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On the Organochlorine pesticide residues in the Marshes, Shatt Al-Arab and the Arabian Gulf system

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Abstract - Organochlorine pesticides are a class of toxic compounds characterized by their relative chemical and biological stability. They are ubiquitous in the environments and are soluble in liquids. Consequently Organochlorine pesticides have been placed on "the top of black list" of potential environmental hazards. Studies on Organochlorine residues have been carried out between 1979-1991. The residues were determined in water, sediments and organisms collected from the Marshes, Shatt Al-Arab and the Arabian Gulf system. Most of the data confirmed the present of ∑DDT, Endrin, Dieldrin, Chlordane and Heptachlor. Their concentrations ranged from nd-336, nd-111, 12-195, nd-57 and nd-79 ng/l in dissolved, from 0.1-2560, nd-217, nd-0.97 , nd-171 and nd-679 µg/kg in particulate and from 0.04-220, nd-47, nd-22, nd from nd-24 µg/kg in sediments from 1-189, nd-154, nd-32, nd-58 and nd-6 ppb in fish, from 0.59-784, 166-540, 2-72, 5-3.1 and nd ppb in mussel and from 0.3-6.3, nd-0.4, nd-0.8, nd and nd-0.3 ppb in shrimp respectively. In the last decade only two studies on Organochlorine residues have been done. One of them in the Mesopotamian wetlands of southern Iraq in which residues of p,p´-DDT have been detected in 100% of the examined samples. This indicates its ability to persist under severe drying of previously exposed surface sediments, long period of more than 10 years of previously exposed surface sediments, high temperature, and intensive solar radiation, and the other by using samples of water, sediment, fish and shrimp from Hor Al-Hammar Marshes of Iraq, high concentration of chlordane in both water and sediments were shown. Therefore further work is needed to establish the baseline levels of Organochlorine residue in the Marshes, Shatt Al-Arab and the Arabian Gulf region.

Keywords: Organochlorine, pesticide, Marshes, Shatt Al-Arab, Arabian Gulf.

Introduction

Chlorinated pesticides enter the marine environment in two ways: In water run-off from agricultural areas and from the atmosphere. The major source of the pesticides in the marine environment is the atmosphere.

The place of origin of the Shatt Al-Arab river starts when Euphrates river (after it flows through the Hor Al-Hammar unit) joins the Tigris river at Garmat Ali. Karun river, the only tributary of the Shatt Al-Arab river, joins its eastern bank south of Basrah city. The Shatt Al-Arab unit end when reaching the Arabian Gulf. The length of the Shatt Al-Arab river from Garmat Ali to its mouth in the Arabian Gulf extends to about 110 km. It's width varies at different points, ranging from 0.4 km at Basra city to 1.5 km at it's mouth.

The water depth increase in general towards the Gulf with maximum of 12.5 m. The water level is affected by the high and low tides of the Arabian Gulf. The Arabian Gulf is a shallow semi-closed water body, several industrial complex have been established along its coast line during the past. The effluent from these, facilities is being discharge into the Gulf. These discharges pose potential hazards to the marine environment is striving to protect the marine environment of the Gulf and has commissioned several studies to assess the damage from the industrial and municipal discharge (Douabul *et al.*, 1988).

The Arabian Gulf water polluted by Chlorinated hydrocarbons via atmospheric deposition (both the wet and dry), tributary inflow, industrial and municipal effluents and sediment resuspension, Losses from the water column include sedimentation outflow, water-to-air and degradation (Douabul *et al.*, 1987 a, b and c). In the water column, chlorinated pesticides distribute between the dissolved and participate phases, and there distribution is operationally defined by the limitations of the filtration process (Danielsson, 1982). The chlorinated pesticides which remain on the filters are defined as participate the fraction of chlorinated pesticides which passes through the filter may be associated with fine particles, colloidal or dissolved organic matter, or may be truly dissolved.

Organochlorine pesticides have been used in Iraq since 1960, because these compounds characterized by their toxicity and relative chemical and biological stability have been officially banned in Iraq since 1976. This study reports all the works done in the Southern of Iraq and NW Arabian Gulf, and to comprise the changes in the concentrations of these compounds in water, sediment, fish, biota and shrimp from 1978 until now.

Residue levels:

Water:

Mesopotamia is predominantly an agricultural region and pesticides control is of great economic significance. Organochlorine pesticides have been used in Iraq and neighboring countries since 1960 (Al-Omar *et al.,* 1985). The parent chemicals and/or their degradation products can enter the aquatic environment by leaching directly to stream or ground water, or by the erosion of contaminated soil. Measurable levels of DDT, endrin, and dieldrin were encountered in environmental samples from the Shatt Al-Arab River, but invariably a significant incremental increase in their concentrations was observed in Hor Al-Hammar Lake, a lake that drains into the Euphrates River and subsequently reaches the Shatt Al-Arab River (DouAbul *et al.,* 1987a). These findings were attributed to the fact that in Hor Al-Hammar pesticides were applied, at various times and in various amounts, close to or over water causing fairly direct contamination (DouAbul *et al.*, 1988).

Organochlorine pesticides enter the Shatt Al-Arab River either adsorbed onto particles or dissolved. Finally, the contaminated particles either settle to the bottom or are transported to the Arabian Gulf. In a preliminary survey of marine samples, Anderlini *et al.* (1981) observed DDT derivatives in the oyster *Pinctada margaratifera* collected from Kuwaiti waters. These observations could serve as evidence that inputs from the Shatt Al-Arab River Delta may indeed be taking place. DouAbul *et al.* (1988) studied the organochlorine pesticides in various phases of the Shatt Al-Arab River and show the major transport mechanisms for organochlorine pesticides entering the Shatt Al-Arab River. Table (1) illuminate sampling Locations (TR, ER, SH1 and SH2).

Table 1: Samples location.

Stations	N	E
TR (Tigris River)	31° 00′ 23.04″	47° 26′ 28.40″
ER (Euphrates River)	30° 35′ 39.02″	47° 41′ 58.44″
SH1 (Shatt Al-Arab River)	30° 34′ 53.30″	47° 46′ 07.18″
SH2 (Shatt Al-Arab River)	30° 27′ 45.08″	48° 00′ 53.09″
St 1 (Khor Al-Zubair)	30° 09′ 20.07″	47° 54′ 07.23″
St 2 (Khor Al-Zubair)	30° 06′ 28.72″	47° 55′ 57.08″
St 3 (Khor Al-Zubair)	30° 03′ 36.60″	47° 57′ 31.78″
St 4 (Khor Al-Zubair)	30° 00′ 48.74″	47° 57′ 25.74″

DDT is one of the most important and most common insecticide in the world as well as in Iraq, where it has been extensively used for insect control by agricultural and public health authorities until 1976 (Al-Omar *et al.,* 1985). DouAbul *et al.* (1988) noted variation in the distribution of ∑DDT residues in the Shatt Al-Arab waters; thus the levels range between 81-168 ng/l (Table 2) and the elevated levels at the origin (SH1) of this river declined to 81 ng/l at the middle of the river (SH2) and attributed to partitioning of dissolved DDT onto particulate matter because the flow of the river at the studies location was constant (919 m^3/s).The tidal nature of the Shatt Al-Arab River increases the probability that some organochlorine pesticides will be retained and deposited.

∑DDT level in particulate matter ranged between 1221 µg/kg dry weight (d.w.) at (SH2) to 143 µg/kg dw at the origin (SH1) of this river (Table 2). Moreover, particulate ∑DDT in SH1comprised of *p,p*`-DDE and *o,p*`-DDT only, whereas in SH2 in addition to these two compounds *p,p*`-DDD was also present. Since DDD residues were encountered dissolved in SH1,its appearance in the suspended particulate matter collected from SH2 should be due to adsorption along the Shatt Al-Arab River course.

Apparently, the Euphrates River was the principal contributor of dissolve DDT group to the Shatt Al-Arab River (concentration of ∑DDT at ER was 407 ng/l). The Tigris River was the major donor of particulate DDT (concentration of ∑DDT at TR was 2560ng/kg dw), due to the variation in the nature of the suspended particulate matter, in ER mainly planktonic organism whereas in TR are enriched by materials such as silt and clay (Abaychi and DouAbul, 1985). Moreover in the ER the most predominant metabolite of DDT was DDD; thus, DDE:DDD:DDT ratio was 1:16:1 (Fig. 1). DDD is a pesticide in the DDT family and a minor component of technical DDT; it is also the major anaerobic breakdown product of *p,p*`-DDT (DouAbul *et al.*, 1988).

The concentration of Organochlorine pesticides in Khor Al-Zubair, North-West Arabian Gulf. particulate was less than Shatt Al-Arab River (Table 2). The most predominant metabolites of DDT was DDD (Fig. 2). Table (1) illuminate sampling Locations (st1, st2, st3 and st4) (Al-Saad and Al-Timari 1991). Also there were variation in the distribution of ∑DDT residues in Khor Al-Zubair elevated levels in water was at the northern region (336 ng/l) but elevated levels in particulate was at the southern region (2.4 µg/kg dw).

Endrin is relatively a short-lived insecticide that has been used on a few occasions in this region. DouAbul *et al.* (1988) found that dissolved endrin residues were encountered in the Euphrates River only (24 ng/l) but endrin residue in

Table 2. Variation in organochlorine pesticide concentration in the particulate (µg/kg dry weight) and the dissolved (ng/l) form in the studied region (1986-2009), and comparison with other areas.

Area	Endrin	Aldrin	Dieldrin	P,P – DDE	P,P – DDD	O,P – DDD	O,P – DDT	P,P – DDT	Σ DDT	lindane	Heptachlor	Cis chloriden	Trans chloriden	ref.
Particulate														
Shatt Al- Arab	85-154	nd	nd	76-204	nd-428	nd	nd	67-589	143-1221		68-186	11-171	nd-32	
Tigris at TR	217	nd	nd	484	1727	nd	nd	349	2560		679	102	10	DouAbul *et al.*, 1988
Euphrates at ER	nd	nd	nd	406	228	nd	nd	55	689		nd	88	nd	
Khor Al-Zuber		0.1-1.06	nd-0.97	nd-0.34		nd-1.63		nd-0.5	0.1-4.5		128-373			Al-saad & Al-Timari, 1991
Al-Hammar	nd-30.94	nd-82.53	nd-3.94	nd-30.18	nd-27.45	nd-23.13	Nd-24.8	nd-133.14	nd-238.7	0.08-339.87	0.08-321.62	nd-240.4		Al-Ali, 2012
Est.& Marine Netherland			5-230											Duursma *et al.*, 1986
Dissolved														
Shatt Al-Arab	nd	18-30	24-66	58-98	12.-28	8.-10	nd	3.-16	81-168		10.-79	25-57	12.-15	
Tigris at TR	nd	2.6	15	15	17	8	nd	2	42		9	38	12	DouAbul *et al.*, 1988
Euphrates at ER	24	7	22	29	351	5	3	19	407		4	37	24	
Khor Al-Zubair	nd-111	nd-31	12-195	nd-52		nd-85		nd-199	nd-336		nd-14			Al-saad & Al-Timari, 1991
Al-Hammar	nd-4.05	0.05-33.03	nd-0.3	nd-5.28	nd-3.15	nd-6.25	nd-3.92	nd-5.11	nd-23.71	0.15-73.78	0.19-59.5	0.24-200.79		Al-Ali, 2012
Nueces Est.-Texas			0.01-0.1						0.21-3.1	0.01-0.1	0.05-0.49	0.03-0.93		Ray *et al.*,1983b
Niagara				0.05-1.5				nd-3.8	0.05-5.3			nd-1		Oliver & Nicol, 1984
North Sea										0.8-6.2				Marchand & Caprais, 1985
Med. Port-Egypt				0.1-108	0.1-177			0.2-57	0.4-342	0.1-7.6		0.1-93		El-Dib & Badawy, 1985
Est.& Marine Netherland			1.-10		2				2					Duursma *et al.*, 1986
Cauvery R. India				nd-960	nd-530			nd-640	nd-2130					Begum *et al.*, 2009

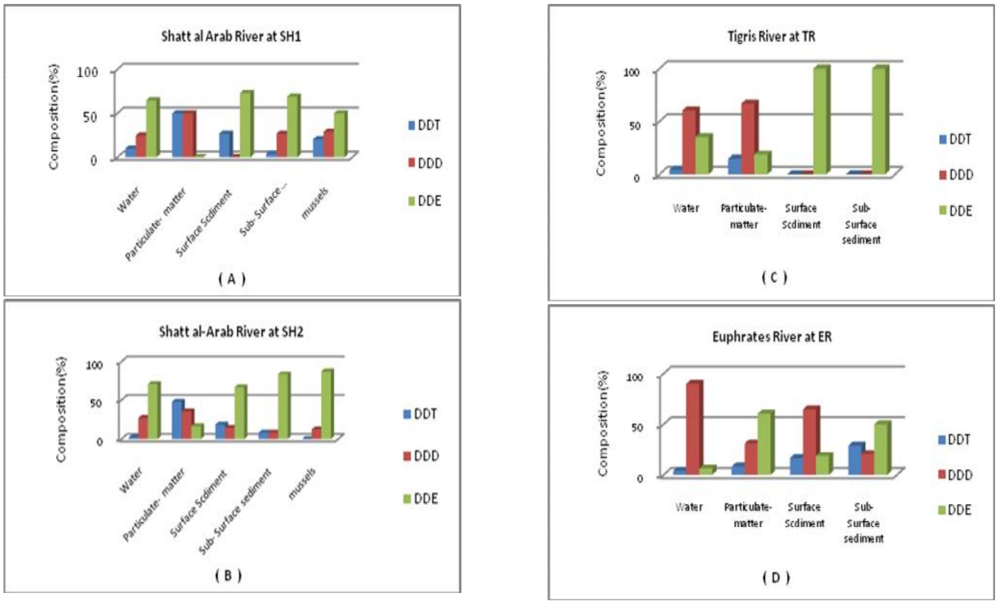

Figure 1. DDT composition in water, particulate matter, surface and sub-surface sediments and Bivalve tissues from Tigris-Euphrates-Shatt Al-Arab Delta (DouAbul *et al.*, 1988).

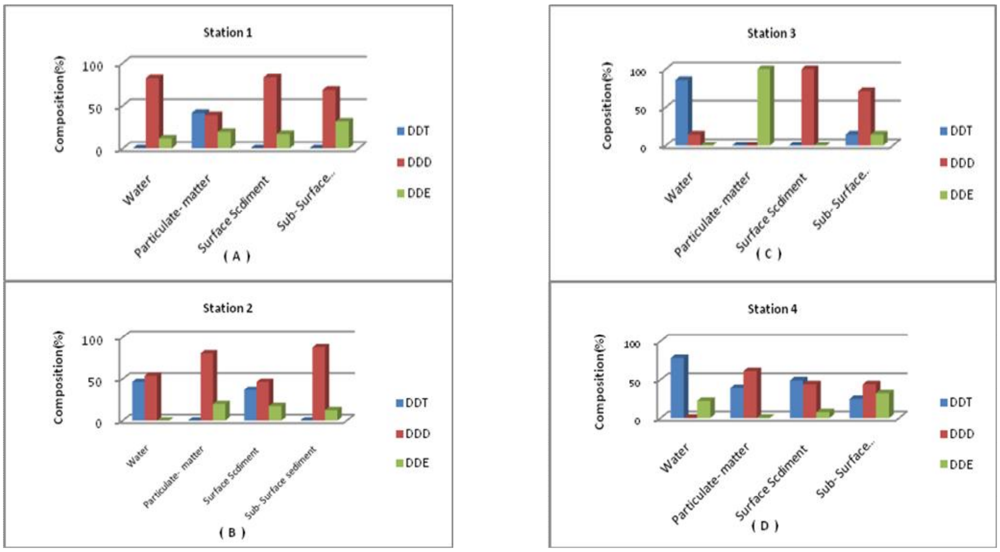

Figure 2. DDT composition in water, particulate matter, surface and sub-surface sediments from Khor Al-Zubair NW Arabian Gulf. (Al-Saad and Al-Timari 1991)

particulate encountered highest levels in the Tigris River (217 µg/kg dw) and lower level was detected in SH2 (84 µg/kg dw). While Al-Saad and Al-Timari (1991) detected endrin residues in the water (nd-111 ng/l) but not in the particulate matter (Table 2).

Although both aldrin and dieldrin have been officially banned in Iraq since 1976, because of their persistency and their lengthy use for agricultural and public health purposes (Al-Omar *et al.*, 1985), Aldrin residues were confirmed in the water sampled from SH1, SH2, TR and ER (30, 18, 26 and 7 ng/l, respectively). Aldrin oxidizes to dieldrin, were detected at SH1, SH2, TR and ER (66, 24, 15 and 22 µg/kg dw, respectively). However, particulate aldrin and dieldrin residues were below the detection limit in the Tigris-Euphrates-Shatt Al-Arab Delta. Since dieldrin residues were detected dissolved but not in the particulate form, it may be concluded that there was no recent input of this contaminant and the sediment act as reservoir giving up traces of dieldrin to the overlying water. While in Khor Al-Zubair, aldrin and dieldrin were detected in dissolved (rang nd-31 and 12-195 ng/l, respectively) and in particulate form (range 0.1-1.1 and nd-0.9 µg/kg dw, respectively).

Technical chlordane is a complex component of which, cis- and trans- are the most abundant and persistent isomers in the Tigris-Euphrates-Shatt Al-Arab Delta, both components were detected in the dissolved form. Thus cis-chlordane concentrations were as follows (expressed in ng/l) 57 in SH1, 25 in SH2, 38 in TR and 37 in ER, that of trans-chlordane were 15 in SH1, 12 in SH2, 12 in TR and 24 in ER. In particulate, cis- chlordane residues were detected also, concentrations in SH1, SH2, TR, and ER were 11, 171, 102 and 88 µg/kg dw, respectively. However, trans-isomers were evident in SH2 and TR only, which may be due to association of this isomer to clay and silt particles. But in Khor Al-Zubair both isomers were not detected in the dissolved and particulate form (Table 2).

Heptachlor has never been used in Iraq (DouAbul *et al.*, 1988). However the detection of its residues may be due to chlordane formulations which are known to contain heptachlor to an extent of 10-11% (Braun and Frank, 1980). Dissolved heptachlor residues exhibited regional variations; average concentrations in SH1, SH2, TR and ER were 10, 79, 19 and 4 ng/l, respectively. Particulate residues followed a similar distribution pattern of the dissolved, they were higher in SH2 (186 µg/kg dw) in comparison to that in SH1 (68 µg/kg dw). This observation may be due to the fact that chlordane insecticide has been used in the City of Basrah for soil treatment or to protect wooden structures against termites (DouAbul *et al.*, 1988). The Euphrates River was free of particulate heptachlor residues. Therefore the Tigris River was a major source of particulate heptachlor to the Shatt Al-Arab River (concentrations in TR was 679 µg/kg dw). Both dissolved and particulate residues were detected in Khor Al-Zubair, concentrations rang from nd-14 ng/l and 0.1-0.4 µg/kg dw respectively.

Sediment:

DouAbul *et al.* (1988) studied the distribution of organochlorine pesticides in the Tigris-Euphrates-Shatt Al-Arab Delta (Table 3), obtained that ∑DDT in surface sediment from SH2 (85 µg/kg dw) being higher in comparison with SH1 (24 µg/kg dw). Furthermore, sediment from SH2 comprised of p,p`-DDE; p,p`-DDT and p,p`-DDD, reflecting that of the suspended particulate matter collected from this location. However, the occurrence of DDD residues in sub-surface sediment from SH1 may be attributed to the production of an anoxic layer in the Shatt Al-Arab

Table 3. Variation in organochlorine pesticide concentration in the sediments (μg/kg dry weight) in the studied region (1984-2009), and comparison with other areas.

Area	Endrin	Aldrin	Dieldrin	P,P` - DDE	P,P` - DDD	O,P – DDD	O,P – DDT	P,P` - DDT	Σ DDT	lindane	Heptachlor	Chloriden	ref.
Shatt Al-Arab	40	__	20	5		nd		nd	5				DouAbul *et al.*, 1987b
Shatt Al-Arab	20	nd	10.-20						10.-15	nd			ROPME, 1986
NW-Arabian Gulf	47	nd	2.-5						20.-23	nd			
Shatt Al-Arab surf.sed.	3. - 16	nd-5	16-20	18-57	nd-12	nd	nd	6.-16	24.-85		nd-24	nd	DouAbul *et al.*, 1988
Shatt Al-Arab sub surf.sed	10.-18	nd-2	13-22	20-185	8.-16	nd	nd	2.-19	30-220		Nd	nd	
Tigris at TR surf.sed.	nd	nd	16	12	nd	nd	nd	12	24		Nd	nd	
Tigris at TR sub surf.sed	nd	nd	nd	28	nd	nd	nd	nd	28		7	nd	
Euphrates. at ER surf.sed.	nd	32	41	18	7	30	6	91	152		Nd	nd	
Euphrates. at ER sub. surf.sed.	11	nd	nd	12	5	nd	nd	7	24		Nd	nd	
Kuwiati coastal	nd-0.02	nd	0.01-0.02	0.01-0.03	0.1-0.2	nd	nd	nd-0.02	0.12-0.27				Al-Timari, 1997
Hor Al-Hammar	0.08	nd	0.04	0-0.6	nd	nd	0.01	nd	0.07				
Khor Al-Zubair	surf	0.01-0.02	0.03-0.1	nd-0.01		0.04-0.1		nd-0.1	0.04-0.21		0.01-0.06		Al-saad & Al-Timari, 1991
	Sub.surf.	nd-0.04	nd-0.03	nd-0.03		0.04-0.11		nd-0.03	0.04-0.17		nd-2.2		
Wetland from Hor Al-Hammar				0.29-2.33					0.29-2.33				DouAbul *et al.*,2009
Hor Al-Hammar	nd-36.45	0.45-98.24	nd-2.52	0.11-24.81	0.02-40.01	0.25-63.51	0.06-25.64	0.07-120.7	0.51-274.67	0.37-190.11	0.48-119.74	1.11-166.77	Al-Ali, 2012
Portland-Maine									0.03-42			0.03-9.8	Ray *et al.*, 1983a
Nueces Est-Texas.			0.01-0.94						0.12-1.5	0.01-0.16	0.06	0.01-3.8	Ray *et al.*, 1983b
Med. port- Egypt	0.1-100			96-600	110-400			0.2-53	206.2-1053	0.1-127		10-50	El-Dib & Badawy, 1985
Coverg -India.				0.09-4.5	nd-4.45			nd-0.98	0.09-9.99				Begum *et al.*, 2009
Indian Marine Env.		nd-3		nd-0.4	nd-0.9			0.3-109.5	0.3-110.5				Pandit *et al.*, 2001

River beneath the hypoxia water column and a surface oxidized layer (Abaychi and DouAbul, 1986). Furthermore, the settlement of the silt and clay particles enriched by organic matter along the Shatt Al-Arab River has enhanced the levels of DDT downriver (DouAbul *et al.*, 1988). DDT is likely degraded to DDD in the highly productive marshes of Southern Iraq and transported to the Euphrates River. It is unlikely that DDDis produced in this river; however, ∑DDT in its surface oxic sediments is composed of p,p`-DDE; p,p`-DDT;p,p`-DDD and o,p-DDT, whereas in the Tigirs River ∑DDT is composed solely of p,p-DDE (Figs. 1c and d). These variation may be due to different biogeochemical processes occurring in either river.

Also Al-Saad and Al-Timari (1991), noted that in Khor Al-Zubair, DDD was the most predominant metabolites of DDT in sediment samples (Fig. 2). Moreover, a significant level of concentration of DDE in coastal sediment may be attributed to the various kinds of degradation of DDT to DDE in marine environment, similar observation was found in Kuwiat coastal water by Al-Timari (1997) (Table 3).

DouAbul *et al.* (1987) suggest that endrin residues in the sediment of the Shatt Al-Arab river may act as a reservoir giving traces of endrin to the water, then DouAbul *et al.* (1988) confirmed that during the study of the Tigris-Euphrates-Shatt Al-Arab Delta, it was apparent that both surface and subsurface layers from SH2 contained relatively higher levels of endrin (concentration were 16 and 18 µg/kg dw, respectively) in comparison to that from SH1 (concentration were 3 and 10 µg/kg dw, respectively). In Kuwait coastal sediment endein was ranging from nd-0.02 µg/kg dw) but not detected in Khor Al-Zubair.

Aldrin residues were at or near the detection limit in the sediment samples from all regions. While, dielrin residues were found in both surface and subsurface sediment sample from the Tigris-Euphrates-Shatt Al-Arab Delta and Arabian Gulf region. This may be attributed to active conversion of aldrin to dielrin.

Both chlordane isomers were not detected in all sediment samples. Heptachlor residues were detected only in surface sediment at SH2 (24 µg/kg dw.) and Khor Al-Zubair range (0.01-0.06 µg/kg dw.) and subsurface sediment at TR (7 µg/kg dw.) and Khor Al-Zubair range (nd-2.2 µg/kg dw).

One of the last studies was done in the Mesopotamian wetlands of southern Iraq (DouAbul et al., 2009) only p,p`-DDE have been detected concentrations ranged from 0.29 to 2.33µg/kg, and it was found in all samples indicating its ability to persist under severe drying of previously exposed surface sediments, high dry of previously exposed surface sediments, high temperature, and intensive solar radiation.

However DDT has been officially banned in Iraq, hence its residues must be originated from more remote source likely from continuing illegal use. While Al-Ali (2012) confirmed the presence of ∑DDT, Endrin, Aldrin, Dieldrin, Clordane and Heptachlor in sediment samples from Hor Al-Hammar Marshes of southern Iraq, high concentration of Chlordane have been detected compared with the previous studies.

Fish:

Only DouAbul *et al.* (1987 a, b, c) and Villeneuvce *et al.* (1987) have studied the concentration of pesticides and their related compounds in fish. Villeneuvce *et al.* (1987) have analyzed 12 samples of different fish species collected from the coast of Kuwait in 1979, p,p`-DDE, p,p`-DDD and p,p`-DDT were detected in the ranges of 1.5-19.5, nd-8.2 and nd-8.9 µg/kg dw, respectively.

DouAbul *et al.* (1987c) examined 13 fish species collected from the NW Arabian Gulf and 4 fish species from Hor Al-Hammar marsh. The ∑DDT in fish obtained from the NW Arabian Gulf was mainly comprised of p,p`-DDT and p,p`-DDE ranged concentrations from nd-6 and 1-5 µg/kg wet weight, respectively. The latter was detected in all the samples analyzed, while the percentage occurrence of p,p`-DDT was approximately 85%. In most cases, conversion of DDT into DDE is initiated by soil micro-organisms immediately after it enters the environment. Other factors such as alkaline pH, light, or heat may also produce chemical changes in the original DDT molecule.

Thus, the above observation may be due to metabolic conversion (Bridges *et al.*, 1963) and/or dehydrochlorination (Hannon *et al.*, 1970) in warm, rather alkaline waters of the Arabian Gulf. Moreover, because the volatility of DDE is several times greater than that of DDT (Spencer, 1975), it is logical to presume that DDE is more readily transferred via the atmosphere of the coastal area of the Arabian Gulf. Technical DDT generally contains <25% o,p-DDT, an impurity, low percentage occurrence of both o,p`-DDT and o,p`-DDD which were 30 and 20%, respectively, because o,p-isomers are less persistent than their p,p-analogs (Fry and Toone, 1981).

Comparison of ∑DDT residues in fish tissue from the NW Arabian Gulf with those from Hor Al-Hammar Lake revealed that the latter retained significantly higher concentration (residue level ranged from 8 µg/kg wet weight to 92 µg/kg in the carp *Cyprinus carpio* (Table 4). The rather elevated values may be due to the fact that Hor Al-Hammar used to be sprayed with technical DDT until recently.

Furthermore, ∑DDT in fishes from the NW Arabian Gulf were an order of magnitude lower than the range of values reported for fish captured from the coastal waters of Oman (Burns *et al.*, 1982). Since the Omani territorial waters are much further down from the Shatt Al-Arab Delta than the NW Arabian Gulf, our previous contention that there is an additional source of DDT to the Arabian Gulf apart from the Shatt Al-Arab Delta and the Tigris-Euphrates River Basin is further supported.

Endrin is considered to be the most toxic of all commercial insecticides to fish and this was the most dominant compound in fishes from the NW Arabian Gulf. Endrin residues were detected in approximately 90% of these fishes, with a mean values ranging from nd to 28µg/kg ww. Slightly higher concentrations of endrin have been detected in fishes from the Hor Al-Hammar Lake with residue levels ranging from 3 to 67µg/kg ww. Endrin has not been detected in the Arabian Gulf before, which may be due to the fact that this insecticide is relatively short-lived and has been used in few occasions in this region.

Residue levels of dieldrin in the fishes from the NW Arabian Gulf were at or near the detection limit. More frequent residues of dieldrin were observed in the Hor Al-Hammar Lake, percentage occurrence was 80% with an average concentration from nd to 3 µg/kg ww. Dieldrin is a metabolite of aldrin and the residue of the latter was below the detection limit, which may be due to the active conversion of aldrin to dieldrin.

Seasonal level of organochlorine pesticide residues in *Tenualosa ilisha* (muscle tissues) captured from the Shatt Al-Arab River was studied by DouAbul *et al.* (1987a) and found that maximum ∑DDT levels (average concentration 131 ppb wet weight) were detected in summer, while minimum value (average concentration 51 ppb wet weight) was recorded in autumn. Contrarily, endrin residue levels were

Table 4. Variation in organochlorine pesticide concentration in fish, µg/kg dry weight, in the studied area (1979-2009), and comparison with other areas.

Area	Fat %	Endrin	Aldrin	Dieldrin	P,P – DDE	P,P – DDD	O,P – DDD	O,P – DDT	P,P – DDT	Σ DDT	Heptachlor	chloriden	lindane	ref.
Kuwiati coastal					1.5-19.5	nd-8.2			nd-8.9	1.5-36.6				Villeneuve *et al.*, 1987
Shatt Al- Arab	3	16		2						13	3	nd		DouAbul *et al.*, 1987a
Al-Hammar	3	154		8						58	6	58		
Shatt Al- Arab	3.-6-2	3.-90		2.-32	4.-64			12.-15	nd-10	16-189				DouAbul *et al.*, 1987b
Al-Hammar	3	20		7	26			131	9	166				
NW-Arabian Gulf	2-1-6-1	nd-28		nd-4	1.-5		nd-3	nd-3	nd-6	1.-21				DouAbul *et al.*, 1987c
Al-Hammar	2-2-5-1	3.-67		nd-3	1.53		nd	3.-10	4.-29	8.-92				
Al-Hammar		nd-3.31	0.05-25.79	nd-0.35	nd-3.74	nd-5.01	nd-8.34	nd-11.1	0.02-33.52	0.02-61.71	0.95-50.63	2.42-99.18	0.23-67.25	Al-Ali, 2012
West Medi.										1.-1792				Albaigés *et al.*, 1987
USA lakes	1.5-18-9			0.29-28	0.88-299	nd-1281			195	0.88-1775	0.98-6.35			Jaffe *et al.*, 1985
Nil R- Egypt		18-31			1.7-22	2.-48	15.-93		35-141	53.7-304				Aly & Badawy, 1984
Med. Port Egypt		0.1-70	0.1		5.-196	0.2-144			0.2-25	5.4-365		0.1-108	0.1-45	El-Dib & Badawy, 1985
Cauvery R. India					0.05-3.28	nd			nd-0.77	0.05-4.05				Begum *et al.*, 2009
Indian Marine Env.	India		nd-0.3		0.2-10.8	0.2-6			nd-2.7	0.4-19.5				Pandit *et al.*, 2001
Rad Sea Yemen										1.1-6.7				Al-Shwafi *et al.*, 2009
Adan Gulf Yemen										1.-6.4				

higher in autumn (average concentration 147 ppb wet weight) (Fig. 3) corresponding to the fat content. However, it has been demonstrated that there was a trend toward a reduction in endrin levels in fishes which were exposed to both DDT and endrin concurrently (Denison *et al.*, 1985). The remaining organochlorine pesticides did not exhibit a definite trend of variation with respect to seasons.

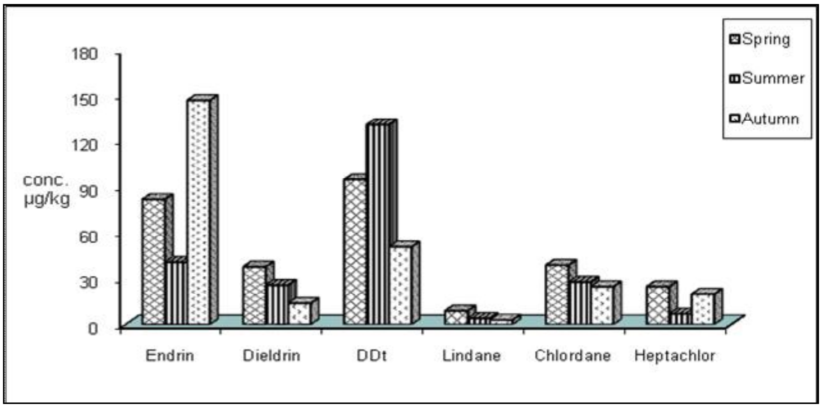

Figure 3. Seasonal variation in the concentration of organochlorin pesticides in fish captured from Shatt Al-Arab River (DouAbul *et al.*, 1987a)

Other Biota:

Organochlorine compounds have surface-seeking properties, they are found adsorbed to suspended matter and sediments (Duinker *et al.*, 1984). Therefore, the concentration of organochlorines present in the tissues of collected species will reflect the contamination of that environment. In a preliminary survey of marine samples, Anderlini *et al.* (1981) observed DDE residues in the oyster *Pinctada margaratifera* collected from Kuwaiti waters (concentration range from 0.59 to 3.8 µg/kg dw) (Table 5). Also Villeneuve *et al.* (1987) detected p,p`-DDE (concentration 1.0-1.3 µg/kg dw) and p,p`-DDD (concentration 0.3-0.4 µg/kg dw) in crab and squid respectively collected from Kuwaiti waters. These observations could serve as evidence that inputs from the Shatt Al-Arab River may indeed be taking place. DouAbul *et al.* (1988) chose the fresh water mussel *Corbicula fluminea*, the most widely distributed filter feeder, to obtain a preliminary assessment of the levels of organochlorine pesticide residues in the Shatt Al-Arab ecosystem. The analysis had detected residues of p,p`-DDE,o,p`-DDD, p,p`-DDT, endrin, alidrin, dieldrin, cis-chlordane and trans-chlordane, with concentration ranges of 43-670, 12-99, nd-15, 161-540, 26-72, 24-493, 6-31 and 5-15 µg/kg ww, respectively. Filter feeders may be expected to absorb chlorinated hydrocarbons from the water and from the particulate matter taken up during feeding. The work had shown that the pesticide pattern of *C. fluminea* closely reflect the pattern of pesticides in the suspended particulate matter. Also the bioconcentration factors (BCF, concentration in organisms on fat weight basis/concentration in water) were calculated in order to know the behavior of organochlorine pesticides for *C. fluminea* (Fig. 4), and found that the BCF of total organochlorine pesticide residues was approximately 2.5 x 106 for this organism.

Table 5. Variation in organochlorine pesticide concentration in biota, µg/kg dry weight, (1978-1986) in the studied area, and comparison with other areas.

Area	Biota	Endrin	Aldrin	Dieldrin	P,P – DDE	P,P – DDD	O,P – DDD	O,P – DDT	P,P – DDT	Σ DDT	Cis-chloriden	Trans-chloriden	ref.
Kuwiati Coastal	oyester				0.59-3.8					0.59-3.8			Anderlini *et al.*, 1981
Kuwiati coastal	squiel				1.3	0.4			nd	1.7			Villeneuve *et al.*, 1987
	crab				1	0.3			nd	1.3			
Shatt Al- Arab	mussel		nd	2.-5						13.-20			ROPME, 1986
Shatt Al- Arab	mussel	161.-540	26.-72	26.-72	43.-670	nd	12.-99	nd	nd-15	55.-784	6.-31	5.-15	DouAbul *et al.*, 1988
Portland-Maine										4.-11	0.03-5.4		Ray *et al.*, 1983a
Antonio Bay- Texas	oyster			nd-48	nd-72					nd-72			Petrocelli *et al.*, 1974
	carbs			nd-44	nd-55					nd-55			
	clams			nd-18	nd-79					nd-79			
Est.& Marine -Netherland	mussel			1.5-60	0.-51	0.-51							Duursma *et al.*, 1986
Costal area Tanzania	Fresh water Biota			nd-2.7	62.42	62.-4.5			2.7-4.4	10.9-57.5			Haji Mwevura *et al.*, 2002
	Marine water Biota			nd	3.1-18.4	6.6-30.8			1.1-2.6	10.8-52.1			
Red Sea	Squid									1.8			Al-Shwafi *et al.*, 2009
Adan Gulf	Squid									1.5			

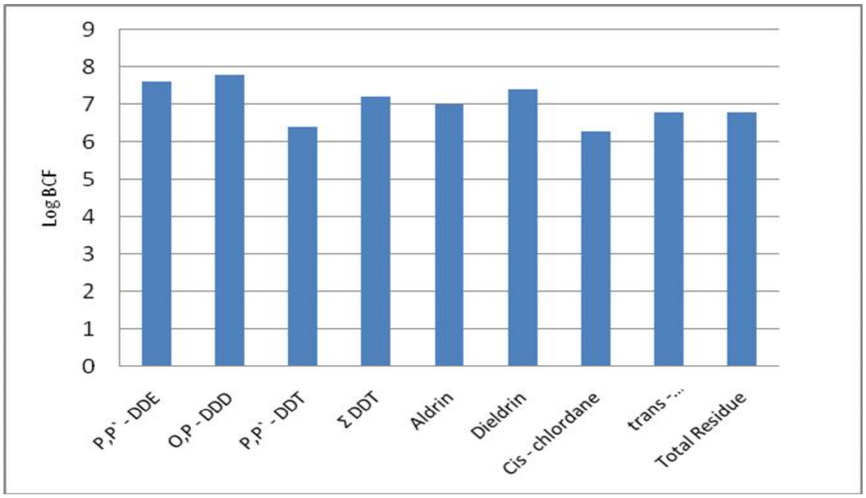

Figure 4. Bioconcentration factor (BCF; concentration in *Corbicula* tissue/concentration in water) of organochlorine pesticides.

Shrimp:

Villeneuve *et al.* (1987) have measured organochlorine pesticide residues in shrimp tissue collected from coastal waters of Kuwait. The concentration range of p,p-`DDE, p,p`-DDD, and p,p`-DDT were 0.3-3.6, nd-1.8 and nd-0.8 µg/kg dw, respectively. While only o,p-`DDT (2 µg/kg dw) was detected in shrimp sample from Shatt Al-Arab (DouAbul *et al.*, 1987b).

Al-Timari (1997) used organochlorine pesticide as a possible marker to differentiate between the *Metapenaeus affinis* from the Southern marshes of Iraq and the Kuwaiti water (if the *Metapenaeus affinis* migrated from the Gulf to the Marsh, it will accumulate organochlorine pesticide distributed in the Marshes), and if there is any interaction between their populations. Also she used *Penaeus semisulcatus* as marine biological sample, which did not migrated to the Iraqi waters. The results showed that DDT in *P. semisulcatus* was mainly comprised of p,p-`DDE, p,p-`DDD and p,p-`DDT, with concentration ranges from 0.2-0.8, 1-2, nd-0.2 and 0.1-0.4 µg/kg dw, respectively. Organochlorine pesticide in *M. affinis* from the Gulf and Marsh area had shown similar pattern except p,p-`DDT (nd-0.3 µg/kg dw) was detected in the Gulf samples only. While other compounds like Endrin, Dieldrin, p,p-`DDE, p,p`-DDD, and o,p`-DDT were detected in *M. affinis* collected from both areas, with concentration ranges from nd-0.2, 0.4-0.8, 0.4-1.5, 1-2.2 and nd-0.8 µg/kg dw, respectively, from Kuwaiti water and 0.4, 0.4, 2.2, 1.2 and 3 µg/kg, respectively, from Iraqi water. Heptachlor residues were detected in all samples, their concentrations were nd-0.2, 0.1-0.3 and 0.3 µg/kg dw, in *P. semisulcatus* and *M. affinis* from the Gulf and *M. affinis* from the Marshes, respectively. p,p`-DDD occurred in sediment (Table 3) and shrimp samples (Table 6) from the Arabian Gulf and in shrimp from Hor Al-Hammar marsh but did not detected in the *M. affinis* from the Arabian Gulf.

The seasonal study of p,p`-DDD in the Arabian Gulf showed higher concentrations in the sediments during spring and autumn but lower in winter.

Table 6. Variation in organochlorine pesticide concentration in the shrimp, µg/kg dry weight, during 1979-2009, and comparison with other areas.

Area	Fat %	Endrin	Aldrin	Dieldrin	P,P – DDE	P,P – DDD	O,P – DDD	O,P – DDT	P,P – DDT	Σ DDT	Heptachlor	ref.
Kuwiati coastal					0.3-3.6	nd-1-8			nd-0.8	0.3-4.4		Villeneu *et al.*, 1987
Shatt Al-Arab	1.1	nd		nd	nd		2		nd	2		Doabul *et al.*, 1987
M. aff. from Al-Hammar	4.-5	0.4	nd	0.4	2.2	1.1-2.2	nd	3	nd	6.3	0.3	Al-Timari 1997
P. semi. Kuwiati coastal	2.6-3.3	nd	nd	nd	0.2-0.8	1.2	nd-0.2	nd	0.1-0.4	1.3-3.4	nd-0.2	
M.aff. from Kuwiati coastal	3.5-4.1	nd-0.2	nd	0.4-0.8	0.4-1.5	1.-2.2	nd	nd-0.8	nd-0.3	1.4-4.8	0.1-0.3	
Al-Hammar		nd-24.28	0.65-27.95	nd-1.34	nd-16.35	nd-27.64	nd-45.11	nd-10.65	0.03-25.56	0.03-125.31	0.67-12.13	Al-Ali, 2012
Cauvery R. India					0.45-3.31	nd-0.63			nd-0.98	0.45-4.92		Begum *et al.*, 2009
Red Sea Yemen										0.9-23		Al-Shwafi *et al.*, 2009
Adan Gulf Yemen										0.3-2.3		

While the concentration of p,p-`DDD in *M. affinis* was higher during winter probably due to the accumulation of organochlorine pesticide from Hor Al-Hammar marsh. In contrast p,p`-DDE in the sediment samples of the marsh was higher than the Arabian Gulf this was reflected in the shrimp samples (Fig. 5) illuminate the seasonal variation in the concentration of organochlorine pesticides (Al-Timari, 1997). The elevated dieldrin levels in the marsh sediment compared with the Arabian Gulf, may be due to the adsorption of this residues to the suspended particulate matter carried by the Euphrates river and deposited in considerable amounts in Hor Al-Hammar marsh (DouAbul *et al.*, 1988), and as was reflected in the shrimp samples, there were lower concentrations in *P. semisulcatus* than *M. affinis*. Adema and Compaan (1975) found that the level of dieldrin in mussels placed in a solution of the chemical did not stabilized after two weeks nither was a 90% elimination achieved in clean water after a mouth. On the contrary, shrimp, which showed a much lower accumulation factor, exhibited a steady state in the same conditions. Satsmadjis and Voutsinou-Taliadouri (1983) suggested that the shrimp would require a few months to come into equilibrium with their environment.

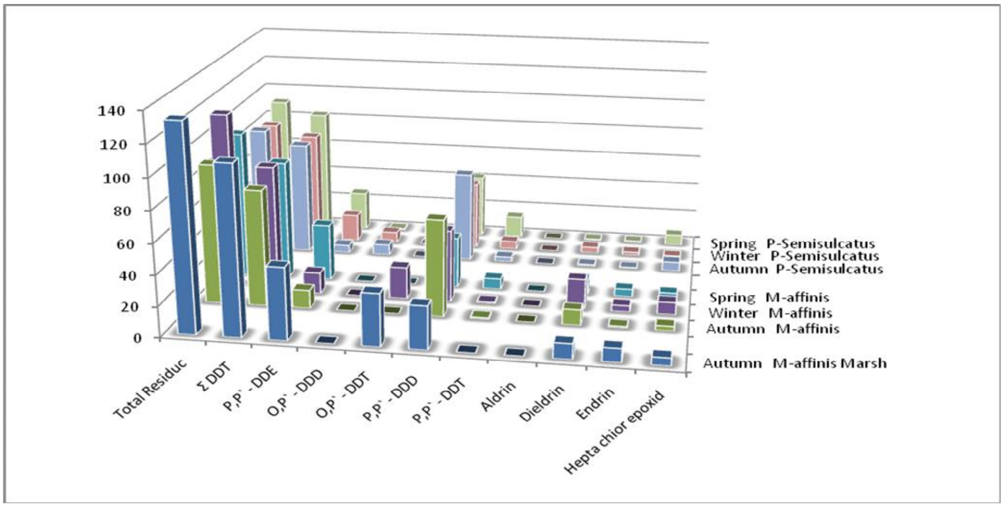

Figure 5. Seasonal Variation in the concentration of organochlorine pesticides in shrimp (ng/g lipid) captured from Hor Al-Hammar Marsh and Arabian Gulf.

Conclusions

1- Application of high-resolution gas chromatography has confirmed residues of p,p´-DDE, o,p´-DDD, o,p´-DDT, p,p´-DDD, p,p´-DDT, endrin, aldrin, dieldrin, cis- and trans- chlordane, and heptachlor in various phases of Shatt Al-Arab Delta and the Arabian Gulf. DDT residues were the most prominent organochlorine pesticides in the area.

2- To the Shatt Al-Arab River, organochlorine pesticide residues in the dissolved form were mainly contributed by the Euphrates River, whereas the particulate form, were principally transported by the Tigris River, and to the variations in the nature as well as the amount of suspended particulate matter carried by either river.

3- Biota tissues reflect the pesticides pattern of living area. Shrimp samples indicate that the populations are marked by their environment and probably have their own life cycle and their own migration pattern.
4- p,p´-DDE residue has survived the drying of the wetlands that took place during the 1990s despite harsh environmental conditions in the intervening period to the present.
5- The contamination levels were relatively low for all the studies except the last one, high concentration of organochlorine pesticides have been detected especially chlordane. Therefore, further studies are needed to be done

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بقايا المبيدات العضوية المكلورة في الاهوار وشط العرب والخليج العربي

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المستخلص - تعتبر المبيدات العضوية المكلورة من المركبات السامة وكذلك تتصف بأستقراريتها ومقاومتها للظروف الكيمياوية والبايولوجية.وهي موجودة في كل مكان في البيئة وتذوب في الدهون ونتيجة لذلك أصبحت في قمة القائمة السوداء للمواد الضارة بالبيئة. عدة دراسات أنجزت حول المبيدات العضوية المكلورة خلال الفترة 1979-1991.وتم قياسها في عينات المياه والرواسب والأحياء والتي جمعت من بيئة الأهوار وشط العرب والخليج العربي. جميع الدراسات أكدت وجود د د ت∑ والاندرين والدي الدرين والكلوردين والهبتاكلور إذ تراوحت تراكيزها بين 336-nd و 111-nd و 12-195 و 57-nd و 79-nd نانوغرام/لتر في الجزء الذائب، وبين 0.1-2560 و 217-nd و 0.97-nd و 171-nd و 679-nd مايكروغرام/كغم في الجزء العالق، و 0.04-220 و 0-47 و 22-nd و nd و 24-nd مايكروغرام/كغم في الرواسب وبين 1-189 و 32-nd و 58-nd و 6-nd جزء من البليون في الأسماك، وبين 0.59-784 و 166-540 و 2-72 و 5-3.1 و nd جزء من البليون في القواقع وبين 0.3-6.3 و 0.4-nd و 0.8-nd و nd و 0.3-nd جزء من البليون في الروبيان على التوالي. أما في العقد الماضي فقد تم القيام بدراستين فقط حول بقايا المبيدات العضوية المكلورة. إحداهما في الأراضي الرطبة لبلاد الرافدين جنوب العراق وتم الكشف عن بقايا بارا بارا- د د ت وبنسبة 100% في عينات الفحص. وهذا يدل على قدرتها للاستمرار في ظروف التجفيف الشديد التي تعرضت لها رواسب المنطقة سابقا ولفترة أكثر من 10 سنوات وارتفاع درجة الحرارة والإشعاع الشمسي المركز. أما الدراسة الأخرى فكانت لعينات مياه ورواسب وأسماك وروبيان من هور الحمار في العراق، وأظهرت تراكيز عالية للكلوردين في عينات المياه والرواسب. ولذلك هناك حاجة إلى مزيد من العمل لتحديد مستويات بقايا المبيدات العضوية المكلورة في منطقة الأهوار وشط العرب والخليج العربي.