

Distribution of butyltins in the waters and sediments along the coast of India

Anita Garg*, Ram M. Meena, Sangeeta Jadhav, Narayan B. Bhosle

Marine Corrosion and Material Research Division, National Institute of Oceanography,

Council of Scientific and Industrial Research, Dona Paula- 403004, Goa, India

*Corresponding author E-mail anita@nio.org; Telephone +918322450202; Fax +91832450602

Abstract

Water and surface sediment samples were analyzed for butyltins (TBT, DBT, MBT) from various ports along the east and west coast of India. The total butyltin (TB) in water samples varied between ~ 1.7 and 342 ng Sn l⁻¹, whereas for sediments it varied between below detection limit to 14861 ng Sn g⁻¹ dry weight of sediment. On an average Chennai port recorded the highest level of butyltins in the sediments while Paradip recorded the highest level of butyltins in the waters. A fairly good relationship between the TB in the sediment and overlying water samples, as well as between organic carbon and TB, implicates the importance of adsorption/desorption process in controlling the levels of TBT in these port areas. In India the data on organotin pollution is very sparse; most of the port areas have been surveyed for butyltins for the first time during this study.

Keywords: antifouling, butyltins, water, sediment, ports, India

The use of Tributyltin (TBT) in antifouling paints increased, mainly due to its longer durability, high efficiency and reasonable cost, especially to control the fouling on recreational, commercial and naval ships and boats. Due to the widespread usage of organotins as an antifouling agent in boat paints, TBT is a common contaminant of the aquatic environment. TBT toxicity is known to have the following effects such as reduced growth and reproduction in organisms, can influence shell fishery, harm algal photosynthesis, decrease spat formation, cell malformation and imposex.

Once released from antifouling paints, TBT is rapidly absorbed by organic material such as bacteria and algae or suspended particles in the water (Luan et al., 2006; Blanca, 2008). Subsequently it is readily incorporated into tissue of filter feeding organisms and eventually, higher organisms such as fish, water birds and mammals where it accumulates (Berge et al., 2004; Ohji et al., 2007; Bangkedphol et al., 2009). Under favorable conditions TBT degrades through successive dealkylation to produce dibutyltin (DBT), monobutyltin (MBT) and ultimately to inorganic tin, via UV radiation, warmer temperature and biological activity, with biological activity being of greater importance. At TBT contaminated sites, water samples may show a low presence of TBT; however, because of bioaccumulation, animal tissue or sediment will show much higher concentrations. TBT has been classified as a persistent organic pollutant due to its persistent nature and bioaccumulative potential (Bangkedphol et al., 2009). The adsorption of TBT to sediment is reversible and contaminated sediment can act as a long term source of dissolved phase contamination to the overlying water column. Wide distribution, high hydrophobicity and persistence of organotin compounds have raised concerns about their bioaccumulation and its potential biomagnifications in the food web and its adverse effects to human health and environment.

The International Maritime Organization (IMO) has totally prohibited the usage of these compounds in antifouling paint formulation (Sonak et al., 2009; Blanca, 2008; Rodriguez et al., 2010). Nevertheless, the present restriction will unfortunately not immediately remove TBT, owing to its hydrophobicity and persistence in sediments, organotins will probably cause problems long after they have been banned, remaining a matter of concern and requiring monitoring for years to come (Bortoli et al., 2003; Blanca, 2008; Bangkedphol et al., 2009; Mukherjee et al., 2009). Monitoring of TBT in developing nations is limited and in most areas the use of TBT is yet to be regulated and there is virtually no data on the antifouling biocides in the marine environment (Mukherjee et al., 2009). In India TBT compounds have been used in antifouling paints and there is no ban as yet on the usage of TBT in antifouling paints. There are very few studies on the

distribution of butyltins in water and sediments (Rajendran et al., 2001; Bhosle et al., 2004, 2006a, Sonak et al., 2009; Meena et al., 2009; Garg et al., 2010).

The objective of this study focuses on the concentration of the antifoulant TBT and its degradation products, dibutyltin (DBT) and monobutyltin (MBT), in the waters and sediments from five ports along the east coast (Kolkata, Paradip, Vishakhapatnam, Chennai and Tuticorin, Fig. 1) and six ports along the west coast (Okha, Porbandar, Veraval, Jafrabad, Marmugao and Karwar, Fig. 2) of India affected by different intensities of maritime traffic.

Surface sediment samples were collected using a Van Veen grab, and transported to the laboratory using a portable icebox. In the laboratory, the samples were lyophilized, powdered and sieved through 120 μm mesh and stored at -20°C until analysis. Surface water (~ 1 m) samples were collected using a clean bucket and immediately transferred to the laboratory and the organotins extracted and stored in refrigerated condition until further processing.

Butyltin compounds from sediment and water samples were extracted and analyzed by GC-MS following the method of Garg et al. (2010). In brief freeze dried and sieved (120 μm mesh) sediment samples (1 g) were spiked with internal standard tripropyltin and extracted twice for 15 minutes with 15 ml of 0.03% of Tropolone solution in methanol and 1 ml of concentrated HCl by vortexing. The extracts were centrifuged for 10 minutes and liquid-liquid- extracted twice in a separating funnel with 15 ml of dichloromethane (CH_2Cl_2) and 100 ml of 5% NaCl in deionised water, (to avoid the formation of emulsion). The organic phase separated was dewatered with sodium sulphate and diluted with 1 ml of Isooctane and then reduced to almost dryness under a stream of nitrogen and treated with the derivatising agent pentylmagnesium bromide (Grignard's reagent) for 5 minutes at room temperature. The excess Grignard's reaction was destroyed by adding carefully, drop-by-drop 2 ml of deionised water. The derivatised butyltins were subsequently extracted twice with 2 ml of n-hexane and 5 ml of 1 M sulphuric acid. Then as a clean up step, the sample was reduced to 0.5 ml under a stream of nitrogen, and eluted with 10 ml of 1:1 hexane-toluene through a 6g column of activated flurosil. The eluate was finally evaporated to 0.5 ml under gentle stream of nitrogen and analyzed with gas chromatography. Separation and quantification of butyltin compounds were performed by GCMS. 1 μl of standard and samples were injected onto the GCMS.

For the water samples, 750 ml of seawater sample was taken in a Teflon separatory funnel, to which 100 ml of 5% NaCl in deionised water was added and the sample pH adjusted to 2 with HCl. The sample was then spiked with internal standard tripropyltin and extracted twice for 15 minutes

with 20 ml of 0.03% of tropolone in dichloromethane by shaking vigorously in a separatory funnel. The extracted organic phase was dewatered with sodium sulphate, to which 1 ml of Isooctane was added and then reduced to almost dryness under a stream of nitrogen and treated with the Grignard's reagent for 5 minutes at room temperature. The rest of the procedure followed was similar to that for the sediments, as mentioned above.

Total organic carbon (OC) was measured by the CN analyzer (Thermo electron corporation, Flash EA, 1112 series). For assessing the degradation of TBT, a degradation index for BT (butyltin) was calculated following the approach of Diez et al., 2006 and Garg et al., 2010.

The mean concentrations of TBT, DBT, MBT and TB in the seawater varied between 2.5 to 32, 1.4 to 110, 3.6 to 72 and 1.7 to 342 ng Sn l⁻¹ respectively (Table 1), wherein the highest value of TB (342 ng Sn l⁻¹) was observed at Paradip port. Chennai port had the highest average TBT concentration (32 ng Sn l⁻¹) in the water sample, especially at station 3 (103 ng Sn l⁻¹), which is most likely associated with the leaching of TBT containing antifouling paints from shipyard activities. This was followed by Kolkata (22 ng Sn l⁻¹), Gujarat (18 ng Sn l⁻¹), Karwar (14 ng Sn l⁻¹) and Tuticorin (2.5 ng Sn l⁻¹). Kolkata, Paradip, Chennai and Tuticorin are international ports whereas the ports of Gujarat and Karwar are small ports having small berthing capacity for large vessels and bigger berthing areas for fishing vessels. Relatively high concentrations of TBT at station 7 (55 ng Sn l⁻¹) of Gujarat and station 6 (40 ng Sn l⁻¹) of Karwar could be attributed to fresh release of TBT containing antifouling paints, as well as slow water flushing rates within these areas of the port. At Chennai, Tuticorin and Karwar it was observed that average TBT concentrations were higher than that of DBT (Table 1), indicating fresh inputs of TBT within these areas, similar to those reported by Rajendran et al. (2001). The TBT degradation index for the water samples varied between 52% to 92%, with Chennai having the lowest value (52%) suggesting a recent input of TBT, while Paradip had the highest (92%) degradation index (Table I), suggesting effective removal of TBT by microbial degradation and/or UV photolysis.

At Kolkata, Paradip and Gujarat ports, the mean values of DBT and MBT were higher than those of TBT (Table 1), probably because TBT is more hydrophobic and is less soluble in water as compared to DBT and MBT (Rajendran et al., 2001) and/or the degradation of TBT to DBT and MBT. Faster degradation of TBT in tropical water may be yet another reason for the abundance of DBT and MBT (Rajendran et al., 2001; Garg et al., 2010).

In recent investigations it has been reported that TBT concentrations in water are declining and maximum concentrations in marine waters rarely exceeds 100 ng l⁻¹ (Blanca, 2008), however we observe TBT value of 103 ng Sn l⁻¹ at station 3 of Chennai port (Table I). Moreover in India there are no water quality guide lines with respect to TBT or any legislation prohibiting the use of TBT based antifouling paints on ship hulls (Sonak et al., 2009, Mukherjee et al., 2009). However in order to determine the toxic effects we compared our results with water quality guidelines from other countries. From the 9 sampling stations, 8 of the sampling sites exceed the UK environment quality standard for seawater, 2 ng Sn l⁻¹ (UK Department of the environment, 1989), USEPA an aquatic life advisory concentration of 10 ng Sn l⁻¹ (US EPA, 1985), the Canadian water quality guideline of 1 ng Sn l⁻¹ (CCME, 1992) and the Dutch government environmental quality limit (9.7 ng TBT l⁻¹) (Rajendran et al., 2001).

The ratio TBT/DBT was used as an estimate of stability of butyltins (Tolosa et al., 1992; Uveges et al., 2007). Lower ratio indicates higher degradation of TBT, and can be used as an indication of old contamination. The average TBT/DBT ratios for Kolkata and Paradip were low between the range of 0.14 to 0.24 (Table 1), indicating old butyltin pollution or faster degradation of TBT, while all the stations at Chennai and a few at Tuticorin, Gujarat and Karwar exhibited higher ratios in the range of 0.8 to 3.85 (Table 1) indicating lesser degradation of TBT. The TBT/DBT ratios for water samples were lower as compared to sediments, likely due to a lower stability of TBT in water and/or a higher affinity for biota and sediments (Tolosa et al., 1992). Bacterial susceptibility to TBT biodegradation in water has been reported by Harino et al. (1998), and also warmer ambient temperature and intense sunlight are likely responsible for higher degradation index of TBT in the waters. The observation from our investigation compares well with many of the investigation reported earlier by others researchers from several other countries (Table 2).

The concentrations of butyltins in the sediments from east coast (Kolkata, Paradip, Vishakhapatnam, Chennai and Tuticorin) are presented in Tables 3, while those from the west coast, (Gujarat, Karwar, and Marmugao) are presented in Table 4. Besides Chennai, Tuticorin and Marmugao ports, butyltins in the other locations have been monitored for the first time during this study. The mean concentrations of TBT, DBT, MBT and TB in the sediments varied between 15 to 3712, 4 to 557, 10 to 554 and 29 to 4823 ng Sn g⁻¹ dw of sediment respectively, (Table 3 and 4). The TBT concentration in the sediments varied widely depending on the locations, from below detection limit to 11871 ng Sn g⁻¹ dw of sediment. The average TBT concentration was highest at Chennai port (3712 ng Sn g⁻¹) with maximum TBT concentration observed at dry docking station (11871 ng Sn

g⁻¹) followed by Ore jetty (4335 ng Sn g⁻¹) and coal jetty (2145 ng Sn g⁻¹) (Table 3). These high concentrations of TBT are probably due to low water current speed and poor water exchange within the Chennai port (Rajendran et al., 2001). At Vishakhapatnam port the concentrations of TBT and TB was very high at fish jetty (6381 and 10968 ng Sn g⁻¹) followed by dry dock (1306 and 2664 ng Sn g⁻¹) (Table 3) and was the second most polluted port, followed by Kolkata, Tuticorin, Karwar, Gujarat, Mormugao and Paradip ports (Table 3 and 4).

Amongst these Kolkata and Vishakhapatnam port are also poorly flushed ports (Mukherjee et al., 2009) and probably could be the reason for higher levels of butyltins in the sediments. This agrees with Kan-Atirekalp et al. (1997) who reported, highest level of TBT in sediments from Thailand coast was in areas where oceanic vessels and large commercial ships operated and also agrees well with Lee et al. (2006) who reported TBT released from ships were not flushed to open waters, but instead was deposited in the port, resulting in high TBT concentrations in sediments, compared to those in well flushed ports. These results also co-inside with higher TBT levels in areas with intense shipping activity with large berthing capacity. It appears that TBT accumulation in sediments from Indian ports was mainly affected by the location, water exchange rates within the port and intensity of shipping activity. The main pathway of TBT entering into marine environment is known to be through direct leaching from antifouling paints into water and then adsorption and partitioning to suspended particulate matter with subsequent sedimentation to sediments (Lee et al., 2006). Thus high levels of TBT in Indian coastal waters suggest its widespread contamination and indicative of usage of TBT based antifouling paints on ships in India

The TBT/DBT ratios for the sediments were higher than that of water samples. The transport of TBT associated with suspended particles in the water column and settling subsequently on the sediment surface could be one of the reasons for high TBT/DBT ratio, as has been suggested by Rajendran et al., (2001). Higher values were observed for the sediment samples from Vishakhapatnam and Marmugao ports (Table 3 and 4) as compared to other ports. This can be explained, since TBT is persistent in sediments where it is protected from photo or biodegradation thus resulting in higher values and could also be due to higher inputs of TBT attributed to the unrestricted usage of TBT in marine paints in India.

High concentration of TBT in sediments from some of the port areas may be of potential risk, for future TBT release due to re-suspension of sediments in the overlaying water. Page et al. (1996) and Axiak et al. (2000) have reported that soluble butyltins were released back into the water from heavily contaminated sediments. In the present study a significant positive correlation was observed

between TBT in the seawater and TBT in the sediments for Kolkata ($R^2 = 0.7359$, $p \leq 0.05$) Chennai ($R^2 = 0.8864$, $p \leq 0.001$), Tuticorin ($R^2 = 0.8708$, $p \leq 0.01$) and Karwar ($R^2 = 0.4457$, $p \leq 0.02$).

TBT exhibits both lipophilic and ionic properties, which means TBT levels will be influenced by the concentrations of organic carbon (Garg et al., 2010). A significant positive correlation was observed between organic carbon and TB for the sediment samples from Kolkata ($R^2 = 0.8756$, $p \leq 0.01$), Chennai ($R^2 = 0.6804$, $p \leq 0.5$), Tuticorin ($R^2 = 0.7813$, $p \leq 0.02$), Vishakhapatnam ($R^2 = 0.6197$, $p \leq 0.01$), Gujarat ($R^2 = 0.8402$, $p \leq 0.01$), Karwar ($R^2 = 0.6169$, $p \leq 0.05$) and Marmugao ($R^2 = 0.8931$, $p \leq 0.001$). Rajendran et al. (2001) similarly reported a good correlation between organic carbon and TB in the sediments from Chennai and Tuticorin ports. Pereira et al. (1999) reported that soil samples with the highest concentration of organic carbon contained the highest levels of butyltins at Mare Island, San Francisco Bay. It is apparent from these relationships in our study that adsorption/desorption played important role in controlling the levels of TBT in waters and sediments of these port areas.

In India there are no sediment quality guidelines for TBT contaminations. However in order to determine the potential ecological toxic effects, we used sediment quality guideline from other countries for comparison. According to Australian sediment quality guidelines TBT values of 5 ng g^{-1} and 70 ng g^{-1} dw of sediment indicate low and high trigger values, respectively (ANZECC/ARMCANZ, 2000), whereas Spanish port authorities have proposed Sediment Quality Guidelines of 10 ng g^{-1} and 200 ng g^{-1} (as Sn) for low and high trigger values (Rodriguez et al., 2010). All the sampled port areas had TBT levels higher than the low trigger value. Even at low levels ($< 10 \text{ ng l}^{-1}$) TBT can cause endocrine disruption, resulting in imposex, especially in gastropods (Hoch, 2001; Blanca, 2008). The observed levels of TBT compounds at all the areas sampled are higher than those known to induce imposex and imposex has been recorded in gastropods collected from Mormugao port (Bhosle, 2007). Bhosle (2006b) reported that oysters, mussels, clams and fish collected from nearby Mormugao port were contaminated with TBT, and TBT can suppress the normal functioning of the immune system of mammals. In view of this, it is possible that the benthic biota especially shellfish, in these areas could be contaminated with TBT and may pose a threat to human health.

For sediments the average butyltin degradation index varied from 34% to 70% with lowest value of 34% observed for Chennai port followed by Marmugao port with 40% (Table 3 and 4). Generally low degradation index values were observed for areas with high TBT levels. The low degradation index values for sediments can be explained based on the sorption/partitioning behavior

of TBT in sediment-water system. It has been reported TBT is highly adsorbed to sediment that are fine textured with abundant organic carbon (Burton et al., 2004) thus minimizing the bioavailable fraction amenable to degradation.

The results from our investigation compares well with many of the investigation reported from several other countries (Table 5). There are reports from a few countries where TBT regulations are being implemented, wherein a decrease in butyltin concentrations has been observed (Rodriguez et al., 2010). Although it would be difficult to draw a comparison with other areas as the field data of each location would be influenced by factors such as geographical location, hydrographic conditions, water temperature, biotic and abiotic factors, intensity of maritime activity etc.

The levels of TBT observed in the waters as well as sediments reflect on the usage of TBT in India. Based on our results the waters as well as the sediments from Chennai port were the most contaminated. Presence of DBT at most of the sampling sites, indicates photochemical and/or biological degradation taking place. Many of the areas have contamination levels high enough to pose a risk to aquatic and benthic organisms. The accumulation of TBT will remain a cause of concern for the aquatic environment for years to come due to its persistence in sediments. This study carries the first report on organotin contamination from most of the port areas sampled in India. Since the usage of TBT in antifouling paints is yet to be regulated in India, it is necessary to form legislations to control the usage of TBT compounds in antifouling applications in order to reduce their level in India.

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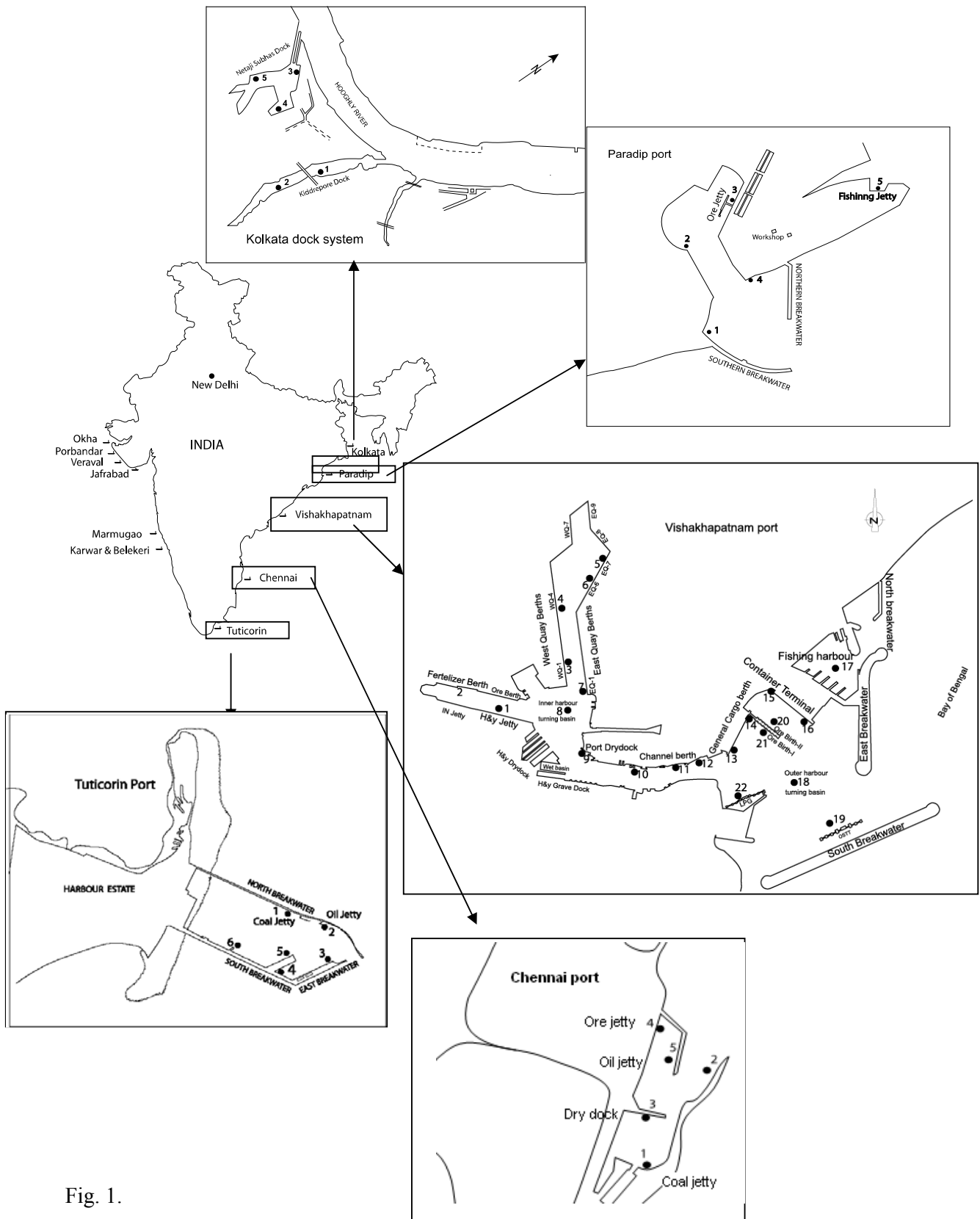


Fig. 1.

Maps showing sampling locations and stations of Kolkata, Paradip, Vishakhapatnam, Chennai and Tuticorin ports, along the east coast of India.

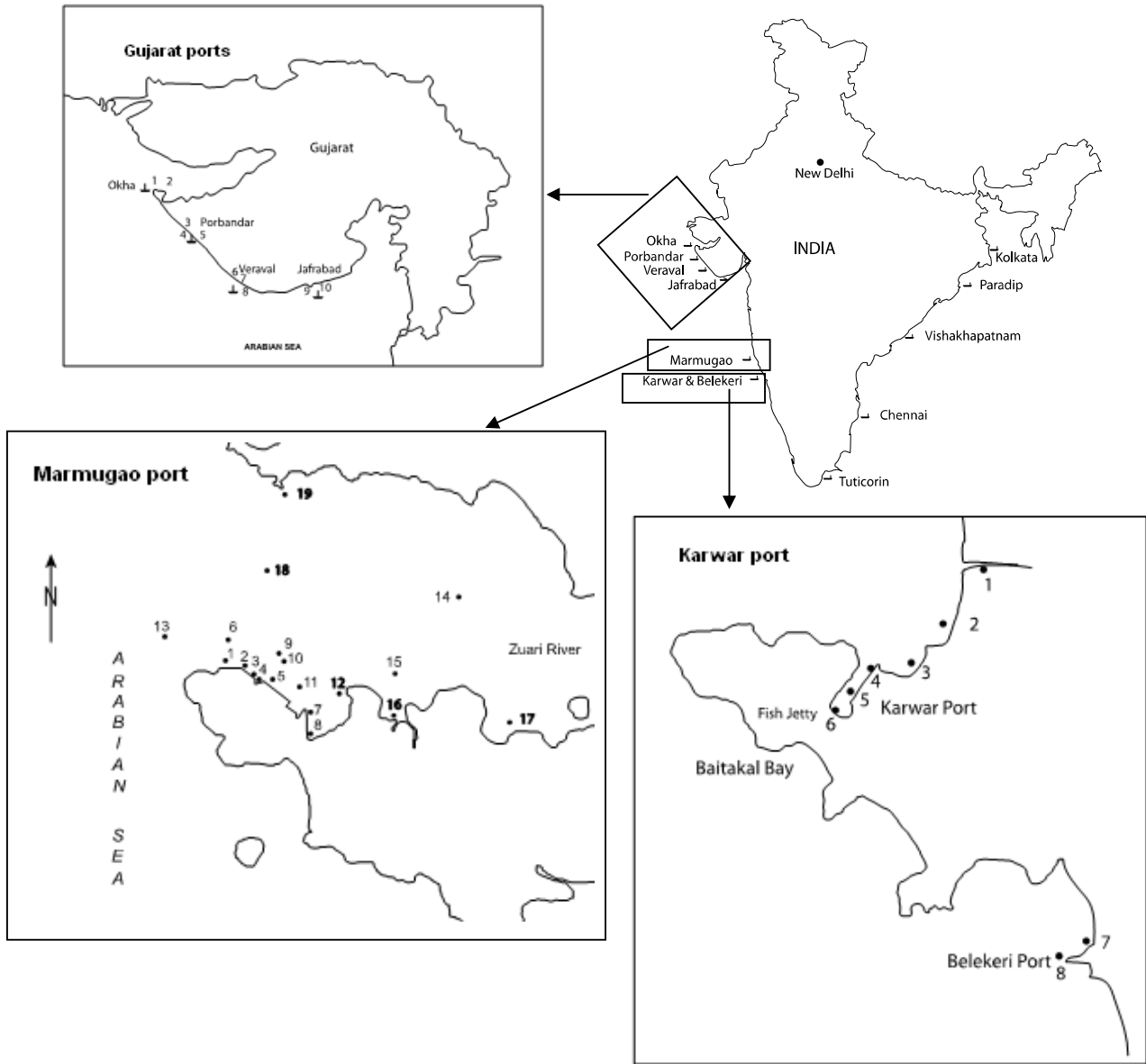


Fig. 2.

Maps showing sampling locations and stations of Gujarat, Karwar and Marmugao ports, along the west coast of India.

Table 1 Concentration of TBT, DBT, MBT, TB (ng Sn l⁻¹), TBT/DBT ratio, and BT deg % in the surface waters analyzed from several port areas along the coast of India.

Port Locations	Stations	TBT	DBT	MBT	TB	TBT/DBT	BT deg%
Kolkata	1 Berth 11	21 ± 1	70 ± 11.6	34 ± 2.1	126	0.30	83
	2 Berth 23	20 ± .3	87 ± 15.2	38 ± 4.1	145	0.24	86
	3 Berth 3	23 ± 5.6	90 ± 10.9	44 ± 2.4	158	0.26	85
	4 Berth 9	21 ± 1.1	98 ± 15.3	41 ± 5.6	159	0.21	87
	5 Berth 19	22 ± 4.5	104 ± 12.2	48 ± 5.4	174	0.21	88
	Mean		22	90	41	152	0.24
Paradip	1 Berth 1	14 ± 1.8	103 ± 13.3	17 ± 0.5	134	0.14	89
	2 Berth 2	16 ± 2.0	105 ± 18.8	41 ± 1.5	162	0.15	90
	3 Berth 3	22 ± 4.3	166 ± 25.3	154 ± 8.7	342	0.13	94
	4 Berth 4	18 ± 3.1	118 ± 19.2	40 ± 11	177	0.16	96
	5 Fish Jetty	7 ± 0.9	59 ± 9.9	93 ± 4.9	159	0.12	90
	Mean		16	110	69	195	0.14
Chennai	1 Coal Jetty	32 ± 1.3	6 ± 1.1	6 ± 1	44	5.33	28
	2 Entrance Channel	4.3 ± 0.8	0.8 ± 0.6	6 ± 0.3	11	5.12	61
	3 Dry Dock	103 ± 1.7	73 ± 0.6	79 ± 0.8	255	1.41	60
	4 Ore Jetty	14 ± 1.0	9 ± 0.9	5 ± 0.6	28	1.56	50
	5 Oil Jetty	6 ± 0.3	6 ± 0.27	4 ± 0.2	16	1.00	63
	Mean		32	19	20	71	2.88
Tuticorin	1 Coal Jetty	4 ± 0.9	0.2 ± 0.1	7 ± 0.6	11.2	17.39	64
	2 Oil Jetty	2.3 ± 0.7	2.3 ± 0.3	2.1 ± 0.3	6.7	1.00	66
	3 Cargo berth	0.1 ± 0.01	1 ± 0.2	0.9 ± 0.2	2.0	0.12	94
	4 Ore Berth	6.4 ± 0.3	2.2 ± 0.12	6.5 ± 1.2	15.1	2.91	58
	5 Berth 5	1.7 ± 0.12	1.7 ± 0.06	1.4 ± 0.7	4.8	1.00	64
	6 Naval Berth	0.7 ± 0.06	1 ± 0.02	ND	1.7	0.70	59
	Mean		2.5	1.4	3.6	6.9	3.85
Gujarat, Okha	1 Ferry Jetty	11 ± 0.9	16 ± 0.1	24 ± 1	51	0.69	78
	2 Fish Jetty	25 ± 0.7	16 ± 0.3	74 ± 3	115	1.56	78
	3 Custom Jetty	13 ± 0.18	24 ± 0.2	49 ± 2	86	0.54	85
Porbandar	4 Ashwoti Ghat	10 ± 0.3	30 ± 0.2	58 ± 8	98	0.33	90
	5 Fish Jetty	7 ± 0.12	29 ± 0.9	38 ± 7	74	0.24	91
	6 Fish Jetty	25 ± 1.2	19 ± 0.1	99 ± 5	143	1.32	83
Veraval	7 Back Ganar off.	55 ± 5	38 ± 0.8	138 ± 12	231	1.45	76
	8 Front Ganar off.	4 ± 0.23	25 ± 1	102 ± 10	131	0.16	97
	9 Fish Jetty	25 ± 2	16 ± 1.2	74 ± 10	115	1.56	78
Jafrabad	10 Light house	7 ± 1	36 ± 1.3	62 ± 3	105	0.19	93
	Mean		18	25	72	115	0.8
Karwar	1 Kodibagh bridge	4 ± 0.1	3 ± 0.2	16 ± 0.2	23	1.33	83
	2 Betkul off Karwar	7 ± 0.2	17 ± 0.5	13 ± 0.2	37	0.41	81
	3 Betkul Fish jetty	11 ± 0.2	14 ± 0.1	18 ± 0.2	43	0.79	74
	4 Karwar port I	18 ± 0.2	4 ± 0.4	12 ± 1.1	34	4.50	47
	5 Karwar port II	11 ± 0.4	8 ± 0.3	16 ± 1.3	35	1.38	69
	6 Karwar fish jetty	40 ± 1.2	12 ± 0.2	23 ± 0.3	75	3.33	47
	7 Bellekeri fish jetty	11 ± 0.1	7 ± 0.1	17 ± 1.0	35	1.57	69
	8 Bellekeri port	12 ± 0.1	5 ± 0.3	23 ± 2.0	40	2.40	70
	Mean		14	9	17	40	1.96

TBT: tributyltin; DBT: dibutyltin; MBT: monobutyltin; TB: total butyltin; BT deg%: butyltin degradation %

Table 2 Butyltin compounds in seawater (ng Sn l⁻¹) reported from several locations around the world.

Sampling		Levels of organotin compounds			Reference
Location	Year	TBT	DBT	MBT	
West and east coast of Canada	1995	b.d.l.–500	b.d.l.–270	b.d.l.– 460	Chau et al. (1997)
Korean coast	1997–1998	b.d.l.– 4.5	b.d.l.–22.3	b.d.l.–13.4	Shim et al. (2005)
North coast of Kyoto, Japan	2003	3.9–27	2.1–13	2.5–23	Ohji et al. (2007)
Marmugao harbour, India	2001	53–127	3.1–17.2	–	Bhosle et al. (2004)
Mandovi Estuary, India	2001	45.5–89.6	b.d.l.	–	Bhosle et al. (2004)
Goa coast, India	2007–2008	0.2–46	0.05–19	b.d.l.–29	Garg et al. (2010)
South east coast of France	1998	0.015–0.12	–	–	Michel and Averty (1999)
Coastal waters of Greece	1998–1999	b.d.l.–70	b.d.l.–159	b.d.l.–19	Thomaidis et al. (2007)
North west coast of Spain	Not provided	0.4–196	0.3 –33.7	0.8–11.6	Rodriguez-Gonzalez et al. (2006)
North coast of Spain	Not provided	0.1–6.2	0.1–0.9	0.2–0.8	Uveges et al. (2007)
Present study					
Karwar coast, India	2007	4–40	3–17	12–23	
Gujarat coast, India	2007	4–55	16–36	24–138	
Kolkata harbour, India	2008	20–23	70–104	34–48	
Paradip harbour, India	2008	7–22	59–166	17–154	
Chennai harbour, India	2008	4–103	1–73	4–79	
Tuticorin Harbour, India	2008	0.1–6	0.2–2.3	1–7	

TBT: tributyltin; DBT: dibutyltin; MBT: monobutyltin; b.d.l.: below detection limit.

Table 3 Concentration of TBT, DBT, MBT, TB (ng Sn g⁻¹), TBT/DBT ratio, BT deg % and OC % in the sediments analyzed from several port areas along the east coast of India.

Port Locations	Stations	TBT	DBT	MBT	TB	TBT/DBT	BT deg%	OC %
Kolkata	1 Berth 11	442 ± 42	344 ± 61	283 ± 17.4	1070	1.3	59	4.2 ± 0.2
	2 Berth 23	186 ± 2.2	143 ± 16	79 ± 4.5	407	1.3	54	2.1 ± 0.06
	3 Berth 3	66 ± 2.4	67 ± 0.6	40 ± 6.1	173	1.0	62	1.6 ± 0.02
	4 Berth 9	87 ± 3.8	74 ± 9.4	45 ± 4.4	206	1.2	58	1.5 ± 0.1
	5 Berth 19	82 ± 0.5	93 ± 0.7	115 ± 0.9	290	0.9	72	0.8 ± 0.02
Mean		173	144	112	429	1.1	61	
Paradip	1 Berth 1	10 ± 1.9	8 ± 2.5	19 ± 2.5	36	1.3	73	0.3 ± 0.01
	2 Berth 2	14 ± 1.9	8 ± 0.5	19 ± 1.3	41	1.7	66	0.7 ± 0.03
	3 Berth 3	20 ± 0.2	7 ± 0.1	13 ± 1.3	41	2.9	50	0.3 ± 0.01
	4 Berth 4	12 ± 0.9	3 ± 0.2	3 ± 0.5	18	4.3	59	0.5 ± 0.03
	5 Fish Jetty	4 ± 0.4	2 ± 0.8	4 ± 1.8	10	2.5	34	0.06 ± 0
Mean		12	5	12	29	2.5	56	
Chennai	1 Coal Jetty	2145 ± 39	394 ± 63	316 ± 51	2855	5.4	25	5.9 ± 0.5
	2 Entrance Chan.	54 ± 4	31 ± 3	25 ± 6	111	1.8	51	0.1 ± 0.01
	3 Dry Dock	11871 ± 176	1512 ± 87	1479 ± 133	14861	7.9	20	7.6 ± 0.1
	4 Ore Jetty	4335 ± 100	787 ± 105	865 ± 123	5988	5.5	28	3.6 ± 0.1
	5 Oil Jetty	157 ± 8	60 ± 11	85 ± 15	302	2.6	48	1.1 ± 0.06
Mean		3712	557	554	4823	4.6	34	
Tuticorin	1 Coal Jetty	149 ± 12	38 ± 2	ND	186	4.0	20	3.1 ± 0.12
	2 Oil Jetty	82 ± 4	41 ± 4	21.5 ± 2.1	144	2.0	43	2.2 ± 0.08
	3 Cargo berth	10 ± 0.5	22 ± 1	7.9 ± 1.2	40	0.4	75	0.6 ± 0.06
	4 Ore Berth	344 ± 33	10 ± 5	39.2 ± 4.5	483	3.4	29	3.6 ± 0.2
	5 Berth 5	35 ± 4	6 ± 1	ND	41	6.3	14	0.7 ± 0.1
	6 Naval Berth	4 ± 0.2	1 ± 0.2	8.6 ± 0.7	13	6.6	70	0.2 ± 0.02
Mean		95	34	19	144	3.8	46	
Vishakhapatnam	1 H & Y Jetty	4 ± 2	4459 ± 697	2998 ± 518	7461	0.0	100	5.2 ± 0.2
	2 Fertilizer Berth	364 ± 2	225 ± 2	496 ± 15	1085	1.6	66	3.3 ± 0.5
	3 WQB-1	144 ± 1	2 ± 0.01	378 ± 10	524	72.0	73	3 ± 0.31
	4 WQB-4	99 ± 1	7 ± 0.2	143 ± 5	249	14.1	60	1.8 ± 0.01
	5 EQB 7	121 ± 11	31 ± 3	162 ± 19	314	3.9	61	2.4 ± 0.3
	6 EQB-6	118 ± 13	72 ± 10	42 ± 5.6	232	1.6	49	2.2 ± 0.1
	7 EQB 1	245 ± 58	136 ± 32	72 ± 17	453	1.8	46	3.1 ± 0.21
	8 Inner Harbour	23 ± 3	4 ± 0.5	129 ± 11	156	5.8	85	0.6 ± .03
	9 Dry Docking	1306 ± 83	36 ± 0.5	1322 ± 27	2664	36.3	51	4.2 ± 0.22
	10 Berth 1	233 ± 10	200 ± 8	102 ± 8	535	1.2	56	2.1 ± 0.11
	11 Berth 2	133 ± 9	6.4 ± 0.1	286 ± 19	425	20.8	69	2.1 ± 0.09
	12 Berth 3	77 ± 2	2 ± 0.08	176 ± 3	255	38.5	70	1.8 ± 0.3
	13 GCBS	51 ± 5	8 ± 0.9	115 ± 5	174	6.4	71	2.7 ± 0.3
	14 GCBN	40 ± 1	4.3 ± 0.1	94 ± 1.2	138	9.3	71	4.4 ± 0.96
	15 CT 1	38 ± 4	7 ± 1	96 ± 9	141	5.4	73	0.6 ± .07
	16 CT 2	28 ± 4	6 ± 0.9	50 ± 6	84	4.7	67	3 ± 0.5
	17 Fishing Jetty	6381 ± 32	3911 ± 30	676 ± 6	10968	1.6	42	6.7 ± 0.23
	18 Outer Harbour	28 ± 5	12 ± 1.1	41 ± 5	81	2.3	65	0.7 ± 0.01
	19 OSTT	5 ± 1	3 ± 1	5 ± 1	13	1.7	62	0.4 ± 0
	20 Ore Berth II	56 ± 5	25 ± 3	25 ± 2	106	2.2	47	1.9 ± 0.01
	21 Ore Berth I	27 ± 2	18 ± 2	24 ± 2	69	1.5	61	1.3 ± 0.2
	22 LPG Berth	19 ± 2	34 ± 3	17 ± 5	70	0.6	73	1.2 ± 0.31
Mean		434	419	339	1191	11	64	

TBT: tributyltin; DBT: dibutyltin; MBT: monobutyltin; TB: total butyltin; BT deg%: butyltin degradation %; OC%: organic carbon %

Table 4 Concentration of TBT, DBT, MBT, TB (ng Sn g⁻¹), TBT/DBT ratio, BT deg % and OC % in the sediments analyzed from several port areas along the west coast of India.

Port Locations	Stations	TBT	DBT	MBT	TB	TBT/DBT	BT deg%	OC %
Gujarat,Okha	1 Ferry Jetty	3 ± 0.2	1.9 ± 0.2	6.5 ± 0.6	11	1.6	74	0.02 ± 0
	2 Fish Jetty	8 ± 1	19 ± 2	14.5 ± 2.1	42	0.4	81	0.1 ± 0
Porbandar	3 Custom Jetty	20 ± 2	18 ± 3	15 ± 2	53	1.1	62	0.12 ± 0
	4 Ashwmoti Ghat	1.3 ± 0.3	7 ± 1	3.3 ± 1	12	0.2	89	0.08 ± 0
Veraval	5 Fish Jetty	49 ± 9	102 ± 16	23 ± 3	174	0.5	72	0.9 ± 0.01
	6 Fish Jetty	78 ± 12	19 ± 1	17 ± 1.2	114	4.1	32	0.35 ± 0
	7 Back Ganar off.	16 ± 4	19 ± 2	31 ± 1.2	66	0.8	76	0.28 ± .02
	8 Front Ganar off.	15 ± 2	17 ± 2	15 ± 2.5	47	0.9	68	0.3 ± 0
Jafrabad	9 Fish Jetty	4.4 ± 0.4	4.4 ± 1	14 ± 2	23	1.0	81	0.24 ± 0
	10 Light house	1.4 ± 0.1	1.5 ± 0.2	16 ± 0.7	19	0.9	93	0.12 ± 0.01
	Mean	23	12	19	54	1.6	70	
Karwar	1 Kodibagh bridge	ND	3 ± 0.1	2 ± 0.1	4	0.0	-	0.01 ± 0
	2 Betkul off Karwar	3 ± 0.2	8 ± 0.6	8 ± 0.5	19	0.4	84	0.09 ± 0
	3 Betkul Fish jetty	4 ± 0.4	4 ± 0.1	4 ± 0.6	12	1.0	67	1.1 ± 0
	4 Karwar port I	49 ± 2	56 ± 6.2	69 ± 8	175	0.9	72	1.3 ± 0
	5 Karwar port II	26 ± 0.3	9 ± 0.7	6 ± 0.2	41	2.9	37	0.08 ± 0
	6 Karwar fish jetty	76 ± 5	52 ± 0.3	112 ± 12	241	1.5	68	1.35 ± 0.2
	7 Bellekeri fish jetty	5 ± 0	5 ± 0.7	10 ± 0.1	20	1.0	75	0.05 ± 0
	8 Bellekeri port	56 ± 6	40 ± 2.6	85 ± 2.3	183	1.5	68	1.03 ± 0
	Mean	43	33	57	132	1.5	64	
Marmugao	1 Berth 1	52 ± 1	11 ± 0.02	23 ± 0.4	86	4.8	40	2.2 ± .021
	2 Berth 3	55 ± 4	5 ± 0.2	61 ± 0.3	121	10.9	54	3.6 ± 0.6
	3 Berth 4	19 ± 2	8 ± 3.2	30 ± 0.7	56	2.5	67	1.8 ± 0.32
	4 Berth 5	128 ± 3	13 ± 3.3	35 ± 2.4	176	9.5	27	3.8 ± 0.15
	5 Berth 8	14 ± 2	8 ± 0.5	12 ± 1.2	33	1.7	59	1.1 ± 0.05
	6 Berth 8 (50m)	5 ± 0.3	1 ± 0.4	3 ± 0.3	9	3.2	50	0.1 ± 0
	7 Berth 9	25 ± 1	2 ± 0.5	ND	26	16.3	6	0.8 ± 0
	8 Berth 10	46 ± 3	13 ± 0.9	37 ± 1	96	3.5	52	2.3 ± 0.04
	9 Berth 11	38 ± 0.4	18 ± 0.5	19 ± 0.2	75	2.2	49	0.9 ± 0.07
	10 Berth 11(50 m)	40 ± 3	6 ± 0.02	8 ± 0.7	54	7.1	26	1.3 ± 0.06
	11 Dolphine I	ND	ND	ND	-	-	-	0.01 ± 0
	12 Dolphine II	6 ± 1	0.3 ± 0.04	4 ± 1.2	10	19.4	43	0.1 ± 0
	13 Marker buoy 6	2 ± 0.3	0.4 ± 0.02	10 ± 0.1	12	4.7	85	0.1 ± 0
	14 Marker buoy 7	4 ± 0.2	0.1 ± 0.03	6 ± 0.7	10	76.0	61	0.1 ± 0
	15 Vasco bay	0.1 ± 0.01	ND	ND	0.12	-	0	0.01 ± 0
	16 Goa shipyard	38 ± 3	3 ± 0.4	ND	40	13.4	7	1.1 ± 0
	17 CCM	34 ± 1	17 ± 0.8	20 ± 0.6	70	2.0	52	1.5 ± 0.03
	18 DP MP	29 ± 4	ND	2 ± 0.1	30	-	6	1.03 ± 0
	19 Dona Paula	8 ± 1	6 ± 0.3	17 ± 2.7	31	1.3	74	0.08 ± 0
	Mean	16	4	10	36	28	42	

TBT: tributyltin; DBT: dibutyltin; MBT: monobutyltin; TB: total butyltin; BT deg%: butyltin degradation %; OC%: organic carbon %

Table 5 Butyltin compounds in sediments (ng Sn g⁻¹ dw) reported from several locations around the world.

Sampling		Levels of organotin compounds			Reference
Location	Year	TBT	DBT	MBT	
West and east coast of Canada	1995	b.d.l.–5100	b.d.l. –1100	b.d.l. –330	Chau et al. (1997)
Crystal Lake, US	2001–2003	1.5–14,000 ^a	59–350 ^a	21–320 ^a	Landmeyer et al. (2004)
Port of Osaka, Japan	1995–1996	10–2100	b.d.l.	b.d.l.	Harino et al. (1998)
Coast of Malaysia	1997–1998	1.03–407 ^a	1.5–124 ^a	2.1–156 ^a	Sudaryanto et al. (2004)
Great Barrier Reef, Australia	1999	b.d.l.–1275	b.d.l. –71	b.d.l. –161	Haynes and Loong (2002)
Alexandria harbour, Egypt	1999	1–2076	0.1–379	<0.1–186	Barakat et al. (2001)
Kochi harbour, India	2000–2001	6–6228	n.a.	b.d.l. –213	Bhosle et al. (2006a)
Mumbai harbour, India	2000–2001	1.6–442	n.a.	b.d.l. –101	Bhosle et al. (2006a)
Goa coast, India	2007–2008	b.d.l.–128	b.d.l. –43	b.d.l.–67	Garg et al. (2010)
Fishing harbours, Taiwan	2001–2004	1–3165	n.a.	n.a.	Lee et al. (2006)
West coast of India	2001	1.8–883	b.d.l.–187	n.a.	Bhosle et al. (2004)
North coast of Kyoto, Japan	2003	1.2–19	2.3–23	4.3–22	Ohji et al. (2007)
Coast, Vietnam	2003	7.8–51	8.1–42.	3.9–30	Nhan et al. (2005)
Sanricu coast, Japan	2005	2–14,000	b.d.l. –3400	b.d.l.–3300	Harino et al. (2007)
West coast of France	1993	7–30	9–29	25–74	Ruiz et al. (1997)
River Thames, UK	1994	1–60	12–219	12–172	Scrimshaw et al. (2005)
South west coast of Spain	1998	1.2–130	2–284	2.5–95	Gomez-Ariza et al. (1998)
Tagus Estuary, Portugal	1998–1999	2–13	n.a.	n.a.	Nogueira et al. (2003)
Danish harbours and marinas, Denmark	1998–1999	37–1852	n.a.	n.a.	Jacobsen (2000)
North west Sicilian coast of Italy	1999–2000	3–27	b.d.l.	b.d.l.	Chiavarini et al. (2003)
North east coast of Spain	1995–2000	51–7673	47–3519	5–1131	Diez et al. (2002)
Coast, Portugal	1999–2000	3.8–12.4	5.3–65	5.2–78	Diez et al. (2005)
North coast of Spain	2000	50–5480 ^a	150–710 ^a	860–2870 ^a	Arambarri et al. (2003)
South west, France	2001	b.d.l.–89	b.d.l.–87	1.0–125	Bancon-Montigny et al. (2004)
Barcelona harbour, Spain	2002	98–4702	67–2607	35–440	Diez et al. (2006)
North west coast of Spain	2005	0.4–303	0.3–357	0.9–32.5	Üveges et al. (2007)
Gulf of Cadiz, Spain	2006	b.d.l.–573	b.d.l.–439	b.d.l.–797	Garg et al. (2009)
Present study					
Marmugao port, India	2007	b.d.l.–128	b.d.l.–18	b.d.l.–61	
Vishakhapatnam port, India	2008	4–6381	2–4459	5–2998	
Karwar coast, India	2007	b.d.l.–76	3–56	2–112	
Gujarat coast, India	2007	1–78	1–102	3–31	
Kolkata port, India	2008	66–422	67–344	40–283	
Paradip port, India	2008	4–20	2–8	3–19	
Chennai port, India	2008	54–11872	31–1512	25–1479	
Tuticorin port, India	2008	4–344	1–100	b.d.l.–39	

TBT: tributyltin; DBT: dibutyltin; MBT: monobutyltin; b.d.l: below detection limit; ^a Wet weight