |  |
| *Kristiansund             | 02/09-93             | 63 06,90               | 07 44,20              | 268           | 1675          |  |
| SØR-TRØNDELAG             | 02/09-93             | 03 00,90               | 07 44,20              | 200           | 1073          |  |
| *Trondheim                | 30/08-93             | 63 26,85               | 10 25,73              | 73            | 415           |  |
| NORD-TRØNDELAG            | 30/06-93             | 03 20,63               | 10 25,75              | 13            | 413           |  |
| *Namsos                   | 30/08-93             | 64 27,85               | 11 29,00              | 37            | 263           |  |
| NORDLAND                  | 30/00-93             | 04 27,03               | 11 29,00              | 31            | 203           |  |
| *Brønnøysund              | 29/08-93             | 65 28,60               | 12 12,52              | 43            | 219           |  |
| *Sandnessjøen             | 28/08-93             | 66 01,50               | 12 12,32              | 147           | 700           |  |
| *Skrova                   | 02/09-94             | 68 10,05               | 14 40,25              | 392           | 2190          |  |
| *Narvik                   |                      | 68 25,55               | 17 25,70              | 31            | 161           |  |
|                           | 27/08-93             |                        | 16 30,70              | 1             | 3019          |  |
| *Ramsund                  | 16/08-94             | 68 29,70               | 16 30,70              | 468           | 3019          |  |
| TROMS                     | 1.6/00.04            | 60 47 74               | 16 22 62              | 726           | 1407          |  |
| *Harstad                  | 16/08-94             | 68 47,74               | 16 33,63              | 736           | 4407          |  |
| Feneset/Grytøya           | 02/09-94             | 68 56,10               | 16 39,00              | 63            | 391           |  |
| *Finnsnes                 | 17/08-94             | 69 13,65               | 17 59,50              | 42            | 247           |  |
| *Tromsø                   | 19/08-94             | 69 39,15               | 18 58,70              | 595           | 3542          |  |
| *Skjervøy                 | 19/08-94             | 70 01,85               | 21 00,50              | 96            | 582           |  |
| FINNMARK                  | 20/00 04             | 60.50.05               | 22.10.00              |               | 207           |  |
| *Alta                     | 20/08-94             | 69 58,85               | 23 18,80              | 51            | 307           |  |
| Elinheim/Sørøya           | 31/08-94             | 70 30,96               | 22 14,803             | 33            | ~200          |  |
| *Hammerfest               | 20/08-94             | 70 40.20               | 23 41,00              | 375           | 2142          |  |
| Smines/Magerøysund        | 30/08-94             | 70 58,35               | 25 48,50              | 7             | 36            |  |
| *Honningsvåg              | 21/08-94             | 70 58,90               | 25 58,20              | 368           | 2056          |  |
| *Mehamn I                 | 22/08-94             | 71 02,45               | 27 50,60              | 112           | 704           |  |
| *Mehamn Y                 | 22/08-94             | 71 02,55               | 27 50,35              | 108           | 688           |  |
| *Berlevåg                 | 22/08-94             | 70 51,45               | 29 05,90              | 68            | 450           |  |
| *Vardø                    | 22/08-94             | 70 22,50               | 31 06,90              | 569           | 3951          |  |
| Skagodden/Ekkerøy         | 02/09-94             | 70 04,15               | 30 10,10              | 40            | 242           |  |
| *Vadsø                    | 24/08-94             | 70 04,13               | 29 44,75              | 281           | 1799          |  |
| *Kirkenes I <sup>3)</sup> | 25/08-94<br>25/08-94 | 69 43,75               | 30 02,80              | 190           | 1397          |  |
| *Virkonas II 3)           |                      |                        |                       |               |               |  |
| *Kirkenes II 3)           | 25/08-94             | 69 43,75               | 30 02,80              | 329           | 2350          |  |

<sup>&</sup>lt;sup>3)</sup>Kirkenes: I=Mussels collected on stones. II=mussels collected on iron plate a few meters from Kirkenes I.