RESEARCH ARTICLE

Risk posed by chlorinated organic compounds in Abu Qir Bay, East Alexandria, Egypt

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Abstract

Introduction In Egypt, the picture of threats to humans and the environment from the exposure to organic pollutants is still incomplete. Thus the objectives of this study were to assess the occurrence and distribution of polychlorinated biphenyls (PCBs), organochlorine pesticides, and chlorpyrifos in sediments and mussels of Abu Qir Bay and their risks for environment and human health.

Materials and methods Twenty-three different compounds organochlorines were determined in 20 surfacial sediment and 10 mussel samples by gas chromatography-electron capture detector. A Screening Level Ecological Risk Assessment (SLERA) and a Human Health Risk Assessment (HHRA) were performed with the data.

Results and discussion Σ DDT (DDT, DDE, DDD) (average concentration 27 µg/kg dw) dominated the detected organic pollutants in the sediments, followed by CHLs (chlordane, heptachlor, heptachloro epoxide), hexachlorocyclohexane, chlorpyrifos, endosulfane, dieldrine, Σ 6 PCBs, aldrine, hexachlorobenzene, pentachlorobenzene, methoxychlor, and mirex. In general, concentrations of Σ 6 PCBs in mussels were higher than their corresponding sediment concentrations reflecting their relatively high bioavailability and bioaccumulative potential. However, concentrations

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M. A. E.-H. Khairy (⊠) · A. R. Mostafa · A. EL-Fiky Department of Environmental Sciences, Faculty of Science, Alexandria University, 21511 Alexandria, Egypt e-mail: mohammed_khairy77@yahoo.com of the organochlorine pesticides in mussels were lower than their corresponding sediment samples. Nevertheless, the SLERA on the bay sediments revealed that adverse ecological effects to benthic species are expected to occur whereas the HHRA showed that adverse health effects are not expected to occur from the consumption of the mussels.

Conclusions With the help of a SLERA, it was possible to indicate which class of chlorinated organic compounds is of highest concern to assess and to improve the environmental quality of the bay. Monitoring of organochlorines and chlorpyrifos would be needed to control the future trend of pollution.

Keywords $Organochlorines \cdot Chlorpyrifos \cdot PCBs \cdot$ Sediment \cdot Mussels \cdot Abu Qir Bay \cdot Risk assessment

1 Introduction

Coastal areas have suffered for decades from the uncontrolled discharges of untreated wastewater from urban activities. Although stringent legislations were developed worldwide in order to protect these highly productive ecosystems, illegal operations and inefficient coastal management plans, especially in developing countries, represent a true threat on the aquatic life and water quality of the marine environment. In Egypt and other developing countries, current data on the origin, production, use, environmental levels, effects, and trends of trace organic pollutants are still deficient or lacking such that the full picture of the threats to humans and the environment from the exposure to these substances is still not well-known (Barakat et al. 2002).

Few studies are available on the status of polychlorinated biphenyls (PCBs) and organochlorine pesticides in Abu Qir Bay (Abdallah 1992; Abdallah et al. 1992; Abdallah and

Abbas 1994). These studies are old and in most cases evaluation of the adverse effects associated with the recorded concentrations were not included. Recent studies (Khairy et al. 2009, 2011) showed that elevated concentrations of PAHs and heavy metals were observed in sediments and mussels of Abu Qir Bay, mostly arising from anthropogenic activities and adverse ecological effects to benthic species were expected from the exposure to Cd, Cr, Hg, and Zn contaminations of the sediments. The objectives of the present investigation were (1) to assess the occurrence and distribution of PCBs, organochlorine pesticides, and the organophosphate insecticide chlorpyrifos in sediments and mussels of Abu Qir Bay and (2) to conduct an environmental risk assessment to evaluate the possibility of occurrence of adverse ecological effects to benthic species from the exposure to chlorinated organic pollutants found in the bay sediments. The study extended to evaluate the possibility of occurrence of adverse health effects from the consumption of the mussel species based on the current ingestion rate.

2 Materials and methods

2.1 Study area

The present study was performed at Abu Qir Bay (Fig. 1). It is a semicircular shallow basin about 35 km east of

Alexandria city with a surface area of 360 km². The shoreline extends about 50 km between 30°40′–30°21′East and 31°30′North. Water depth ranges from less than 1 m along the coast to a maximum depth of about 15 m offshore (Nasr et al. 2004). Marine pollution originates from several land-based sources, mainly: Abu Qir Drain (AQD), which receives industrial wastes and brackish drainage water from agriculture of the northeastern part of the Nile Delta, Lake Edku via a channel called Maadeya outlet, which receives runoff water from agriculture and Rosetta Nile Branch (Fig. 1; Saad et al. 1980). Further details about the study area are given in Khairy et al. (2009).

2.2 Sampling

In April 2006, 20 surfacial sediment samples were collected from Abu Qir Bay using an Ekman grab sampler covering the whole western part of the bay till Maadeya Outlet (Fig. 1). The station 5 in the northern part of the bay near the open sea, 6 km away from land-based activities was selected as the reference site. In parallel, equal size specimens of mussel samples were collected by scuba divers at 10 of the 20 sampling sites as they were not available at all the sites. The mussels belonged to two different species: *Mactra corallina* (collected from seven sites in the middle and eastern part of the bay) and *Tapes decussata* (collected from three sites in the western part of



Fig. 1 Study area (Abu Qir Bay) with sampling sites

the bay). *T. decussata* was collected from the western part as *M. corallina* was not available in this area. All samples were placed in hexane-rinsed glass jars and were frozen at -20° C until sample preparation.

2.3 Determination of the physical-chemical properties of the sediment samples

Aliquots of sediment samples were oven dried at 100°C for 24 h to determine the dry mass. Sediment grain size analysis was performed according to Sweet et al. (1998). Total organic carbon (TOC) was determined in the dried sediment samples using DC-90 Total Organic Carbon Analyzer (Dohrmann, USA).

2.4 Extraction and clean up of the chlorinated organic pollutants

Homogenized wet sediment samples (60–70 g) mixed with 70–90 g anhydrous Na₂SO₄ p.a. (heated for 4 h at 500°C) and 70 g sea sand were ground in a porcelain mortar until a flowing powder was obtained. This mixture was extracted in a glass column (5 cm i.d. and 50 cm length) with 450 mL *n*-hexane/acetone (pesticide grade) (2:1 ν/ν) at a flow rate of about 1 mL/min according to Batarseh et al. (2003). The extract was rotary evaporated at 40°C and the solvent was exchanged to *n*-hexane. The extract was brought to a final volume of 1 mL under a gentle stream of nitrogen.

Approximately 10 g of each thawed mussel sample was homogenized with 70 g anhydrous Na_2SO_4 and 40 g sea sand as described for the sediments. The mixture was extracted overnight on an orbital shaker with 225 mL *n*hexane/acetone (pesticide grade; 2:1v/v). The lipid content of each sample was determined according to Quian et al. (1998). Extracts were concentrated to 1 mL as described for the sediment samples.

Sediment and mussel extracts were first cleaned up by column chromatography (2 cm i.d., 30 cm length) with 10 g Alumina 90 (neutral, 70-230 mesh activated at 185°C overnight and deactivated with 12.5% water). The elution of the organochlorines was performed with 100 mL nhexane/ethylacetate (7:3). The solvent of the eluate was exchanged to n-hexane and concentrated to 1 mL. A second clean up was performed with 10 g silica gel 60 (70-230 mesh, activated at 185°C over night) in a glass column (2 cm i.d., 30 cm length). PCBs, pentachlorobenzene, hexachlorobenzene, mirex, aldrine, a portion of heptachlor, and *p*,*p*'-DDE were eluted with 75 mL *n*-hexane (fraction 1). All other organochlorines were eluted in the second fraction (fraction 2) with 70 mL *n*-hexane/ethylacetate (1:1, v/v) together with PAHs (Khairy et al. 2009). Each fraction was evaporated, 1,2,3,4-tetrachloronaphthalene (TCN) was added as internal standard and the volume was adjusted to 1 mL. In order to remove sulfur that interferes with gas chromatography-electron capture detector (GC-ECD) analysis activated copper powder was added in excess to the two fractions.

2.5 Instrumentation

The prepared samples were analyzed for the following organochlorine pesticides: hexachlorocyclohexanes (HCHs; α -, β -, γ -, and δ -hexachlorocylohexane), CHLs (*cis*- and trans-chlordane, heptachlor, cis- and trans-heptachloro epoxide), \sum DDT (*o*,*p*'-DDT, *p*,*p*'-DDT, *o*,*p*'-DDD, *p*,*p*'-DDD, o,p'-DDE, p,p'-DDE), endosulfane (α - and β isomers), aldrine, dieldrine, methoxychlor, mirex, pentachlorobenzene (PCBz) and hexachlorobenzene (HCBz), chlorpyrifos and six indicator NDL-PCBs (nondioxin-like; PCB 28, 52, 101, 153, 138, 180) by using a Hewlett Packard HP 5890 series II GC equipped with ⁶³Ni ECD. The GC was equipped with a DB-5 fused silica capillary column (60 m length, 0.25 µm film thickness, 0.32 mm i.d. from J&W Scientific, Folsom, USA). For the analysis of both fractions, column temperature initially remained at 60°C for 5 min and then increased from 60°C to 160°C at 10°C/min and held at this temperature for 3 min. Then the temperature was programmed to 220°C at 2°C/min. Finally, the temperature was raised to 280°C at a rate of 8°C/min and held for 15 min. Helium was used as carrier gas with a flow rate of 2 mL/min. Nitrogen was used as the makeup gas and the total flow was set at 60 mL/min. The injector and detector temperatures were kept at 250°C and 300°C, respectively. By using a 60 m DB-5 columns problems of coelution were minimized. The identification of the peaks was confirmed by GC-ECD analysis with DB-1701 fused silica capillary column (45 m×1.0 µm×0.32 mm i.d. from J&W Scientific, Folsom, USA).

2.6 Quality assurance

To assure that no cross-contamination and interferences were caused during the analytical procedure, blank samples and pure standard samples were analyzed in each sample series. Generally, all the investigated analytes were below the limit of detection in the blanks. The calibration was performed by injection of standard mixtures at seven concentration levels using 1,2,3,4-TCN as internal standard. Recovery experiments were performed to check the extraction and clean up procedure. For that purpose, sediment and mussel samples were spiked with two concentration levels: 2.00 and 25.0 μ g/kg dw for sediments (from reference site, station 5), and 5.50 and 55.0 μ g/kg ww for mussel tissues (*Mytilus edulis*, from a German supermarket). For each concentration level, four spiked sediment samples were analyzed and the mean value was

calculated. The recoveries were all in the acceptable range of 70–110% (Table 1). The method detection limits (MDL) for the investigated organic pollutants were determined according to US Environmental Protection Agency (USEPA 1984; Table 1). For the quantification limit, we used the lowest concentration of seven matrix-spiked standard replicates with acceptable recoveries (70–110%) and percentage relative standard deviation (RSD%; \leq 20%). The relative standard deviation of the replicates in the fortification experiments and MDL determination was always lower than 20%. In case of p,p'-DDT and o,p'-DDT, it has to be considered that both compounds can be partly decomposed in the hot inlet system. So it was regularly checked by means of standard solutions that the thermal decomposition was below an accepted grade of 20%. But nevertheless, it must be considered that the degradation is matrix dependent and thus might change in samples with different matrix.

2.7 Correlation analyses

In order to examine whether TOC content and/or sediment grain size were the major factors controlling the distribution of PCBs and organochlorine pesticides in Abu Qir Bay, correlation analyses were performed using statistic software from SPSS statistics, version 13.

Table 1 Recovery rates of organochlorine compounds from sediment and mussels spiked at two levels and method detection limit data (MDL)

	Sedime	ents				Mussels				
	2 μg/kg	g dw (<i>n</i> =4)	25 μg/k	g dw (<i>n</i> =4)		5.5 μg/ł	cg ww (<i>n</i> =4)	55 μg/k	ag ww (<i>n</i> =4)	
	Mean	RSD (%)	Mean	RSD (%)	MDL (<i>n</i> =7)	Mean	RSD (%)	Mean	RSD (%)	MDL (<i>n</i> =7)
PCBz	91	1.1	84	4.5	0.083	83	9.7	96	9.4	0.21
HCBz	96	5.3	90	4.6	0.10	81	6.8	86	4.8	0.26
PCB 28	103	3.6	95	4.2	0.15	96	7.6	89	4.4	0.17
PCB 52	92	5.9	106	0.36	0.18	97	2.7	89	7.1	0.37
PCB 101	98	7.1	97	9.1	0.082	96	12	88	4.1	0.54
PCB 153	105	4.2	98	13	0.17	98	7.4	90	8.5	0.42
PCB 138	93	1.9	99	6.2	0.11	95	2.1	89	9.3	0.31
PCB 180	103	2.1	96	2.4	0.10	89	7.0	93	8.4	0.21
$\alpha ext{-HCH}$	80	8.5	92	4.0	0.15	95	2.3	90	1.1	0.42
β -HCH	94	10	109	0.96	0.23	102	3.0	100	0.87	0.25
γ -HCH	82	7.1	99	4.1	0.17	97	3.3	99	1.2	0.46
δ -HCH	85	8.3	106	3.3	0.15	104	3.5	99	3.1	0.39
Heptachlor	114	2.7	92	10	0.22	105	2.5	103	9.1	0.17
Chlorpyrifos	80	13	90	10	0.38	87	9.3	105	0.47	0.28
Aldrine	90	5.2	99	5.3	0.18	84	13	97	7.4	0.31
cis-Epoxide	90	10	105	7.0	0.25	81	1.8	ND	ND	0.32
trans-Epoxide	100	10	100	2.6	0.23	98	9.9	ND	ND	0.27
trans-Chlord.	99	7.7	103	5.3	0.37	92	2.4	87	1.5	0.38
cis-Chlordane	92	11	93	6.3	0.27	99	2.5	96	1.6	0.41
$\alpha ext{-Endosulfane}$	91	10	104	3.9	0.19	103	3.6	100	0.51	0.43
β -Endosulfane	88	1.6	102	6.1	0.13	101	2.6	104	5.5	0.28
o,p'-DDE	91	11	91	4.6	0.26	77	2.0	78	1.3	0.46
<i>p,p'</i> -DDE	84	2.6	90	2.8	0.18	87	2.9	105	2.7	0.25
Dieldrine	75	4.9	99	3.8	0.26	88	2.3	97	1.5	0.32
o,p'-DDD	86	12	91	7.6	0.16	94	4.3	105	1.1	0.30
<i>p,p'</i> -DDD	91	10	99	4.1	0.071	85	3.5	99	1.2	0.38
o,p'-DDT	97	6.0	91	7.4	0.14	92	3.3	93	3.4	0.64
<i>p,p'</i> -DDT	55	11	96	3.5	0.072	97	6.2	89	5.7	0.80
Methoxychlor	82	14	97	8.9	0.071	97	9.0	104	4.0	0.74
Mirex	94	5.4	96	3.6	0.12	94	9.2	99	10	0.34

ND not determined

2.8 Risk assessment

2.8.1 Ecological risk assessment

For evaluating the possibility of occurrence of adverse ecological effects from the exposure to chlorinated organic compounds in the sediments, a Screening Level Ecological Risk Assessment (SLERA) was performed

 $HQ = \frac{\text{maximum concentration of the organochlorine in sediments}(\mu g/kg \text{ dw})}{\text{sediment quality guideline of the organochlorine}(\mu g/kg \text{ dw})}$

However, several differing sets of SQGs for chlorinated organic compounds are reported in the literature. In consequence, consensus-based SQGs were used by calculation the geometric mean of the different reported SQGs according to the approach of MacDonald et al. (2000a, b). Furthermore, the consensus-based SQGs were divided in two groups: SQGs that are based on threshold effect concentrations (TEC) and thus intend to identify concentrations below which harmful effects are rarely observed, comparable with the NOAEL approach. For this group, the HQ (TEC HQ) was calculated according to the following equation:

TEC HO -	maximum concentration of the organochlorine compound in sediments ($\mu g/kg dw$)
TEC IIQ –	TEC (μ g/kgdw)

According to this equation, adverse ecological effects are only considered when TEC HQ >1.

2. SQGs that are based on probable effect concentrations (PEC) and thus intend to identify concentrations above

which harmful effects are frequently observed. This represents the worst case with a higher level of risk than the first group. The PEC HQ was calculated as follows:

PECHO -	maximum concentration of the organochlorine compound in sediments (μ g/kg dw)	
$\Gamma LC \Pi Q =$	PEC (μ g/kg dw)	

When PEC HQ>1, effects on sediment dwelling organisms are likely to be frequently observed.

When no consensus-based sediment quality guidelines were calculable due to the lack of guidelines, pollutants were evaluated by comparing the maximum detected concentration of each pollutant with screening concentrations developed by the USEPA (2004) and the Canadian Sediment Quality Guidelines (2003). Pollutants evaluated by this approach included α -HCH, chlorpyrifos, α - and β endosulfane, *cis*- and *trans*-heptachloro-epoxide, methoxychlor and PCBz.

2.8.2 Human Health Risk Assessment

Human Health Risk Assessment (HHRA) was performed according to USEPA (1989, 1997b) to assess the occurrence of adverse health effects (carcinogenic and noncarcinogenic)

as a result of the consumption of the mussels. The non-Carcinogenic Daily Intake or dose (CDI) for the ingestion of mussels was calculated based on the following equation:

$$CDI_{mussels-nc-ing} = \frac{C_{tis} \bullet EF \bullet ED \bullet IR \bullet C_{f}}{BW \bullet AT}$$

according to the framework of the USEPA (1992, 1997a, 1998). The protection of benthic species was selected as the assessment endpoint. For this purpose, the risk was estimated by comparing the maximum concentration of each organochlorine compound with data of Sediment Quality Guidelines (SQG, toxicity reference values) in form of hazard quotient (HQ) according to the following equation:

BW	body weight (70 kg)
AT	average time of exposure (365×30 days)
Cf	conversion factor of units $(10^{-3}/\text{kg})$

All used values were default values according to USEPA (1997b). Potential noncarcinogenic effects were evaluated by calculating a chronic HQ. For a single compound, the HQ is the ratio of the CDI to a reference dose (RfD):

$$HQ = \frac{CDI}{RfD}$$

RfD is a threshold dose or intake, which is conservatively chosen, so there is almost no possibility of an adverse health effect if the estimated intake is less than the reference dose (HQ<0.1). However, if the intake exceeds the reference dose (HQ>0.1) adverse noncarcinogenic effects could be expected.

In order to estimate the cancer risk, the CDI was multiplied by the cancer oral slope factors of the chemical investigated as shown in the following equation:

 $Risk = CDI \cdot OSF$

The cancer risk of 10^{-6} to 10^{-4} was taken as the critical range for a risk management decision (USEPA 1996). Further information about the formulated risk assessment can be found in Khairy et al. (2009, 2011).

3 Results and discussion

3.1 Sediments

Concentrations of PCBs and organochlorine pesticides in the sediment samples of Abu Qir Bay as well as TOC and clay content are summarized in Table 2.

According to the average concentrations, investigated organic pollutants can be arranged in the following descending order: \sum DDT (27 µg/kg dw)>CHLs (13 µg/kg dw)>HCHs (7.7 µg/kg dw)>chlorpyrifos (6.1 µg/kg dw)>endo-sulfane (3.9 µg/kg dw)>dieldrine (3.8 µg/kg dw)> \sum 6 PCBs (3.4 µg/kg dw)>aldrine (1.8 µg/kg dw)>HCBz (1.4 µg/kg dw)>PCBz (0.58 µg/kg dw)>methoxychlor (0.59 µg/kg dw)>mirex (0.15 µg/kg dw).

For all the investigated chlorinated organic compounds, the highest concentrations were always observed in the nearshore stations in front of the Paper Company, Abu Qir Drain, and the Fertilizer Company (stations 1, 2, 14, and 15), and to a lesser extent, in front of the Maadeya Outlet (stations 4 and 6). At these specific locations, also TOC contents of the sediments were higher than at the rest of the bay. As already presented in Khairy et al. (2011), TOC contents had a large variation ranging from <0.01% (station 7) to 12% (station 15) with the highest values also in front of Abu Qir Drain, Paper Company, and AQ Fertilizer Company (station 1, 2, 14, and 15). These are locations where settlement and accumulation of allochthonous materials occur which is influenced by the discharge of effluents from the land-based sources introducing TOC into the bay.

Relatively high concentrations of most of the investigated chlorinated organic pollutants and a comparable high TOC content of 2.4% were also observed in station 10 in front of the Wepco Petroleum Company in the seaward direction, which could possibly be related to the water circulation pattern inside Abu Qir Bay, where the water discharged from Lake Edku via Maadeya Outlet flows most of the year in the northern and the northwestern direction (location of station 10; Alam EL-Din and AL-Hogaraty 2001). Correlations of TOC with PCB and pesticides are presented in Sections 3.1.1 and 3.1.2.

In all the other locations, concentrations of the chlorinated organic compounds and TOC were generally lower or below the limit of detection (Table 2). These findings show the known influence of TOC on the distribution of organochlorine compounds in surface sediments as observed in several studies (Hu et al. 2009; Malik et al. 2009; Tang et al. 2007; Doong et al. 2002).

3.1.1 PCBs

PCB concentrations given as the sum of the six indicator PCB congeners (EFSA 2010) ranged from <MDL to 12 μ g/kg dw (stations 1 and 2) with an average concentration of 3.4 μ g/kg dw. None of the PCBs were detected in the sediments at the reference site (station 5), the western part of the bay (stations 18, 19, 20) and at stations 8 and 17. The spatial distribution of PCBs in the bay sediments revealed that PCB contamination is spread in the whole bay except for the western part.

No published data are available about the total release of PCBs into the bay and it is also not known whether any point source exists (Barakat et al. 2002). However, the widespread use of PCBs in fluids of transformers, electrical equipments, ship paints, and several other industrial products is supposed to be responsible for their entry into the sediments of Abu Qir Bay.

An intermediate degree of correlation was observed between concentrations of PCBs in sediments of Abu Qir Bay and TOC (r=0.67 at p<0.05) and clay (r=0.71 at p<0.05). Consequently, the highest concentrations of PCBs were observed in the sediment samples with the highest TOC and also relatively high clay content at stations 1 and 2. Therefore, the spatial distribution of PCBs in the bay sediments is influenced by the physical and chemical properties of the sediments to a great extent.

PCB congeners in sediments of Abu Qir Bay were mainly dominated by penta, hexa and hepta chlorinated

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Station no.	PCBs ^a	$\Sigma DDT^{\rm b}$	HCHs ^c	Chlorpyrifos	CHLS ^d	Aldrine	Dieldrine	Endosulfane ^e	Methoxychlor	Mirex	PCBz	HCBz	TOC (%)	Clay (%)
1	12	128	35	30	54	9.6	18	13	<mdl< td=""><td><mdl< td=""><td>0.39</td><td>0.20</td><td>5.8</td><td>14</td></mdl<></td></mdl<>	<mdl< td=""><td>0.39</td><td>0.20</td><td>5.8</td><td>14</td></mdl<>	0.39	0.20	5.8	14
2	12	116	30	42	54	8.3	12	12	<mdl< td=""><td><mdl< td=""><td>0.58</td><td>0.13</td><td>6.2</td><td>32</td></mdl<></td></mdl<>	<mdl< td=""><td>0.58</td><td>0.13</td><td>6.2</td><td>32</td></mdl<>	0.58	0.13	6.2	32
3	0.91	1.4	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.47</td><td>18</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.47</td><td>18</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.47</td><td>18</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.47</td><td>18</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.47</td><td>18</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.47</td><td>18</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.47</td><td>18</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.47</td><td>18</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.47</td><td>18</td></mdl<></td></mdl<>	<mdl< td=""><td>0.47</td><td>18</td></mdl<>	0.47	18
4	8.1	64	19	17	37	7.3	<mdl< td=""><td>116</td><td>5.2</td><td><mdl< td=""><td>5.7</td><td>22</td><td>0.59</td><td>15</td></mdl<></td></mdl<>	116	5.2	<mdl< td=""><td>5.7</td><td>22</td><td>0.59</td><td>15</td></mdl<>	5.7	22	0.59	15
5	<mdl< td=""><td>0.21</td><td><mdl< td=""><td>0.50</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>1.2</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.093</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	0.21	<mdl< td=""><td>0.50</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>1.2</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.093</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	0.50	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>1.2</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.093</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>1.2</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.093</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>1.2</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.093</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td>1.2</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.093</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	1.2	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.093</td><td>0.60</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.093</td><td>0.60</td></mdl<></td></mdl<>	<mdl< td=""><td>0.093</td><td>0.60</td></mdl<>	0.093	0.60
9	3.1	17	2.6	1.1	7.0	1.4	4.6	3.4	<mdl< td=""><td>0.30</td><td><mdl< td=""><td><mdl< td=""><td>1.1</td><td>14</td></mdl<></td></mdl<></td></mdl<>	0.30	<mdl< td=""><td><mdl< td=""><td>1.1</td><td>14</td></mdl<></td></mdl<>	<mdl< td=""><td>1.1</td><td>14</td></mdl<>	1.1	14
7	0.79	1.1	0.32	0.64	1.2	<mdl< td=""><td><mdl< td=""><td>0.33</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>3.8</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td>0.33</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>3.8</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	0.33	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>3.8</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>3.8</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><0.010</td><td>3.8</td></mdl<></td></mdl<>	<mdl< td=""><td><0.010</td><td>3.8</td></mdl<>	<0.010	3.8
8	<mdl< td=""><td>21</td><td>3.1</td><td><mdl< td=""><td>9.5</td><td>2.7</td><td>4.5</td><td>1.3</td><td><mdl< td=""><td>0.21</td><td><mdl< td=""><td><mdl< td=""><td>0.023</td><td>4.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	21	3.1	<mdl< td=""><td>9.5</td><td>2.7</td><td>4.5</td><td>1.3</td><td><mdl< td=""><td>0.21</td><td><mdl< td=""><td><mdl< td=""><td>0.023</td><td>4.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	9.5	2.7	4.5	1.3	<mdl< td=""><td>0.21</td><td><mdl< td=""><td><mdl< td=""><td>0.023</td><td>4.3</td></mdl<></td></mdl<></td></mdl<>	0.21	<mdl< td=""><td><mdl< td=""><td>0.023</td><td>4.3</td></mdl<></td></mdl<>	<mdl< td=""><td>0.023</td><td>4.3</td></mdl<>	0.023	4.3
6	1.4	2.3	0.36	0.47	1.1	<mdl< td=""><td>0.30</td><td>0.78</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.052</td><td>8.0</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	0.30	0.78	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.052</td><td>8.0</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.052</td><td>8.0</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.052</td><td>8.0</td></mdl<></td></mdl<>	<mdl< td=""><td>0.052</td><td>8.0</td></mdl<>	0.052	8.0
10	4.5	12	5.8	1.2	7.6	1.4	3.9	2.0	1.6	0.34	<mdl< td=""><td><mdl< td=""><td>2.4</td><td>2.0</td></mdl<></td></mdl<>	<mdl< td=""><td>2.4</td><td>2.0</td></mdl<>	2.4	2.0
11	0.52	8.3	1.7	<mdl< td=""><td>4.8</td><td>1.0</td><td>1.7</td><td>2.1</td><td>1.3</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.030</td><td>15</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	4.8	1.0	1.7	2.1	1.3	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.030</td><td>15</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.030</td><td>15</td></mdl<></td></mdl<>	<mdl< td=""><td>0.030</td><td>15</td></mdl<>	0.030	15
12	1.5	5.2	0.30	<mdl< td=""><td>5.0</td><td><mdl< td=""><td>0.62</td><td>0.51</td><td>1.9</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.034</td><td>4.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	5.0	<mdl< td=""><td>0.62</td><td>0.51</td><td>1.9</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.034</td><td>4.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	0.62	0.51	1.9	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.034</td><td>4.3</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.034</td><td>4.3</td></mdl<></td></mdl<>	<mdl< td=""><td>0.034</td><td>4.3</td></mdl<>	0.034	4.3
13	4.5	22	2.1	6.1	15	5.9	<mdl< td=""><td>1.5</td><td><mdl< td=""><td><mdl< td=""><td>4.7</td><td>4.9</td><td>0.29</td><td>8.4</td></mdl<></td></mdl<></td></mdl<>	1.5	<mdl< td=""><td><mdl< td=""><td>4.7</td><td>4.9</td><td>0.29</td><td>8.4</td></mdl<></td></mdl<>	<mdl< td=""><td>4.7</td><td>4.9</td><td>0.29</td><td>8.4</td></mdl<>	4.7	4.9	0.29	8.4
14	7.6	42	23	8.106	27	<mdl< td=""><td>15</td><td>10</td><td><mdl< td=""><td>0.82</td><td>0.085</td><td><mdl< td=""><td>1.5</td><td>12</td></mdl<></td></mdl<></td></mdl<>	15	10	<mdl< td=""><td>0.82</td><td>0.085</td><td><mdl< td=""><td>1.5</td><td>12</td></mdl<></td></mdl<>	0.82	0.085	<mdl< td=""><td>1.5</td><td>12</td></mdl<>	1.5	12
15	6.9	82	26	8.8	27	<mdl< td=""><td>11</td><td>15</td><td><mdl< td=""><td>0.36</td><td><mdl< td=""><td>0.17</td><td>12</td><td>19</td></mdl<></td></mdl<></td></mdl<>	11	15	<mdl< td=""><td>0.36</td><td><mdl< td=""><td>0.17</td><td>12</td><td>19</td></mdl<></td></mdl<>	0.36	<mdl< td=""><td>0.17</td><td>12</td><td>19</td></mdl<>	0.17	12	19
16	1.7	3.5	0.48	2.7	2.2	0.74	1.7	1.2	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.16</td><td>5.1</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.16</td><td>5.1</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.16</td><td>5.1</td></mdl<></td></mdl<>	<mdl< td=""><td>0.16</td><td>5.1</td></mdl<>	0.16	5.1
17	<mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.37</td><td>5.3</td></mdl<></td></mdl<>	<mdl< td=""><td>0.37</td><td>5.3</td></mdl<>	0.37	5.3
18	<mdl< td=""><td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></td></mdl<>	<td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.014</td><td>3.2</td></mdl<></td></mdl<>	<mdl< td=""><td>0.014</td><td>3.2</td></mdl<>	0.014	3.2
19	<mdl< td=""><td><mdl< td=""><td>0.40</td><td><mdl< td=""><td>0.47</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.031</td><td>2.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td>0.40</td><td><mdl< td=""><td>0.47</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.031</td><td>2.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	0.40	<mdl< td=""><td>0.47</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.031</td><td>2.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	0.47	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.031</td><td>2.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.031</td><td>2.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.031</td><td>2.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.031</td><td>2.3</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.031</td><td>2.3</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.031</td><td>2.3</td></mdl<></td></mdl<>	<mdl< td=""><td>0.031</td><td>2.3</td></mdl<>	0.031	2.3
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Min.	<mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><0.010</td><td>0.60</td></mdl<></td></mdl<>	<mdl< td=""><td><0.010</td><td>0.60</td></mdl<>	<0.010	0.60
Мах.	12	128	35	42	54	9.9	18	15	5.2	0.82	5.7	22	12	32
Mean ^f	3.4	27	7.7	6.1	13	1.8	3.8	3.9	0.59	0.15	0.58	1.4	1.6	9.5
S.d. ^f	3.9	40	12	11	18	2.9	5.8	5,2	1.2	0.19	1.6	5.0	3.1	8.0
<mdl below<="" td=""><td>v detection</td><td>limit</td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td></mdl>	v detection	limit												

^a PCBs represent the sum of six investigated congeners (PCB 28, 52, 101, 153, 138, 180)

^b \sum DDT is the sum of a_p '-DDT and p_p '-DDT and their breakdown products a_p '-DDD, a_p '-DDE, p_p '-DDD, and p_p '-DDE

 $^{\rm c}$ HCHs is the sum of α -, β -, γ - and δ -hexachlorocyclohexane

^d CHLs is the sum of cis- and trans-chlordane, heptachlor and cis- and trans-heptachloro epoxide

 $^{\rm e}$ Endosulfane is the sum of $\alpha\text{-}$ and $\beta\text{-}$ isomers

^fCalculations based on half the method detection limit for unquantified samples

biphenyls indicated by PCB 101, 153, 138, and 180 (Fig. 2a). In case of stations 3, 7, and 11, hexa and hepta chlorinated biphenyls (PCB 153, 138, and 180) dominated. In these sediment samples, the lowest concentrations of all samples (<1.0 µg/kg dw) were observed. In case of stations 5, 8, 17, 18, 19, and 20 all concentrations were below the MDL, so no distribution was calculated. Dominance of the higher chlorinated biphenyls over the lower chlorinated ones in sediments of Abu Qir Bay is supposed to be due to the higher water solubility of lower chlorinated congeners (Edgar et al. 2003). However, this could possibly be also due to the different sources of PCBs at these specific locations as PCB technical mixtures differ greatly in their composition with respect to the degree of chlorination and these technical mixtures are subjected to great variations when they enter the environment. Absence of any information about the possible point sources and possible uses of PCBs in Egypt makes it difficult to rely on one explanation for the dominance of the higher chlorinated PCBs at these locations. Similar pattern of the different PCB congeners were obtained at stations 1, 2, 4, 10, 13, 14, and 15 with the highest PCB concentrations (range, 4.5-

Fig. 2 Contribution (%) of the single NDL-PCBs to the overall sum in surfacial sediments (a) and mussel samples (b) of Abu Qir Bay. *M. corallina*, stations 4–13; *T. decussata*, stations 18–20. Half the MDL was used to present the concentrations <MDL. In case of stations 5, 8, 17, 18, 19, and 20 no distribution is shown as all PCB concentrations are <MDL

 $12 \mu g/kg dw$). These stations are mainly nearshore and influenced by Abu Qir Drain or Maadeya outlet with the exception of station 10.

3.1.2 Organochlorine pesticides

 Σ DDT (DDT isomers and metabolites) were detected in 85% of the investigated sediment samples but the contribution of individual metabolites showed differences. No Σ DDT were detected at station 17 and stations 18 and 19 in the western part of the bay (Table 2). The concentrations of Σ DDT in sediments varied from <MDL to 128 µg/kg dw (station 1) with an average concentration of 27 µg/kg dw.

Based on the average concentrations, the single compounds were in the following order of predominance: p,p'-DDD (<MDL-45 µg/kg dw; average, 9.4 µg/kg dw)>o,p'-DDD (<MDL-38 µg/kg dw; average, 7.8 µg/kg dw)>p,p'-DDE (<MDL-335 µg/kg dw; average, 5.9 µg/kg dw)>o,p'-DDE (<MDL-17 µg/kg dw; average, 1.7 µg/kg dw)>o,p'-DDT (<MDL-8.0 µg/kg dw; average, 1.3 µg/kg dw)>o,p'-DDT (<MDL-5.5 µg/kg dw; average, 0.39 µg/kg dw). Over 70% of the Σ DDT in sediments from stations 1, 2, 4,



8, 9, 10, 11, and 13 and all the \sum DDT observed in sediments from stations 3, 5, 6, 7, 14, 15, and 16 were present as DDT metabolites (DDD and DDE). This suggests that DDT was not recently released into those locations because of the half-life time of DDT in sediments that was estimated, e.g., to be 4.1 years for Mexico (Tavares et al. 1999) and in the range of 4.7–6.4 years for Japan (Morisawa et al. 2002).

Only stations 12 and 20 showed a higher contribution of $p_{,p}$ '-DDT (48–77%) that indicates a slow degradation of DDT or even recent inputs at these locations (Tavares et al. 1999; Yuan et al. 2001). The dominating pathway of degradation was the reductive dechlorination of DDT to DDD that is the expected pathway under anaerobic conditions in sediments (Baxtor 1990). DDE isomers dominated only at stations 3 that might be a consequence of an aerobic milieu at this nearshore location.

DDT was widely used in Egypt on a variety of agricultural crops and for the control of disease vectors. The largest agricultural use of DDT in Egypt was on cotton fields, which accounted for more than 80% of the use before its ban. Although its usage was banned in 1988 (Barakat et al. 2002), its detection along with the detection of its transformation products in sediments is expected because of their long environmental half-life time.

HCH isomers were detected in most of the investigated sediment samples with different contributions of the respective individual isomers. Based on the average concentrations, the isomers appeared in the following descending order: γ -HCH (lindane; <MDL-222 µg/ kg dw; average, 3.7 μ g/kg dw)> α -HCH (<MDL-10 μ g/ kg dw; average, 2.2 μ g/kg dw)> β -HCH (<MDL-12 μ g/ kg dw; average, 1.3 μg/kg dw)>δ-HCH (<MDL-2.5 μg/ kg dw; average, 0.62 µg/kg dw). For the technical HCH mixtures, the relative proportions of the four major components were α (65–70%)> γ (12–14%)> δ (6.0%) and β (5.0–6.0%; Zhang et al. 2004). The insecticide lindane consists of 99% y-HCH. For residues of technical HCH in sediments, the ratio of α -HCH/ γ -HCH was reported to be 4.6-5.8 (Zhang et al. 2004) or 4.0-15 (Wurl and Obbard 2005) and for contaminations with lindane between 0.20 and 1.0 (McConnell et al. 1998). Calculated values of α -HCH/ γ -HCH ratio in sediments of Abu Qir Bay ranged from 0.29 to 1.1 indicating lindane as probably the main HCH source with possibly also minor contribution of the technical HCH mixtures. This agrees with the usage data given by EL-Sebae (1989), as lindane was intensively used in Egypt in the period from 1952 to 1978. About 11,300 t were used in agriculture. No data are available for the use of lindane or HCHs in Egypt after this time period.

cis-Chlordane, trans-chlordane, heptachlor, cis-heptachloro epoxide, trans-heptachloro epoxide, aldrine, dieldrine, α -

endosulfane. β -endosulfane, and mirex were the investigated cyclodiene pesticides in surfacial sediments of Abu Oir Bay. According to the calculated average concentrations, cyclodiene insecticides can be arranged according to the following descending order: trans-chlordane (<MDL-29 µg/kg dw; average, 6.3 µg/kg dw)>dieldrine (<MDL-19 µg/kg dw; average, 3.8 µg/kg dw)>cis-chlordane (<MDL-17 µg/kg dw; average, 3.5 μ g/kg dw)> α -endosulfane (<MDL-11 μ g/ kg dw; average, 2.5 µg/kg dw)>aldrine (<MDL-9.3 µg/ kg dw; average, 1.8 μ g/kg dw)> β -endosulfane (<MDL-6.3 μg/kg dw; average, 1.5 μg/kg dw)>heptachlor (<MDL-4.5 µg/kg dw; average, 0.97 µg/kg dw)>trans-heptachloro epoxide (<MDL-3.9 µg/kg dw; average, 0.67 µg/kg dw)> cis-heptachloro epoxide (<MDL-4.1 µg/kg dw; average, 0.58 µg/kg dw)>mirex (<MDL-0.82 µg/kg dw; average, 0.15 µg/kg dw). Most of the cylcodiene insecticides could not be found in the western part of the bay except for cischlordane, α -endosulfane, and dieldrine that were detected at station 20 in the northwestern part of the bay in concentrations below 1.0 µg/kg.

Concentrations of CHLs (*cis-*, *trans*-chlordane, heptachlor, and heptachloro epoxide) ranged from $<MDL-54 \mu g/kg$ dw with an average concentration of 13 $\mu g/kg$ dw (Table 2). Concentrations of *trans*-chlordane were generally higher than of *cis*-chlordane in most of the sediment samples. This is expected as the concentration of the *trans*-isomer in the technical mixture is higher than of the *cis*-isomer (Dearth and Hites 1991). About 50 t of formulated Chlordane was applied in 1985 in Egypt (EL-Sebae 1989). According to the United Nations Environment Programme, the use of chlordane is severely restricted and limited to non-agricultural uses in Egypt.

Concentrations of aldrine ($\langle MDL-9.9 \ \mu g/kg \ dw$) and dieldrine ($\langle MDL-18 \ \mu g/kg \ dw$) were relatively low in sediments of Abu Qir Bay (Table 2). Concentrations of dieldrine were generally higher than of aldrine in most of the sediment samples. This could be attributed to the fact that aldrine is converted into dieldrine in the sediments by epoxidation. At the same time, dieldrine is known to be more persistent than aldrine (Chiu et al. 2005 and references therein).

Concentrations of endosulfane (α - and β -isomer) in sediments of Abu Qir Bay ranged from <MDL-15 µg/ kg dw with an average concentration of 3.9 µg/kg dw. Concentrations of α -endosulfane were generally higher than β -endosulfane, which can be related to the higher contribution of the α -isomer in the pesticide formulations (the ratio of α -endosulfane to β -endosulfane is about 7:3; Shetty et al. 2000). In addition, β -endosulfane can be converted into α -endosulfane (Schmidt et al. 2001).

Mirex and methoxchlor were rarely detected in Abu Qir Bay. Mirex was found only in five sediment samples at locations in a certain distance to the shoreline (<1.0 μ g/

kg dw) and also methoxychlor was detected only in five samples (1.2–5.2 μ g/kg dw; Table 2).

PCBz and HCBz were detected in only four nearshore sediment samples (stations 1, 2, 4, and 13). The highest concentrations of 5.7 and 22 μ g/kg dw were found at station 4 in front of the Maadeya Outlet.

Chlorpyrifos was the only investigated organophosphorus insecticide in sediments of Abu Qir Bay. It was selected since it was previously detected in sediments along the Coast of Alexandria and being known to be very highly toxic for aquatic organisms (Pait et al. 1992). Concentrations ranged from <MDL to 42 µg/kg dw with an average concentration of 6.1 µg/kg dw (Table 2). Despite its relatively high degradation rate ($t_{1/2}$ =60–120 days; Pait et al. 1992), chlorpyrifos was detected in 65% of the sediment samples in obviously higher concentrations than the other investigated pesticides. This finding can be attributed to the use of organophosphorus insecticides in agriculture after banning the persistent organochlorine insecticides.

Significant positive correlation was observed between the concentration of TOC and the concentrations of Σ DDT, HCHs, *cis*-heptachloro epoxide, α - and β -endosulfane (*r* ranged from 0.70 to 0.87 at p<0.05). Significant positive correlation was also observed between the clay content and the concentrations of Σ DDT, *cis*-heptachloro epoxide, β endosulfane, *trans*-chlordane, total chlordane, and chlorpyrifos (*r* ranged from 0.72 to 0.76 at p<0.05) in the bay sediments. However, it must be pointed out that the occurrence of many of the investigated organochlorine pesticides below the limit of detection in several sediment samples greatly affected the degree of correlation.

3.2 Mussels

Concentrations of organochlorines and chlorpyrifos in mussels of *M. corallina* and *T. decussata* are summarized in Table 3.

Concentrations of $\sum 6$ PCBs in both species (*M. corallina*: 0.96–56 µg/kg dw, average: 24 µg/kg dw; *T. decussata*: 0.54–10 µg/kg dw, average: 4.5 µg/kg dw) were the highest among all the investigated chlorinated organic compounds, followed by $\sum DDT$ (*M. corallina*: 4.9–31 µg/kg dw, average: 14 µg/kg dw; *T. decussata*: 0.94–4.4 µg/kg dw, average: 2.4 µg/kg dw). Based on the average concentrations in *M. corallina*, chlorinated organic compounds can be arranged in the following descending order: PCBs> $\sum DDT>CHLs>$ chlorpyrifos>HCHs>dieldrine> endosulfane>Mirex>HCBz>methoxychlor>PCBz, and in *T. decussata*: PCBs> $\sum DDT>CHLs>$ chlorpyrifos>HCHs, dieldrine, endosulfane, mirex, and PCBz in all samples of *T. decussata* were below the detection limit. Aldrine, *o.p'*-

DDT, p,p'-DDT and β -HCH were completely absent in both species. The absence of aldrine, o,p'-DDT and p,p'-DDT can be related to the metabolic activity of the bivalve species.

The highest concentrations of organochlorines were found at stations 4 and 6 in front of Maadeya outlet and at stations 13 and 12, with a trend decreasing in the offshore direction (stations 4-6 and 13-12). No mussel data can be reported for the sediment's hot spot stations (1, 2, 14, and 15) in front of Abu Qir Drain as no mussels were found at these locations. According to Fishar and EL-Haweet (2005), the pollution problem in the bay has caused both the change and loss of the habitat of marine fauna and change in the water quality of the bay. They concluded that there was a remarkable decrease in the biodiversity of fishes and benthic fauna as a result of the intensive human impacts. The absence of mussels at these stations could thus be assumed among other reasons to be caused by the overall relatively high contamination of the respective sediments with organochlorines (present work) correlated with relative high TOC values, PAHs (Khairy et al. 2009), and heavy metals (Khairy et al. 2011) and/or high ammonia levels which were detected in front of AQD as a result of the effluents discharged from the Fertilizer Company (Nasr et al. 2004). Also further pollutants or water quality parameters might have contributed.

3.2.1 PCBs

Concentrations of PCBs in *M. corallina* (average, $24 \mu g/kg dw$) were generally about sixfold higher than those in *T. decussata* (average, 4.5 $\mu g/kg dw$). The reason for this difference can be related to sampling locations of both species, where *Mactra* sp. was sampled in an area with high TOC content influenced by discharges from the major sources of pollution in the bay whereas the sediment at the sample locations of *T. decussata* was characterized by much lower levels of TOC and accordingly, lower contamination levels.

In *M. corallina*, the contribution of the lower chlorinated congeners PCB 28, 52, and 101 to the total PCB concentration was generally higher than in the corresponding sediment samples (Fig. 2b; stations 4, 6, 7, 9, 12, 13). The reason might be that lower chlorinated biphenyls are less sorbed on the sediment particles and, therefore, are more available to aquatic organisms (De Boer et al. 2001). In *T. decussata*, PCB 28 showed slightly elevated contribution of the total PCB especially at station 19, where PCB 28 was the only detected PCB congener and the concentration of the PCBs in the sediments were below the MDLs. In the other two samples, PCB 28 was also detected but to a lower extent of 17.6% and 31%.

Station no.	PCBs ^a	ΣDDT^{b}	HCHs ^c	Chlorpyrifos	CHLs ^d	Dieldrine	Endosulfane ^e	Methoxychlor	Mirex	PCBz	HCBz	Lipid%	DW/WW
M. corallina													
4	56	26	11	10	35	12	9.3	4.0	<mdl< td=""><td>0.33</td><td>0.50</td><td>5.2</td><td>0.19</td></mdl<>	0.33	0.50	5.2	0.19
9	36	31	8.1	14	15	2.6	4.3	1.3	<mdl< td=""><td>0.39</td><td>0.39</td><td>4.8</td><td>0.17</td></mdl<>	0.39	0.39	4.8	0.17
7	0.96	4.9	<mdl< td=""><td>3.3</td><td>1.1</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.3</td><td>0.22</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	3.3	1.1	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.3</td><td>0.22</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.3</td><td>0.22</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.3</td><td>0.22</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.3</td><td>0.22</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>4.3</td><td>0.22</td></mdl<></td></mdl<>	<mdl< td=""><td>4.3</td><td>0.22</td></mdl<>	4.3	0.22
8	1.8	6.1	<mdl< td=""><td>4.6</td><td>0.91</td><td>2.4</td><td><mdl< td=""><td><mdl< td=""><td>0.89</td><td><mdl< td=""><td><mdl< td=""><td>4.1</td><td>0.18</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	4.6	0.91	2.4	<mdl< td=""><td><mdl< td=""><td>0.89</td><td><mdl< td=""><td><mdl< td=""><td>4.1</td><td>0.18</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td>0.89</td><td><mdl< td=""><td><mdl< td=""><td>4.1</td><td>0.18</td></mdl<></td></mdl<></td></mdl<>	0.89	<mdl< td=""><td><mdl< td=""><td>4.1</td><td>0.18</td></mdl<></td></mdl<>	<mdl< td=""><td>4.1</td><td>0.18</td></mdl<>	4.1	0.18
6	8.0	7.0	<mdl< td=""><td>6.8</td><td>2.3</td><td>2.9</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.46</td><td>4.3</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	6.8	2.3	2.9	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.46</td><td>4.3</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.46</td><td>4.3</td><td>0.19</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.46</td><td>4.3</td><td>0.19</td></mdl<></td></mdl<>	<mdl< td=""><td>0.46</td><td>4.3</td><td>0.19</td></mdl<>	0.46	4.3	0.19
12	25	17	6.5	15	14	<mdl< td=""><td><mdl< td=""><td>2.9</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.6</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td>2.9</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.6</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	2.9	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.6</td><td>0.19</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>4.6</td><td>0.19</td></mdl<></td></mdl<>	<mdl< td=""><td>4.6</td><td>0.19</td></mdl<>	4.6	0.19
13	37	6.6	2.1	3.3	4.4	<mdl< td=""><td>0.63</td><td><mdl< td=""><td>0.46</td><td><mdl< td=""><td><mdl< td=""><td>4.8</td><td>0.18</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	0.63	<mdl< td=""><td>0.46</td><td><mdl< td=""><td><mdl< td=""><td>4.8</td><td>0.18</td></mdl<></td></mdl<></td></mdl<>	0.46	<mdl< td=""><td><mdl< td=""><td>4.8</td><td>0.18</td></mdl<></td></mdl<>	<mdl< td=""><td>4.8</td><td>0.18</td></mdl<>	4.8	0.18
Min	0.96	4.9	<mdl< td=""><td>3.3</td><td>0.91</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.1</td><td></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	3.3	0.91	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.1</td><td></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.1</td><td></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.1</td><td></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>4.1</td><td></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>4.1</td><td></td></mdl<></td></mdl<>	<mdl< td=""><td>4.1</td><td></td></mdl<>	4.1	
Max	56	31	11	15	35	12	9.3	4.0	0.89	0.39	0.50	5.2	
Mean ^f	24	14	4.3	8.1	10	2.9	2.2	1.4	0.31	0.17	0.27	4.6	
SD^{f}	21	11	4.2	4.9	12	4.2	3.4	1.5	0.28	0.13	0.17	0.37	
T. decussata													
18	3.0	1.9	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.39</td><td>8.2</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.39</td><td>8.2</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.39</td><td>8.2</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.39</td><td>8.2</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.39</td><td>8.2</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.39</td><td>8.2</td><td>0.19</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.39</td><td>8.2</td><td>0.19</td></mdl<></td></mdl<>	<mdl< td=""><td>0.39</td><td>8.2</td><td>0.19</td></mdl<>	0.39	8.2	0.19
19	0.54	0.94	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>8.0</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>8.0</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>8.0</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>8.0</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>8.0</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>8.0</td><td>0.19</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>8.0</td><td>0.19</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>8.0</td><td>0.19</td></mdl<></td></mdl<>	<mdl< td=""><td>8.0</td><td>0.19</td></mdl<>	8.0	0.19
20	10	4.4	<mdl< td=""><td>0.33</td><td>0.36</td><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.40</td><td>8.8</td><td>0.18</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	0.33	0.36	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.40</td><td>8.8</td><td>0.18</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.40</td><td>8.8</td><td>0.18</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>0.40</td><td>8.8</td><td>0.18</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>0.40</td><td>8.8</td><td>0.18</td></mdl<></td></mdl<>	<mdl< td=""><td>0.40</td><td>8.8</td><td>0.18</td></mdl<>	0.40	8.8	0.18
Min	0.54	0.94		<mdl< td=""><td><mdl< td=""><td></td><td></td><td></td><td></td><td></td><td><mdl< td=""><td>8.0</td><td></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td></td><td></td><td></td><td></td><td></td><td><mdl< td=""><td>8.0</td><td></td></mdl<></td></mdl<>						<mdl< td=""><td>8.0</td><td></td></mdl<>	8.0	
Max	10	4.4		0.33	0.36						0.40	8.8	
Mean ^f	4.5	2.4		0.20	0.87						0.31	8.3	
SD^{f}	4.9	1.8		0.11	0.16						0.15	0.44	
ADL below ^a PCBs repres	detection 1: ent the sum	imit, <i>DW/WW</i>	7 dry weight tigated cong	fraction of the we eners (PCB 28, 52	t mussel tiss.	ues 38, and 180)							
)			,							

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^b \sum DDT is the sum of a_p '-DDT and p_p '-DDT and their transformation products a_p '-DDD, a_p '-DDE, p_p '-DDD, and p_p '-DDE

^d CHLs is the sum of *cis-* and *trans-chlordane*, heptachlor, and its epoxides

° HCHs is the sum of α -, β -, γ - and δ -hexachlorocyclohexane

^fCalculations based in half the MDL for unquantified samples

 $^{\rm e}$ Endosulfane is the sum of $\alpha\text{-}$ and $\beta\text{-}{\rm isomers}$

Table 3 Concentrations of PCBs and chlorinated pesticides (µg/kg dw) in mussel samples of Abu Qir Bay

Detected concentrations of PCBs in the mussel samples were significantly higher than their corresponding sediment samples (Mann–Whitney rank sum test at p=0.017) reflecting a great bioaccumulative potential and bioavailability.

3.2.2 Organochlorine insecticides and chlorpyrifos

DDE and DDD were detected in all investigated mussel samples with total concentrations ranging from 0.94 µg/ kg dw (*T. decussata*, station 19) to 31 µg/kg dw (*M. corallina*, station 6; Table 3). The parent compound DDT was not found in the tissues of both species. p,p'-DDE was the dominant isomer in both species representing from 49% to 100% of the total detected DDTs (Fig. 3b). This pattern was different from the corresponding sediment samples (Fig. 3a), where p,p'-DDD was the dominant isomer, and o,p'-DDT and p,p'-DDT were also present. Several authors have observed preferential accumulation of DDT transformation products in aquatic organisms (Thompson et al. 1999; Qiang 2004; Sapozhnikova et al. 2004). This phenomenon is due to uptake, metabolism, and excretion mechanisms of DDT derivatives in mussels (Qiang 2004). Indeed, bivalves are known for a certain capacity to metabolize organic contaminants (Livingstone 1994).

CHLs ranged from <MDL to 35 μ g/kg dw in the mussel samples. *cis*-chlordane (<MDL–17 μ g/kg dw) was only detected in two samples (stations 4, 6), whereas *trans*-chlordane (<MDL–15 μ g/kg dw) was detected in all the samples of *M. corallina* (Table 3). Similar to the sediment samples, concentrations of the *trans*-isomer were higher in the mussels than the *cis*-isomer.

Dieldrine was detected only in four samples of *M. corallina* (stations 4, 6, 8, and 9) with concentrations ranging from <MDL to 12 μ g/kg dw. The concentration detected in the sample from station 4 in front of the Maadeya Outlet was obviously higher than in the other samples due to the influence of the water discharged from Maadeya Outlet.

Only α -, γ -, and δ - isomers of HCHs were observed in four of the mussel samples (stations 4, 6, 12, and 13) of *M. corallina* (<MDL-11 µg/kg). Similar to sediments, α - and



DDT and its metabolites in surfacial sediments (**a**) and mussel samples (**b**) of Abu Qir Bay. *M. corallina*, stations 4–13;

T. decussata, stations 18-20

Fig. 3 Percent contribution of

Location	Sampling year	Species type	PCBs	<i><u>SDDT</u></i>	HCHs	HCBz	Endosulfane	Heptachlor	Reference
Abu Qir Bay, Egypt	2006	M. corallina T. decussata	0.21-11 (4.3) 0.11-1.8 (0.83)	$\begin{array}{c} 1.1{-}5.4 \ (2.6) \\ 0.18{-}0.80 \ (0.45) \end{array}$	<dl-2.0 (0.72)<br="">ND</dl-2.0>	<dl-0.093 (0.035)<br=""><dl-0.073 (0.049)<="" td=""><td>0.030–2.2 (0.52) ND</td><td><dl-0.28 (0.091)<br=""><dl-0.066 (0.022)<="" td=""><td>Present study</td></dl-0.066></dl-0.28></td></dl-0.073></dl-0.093>	0.030–2.2 (0.52) ND	<dl-0.28 (0.091)<br=""><dl-0.066 (0.022)<="" td=""><td>Present study</td></dl-0.066></dl-0.28>	Present study
Alfacs Bay, Spain Fangar Bay, Spain	1991 1992	Mytilus galloprovincialis	9.7 6.0	7.5 1.1	0.10 ND	0.60 ND	NA NA	NA NA	Sole et al. 1994
Ebro Delta, NW Mediterranean	1989–1991	Mytilus galloprovincialis Rotala decussata	7.0–70 1.0–18	2.0–144 0.60–7.0	NA NA	NA NA	NA NA	NA NA	Sole et al. 2000
Adriatic Sea, Italy	NA	Mytilus galloprovincialis	ND-20	ND-6.6	NA	ND-0.50	NA	NA	Marcotrigiano and Storelli 2003
Monterey Harbour, California, USA	1999	Mussel	16	12	0.99	NA	NA	NA	Kannan et al. 2004
Abu Qir Bay, Egypt		Donax trunculus	37	22	NA	2.1	1.1	ND	Abd-Allah et al. 1998
Location	Sampling year	Species type	Chlorpyrifos	Dieldrine	Σ CHLs	Heptachloro epoxide	Methoxychlor	Mirex	Reference
Abu Qir Bay	2006	M. corallina T. decussata	0.60–2.9 (1.5) <dl-0.059 (0.020)<="" td=""><td><dl-2.1 (0.51)<br="">ND</dl-2.1></td><td>0.17–5.9 (1.6) ND</td><td><dl-0.42 (0.17)<br="">ND</dl-0.42></td><td><dl-0.75 (0.22)<br="">ND</dl-0.75></td><td><dl-0.16 (0.036)<br="">ND</dl-0.16></td><td>Present study</td></dl-0.059>	<dl-2.1 (0.51)<br="">ND</dl-2.1>	0.17–5.9 (1.6) ND	<dl-0.42 (0.17)<br="">ND</dl-0.42>	<dl-0.75 (0.22)<br="">ND</dl-0.75>	<dl-0.16 (0.036)<br="">ND</dl-0.16>	Present study
Monterey Harbour, California, USA	1999	Mussel	NA	0.96	2.2 ^z	0.10	NA	NA	Kannan et al. 2004
Salton Sea Lake, CA, USA	2001	Tilapia mossambique	0.30–11	0.40–55	NA	NA	NA	NA	Sapozhnikova et al. 2004
Abu Qir Bay, Egypt	NA	Donax trunculus	NA	0.60	NA	NA	ND	2.2	Abd-Allah et al. 1998

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 γ -HCH were the most abundant isomers with higher concentrations of the γ -isomer (<MDL–6.7 µg/kg dw; average, 2.1 µg/kg dw) compared with the α -HCH isomer (<MDL–4.1 µg/kg dw; average, 1.6 µg/kg dw).

Chlorpyrifos was detected in relatively high concentrations $(3.3-15 \ \mu g/kg \ dw)$ in all samples of *M. corallina*, whereas this organophosphate insecticide was detected in only one sample of *T. decussata* (station 20) but with a tenfold lower level than the lowest concentrations recorded for *M. corallina*. Although chlorpyrifos was not found in some sediment samples (stations 8 and 12), it was detected in tissues of their corresponding mussel samples likely through the direct uptake of the water-soluble chlorpyrifos from the water phase.

All the other investigated organochlorine pesticides (endosulfane, methoxychlor, PCBz, HCBz, and mirex) were either detected in low concentrations compared to the rest of the investigated pesticides or found below the limit of detection (Table 3). No statistical significant differences were observed between concentrations of pesticides in sediments and mussels of Abu Qir Bay (Mann-Whitney rank sum test at p=0.132-0.910). This finding was in opposite to the statistic differences in case of the PCBs (see Section 3.2.1) and can be attributed to a lower bioavailability and/or a elimination of pesticides by mussels metabolic activities. Concentrations of pesticides in tissues of Mactra sp. were apparently higher than concentrations in tissues of Tapes sp. This difference might be mainly influenced by the sampling stations of Tapes sp. with low pesticide concentrations combined with low TOC contents (compare Section 3.2.1).

Concentrations of the chlorinated organic compounds in the tissues of the two species investigated in the present study were compared with concentrations in different mussel species worldwide and in the previous studies performed along the Egyptian Coast. But most other studies did not include the whole range of compounds that were investigated in the present study. As shown in Table 4, 807

detected concentrations in the present study were generally in a similar range as most of the concentrations recorded in other mussel species worldwide. At the same time, concentrations were generally lower than concentrations recorded in mussel samples investigated previously in Abu Qir Bay by Abd-Allah et al. (1998). It should be noted that the difference in the species, sampling locations, and/or difference in the sampling season could have a great influence on the obtained concentrations, and comparisons should be interpreted with care.

3.3 Risk assessment of chlorinated organic compounds in Abu Qir Bay

3.3.1 SLERA

Results obtained from the risk assessment of the maximum concentration of the chlorinated organic compounds in sediments of Abu Qir Bay are given in Fig. 4.

The figure shows that several organochlorine pesticides (γ -HCH, chlordane, dieldrine, p,p'-DDE, p,p'-DDD, p,p'-DDT, DDD (p,p'- and o,p'-DDD), DDT (p,p'- and o,p'-DDD)DDT), and Σ DDT) had TEC HQ values higher than 1, which means that adverse ecological effects to benthic species are likely to be observed. No guidelines exist for o,p'isomers of the DDT-group, thus p,p'- and o,p'-isomers were assessed together. In consequence to the calculated TEC HQ values, these contaminants are of potential ecological concern. In addition, γ -HCH, heptachlor, chlordane, dieldrine, p,p'-DDD, DDD, DDT, and Σ DDT had also PEC HQ values higher than one indicating that for these pollutants, the occurrence of adverse ecological effects to benthic species is likely to be frequently observed. For the compounds with maximum TEC and PEC HQ values above 1, the calculated values for all sediment samples are marked in the map of Abu Qir Bay (Fig. 5). The calculated

Fig. 4 TEC HQ and PEC HQ for the maximum detected concentrations of chlorinated organic compounds in sediments of Abu Qir Bay. *PCBs* polychlorinated biphenyls, *HCBz* hexachlorobenzene, *DDD o.p'*-DDD+*p.p'*-DDD, *DDT o.p'*-DDT+*p.p'*-DDT, ×*DDT* sum of the six isomers of the DDT and its metabolites, *PCBz* pentachlorobenzene





Fig. 5 Ecological risk assessment of selected chlorinated organic compounds in sediments using HQ. *TEC* threshold effect concentration, *PEC* probable effect concentration, *DDTs* sum of *o*,*p*'-DDT, *p*,*p*'-DDT and their metabolites

maximum TEC HQ and PEC HQ were always located in front of the major sources of pollution (Abu Qir Drain or the Maadeya Outlet), and thus these sites can be regarded as hot spots. In case of dieldrine, lindane, and chlordane also station 10 offshore emerged as hot spot. This finding is suspected to be attributed to the fact that water discharged through the Maadeya outlet will be moved by circulation in northern and northwestern direction during spring, summer, and autumn.

For HCBz, heptachlor, and aldrine no TEC levels were available. When the maximum concentration of HCBz and aldrine were compared with the PEC, the resulting PEC HQs were lower than one in both cases. But this only indicates that adverse ecological effects are not frequently expected. Thus, further investigations are required to determine the extent of adverse ecological effects associated with the exposure to HCBz and aldrine in the bay sediments.

As SQGs were not available for all investigated compounds, some of the organochlorine pesticides (α -HCH, chlorpyrifos, endosulfane, heptachloro epoxide, methoxychlor, and PCBz) were evaluated by comparing the maximum concentrations with sediment screening concentrations of USEPA (2004) and the Canadian sediment quality guidelines (CCME 2003). It was found that all the concentrations of α -HCH, methoxychlor, and PCBz in sediments of Abu Qir Bay were lower than the respective screening concentrations (Fig. 4). This indicates that adverse ecological effects are unlikely to occur by these substances. But the concentrations of endosulfane in the bay sediments were much higher than the screening concentration of 0.11 µg/kg dw. This finding indicates that adverse ecological effects are likely to occur to benthic

Table 5 Calculated HQs andcancer risks in the performedhuman health risk assessment inAbu Qir Bay

organisms by endosulfane. Also for the heptachloro epoxide isomers, most of the investigated samples showed higher values than the screening concentration of 0.60 μ g/kg dw and in the case of chlorpyrifos four of the investigated sediment samples (in front of Abu Qir Drain and the Maadeya Outlet) showed higher concentrations than the screening benchmark of 8.30 μ g/kg dw. Thus, it can be concluded that also in case of chlorpyrifos and heptachloro epoxides adverse ecological effects may be expected. As no higher screening values (equal to PEC) are available for these compounds it is not possible to specify the extent of adverse effects that have to be expected.

3.3.2 Human Health Risk Assessment of chlorinated organic compounds in Abu Qir Bay

The results of the calculated HQs and cancer risks for the chlorinated organic compounds of both species are presented in Table 5. No adverse health effects (either carcinogenic or noncarcinogenic) can be deduced by this risk assessment for the consumption of the investigated mussel species.

4 Conclusions

With the help of a SLERA, it was possible to indicate which class of chlorinated organic compounds are of highest concern to assess and to improve the environmental quality of the bay. Adverse ecological effects to benthic species that are likely to be frequently observed were found to be associated with the organochlorine insecticides

Analyte	M. corallina		T. decussata	
	HQ	Cancer risk	HQ	Cancer risk
PCBs	3.9×10^{-3}	7.8×10^{-5}	6.7×10^{-4}	1.4×10^{-5}
PCBz	1.9×10^{-7}	NA	ND	NA
HCBz	3.1×10^{-7}	4.2×10^{-10}	3.4×10^{-7}	4.5×10^{-10}
α -HCH	NA	1.3×10^{-8}	NA	ND
Lindane (γ -HCH)	9.0×10^{-6}	3.7×10^{-9}	ND	ND
Heptachlor	1.3×10^{-6}	3.1×10^{-9}	4.8×10^{-7}	1.1×10^{-9}
Heptachloro epoxide	8.3×10^{-5}	1.0×10^{-8}	ND	ND
Chlorpyrifos	2.7×10^{-6}	NA	7.3×10^{-8}	NA
Chlordane	2.4×10^{-5}	4.3×10^{-9}	ND	ND
Dieldrine	7.9×10^{-5}	6.6×10^{-8}	ND	ND
Endosulfane	6.4×10^{-7}	NA	ND	NA
<i>p,p'</i> -DDE	NA	2.5×10^{-9}	NA	$7. \times 10^{-9}$
Methoxychlor	3.3×10^{-7}	NA	ND	NA
Mirex	1.6×10^{-6}	5.8×10^{-10}	ND	ND

NA not available, *ND* not detected

dieldrine, lindane, chlordane, and Σ DDT in the bay sediments. Thus, further detailed studies involving the fate and behavior of these contaminants are deemed important for the future improvement of the environmental quality of Abu Oir Bay. Hot spots of contamination could be located in front of Abu Qir Bay and Maadeya Outlet indicating that the discharges of both outlets into the bay have to be controlled for improving its environmental quality. With regard to the bioaccumulation potential of pollutants in mussels, PCBs, Σ DDT, and chlorpyrifos were found highly enriched in the mussel tissues. However, the HHRA showed that no adverse health effects could be expected from the consumption of the mussels so far. Monitoring of organochlorines and chlorpyrifos in sediments and mussels would be needed to control the future trend of pollution. This should be supplemented by the analyses of further pollutants that were identified by means of SLERA to be of major concern for the Abu Qir bay in a previous paper (Khairy et al. 2011).

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