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Organochlorine contaminants in eggs of the yellow-legged gull (*Larus cachinnans michahellis*) in the North Eastern Mediterranean: is this gull a suitable biomonitor for the region?

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"Capsule": *The eggs of the yellow-legged gull are not good biomonitors for organochlorines*

Abstract

Levels of eight PCB congeners and thirteen organochlorine pesticides were measured in eggs sampled at four yellow-legged gull colonies from the Aegean Sea (NorthEastern Mediterranean) in 1997. There were no significant differences among colony areas in the median concentrations in any of the pollutants whereas cluster analyses did not generally reveal reasonable pollution patterns. The maximum concentrations of four congeners were found at Kinaros colony and of nine compounds were found at Lipsos colony. Fingerprints in both groups were similar in all areas. Of PCBs, congener 28, 118, 138, 180 and of pesticides β-BHC and 2,4′-DDD were prominently dominant suggesting a particular pollution pattern in this region. Statistically significant correlations were found between most of the higher PCBs in all areas studied. The DDT metabolites correlated mostly with other OCs. We suggest that regional pollution by both groups is not adequately reflected in the eggs of this gull probably due to its extensive scavenging habits and, though information is needed from more colonies, it seems to be a poor biomonitor for organochlorines in this region.

Keywords: PCBs; Organochlorine pesticides; Eggs; *Larus Cachinnans*; Aegean Sea

1. Introduction

Polychlorinated biphenys (PCBs) and organochlorine pesticides (OCs) are two groups of chemicals commonly detected in environmental samples and biota. Their long ago banning from most part of the world seems to have reduced levels (Bishop et al., 1996; Hebert et al., 1999; Aurigi et al., 2000) but their persistence in biota is mainly due to their lipophilic structure and bioaccumulative properties (Tanabe et al., 1987; Harding et al., 1997). Though their levels dropped, monitoring must continue due to their hamful effects to man and wildlife (Gilbertson et al., 1991; Longnecker et al., 1997). As many studies indicated, in the Mediterranean these organochlorines can be found in a great variety of biota (Focardi et al., 1988b; Lopez-Martin et al., 1995; Swindlehurst et al., 1995; Fasola et al., 1998). Waterbird eggs especially those of some pelecaniform (*Pelecanus* sp., *Phalacrocorax* sp.) and larid (*Larus* sp., *Sterna* sp.) species have been proved to be particularly useful as bioindicators of organochlorines in the aquatic environment (Vermeer and Reynolds, 1970; Weseloh et al., 1989; Scharenberg, 1991; Pastor et al., 1995b). Moreover, egg pollutant concentrations enable the assessment of hazards faced by embryos during development. The eggs of the herring gull (*Larus argentatus*) have been used in long-term studies in the Canadian Great Lakes and elsewhere (Oxynos et al., 1993; Weseloh et al., 1995). In the Great Lakes, the herring gull has been selected being a top predator in the food web and a primarily fish eater (having a stable diet), a year round

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resident, nesting colonially (easy to locate count and sample) and a cosmopolitan species allowing for comparisons over broad geographical areas (Mineau et al., 1984).

In the Mediterranean the closest relative species to the herring gull is the yellow-legged gull (*Larus cachinnans michahellis*, formerly *L. argentatus michahellis*, Cramp and Simmons, 1983). Some studies in the western Mediterranean revealed considerable organochlorine levels in its eggs (Lambertini and Leonzio, 1986; Focardi et al., 1988a; Pastor et al., 1995a). Although the yellow-legged gull is a very common and widespread breeding species in the eastern Mediterranean and in the Aegean Sea in particular (Handrinos and Akriotis, 1997), its use as an organochlorine biomonitor has never been attempted so far. The scope of this study was to investigate the levels of some environmentally important PCBs and organochlorine pesticides in eggs of the yellow-legged gull from some Aegean colonies and to investigate the usefulness of the species is a biomonitor for this region.

2. Materials and methods

2.1. Study areas

The general location of the colonies sampled is shown in the Fig. 1. In the Dodecanese area, Aegean Sea, the yellow-legged gull colonies were situated on small-uninhabited rocky islands varying in extent, vegetation composition, cover and slope. The name given for each colony site corresponds to the nearest big island. The distance between Lipsos and Arki colonies is c. 8.5 km whereas between Kinaros and Lipsi (being the closest) is c. 70.5 km. The Evros Delta is far nothern c. 370 km from the Dodecanese study area.

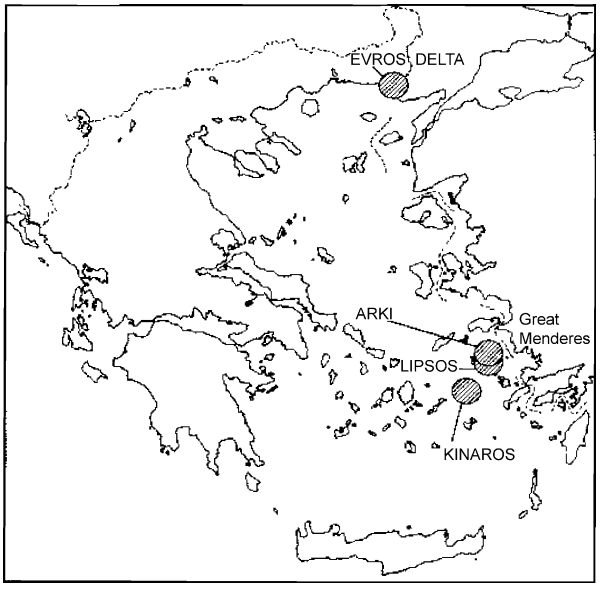

Fig. 1. Map of Greece indicating the colony areas studied.

2.2. Egg sampling

In April 1997, freshly laid eggs with uncracked shells were collected under licence from three Dodecanese colonies (within the frame work of a project on the Audouin's gull (*Larus audouinii*) (Goutner et al., 2001) and from a colony in the Evros Delta (Fig. 1). The latter collection aimed in comparing pollutant levels from absolutely different areas and habitats (islands–wetland) where we predicted that levels should be different due to different pollution regimes. The eggs were placed in cotton-spread cartons (of these used to transport hen's eggs) and were opened as soon as possible after collection, their contents poured in chemically cleaned glass containers and were subsequently deep frozen to −20 °C. Then the material was transported to the University of Ioannina for chemical analyses.

2.3. Materials

In total, the following eight PCB congeners were analysed: PCB 8, 20, 28, 52, 101, 118, 138, 180. Of these congeners five (in terms of the PCB IUPAC numbers 28, 52, 101, 138, 180) belong to the group known as "target" or "indicator" PCBs (Scrimshaw and Lester, 1995; Bachour et al., 1998). PCB 153 is also included in this list but we did not analysed it due to lack of standard. In addition, we measured the levels of PCB congeners 8 and 20 on the basis of their presence in the environment and representativeness of different substitution patterns (dichloro to octachlorobiphenyl). Furthermore the determination of PCBs 8, 20, 28, is relevant with the metabolizable fraction of PCBs congeners in which the structure permits the formation of an epoxide. Although di, tri and tetrachlorobiphenyls exhibit low accumulation in bird body tissues in comparison to highly chlorinated congeners (Gagnon et al., 1990; Metcalfe and Metcalfe, 1997), their bioaccumulation in fish and marine birds is largely mediated by food and particulate ingestion (Stranberg et al., 1998) and therefore we were stimulated to examine this in the present study. The concentrations of several di and tri PCBs has also been reported in various waterbird species (Henriksen et al., 1996; Zimmermann et al., 1997; Stranberg et al., 1998; Van der Brink and Bosveld, 2001). The organochlorine pesticides analysed in this study were α-BHC, β-BHC, lindane, heptachlor, heptachlor epoxide, aldrin, dieldrin, endrin, 2,4′-DDT, 2,4′-DDD, 4,4′-DDT 4,4′-DDD 4,4′-DDE.

PCB-standards were obtained from Dr. Ehrensdorfer GmbH laboratory in concentrations of 10 mg/ml. Supelco No. 4-9151 organochlorine pesticides mixture standard in isooctane was used in concentrations of

mg/ml for the chromatographic analysis. All solvents used (hexane, acetone, petroleum ether), were pesticide residue analysis grade, purchased from Pestiscan (Labscan Ltd, Dublin, Ireland). Florisil (50–100 mesh) and sodium sulfate (pro-analysis) were from Merck (Darmstadt, Germany). Glassware was soaked, cleaned with chromic solution, thoroughly rinsed with distilled water and acetone and heated at 150 °C for 12 h.

2.4. Analytical procedures

Egg contents were homogenized in a blender and a part from each sample (1–2 g) was transferred to a watch glass and then to an oven for drying to constant weight in 60 °C for estimating moisture content. A mean egg moisture content of 74.7% (S.D.: 2.5) was found. Mean lipid content, determined with the Bligh and Dyer method, was 26.2% of dry weight. An aliquot of 5–10 g was homogenized again with 20–30 g of sodium sulphate in glass tubes of 100 ml. The mixture was extracted firstly with 20 ml, and followed by two more times with 10 ml with hexane: petroleum ether (1:1) mixture using a vortex (2 min), sonication bath (5 min) and manual mixing (5 min) with a glass rod. The extracts were collected in polypropylene centrifugation tubes of 50 ml, centrifuged in 4000 rpm for 5 min. The supernatant was evaporated in a rotary evaporator to 10 ml and lipids were then removed by treating the extracts with 0.5–0.8 ml aliquots of concentrated sulfuric acid. The procedure was repeated until the acid layer remained colorless (Pavoni et al., 1991). The clean-up was completed by adsorption chromatography, eluting the colorless layer through a chromatography glass column 1 cm i.d., 20 cm length, provided with o teflon stopcock. The column was packed as follows: 5 cm Florisil slurry was added first under gentle tapping of the column and keeping stopcock open, to avoid bubbles, and then 2 cm dried sodium sulfate were added. The column was washed with 20 ml n-hexane. All solvents used for packing the column were degassed in sonication bath (Fytianos et al., 1997). The purified sample was evaporated in a rotary evaporator to a ca. 5 ml and in gentle N_2 stream at 35 °C to a ca. 0.5 ml, then samples were stored in silanized vials in a refrigerator (−20 °C). Mean recoveries and method detection limits for each congener and compound are given in Konstantinou et al. (2000).

2.5. Chromatographic conditions

2.5.1. GC-ECD

A Shimadzu 14B gas chromatograph equipped with ^{63}Ni electron capture detector (ECD) was used for the organochlorine residue analysis. The capillary column used was a DB-5, 30 m×0.32 mm i.d., contained (5% phenyl) methyl polysiloxane (J and W Scientific, Folsom, CA) followed the temperature program: 150 °C (2 min), 150-200 °C (5 °C/min), 200 °C (45 min), 210–270 °C (10 °C/min), 270 °C (3 min). The temperatures were set at 250 °C for the injector and 300 °C for the detector. Helium was used as the carrier and nitrogen as the make-up gas. Pure reference standard solutions were used for instrument calibration, recovery, quantification and confirmation using internal standard method (Internal Standard: pentachlorocyclohexane). The splitless mode was used for injection of 1 μl volume, with the valve opened for 30 s.

2.5.2. GC-MS

The confirmation of organochlorine residues was performed by using a GC-MSD, QP 5000 Shimadzu equipped with DB-5 capillary column, 30 m×0.32 mm i.d. contained (5% phenyl) methyl polysiloxane (J and W Scientific, Folsom, CA) was used at the following chromatographic conditions: Injector temperature 220 °C, column programme of temperatures 55 °C (2 min), 55–210 °C (5 °C/min), 210 °C (20 min), 210–271 °C (20 °C/min), 270 °C (4 min). Helium was used as the carrier gas at 14 psi. The interface was kept at 270 °C. The spectra were obtained at 70 eV. The splitless mode was used for injection of 1 μl volume, with the valve opened for 30 sec. Two ions (M^+, M^{+2}) for each pesticide and biphenyl were chosen for screening analysis in selected ion monitoring mode (SIM). Ions were selected after injecting a concentrated solution of compounds and recording the "total ion chromatogram". The ions' traces were divided into five groups that were recorded sequentially during the injection, on the basis of the retention times of the single substances. For the PCBs identification the selected ions for each degree of chlorination were confirmed by checking the ratios of intensities of ions belonging to the same cluster (Raccanelli et al., 1994; Singh et al., 1998).

2.6. Statistical procedures

Concentrations of pollutants were not normally distributed, thus the median of each pollutant, total median concentrations of PCBs (hereafter ΣPCBs) and Ocs (ΣOCs) were compared. We used the Kruskal–Wallis χ^2 test to compare concentrations from the four areas. Correlations between contaminants were tested using Spearman rank correlations. We calculated the ratio ΣOCs/ΣPCBs in samples as a measure of agrochemical or industrial pollution (Fossi et al., 1984; Pastor et al., 1995b) and we then compared the median values as previously specified. Cluster analysis (with Euclidean distances as distance measure and single linkage as a linkage rule), separately for PCB and OC percentage levels, were used to evaluate differences in pollution patterns among areas studied.

3. Results

3.1. Polychlorinated biphenyls

All eight congeners analysed were detected in the samples from all areas. Nevertheless mean (median) concentrations of all congeners were very low (Table 1). Against our prediction, there were no significant differences among areas in the median concentrations in any congener. The maximum concentrations of four out of the eight congeners were found at Kinaros (PCB 28, 52, 138, 180), of two at Lipsos (PCB 20, 101) and of one at both Arki (PCB 8) and Evros Delta (PCB 118). The proportions of the eight individual congeners were similar among the areas studied resulting in a similarity in the fingerprints (Fig. 2). In these fingerprints the higher PCBs dominated with the sequence PCB 138 > 118 > 180. The proportion of PCB 28 was considerably higher than the remaining congeners.

The cluster analysis separated Kinaros from the other areas and joined Arki and Lipsos separately from the Evros Delta (Fig. 3).

Statistically significant Spearman Rank Correlations between PCB congeners were mainly found between higher PCBs (101 and above) and other either low or high PCBs (13 out of a total of 14 significant correlations, Table 2). Common correlations found in all island areas were between PCB 20 and 101, in two between 118 and 138 and between PCB 8 and 118. PCB 28 and 52 did not correlated significantly in any area. In the Evros Delta none significant correlation was found.

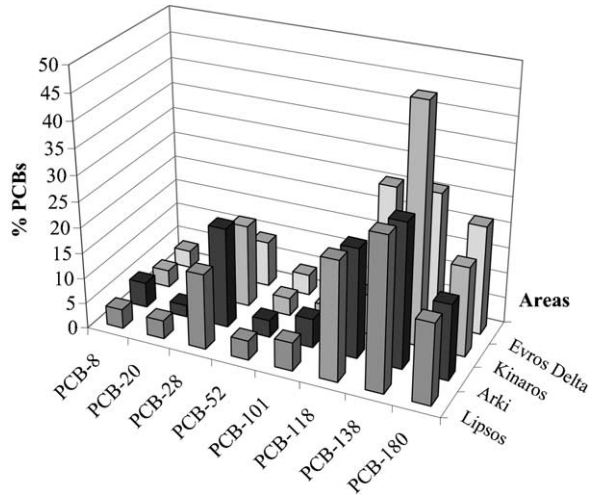

Fig. 2. Fingerprint of PCB congeners detected in yellow-legged gull eggs in the Aegean colonies studied.

Table 1
PCB and organochlorine concentrations (ppb wet weight) in yellow-legged gull eggs from Aegean colonies

	Lipsos ($N=11$)				Arki ($N=15$)				Kinaros ($N=12$)				Evros Delta ($N=7$)				K-Wallis	
	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	$\chi 2$	P
PCBs																		
PCB8	1.59	0.09	0.00	6.32	2.14	0.91	0.00	8.68	2.93	1.67	0.00	8.49	1.43	0.00	0.00	6.12	1.34	n. s.
PCB20	1.50	0.76	0.00	6.46	1.00	0.97	0.00	2.50	1.28	1.14	0.00	3.54	1.48	1.69	0.31	2.52	1.34	n. s.
PCB28	6.22	3.83	0.00	21.42	8.51	2.97	0.00	38.27	14.10	2.87	0.00	65.02	3.70	0.00	0.00	16.95	1.54	n. s.
PCB52	1.58	1.31	0.00	6.27	1.45	1.07	0.57	4.83	3.05	1.64	0.00	16.06	1.67	1.17	0.60	3.61	1.15	n. s.
PCB101	2.33	1.10	0.76	13.99	2.45	1.33	0.81	11.50	3.37	1.72	0.00	12.31	2.95	2.15	0.00	10.32	2.87	n. s.
PCB118	9.81	10.62	0.00	21.13	9.28	10.00	0.00	25.69	6.84	3.54	0.00	20.70	10.49	8.84	0.00	26.99	2.34	n. s.
PCB138	12.53	10.35	0.00	26.62	12.22	12.54	0.00	32.59	40.43	18.67	0.00	156.52	10.67	10.45	0.00	22.31	2.15	n. s.
PCB180	6.64	6.12	3.53	11.27	6.40	5.83	1.62	14.07	15.24	12.80	1.01	57.00	8.76	7.64	3.84	19.15	5.61	n. s.
∑PCBs	42.21	39.43	17.15	69.77	43.47	37.48	11.38	79.86	87.25	71.85	10.92	204.19	41.16	39.91	14.60	74.30	1.14	n. s.
Organochlorines																		
α-BHC	0.27	0.13	0.00	1.27	0.15	0.03	0.00	0.93	0.24	0.18	0.00	0.75	0.84	0.13	0.00	4.85	2.21	n. s.
β-BHC	124.12	38.60	0.00	786.38	49.30	41.53	22.43	112.58	72.16	60.78	18.75	186.34	62.09	57.50	26.41	98.06	3.02	n. s.
Lindane	8.58	0.09	0.00	92.07	3.89	0.67	0.00	26.19	2.58	0.06	0.00	17.54	1.52	0.78	0.00	6.77	4.47	n. s.
Aldrin	0.17	0.17	0.00	0.45	0.24	0.24	0.00	0.46	0.58	0.22	0.00	3.53	0.41	0.45	0.00	0.71	5.89	n. s.
Dieldrin	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	2.05	n. s.
Endrin	14.02	0.76	0.00	141.26	0.65	0.70	0.00	1.28	1.05	0.00	0.00	5.56	0.44	0.00	0.00	1.38	4.88	n. s.
Heptachlor	0.70	0.14	0.00	5.83	0.24	0.21	0.00	0.69	0.37	0.28	0.00	1.47	0.22	0.18	0.00	0.53	5.15	n. s.
Hept. Epoxide	5.62	3.91	0.00	27.55	3.21	2.62	1.22	7.67	5.31	3.56	0.16	18.21	3.27	2.91	1.10	7.94	0.64	n. s.
4,4′-DDE	1.19	0.72	0.00	7.22	0.90	0.82	0.00	3.06	1.24	0.60	0.00	3.86	1.04	0.97	0.10	2.11	2.48	n. s.
2,4′-DDD	206.06	172.34	37.18	564.86	235.45	267.08	32.28	555.66	162.21	164.39	13.32	287.62	211.58	144.99	63.69	417.40	1.15	n. s.
2,4′-DDT	0.91	0.34	0.00	6.52	0.49	0.59	0.00	1.05	0.35	0.00	0.00	1.39	0.38	0.00	0.00	1.39	2.15	n. s.
4,4′-DDD	0.15	0.00	0.00	1.65	0.00	0.00	0.00	0.00	0.22	0.00	0.00	2.60	0.30	0.00	0.00	1.55	4.82	n. s.
4,4′-DDT	1.81	0.00	0.00	5.59	2.65	2.85	0.00	5.91	2.85	1.31	0.00	10.36	1.69	0.00	0.00	6.06	1.96	n. s.
∑OCs/∑PCB	8.92	6.84	1.22	33.01	7.34	6.55	1.28	14.71	4.25	3.74	1.00	8.37	7.87	6.63	2.36	14.68	4.01	n. s.

Means are given for comparisons with the literature.

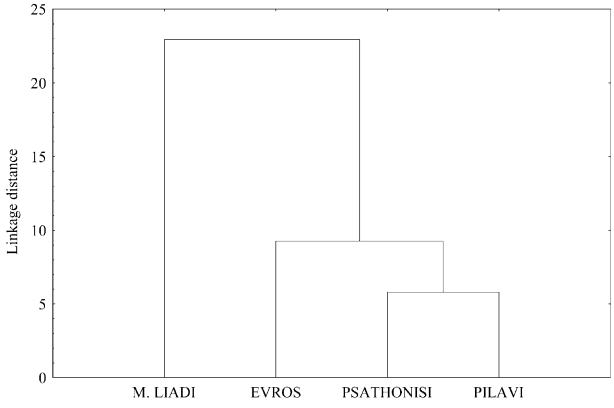

Fig. 3. Cluster indicating the relationship of areas studied in% levels of PCB congener in yellow-legged gull eggs. Euclidean distances as distance measure and single linkage as a linkage rule were used.

3.2. Organochlorine pesticides

In the study area as a whole, all 13 compounds analysed were detected in the egg samples (Table 1). Dieldrin was only found at Arki in very low concentrations. Aldrin was found in all areas but even its maximum concentration detected (3.53 ppb, Kinaros) was too low. Of drins, endrin was found at Lipsos in maximum concentrations by far greater than the other drins in the study area (141.26 ppb). As in the case of PCBs, the median concentrations of all compounds was not significantly different among the areas studied. The maximum concentrations of nine compounds were found at Lipsos, of only two at Kinaros and of one at Arki and Evros respectively. In all areas the relative proportions of β-BHC and 2,4′-DDD predominated over all other

Table 2
Spearman Rank Correlation Coefficients (lower part of tables) and statistically significant values (upper part of tables) of PCB congener concentrations in areas studied

Lipsos	PCB8	PCB20	PCB28	PCB52	PCB101	PCB118	PCB138	
PCB8								
PCB20	0.491				0.0331			
PCB28	0.257	0.101						
PCB52	−0.296	0.174	0.218					
PCB101	0.296	0.642	−0.318	0.400				
PCB118	0.506	0.106	0.118	−0.260	0.364		0.053	
PCB138	0.505	−0.055	0.218	−0.073	0.118	0.597		
PCB180	−0.029	0.257	0.018	0.018	0.373	0.579	0.309	
Arki	PCB8	PCB20	PCB28	PCB52	PCB101	PCB118	PCB138	PCB180
PCB8		0.003				0.001	0.002	
PCB20	0.703				0.040			
PCB28	0.404	0.465						
PCB52	0.073	0.063	0.314					
PCB101	0.295	0.534	0.154	−0.100				
PCB118	−0.748	−0.353	−0.295	−0.135	−0.342		0.000	0.001
PCB138	−0.726	−0.322	−0.353	−0.127	−0.335	0.948		0.000
PCB180	−0.487	−0.300	0.004	−0.032	−0.332	0.767	0.804	
Kinaros	PCB8	PCB20	PCB28	PCB52	PCB101	PCB118	PCB138	PCB180
PCB8						0.002		
PCB20	0.175				0.025		0.023	0.049
PCB28	−0.055	0.093						
PCB52	0.435	0.056	−0.203					
PCB101	0.377	0.641	−0.164	−0.105				
PCB118	−0.805	0.218	0.388	−0.284	−0.224			
PCB138	−0.074	0.645	−0.145	0.320	0.470	0.346		0.055
PCB180	−0.080	0.577	0.384	0.084	0.014	0.452	0.566	
Evros	PCB8	PCB20	PCB28	PCB52	PCB101	PCB118	PCB138	
PCB8								
PCB20	0.355							
PCB28	−0.685	−0.039						
PCB52	0.020	0.250	−0.020					
PCB101	0.236	0.250	−0.236	0.679				
PCB118	−0.143	0.371	0.511	−0.556	−0.185			
PCB138	−0.450	−0.037	0.695	−0.111	−0.667	0.192		
PCB180	0.197	0.321	0.059	0.429	−0.143	−0.296	0.593	

compounds providing characteristic and similar fingerprints (Fig. 4). Additionally, the relative proportions of heptachlor hepoxide were higher than those of heptachlor. In most areas 4,4′-DDT and 4,4′-DDE concentrations followed those of 2,4′-DDD (Table 1).

The cluster analysis separated two groups, one including Evros and Arki (but with a low linkage distance) and another with Kinaros and Lipsos (Fig. 5).

Of a total of 55 significant Spearman Rank Correlations found between compounds (Table 3), 40 (72.7%) were between DDT metabolites (DDTs) and other compounds. Specifically, 12 (21.8%) were among DDTs, 13 (23.6%) were between DDTs and drins, 11 (20.0%) between DDTs and heptachlor or heptachlor epoxide and 4 (7.3%) between DDTs and α-BHC. Of the remaining significant correlations, 5 (9.1%) were between heptachlor (or heptachlor epoxide) and drins, 4 (7.3%) between heptachlor (or heptachlor epoxide) and other compounds, 4 (7.3%) between BHCs and 4 (7.3%) other correlations. In total, heptachlor or heptachlor epoxide indicated 20 (36.4%) significant correlations with other compounds. In none of the areas the correlations between ΣPCBs and ΣOCs were significant.

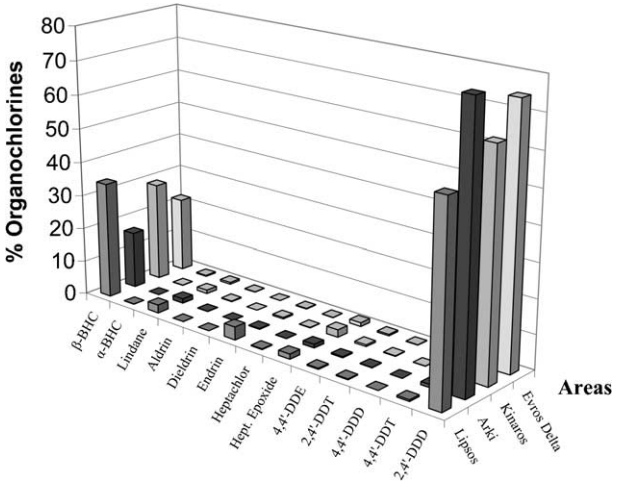

Fig. 4. Fingerprint of organochlorine pesticides detected in yellow-legged gull eggs in the Aegean colonies studied.

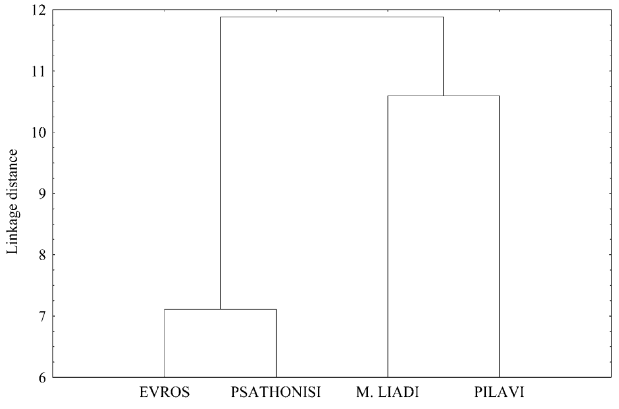

Fig. 5. Cluster indicating the relationship of areas studied in% levels of organochlorine pesticides in yellow-legged gull eggs. Euclidean distances as distance measure and single linkage as a linkage rule were used.

4. Discussion

Of PCBs detected in our samples, congeners 118, 138 and 180 were the most elevated in all colonies. These compounds, and especially the last two, were also found in high concentrations in yellow-legged gull eggs in other parts of the Mediterranean (Focardi et al., 1988a; Gonzalez et al., 1991; Pastor et al. 1995a); also in other waterbirds' eggs in the Mediterranean, with levels depending on the species (Focardi et al., 1988b; Konstantinou et al., 2000; Goutner et al., 2001). The higher concentrations of the above mentioned three congeners and also of PCB 28 seem to be due to both their persistency in the environment and bioaccumulative properties (Gagnon et al., 1990; Scrimshaw and Lester, 1993; Hong et al., 1998). Their bioaccumulative properties are due to their structure characterised by the presence of chlorine atoms in positions *ortho*, *meta* and/or *para* of at least one biphenyl ring (2,4,5 substitution pattern) (Gagnon et al., 1990; Metcalfe and Metcalfe, 1997).

We did not detect significant differences in none of the median congener concentrations. Whereas the overall pollution pattern by PCBs was probably expected to be similar between Lipsos and Arki due to the vicinity of the areas, similarities of this Dodecanese area with the Evros Delta are unreasonable. The nearest major pollution source for these two Dodecanese colonies is the Büyük (Great) Menderes river whose tributary lies about 30 km northeastern on the Turkish coast (Fig. 1). This river occupies a drainage area of 24.976 km^2, including tributaries of some other rivers and streams, many populated towns within its limits and is also polluted by an increasing number of geothermal energy production plants (Environmental Foundation of Turkey, 1995). Indications that Menderez River is a much probable source of pollution in the Dodecanese came from a recent study using the Audouin's gull as bioindicator and from a monitoring study of OC pesticide in Menderez river (Goutner et al., 2001; Turgut, 2003). The Evros Delta, crossing the border between Greece, Turkey and Bulgaria, receives considerable transboundary pollution. In a study based on cormorant eggs, the Evros Delta apparently was not the most polluted area (Konstantinou et al., 2000), but in another, based on the analysis of charadriiform bird eggs, the highest concentrations of most congeners were detected in the Evros Delta (Goutner et al., in press). These discrepancies are due to the different diets of the bird species, therefore reflect the choice of the bioindicator species rather than the pollution of the area per se. Organochlorine contaminants enter the egg through

Table 3
Spearman Rank Correlation Coefficients (lower part of tables) and statistically significant values (upper part of tables) of organochlorine pesticide concentrations in areas studied

Lipsos	α-BHC	β-BHC	Lindane	Heptachlor	Hept. Epoxide	Aldrin	Endrin	4,4′-DDE	2,4′-DDD	2,4′-DDT	4,4′-DDD	4,4′-DDT
α-BHC												
β-BHC	0.256											
Lindane	−0.093	−0.484			0.042							
Heptachlor	0.061	−0.376	−0.277			0.052	0.025		0.009			0.053
Hept. Epoxide	0.451	−0.318	0.619	−0.266						0.022		
Aldrin	0.112	0.438	0.117	−0.598	0.019		0.027					0.001
Endrin	0.031	0.237	0.362	−0.667	0.502	0.661			0.002			0.001
4,4′-DDE	−0.085	−0.532	0.559	−0.009	0.395	−0.080	0.160					
2,4′-DDD	−0.307	0.255	0.191	−0.743	0.164	0.569	0.819	−0.110				0.012
2,4′-DDT	0.221	−0.092	0.545	−0.102	0.679	0.184	0.568	0.111	0.248			
4,4′-DDD	−0.358	0.500	−0.358	−0.404	−0.500	0.513	0.307	−0.404	0.500	−0.404		
4,4′-DDT	0.020	0.059	0.360	−0.595	0.382	0.842	0.857	0.100	0.724	0.475	0.327	

Arki	α−BHC	β−BHC	Lindane	Heptachlor	Hept. Epoxide	Aldrin	Dieldrin	Endrin	4,4′−DDE	2,4′−DDD	2,4′−DDT	4,4′−DDT
α-BHC					0.003			0.003	0.000	0.019	0.001	0.000
β-BHC			0.004									
Lindane	−0.228	0.693		0.002								
Heptachlor	−0.413	0.296	0.741									
Hept. Epoxide	0.716	−0.054	−0.318	−0.631					0.004		0.006	0.000
Aldrin	0.273	0.340	0.443	0.361	0.059					0.003		
Dieldrin	0.128	−0.062	−0.062	−0.125	−0.124	0.155						
Endrin	0.712	0.284	−0.147	−0.433	0.495	0.186	−0.063			0.016	0.003	0.012
4,4′-DDE	0.789	0.057	−0.091	−0.353	0.700	0.361	0.124	0.382		0.054	0.008	0.000
2,4′-DDD	0.596	0.293	0.120	−0.094	0.493	0.717	0.186	0.607	0.507		0.002	0.000
2,4′-DDT	0.761	0.306	0.015	−0.303	0.673	0.368	0.063	0.707	0.658	0.720		0.000
4,4′-DDT	0.892	0.113	−0.184	−0.442	0.829	0.403	0.126	0.630	0.822	0.771	0.844	

Kinaros	α−BHC	β−BHC	Lindane	Heptachlor	Hept. Epoxide	Aldrin	Endrin	4,4′−DDE	2,4′−DDD	2,4′−DDT	4,4′−DDD	4,4′−DDT
α-BHC		0.033				0.026						
β-BHC	0.616											
Lindane	−0.397	−0.388							0.037			
Heptachlor	−0.196	0.042	−0.254				0.008			0.011		
Hept. Epoxide	−0.132	−0.154	0.075	−0.242			0.010			0.028		0.003
Aldrin	0.638	0.061	−0.245	−0.378	0.452							0.016
Endrin	0.056	−0.109	0.312	−0.723	0.710	0.373			0.000	0.000		0.013
4,4′-DDE	0.305	0.289	−0.026	0.332	−0.028	0.133	−0.149					
2,4′-DDD	−0.178	−0.084	0.605	−0.543	0.538	−0.007	0.873	−0.063		0.000		
2,4′-DDT	0.071	−0.047	0.345	−0.699	0.632	0.357	0.965	−0.086	0.889			0.016
4,4′-DDD	−0.356	−0.218	0.420	0.044	−0.306	−0.356	−0.243	−0.396	−0.044	−0.243		
4,4′-DDT	0.321	−0.011	0.112	−0.280	0.773	0.674	0.691	0.132	0.470	0.674	−0.280	

Evros Delta	α−BHC	β−BHC	Lindane	Heptachlor	Hept. Epoxide	Aldrin	Endrin	4,4′−DDE	2,4′−DDD	2,4′−DDT	4,4′−DDD	4,4′−DDT
α-BHC												
β-BHC	0.074				0.023							
Lindane	0.561	−0.559										
Heptachlor	−0.561	0.090	−0.136				0.033			0.033		0.010
Hept. Epoxide	0.408	0.821	−0.144	0.090								
Aldrin	−0.111	0.214	0.252	0.685	0.214							
Endrin	0.532	0.256	−0.040	−0.795	0.039	−0.335			0.005			0.022
4,4′-DDE	0.334	0.643	−0.126	−0.360	0.464	0.250	0.591					
2,4′-DDD	0.704	0.393	0.018	−0.595	0.286	−0.143	0.906	0.571		0.005		0.053
2,4′-DDT	0.532	0.256	−0.040	−0.795	0.039	−0.335	1.000	0.591	0.906			0.022
4,4′-DDD	−0.370	0.134	−0.584	−0.045	−0.134	−0.535	0.147	−0.401	0.045	0.147		
4,4′-DDT	0.613	0.020	0.040	−0.875	−0.039	−0.493	0.826	0.512	0.749	0.826	−0.147	

contaminated food and their composition in the egg reflects the trophic level and dietary changes in birds (Fasola et al., 1987; Weseloh et al., 1995; Hebert et al., 1997). Dietary data on the yellow-legged gull are not available but numerous observations of birds following fishing boats in the Dodecanese suggests that a considerable part of their diet constitute of fish. At least during the egg laying, big numbers of olive stones were found around nests in the Dodecanese colonies indicating that a part of the diet constituted of terrestrial food. It seems unreasonable that the Dodecanese yellow-legged gull have had similar habits to those in the Evros Delta. Similarities in pollution PCB patterns rather reflect extensive scavenging habits. Among several Mediterannean water-birds, the fish eating birds used to exhibit the highest amounts of PCBs (Focardi et al., 1988a) because an additional trophic level transfer increases the potential of bioaccumulation. Accordingly, levels of most PCBs in the sympatric Audouin's gull in the Dodecanese were considerably higher than in Yellow-legged Gull. This is in agreement with the results of Pastor et al. (1995a), showing that differences in concentrations between specialized feeders (i.e. Audouini Gull) and scavenging species (i.e. Yellow-legged Gull) can be enhanced by the fact that detoxifying systems seems to be deficiently developed in the first ones (Walker, 1990; Fossi et al., 1995). The most polluted colony of yellow-legged gulls in this study appeared to be at Kinaros probably affected by tanker-washouts being situated in the vicinity of international ship traffics.

Even the maximum total PCB levels (204 ppb) found at Kinaros, were too low. Levels detected in other parts of the Mediterranean were extremely higher (wet weight unless specified). In Capraia island, Italy, mean levels were 2.940 ppb (dry weight, Focardi et al., 1980) whereas in the Rhone Delta ranged from 16.000 to 160.000 ppb (Lambertini and Leonzio, 1986). In the seventies, in the Balearic islands, Spain, mean levels were 16.750 ppb whereas in Chafarinas islands ranged from 3.100 to 4.200 ppb (Lambertini and Leonzio, 1986) dropping to 2.450 ppb in the early nineties (Gonzalez et al., 1991). In the Ebro Delta mean levels in 1992 were 897 ppb (dry weight, Pastor et al., 1995a, b). In the seventies, in Crete mean levels were 5.320 ppb and in Cyprus ranged from 1.200 to 4.200 ppb (Lambertini and Leonzio, 1986) indicating lower levels in the Eastern Mediterranean. In the herring gull, levels that can affect egg hatchability vary among colonies. In the Lake Erie, Canada, levels ranging from 35 to 140 ppm seemed to had normal hatching success (Weseloh et al., 1990) though in the same region earlier studies suggested that adverse effects in some colonies were associated with levels above 70 ppm (Gilman et al., 1977; Weseloh et al., 1979). Therefore the levels detected in our study are too low to have any biological consequences to the breeding success of the yellow-legged gulls in the Aegean.

Regarding OCs, DDE is an important compound due to the well established inverse relationship between DDE content and eggshell thickness and consequences on the breeding productivity of waterbird populations (Gilbertson, 1974; Blus, 1984; Custer et al., 1999). But in comparison to other waterbirds where the critical threshold for eggshell thinning is between 4 ppm and 10 ppm (Custer et al., 1983; Blus 1984; Leonzio et al., 1989), these effects are minimal in the herring gull (Weseloh et al., 1979) thus cracking and flacking of eggshells has been observed at DDE concentrations of 100–200 ppm in Lake Michigan colonies (Gilman et al., 1977). Mean levels ranging from 2.8 to 9.4 ppm, have not seem to actually affect the breeding productivity and eggshell thickness of the herring gull in the Great Lakes (Weseloh et al., 1990). Thus, levels found in the eggs in the Aegean are too low to affect eggshell thickness.

DDT metabolites have been found in previous studies in the areas studied (Konstantinou et al., 2000; Goutner et al., 2001, in press) and may also originate both from local sources but also in part from the wintering or migration grounds. The elevated amounts of 2,4′-DDD in comparison to 4,4′-DDE is probably owed to its presence in zooplankton and in water column. In zooplankton the concentrations of 2,4′-DDD have been found to be comparable with the concentrations of 4,4′-DDE. Zooplankton possibly acquires DDTs from sediments or from the water column where the DDD form constitutes the major fraction of DDTs (Stranberg et al., 1998). Elevated 2,4′-DDD levels may also indicate that this compound was a major constituent in a technical mixture used in the region. Furthermore, higher concentrations of DDD were also found among DDTs in water samples of Menderes river (Turkey) the tributaries of which lie close to the Dodecanese colonies of our study (Turgut, 2003). The characteristic similarity of fingerprints for organochlorines in all areas and species was also found in our above mentioned studies in Greece on other waterbirds. These suggest a particular pollution pattern with organochlorine pesticides in Greece.

In previous studies, drins were found in avocet eggs in the Evros Delta, in eggs of common terns (*Sterna hirundo*), little terns (*Sterna albifrons*) and squacco herons (*Ardeola ralloides*) in the Axios Delta, northeastern Greece (Albanis et al., 1996; Goutner et al., 1997, in press) and also in the Audouin's gull (Goutner et al., 2001) being sympatric to the yellow-legged gull in the study area. We suggested that the occurrence of drins in these birds' eggs, being all distant migrants, was due to accumulation during migration to African wintering quarters. There is no information of the movements of the yellow-legged gulls breeding in our study area and the occurrence of drins may be due to scavenging habits rather than long migration.

β-BHC and lindane had predominent concentrations in our egg samples. The dominance of β-BHC in the

samples from all areas seem to be due to its high stability against metabolism pointing to chronic rather than acute contamination (Oxynos et al., 1993). Lindane is not harmful to birds and the toxic heptachlor and heptachlor epoxide being lethal for birds in concentrations about 9 ppm (Blus et al., 1985) have been found in our samples in concentrations too low to have adverse effects on the yellow-legged gull. The presence of benzenehexachloride derivatives (BHC), drins, heptachlor and heptachlor epoxide were also detected in water samples from Menderes river. Similar patterns i.e. heptachlor epoxide > heptachlor and DDD > other DDTs were also found (Turgut, 2003).

Above all, the similarities of OC fingerprints with 2,4′-DDD and β-BHC dominating both in this study and the previous mentioned above carried out in a variety of species and areas, suggest a particular organochlorine contamination pattern in the North Eastern Mediterranean.

The cluster analysis associated areas being theoretically unrelated in their OC pollution patterns (Evros Delta to Arki and Kinaros to Lipsos). Colonies situated at closer distances should have greater similarities when affected by a nearby pollution source, as a previous study in the Audouin's gull suggested (Goutner et al., 2001). These mean that the yellow-legged gull eggs do not seem to reflect the pollution around the colonies sampled.

The tendency of the higher PCB congeners to significantly correlate with other congeners has been documented in other studies too (Konstantinou et al., 2000; Goutner et al., 2001, in press). Organochlorines, especially DDT metabolites have also been found to make significantly correlations of the type described in this paper (the above mentioned studies, also Weseloh et al., 1990). Nevertheless, in contrast to the findings of many studies (Blus et al., 1974; Gilbertson, 1974; Gabrielsen et al., 1995; Custer et al., 1999) there was none significant correlation between the total concentrations of both groups in the eggs of the yellow-legged gull. This probably means that the source and pathways of these two groups of pollutants in the food chain of the yellow-legged gull are different. The median ratio of ΣOCs/ΣPCBs was > 1 in all colonies showing that the agrochemical dominates over the industrial pollution in this part of the region, in agreement with previous studies (Focardi et al., 1988; Crivelli et al., 1989; Konstantinou et al., 2000; Goutner et al., 2001). The insignificant differences in the median ratio of ΣOCs/ΣPCBs seem to suggest a rather uniform type of pollution in the northeastern Aegean depending on the incidence of pollutants in this region. This was also concluded by a relative study on the Audouin's gull in the Aegean (Goutner et al., 2001) but this seem to be rather unreasonable because pollution is not so predictable, simply its variability does not seem to be reflected in the eggs of these gulls.

5. Conclusions

The use of the yellow-legged gull eggs for the detection of pollution by polychlorinated biphenyls and organochlorine pesticides in the Aegean Sea revealed, in conjuction with some recent studies, a particular pollution pattern in this part of the region and, verified that some of these substances especially those with high stability and bioaccumulative properties still occur in the food chains. Nevertheless, the concentrations of these chlororganics are probably the lowest detected in the world and it seems improbable to have any adverse effect in the biology of the yellow-legged gull and associated biota. Nevertheless, this study failed to detect regional differences in the pollution patterns probably because the pollution in the aquatic environment is not reflected in the egg of the yellow-legged gull rather due to its highly scavenging habits. This is an inconclusive decision due to the small number of colonies sampled but based on the present study, the yellow-legged gull seems to be an unsuitable biomonitor for these chlororganics in the northeastern Mediterranean. Species such as Larus Audouini and Cormorants exhibited higher concentrations of organochlorines and PCBs and thus could be classified as better biomonitors for the region under study.

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