ROPME/GC-16/3

ROPME MUSSEL WATCH PROGRAMME 2014

Technical Report: No.3

ORGANOTIN COMPOUNDS SCREENING

Prepared by:

MESL/IAEA
Monaco, December 2015

For:

REGIONAL ORGANIZATION FOR THE PROTECTION OF THE MARINE ENVIRONMENT

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1. INTRODUCTION

Under the ROPME-IAEA Contaminant Screening Project (1994-2005) and the ROPME Mussel Watch Programme (2011), surveys of heavy metal and organic contaminants have taken place in coastal areas of the Inner RSA and the Sea of Oman, collectively known as ROPME Sea Area (RSA).

The aim of the survey undertaken in February-March 2014 was to screen for Organotin compounds and organic contaminants in key coastal areas of Bahrain, I.R. Iran, Iraq, Oman, Saudi Arabia and the United Arab Emirates (UAE) and to compare the results with those from earlier surveys from the same areas. This report summarizes the results of organotin compounds in the sediments and bivalves in the RSA. This report should therefore be considered as a follow up report to the ROPME 2001 and 2013 monitoring reports.

2. SAMPLING METHODOLOGY

The following report concerns samples collected from February to July 2014 as part of the Mussel Watch Project, along the coast of Bahrain, I.R Iran, Iraq, Oman, Saudi Arabia and United Arab Emirates (UAE). The sampling stations, locations and the types of samples collected are given in Table 1, Figures 1 and 2.

Table 1. Sediment and biota sampling sites in the ROPME Sea Area (2014)

Country	Date	Site Name	Code	Station	Latitude	Longitude	Sample type		Remarks
K.Bh	2014-02-18	Askar *	Bah-5	1	26°3' N	50°37' E	Pearl Oyster	Sediment	
	2014-02-17	Marwada	Bah-9	1	26°18' N	51°26' E	Pearl Oyster	Sediment	
I.R.Iran	2014-02-17	Bushehr *	IRAN-2	1	28°49' N	50°52' E	Rock Oyster	Sediment	
	2014-02-19	Dayer	IRAN-2-1	1	27°49' N	51°54' E	Rock Oyster	Sediment	
	2014-02-20	Gavbandi	IRAN-2-2	1	27°7' N	53°1' E	Rock Oyster		
	2014-02-21	Chiru (new site)		1	26°43' N	53°47' E	Rock Oyster		
	2014-02-22	Lengeh *	IRAN-4	1	26°31' N	54°50' E	Rock Oyster		
	2014-02-24	Qeshm Island	IRAN-4-1	1	26°50' N	56°8' E	Rock Oyster		
Iraq	?/07/2014	Shat Al Arab		1			Pearl Oyster		10 Km offshore
OMAN	2014-04-01	Mirbat *	OMAN-8	1	17°0' N	54°40' E	Rock Oyster	Sediment	
	2014-03-14	Masirah	OMAN-8-1	1	20°40' N	58°50' E	Rock Oyster	Sediment	
	2014-02-08	Qalhat	OMAN-8-2	1	22°45' N	59°20' E	Rock Oyster	Sediment	
	2014-02-06	Mina Al Fahal *	OMAN-2	1	23°37' N	58°31' E	Rock Oyster	Sediment	
	2014-02-10	Sohar	OMAN-2-1	1	24°23' N	56°44' E	Rock Oyster	Sediment	
	2014-04-08	Khasab	OMAN-2-2	1	26°11' N	56°14' E	Rock Oyster	Sediment	
KSA	2014-03-06	Ras Tanura *	KSA-3	1	26°33' N	50°12' E	Pearl Oyster	Sediment	
	2014-03-08	Jubail	KSA-2-1	1	27°8' N	49°34' E	Pearl Oyster	Sediment	pleasance harbour (Fanateer)
	2014-03-09	KSA-2	KSA-2-2	2	27°18' N	49°38' E	Pearl Oyster	Sediment	30 km N. Jubail (Ras abu Ali)
	2014-03-09	Al Khafji	KSA-1	1	28°30' N	48°29' E	Pearl Oyster	Sediment	little No. of oysters
UAE	2014-02-13	Umm Al-Quwain *	UAE-7-1	1	25°35' N	55°33' E	Rock Oyster	Sediment	
		UAE-7	UAE-7-1	2			Rock Oyster	Sediment	
			UAE-7-1	3			Rock Oyster	Sediment	
			UAE-7-2	1			Pearl Oyster	Sediment	50 m offshore of R. Oyster stations
			UAE-7-2	2			Pearl Oyster	Sediment	
			UAE-7-2	3			Pearl Oyster	Sediment	
	2014-02-18	Dubai (Jebal Ali)	UAE-3	1	25°20' N	55°20' E	Rock Oyster	Sediment	

*Location sampled during the ROPME Contaminant Screening Programme (1994-2005) and Mussel Watch Programme 2011

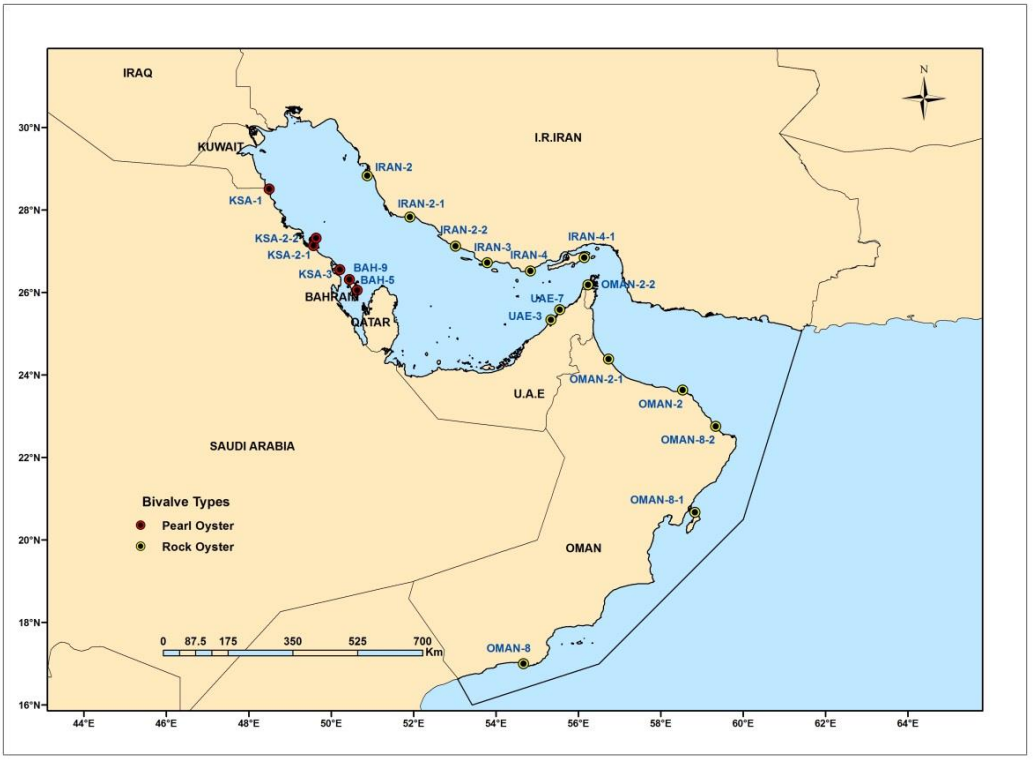

Figure 1. Sampling stations (ROPME Mussel Watch 2014) by code number

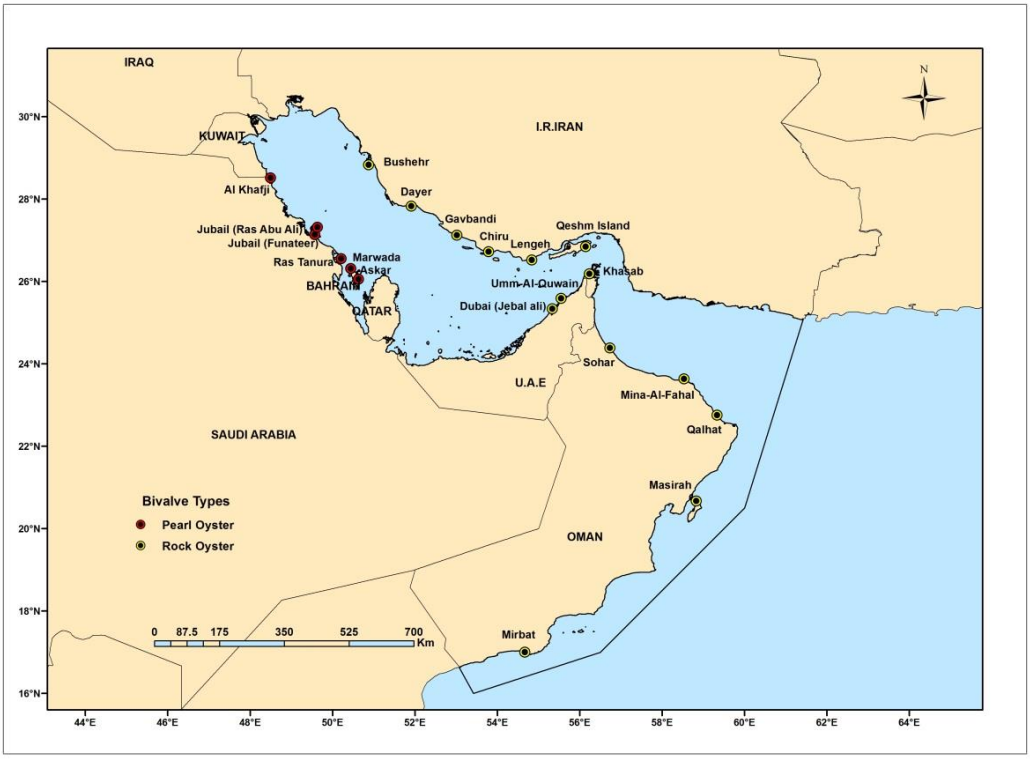

Figure 2. Sampling stations (ROPME Mussel Watch 2014) by name

3. ANALYTICAL PROCEDURES

The analytical protocols for measuring organotin compounds are detailed in this section.

3.1 SEDIMENT SAMPLES

Deuterated surrogates (Mono-n-butyl trichloride-D9 and Tributyltin-chloride-D27) were added to 3 to 5 grams of freeze-dried sediment samples. The slurry was leached by shaking with acetic acid, centrifuged and filtered. Using buffer and ammonia, the pH was stabilized between 5 and 6. The organotin compounds were simultaneously derivatized and extracted using sodium tetraethylborate (NaBEt4) and n-hexane. Elemental Hg was used to eliminate sulfur, and Florisil cartridges were used to clean up the samples. The purified samples were concentrated to about 0.5 ml (prior a solvent change from n-hexane to isooctane) and injected into a gas chromatograph mass spectrometer (GC/MS). The samples were quantified using a calibration curve made out of ethyl derivatives target compounds and deuterated ethyl derivatives surrogates (Mono-n-butyl triethyltin-D9, Ethyltributyltin-D27). Tetra-n-propyltin- D7 was used as an internal standard to quantify recoveries of surrogates.

Appropriate blanks were analyzed with each set of analyses and in addition, reference materials NRC SOPH-1 and BCR-462 (marine sediments), containing certified concentrations of organotin compounds were analyzed simultaneously along with each analytical batch. The results obtained for the certified reference materials were:

	SOPH-1 #1	***SOPH-1***	**BCR-462 *#1***	**BCR-462**
Compound	**ng/g as Sn**	**Certified as ng/g Sn**	**ng/g as Sn**	**Certified as ng/g Sn**
MBT	181		158	
DBT	180.3	174±9	52	34 ± 6
TBT	151.6	125±7	20	22 ± 6

3.1.1 Total organic carbon and carbonates

The contents of total carbon (TC) and total organic carbon (TOC) were determined by using a CHN analyzer (Vario EL Cube, Elemental Analyze system GmbH, Germany). Approximately 4-18 mg of dry sediment was encapsulated into a tin foil cup and introduced into a combustion furnace. Organic carbon was determined after treating the samples with concentrated H_3PO_4 to remove inorganic carbon. Concentration of total organic carbon is expressed as the percentage of total dry weight. Carbonate content was calculated by subtracting the TOC from the TC content. The quality control was realized by measuring TC and TOC levels in two certified reference material, SMR 1941b from NIST and a synthetic mixture for soil #4 from Eurovector.

Quality control for TC and TOC

	TC Certified value	TC Measured value	TOC Certified value	TOC Measured value
Soil#4	2.417%	2.31%	0.328%	0.38%
SMR 1941b	3.3%	3.11%±0.02	2.99±0.24	2.61±0.15 (n=2)

3.1.2 Grain size

Preparation of samples

The samples were sieved at 300 µm. Most of the samples have very small material below 300 µm. The portion of material above 300µm was recorded (by weighing).; the samples of Oman-8 and Oman-2-2 could not be prepared as not enough material was available.

Approximately an aliquot of 1 g (or less when not enough material was available, for a large number of samples it was a lot less which implies more uncertainty in the measurements) of sediment was put in a 10 ml tube. 5 ml of MilliQ water was added and the tube was shaken in order to separate the silt particles properly. An equilibration period of about 12 hours was used to insure that the sample was uniformly wet before analysis.

Particle Size Analysis

The particle size distribution was determined using a Malvern Instrument Mastersizer device. The principle of this device is that small particles cause incident light to be diffracted through a large angle whereas large particles will diffract incident light through a small angle. Particle size information is derived by deconvulation of the diffraction data obtained by the instrument.

Apparatus used

The MALVERN Mastersizer Micro v2.12 is designed to analyze particle size of silty sediments (<300 µm), the particles need to stay in suspension during the measurement process (this device is not suited for the analysis of coarse sandy material).

Protocol used

The analysis of the particles is achieved by slurring a sediment sample into a beaker containing 500 ml of water. The mixture is pumped through a cell which is interrogated by the instrument's laser beam. The particle size distribution is determined from the resulting diffraction pattern.

Results of grain size analysis

The size distribution in percentage for each sample is reported as:

% Sand = ∑ percentage of particulates between 300 µm and 63 µm

% Silts = ∑ percentage of particulates between 63 µm and 3.9 µm

% Clay = ∑ percentage of particulates below 3.9 µm

% Mud = ∑ % Clay and % Silt

3.2. BIOTA SAMPLES

The deuterated surrogates (Mono-n-butyl trichloride-D9 and Tributyltin-chloride-D27) were added to 0.5 g of freeze-dried biological samples. The slurry was dissolved (alkaline digestion) by sonication in an ultrasonic bath with tetra methyl ammonium hydroxide, TMAH (25% solution in water). Using buffer and acetic acid, the pH was stabilized between 5 and 6. The organotin compounds were simultaneously derivatized and extracted using sodium tetraethylborate (NaBEt4) and n-hexane. Florisil cartridges were used to clean up the samples. The purified samples were concentrated to about 0.5 ml (prior a solvent exchange from n- hexane to isooctane) and injected into a gas chromatograph mass spectrometer (GC/MS). Samples were quantified using a calibration curve made out of ethyl derivatives target compounds and deuterated ethyl derivatives surrogates (Mono-n-butyl triethyltin-D9 and Ethyltributyltin-D27). Tetra-n-propyltin-D7 was used as an internal standard to quantify recoveries of surrogates.

The appropriate blanks were analyzed with each set of analyses and in addition, the reference material ERM-CE477 (mussel tissue) and BCR-710 containing certified concentrations of organotin compounds was analyzed simultaneously along with each analytical batch. The results obtained for the certified reference materials are presented in Table 2.

Table 2. Results of the CRMS analysis

	BCR-710 #1	**BCR-710 #2**	**BCR-710**	**BCR-477 #1**	**BCR-477 #2**	**BCR-477**
Compound	**ng/g as Sn**	**ng/g as Sn**	**Certified as Sn**	**ng/g as Sn**	**ng/g as Sn**	**Certified as Sn**
MBT	56.4	48		1558	1534	1011 ± 182
DBT	35.7	48	41.76 ± 7.6	866	833	784 ± 61
TBT	88.9	84	54 ± 7.7	1150	1028	900 ± 77

GC-MS conditions used were as follows:

The organotin compounds were analyzed using selective ion monitoring (SIM) to enhance sensitivity. GC-MS conditions are presented in Table 3. Details of the acquisition are provided in the Table 4.

Table 3. GC-MS conditions

Gas Chromatograph	Agilent 6890 N
Detector	MSD 5975
Injection mode	Splitless
Carrier gas	Helium 1.6 ml min^{-1}
Column	DB-5 30 m x 0.25 mm i.d. x 0.25 µm film thickness
Injection specifications	inj. press.: 13 psi, Constant flow on 13 psi at 60°C, Temp. injector 270°C
Transfer line	280°C
Ion source	240°C
Analyser	100°C
Oven temperature program	60°C initial, 60°C to 120°C at 10°C min^{-1}, 120°C to 310°C at 4°C min^{-1}

Table 4. Selected masses for the analysis of ethylated organotin in GC-MS-SIM

Group	**Time** (min)	**Compounds** (m/z monitored)	**Cycles** (s^{-1})	**Ions** (m/z)
1	6-9	MBT-D9 (121, 179, 180, <u>244</u>)	3.65	179, 235, 121, 180, 244
		MBT (121, <u>179</u>, 235)		
2	9-12	TePrT-D27 (207, 121, 214, <u>256</u>)	3.07	121, 235, 263, 207, 214, 256
		DBT (121, <u>207</u>, 235, 263)		
3	12-15	TBT-D27 (121, 318, <u>217</u>)	1.72	121, 318, 217, 235, 263, 291
		TBT (235, 121, <u>263</u>, 291)		

Note: m/z used for quantification are underlined

4. RESULTS AND DISCUSSION

4.1. ORGANOTIN COMPOUNDS

4.1.1. Sediment samples

The concentrations of butyltins (BTs) compounds in sediments collected in February-March 2014 period (as part of the ROPME Mussel Watch Project, along the coast of Bahrain, I.R Iran, Oman, Saudi Arabia and UAE) are reported in the Table 5.

Table 5. Total organic carbon, carbonate content, percentage of fine particles (silt + clay <63 μm) and organotin compounds concentrations in sediment sample (ng g^{-1} as An)

Station	TOC (%)	Carbonates (%)	<63 μm (%)	MBT	DBT	TBT	ΣButyltins
BAH-5	0.97	9.43	5.8	7.1	<0.2	<0.2	7
BAH-9	0.61	10.59	0	<0.2	<0.2	<0.2	<0.6
IRAN-2	0.07	8.43	0.4	<0.2	<0.2	<0.2	<0.6
IRAN-2-1	0.1	7.2	63	<0.2	<0.2	<0.2	<0.6
OMAN-2	0.1	8.2	0	<0.2	<0.2	<0.2	<0.6
OMAN-2-1	0.06	1.64	0.6	<0.2	<0.2	<0.2	<0.6
OMAN-8-1	0.38	8.22	1	<0.2	<0.2	<0.2	<0.6
KSA-1	0.69	9.81	0	<0.2	<0.2	<0.2	<0.6
KSA-2-1	0.46	3.24	10.1	63	49	66	178
KSA-2-2	0.6	9.8	0	<0.2	<0.2	<0.2	<0.6
KSA-3	0.15	6.55	0	<0.2	<0.2	<0.2	<0.6
UAE-7-1	0.13	3.97	0	10	3.5	5.4	19
UAE-7-1-2	0.47	7.23	1.9	25	9.9	19	54
UAE-7-1-3	0.13	4.17	0.1	14	4.8	9	28
UAE-7-2	0.12	3.58	0	11	2.5	6.5	20
UAE-7-2-2	0.36	7.04	0.9	26	16	30	72
UAE-7-2-3	0.29	8.21	0.1	<0.2	<0.2	<0.2	<0.6
UAE-3	0.23	8.87	3.6	<0.2	<0.2	<0.2	<0.6

The concentration of butyltins were low in most sediments collected in I.R. Iran and Oman where the total concentration values were reported as <0.6 ng g^{-1} as Sn. On a total of 18 sediments analyzed, only the site of the pleasance harbor of Jubail in Saudi Arabia (KSA-2-1) and most of the sediments collected in Umm Al-Quwain presented concentrations of total butyltins ranging from 19 to 178 ng g^{-1} as Sn (Figure 3). Also the sediment from Askar in Bahrain exhibited a measurable concentration of monobutyltin as 7 ng g^{-1} as Sn.

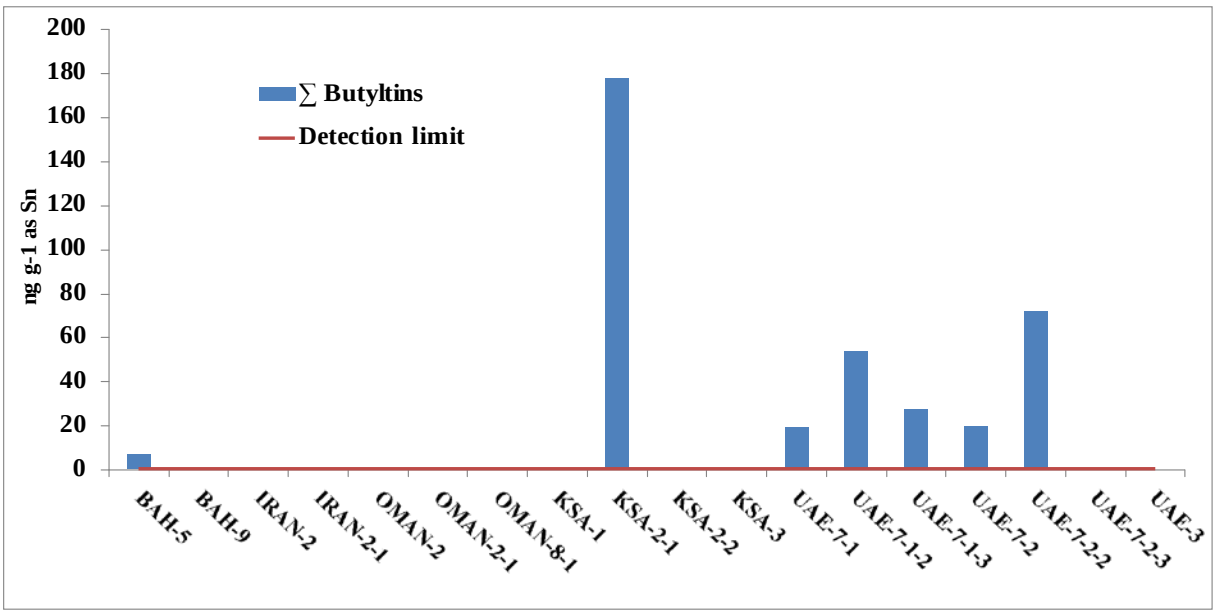

Figure 3. Total butyltins in sediments samples

The station of Jubail at the pleasance harbor of Fanateer (KSA-2-1) in Saudi Arabia and most of the samples from Umm Al Quwain, (excepting the UAEA-7-2-3 and UAE-3), were the sediments that could be considered contaminated with respect to TBT (i.e. TBT>1.3 ng g^{-1} as Sn) according to the classification scheme of Dowson *et al.* (1993). Taking in consideration the distribution of butyltin species within these samples (Figure 4) and the fact that TBT degrades only slowly in marine sediments (Stewart and de Mora, 1990) the high relative percentage of TBT in the site of Jubail in Saudi Arabia and UAE indicates that there has been a relatively recent input of TBT into the marine environment of these locations.

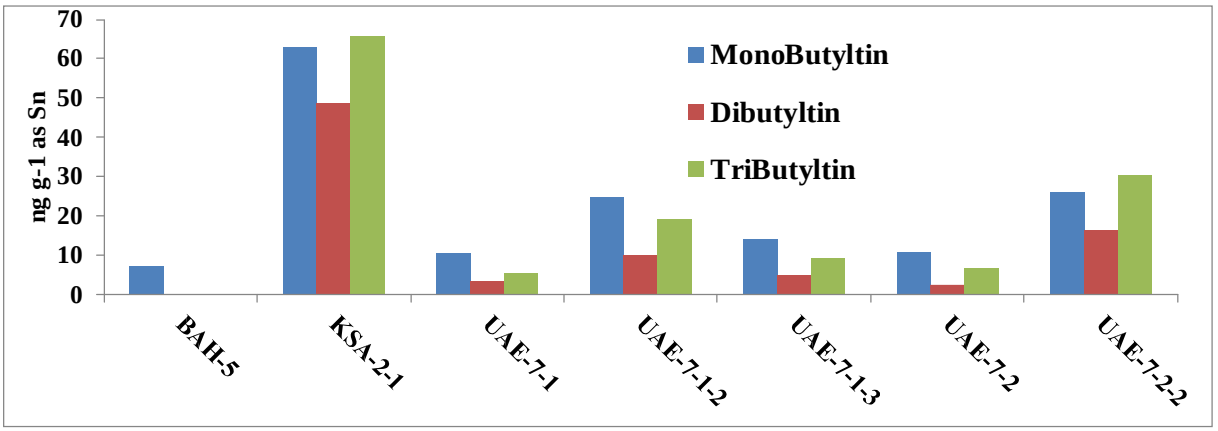

Figure 4. Butyltin species distribution in selected sediment samples

According to the Australian sediment quality guideline (SQG), TBT levels in sediments of 5 and 70 ng g^{-1} as Sn indicate low and high trigger values, respectively (Burton *et al*., 2005). As shown in Fig. 2, TBT concentration in all samples did not exceeded the Australian high trigger value of 70 ng g^{-1} as Sn, and were far lower than the literature values shown to have adverse effects on populations of sediment-dwelling bivalves (e.g. 300 ng g^{-1} as Sn, (Ruiz *et al*., 1997).

Previous studies (de Mora *et al.*, 2003; ROPME, 2013) reported very similar concentrations of butyltins, below and close to the detection limit, in sediments from Askar (Bahrain), Jebel Ali in UAE and Mina Al Fahal in Oman. Only a few samples collected in 2000-2001 close to the industrial complex in Bahrain (BAPCO), and in Oman (Hilf), close to a ferry terminal for small vessels, exhibited concentrations of total BTs of 80 and 72 ng g^{-1} as Sn respectively, similar to those measured in the Fanateer marina of Jubail in Saudi Arabia.

In conclusion the concentrations of total butyltins measured in most of the stations were very low, (below the detection limit of 0.2 ng g^{-1} as Sn). In some stations, relatively increased concentrations were detected, indicating local inputs, but the levels were relatively low compared to polluted European and Asian coastal sediments in which levels of 1000 to over 70000 ng g^{-1} as Sn are found especially in ports and marinas (Cassi *et al*., 2008, Kim *et al*., 2015).

4.1.2. Biota

The concentrations of BTs in bivalves collected in February and March 2014 as part of the ROPME Mussel Watch Project along the coast of Bahrain, I.R.Iran, Iraq, Oman, Saudi Arabia and UAE are reported in the Table 6.

Only 38% of the bivalve samples analyzed presented measurable concentrations of butyltins with maximum concentrations ranging from 40 to 46 ng g^{-1} as Sn in oysters from Askar (BAH-5), Fanateer (KSA-2-1) and Umm Al Quwain (UAE-7). Lower concentration levels were measured in rock oysters from Bushehr (18 ng g^{-1} as Sn in IRAN-2); and concentrations near the detection limit were found in Masirah (OMAN-8-1), as represented in the Figure 5. The total butyltin concentrations in oysters generally mirror those found in their respective sediments (Fig. 3), excepting for the site of Bushehr (IRAN-2), where organotins were only detected in the rock oysters and were not measurable in the sediment. Also, small amounts of organotins were detected in the rock oysters from Masirah (OMAN-8-1), but not in their sediment sample.

Table 6. Concentrations of lipids (mg g^{-1} dry wt) and organotin compounds in biota samples (ng g^{-1} as Sn)

Sample name	Lipids	MBT	DBT	TBT	∑ Butyltins
BAH-5	28	10.0	<1.2	36.0	46.0
BAH-9	47	<1.2	<1.2	<1.2	0.0
IRAN-2	43	<1.2	<1.2	17.9	17.9
IRAN-2-1	84	<1.2	<1.2	<1.2	0.0
IRAN-2-2	94	<1.2	<1.2	<1.2	0.0
IRAN	115	<1.2	<1.2	<1.2	0.0
IRAN-4	99	<1.2	<1.2	<1.2	0.0
IRAN-4-1	67	<1.2	<1.2	<1.2	0.0
IRAQ	86	<1.2	<1.2	<1.2	0.0
OMAN-8	82	<1.2	<1.2	<1.2	0.0
OMAN-8-1	114	1.8	<1.2	2.2	4.0
OMAN-8-2	74	<1.2	<1.2	<1.2	0.0
OMAN-2	72	<1.2	<1.2	<1.2	0.0
OMAN-2-1	56	<1.2	<1.2	<1.2	0.0
OMAN-2-2	136	<1.2	<1.2	<1.2	0.0
KSA-1	54	<1.2	<1.2	<1.2	0.0
KSA-2-1	57	18.6	<1.2	24.2	42.8
KSA-2-2	41	<1.2	<1.2	<1.2	0.0
KSA-3	77	<1.2	<1.2	<1.2	0.0
UAE-7-1	107	1.5	<1.2	16.5	17.9
UAE-7-1-2	110	8.1	10.0	17.7	35.9
UAE-7-1-3	95	3.3	<1.2	17.2	20.6
UAE-7-2	56	5.3	<1.2	34.7	40.0
UAE-7-2-2	47	3.3	<1.2	28.4	31.7
UAE-7-2-3	72	2.7	<1.2	26.0	28.7
UAE-3	103	<1.2	<1.2	<1.2	0.0

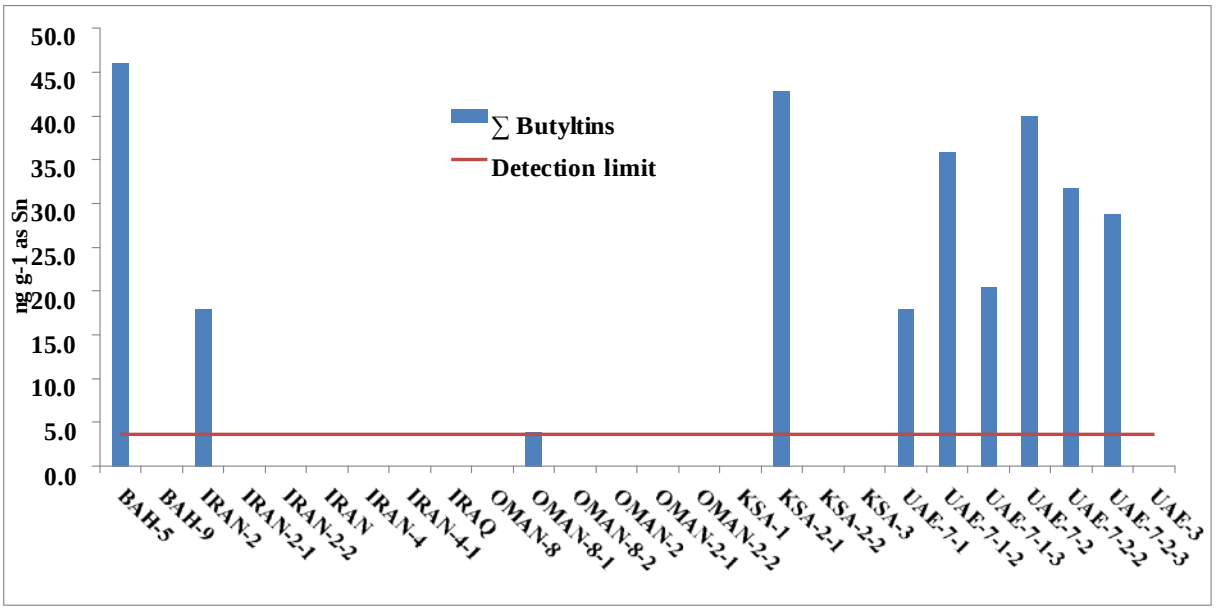

Figure 5. Total butyltins in biota samples

The Figure 6 reports the distribution of the BTs species in samples that presented measurable concentrations.

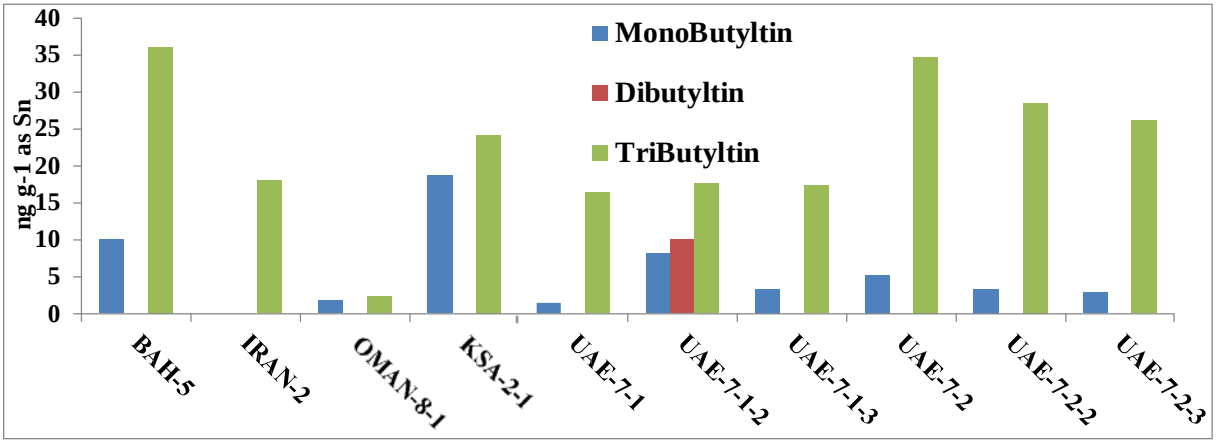

Figure 6. Butyltin species distribution in selected biota samples

Once released in the environment, butyltins are subjected to both biotic and photochemical debutylation from parent compounds (TBT) to their degradation products (DBT and MBT). Whereas in the water column microbial degradation and photolysis are the main debutylation processes, only anaerobic biodegradation can occur in sediments (Amouroux *et al.*, 2000). The degradation of TBT in water takes place within days to weeks (Stewart and de Mora, 1990), while the half-life was

half-life was estimated to be about 2 years in the sediments underlying a seawater marina (de Mora *et al.*, 1989).

Oysters are filter feeders organisms exposed to pollutants coming from the water (either dissolved or adsorbed on fine particles) and pollutants re-suspended from the surrounding sediments. Looking at the ratio between TBT and its main metabolites DBT and MBT, it appears evident that an exposure to contaminated seawater and thus, a relatively fresh input of TBT in the environment occurs in Bahrain, I.R. Iran, Saudi Arabia and UAE. Because of its long half-life, sediments polluted by TBT, could act as a constant secondary source of TBT to the water column and aquatic organisms for several years or even decades, despite the ban on its use (Shim *et al*., 2002; Viglino *et al*., 2004; Kim *et al*., 2014b). However, sediments from Askar, Bahrain, only exhibited the MBT metabolite and TBT and DBT concentrations were not detected. These results support a relatively recent input of TBT in marine waters.

In the previous contamination surveys undertaken in 2000 and 2011, much higher concentrations of butyltins from 140 to almost 500 ng g^{-1} as Sn were reported for the most contaminated sites of Abu Dhabi and Dubai in UAE and Hilf in Oman (de Mora *et al.*, 2003; ROPME, 2013)

Considering the range of butyltin concentrations in oysters from coastal sites in various regions of the world (Alzieu, 1996), the levels of BTs in bivalves from the ROPME Sea Area generally fall in the lower end of the range of typical concentrations.

Recently the OSPAR Commission (OSPAR, 2009, Nyberg *et al*, 2013) developed Environmental Assessment Criteria (EACs) for TBT concentrations in biota, based on ecotoxicological studies. The OSPAR proposed EAC value for mussels and oysters is 12 ng g^{-1} TBT (dry weight) (4.92 ng g^{-1} as Sn). EAC represents the contaminant's concentration in biota, below which no chronic effects are expected to occur in marine species, including the most sensitive ones. However, caution should be exercised when using these generic environmental assessment criteria in specific cases. EACs are based on ecotoxicological studies that do not take into account specific long-term biological effects such as carcinogenicity, genotoxicity and reproductive disruption due to hormone imbalances, and do not include combination toxicology. Also the OSPAR EACs have been developed using available scientific data that may not be directly applicable in all regions because of differences in local species and environmental conditions.

Half of the biological samples analyzed in this study were found to be below or close to the detection limit (TBT <1.2 ng g^{-1} as Sn) and thus below the EAC value for bivalves. However, TBT concentrations above the OSPAR EAC value were found in bivalves from stations at Bahrain, I.R. Iran, Saudi Arabia and UAE, indicating that biological effects are likely to take place in some of these areas. However, regarding the possible impact on public health from oyster consumption, the World Health Organization (WHO) limit for TBT uptake by food is set to 0.25 μg per kg of human weight per day (WHO, 1990), which corresponds to 17,500 ng of TBT per day for an average person of 70 Kg. Therefore, in order to reach the maximum safe uptake of TBT from oysters collected from the most TBT contaminated station at BAH-5 (36 ng g^{-1} dry weight as Sn, i.e. 87.8 ng g^{-1} dry weigh as TBT caution, which stands for 20 ng g^{-1} wet weight as TBT, using a dry/wet ratio of 0.23) an average person has to consume daily 0.87 Kg of fresh oyster flesh, which is not likely to happen. From this observation, it can be concluded that organotin concentrations in edible bivalves from the countries investigated seem not to pose immediate public health concerns. However, in order to ensure effective protection of public health from seafood contamination, the regular monitoring of TBT concentrations in oysters should continue, especially in areas where enhanced TBT concentrations were detected in locally grown oysters.

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