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Organochlorine Chemical Residues in Herring Gulls, Ring-billed Gulls, and Common Terns of Western Lake Superior

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Abstract. Residues of polychlorinated biphenyls (PCBs), DDE, DDT, and hexachlorobenzene (HCB) were analyzed for three age classes (e.g., pre-fledge muscle and blood, and post-fledge muscle) of the herring gull, ring-billed gull, and common tern for samples collected in the western end of Lake Superior in 1977. Concentrations of PCBs were highest and HCB concentrations were lowest of the chemicals analyzed in all four sample types and for all three species. Without exception, concentrations of all residues increased from the pre-fledge to the post-fledge stage despite this being a period of dilution. Residue concentrations of PCBs, DDE, and DDT in the eggs were higher ($P < .01$) for the herring gull than for the ring-billed gull and common tern, but concentrations in the latter two species were similar. All residue concentrations in the pre-fledge samples of the common tern and herring gull were similar ($P > 0.05$); both species had higher concentrations of PCBs and DDT than the ring-billed gull. The herring gull had higher residue concentrations of HCB and DDE than the ring-billed gull, but the common tern did not differ from the ring-billed gull for HCB or DDE. Residue concentrations among the three species were similar in the post-fledge samples, however, there was an increasing pattern of residue concentrations in the common tern relative to the gull species. Principal component analysis revealed that residue concentrations of PCBs, DDE, and DDT were highly correlated ($r > .76$) in all three sample types and the first principal component could express a high proportion of the variation (>65%) in residue concentrations. In contrast, HCB residues were more independent and correlated ($r > .53$) with the second principal component. There was little correspondence between blood residue samples and the pre-fledge muscle samples. Residue concentration differences among the species are probably related to different life histories. The concentrations are below those that affect reproduction in the gull species, but potential impairment cannot be excluded for the common tern.

Although several of the U.S. Great Lakes have moderate to serious water quality problems, the waters of Lake Superior have generally remained high in quality (Veith *et al.* 1977). This is due to several factors such as the small watershed area relative to the lake size and the undeveloped shoreline (Veith *et al.* 1977). However, Lake Superior is contaminated in localized areas, particularly in nearshore areas and in major harbors (Glass and Poldoski 1974). One of the most troublesome areas has been the Duluth-Superior Harbor which lies at the extreme western end of Lake Superior. This harbor has a history of water quality problems due to long-term inputs of inadequately treated municipal and industrial effluents. High phosphorous loading and sediments with unacceptable levels of nitrogen, oil and grease, biochemical oxygen demand, metals (*e.g.*, zinc and mercury), and select organochlorine chemical compounds have been found throughout the harbor (US EPA 1977).

Because birds are terminal consumers in aquatic communities, they are ideal subjects for examinations on the bioaccumulation of residues, and in many cases the residue concentrations have been linked to impaired reproductive success (Blus *et al.* 1972a, 1972b; Cooke 1973; Stickel 1973; Ohlendorf *et al.* 1979). Several broad-scale studies on pollutant levels have been published (Hickey and Anderson 1968; Vermeer and Reynolds 1970, Ohlendorf *et al.* 1979), but we are aware of only two studies that include data from Lake Superior (Ryder 1974; Gilman *et al.* 1977). In this report, information is presented on the levels of organochlorine chemical compounds found in the tissues of three colonial birds that nest in the Duluth-Superior Harbor and adjacent to Lake Superior. The objectives were to (1) determine the levels of selected organochlorine compounds present in eggs and young-of-the-year gulls and terns, (2) determine if there are similar organochlorine residues among larid species that commonly occur in the study area, and (3) examine whether blood residues were correlated with those in pectoral muscle.

Methods

Study Area

Tissue and blood samples of the ring-billed gull and common tern were collected from the Duluth-Superior Harbor which is located at the extreme western end of Lake Superior. Herring gull samples were collected from Lake Superior's Knife Island which lies 37 km northeast of Duluth.

Sample Collection

Samples were collected at three different stages during the 1977 nesting season for each species: egg, pre-fledge, and post-fledge. Single eggs were taken at the peak of incubation from three-egg clutches. The eggs were immediately placed in a cooler and transported to the U.S. Environmental Protection Agency's Environmental Research Laboratory in Duluth, Minnesota. Their weight, length, and breadth were determined and then placed in a freezer until chemical analyses.

Two types of samples were taken from each pre-fledge chick: (1) pectoral muscle, and (2) blood drawn from the alar vein of the live bird (Dieter *et al.* 1976). The muscle samples were individually wrapped in acetone-rinsed aluminum foil and frozen in a cooler of dry ice. As with the eggs, these samples were immediately transported to the laboratory and placed in a freezer until chemical analyses. Blood samples were drawn from the alar vein of the live bird using the procedures outlined by Dieter *et al.* (1976). Blood samples were refrigerated upon transferring to the laboratory and were processed within two days of collection. After the pre-fledge samples were collected, as many of the remaining chicks as feasible were banded with standard U.S. Fish and Wildlife Service leg bands. This allowed positive field identification of these birds and eventual collection of post-fledge samples later in the season. Post-fledge samples consisted of pectoral muscles taken from birds shot in the vicinity of the colonies in late summer.

Sample Preparation and Analysis

Polychlorobiphenyls (PCBs), DDE, DDT, and HCB were measured, because previous exploration studies using mass spectrometry have shown that these chemicals are the primary contaminants in western Lake Superior (Veith *et al.* 1977; 1979). The analytical procedures have been reported previously (Veith 1975; Veith *et al.* 1979). Briefly, samples were homogenized with anhydrous sodium sulfate and extracted in a Soxhlet extractor with a 1:1 mixture of hexane and methylene chloride. After lipid determination, Florisil® was used for sample cleanup and separation of DDT from the other chemicals.

The fractions were analyzed by a Hewlett-Packard Model 5700A gas chromatograph equipped with a Model 3352B Lab Data System and a linearized argon-methane detector. The analytical column was 2 m × 2 mm T.D. packed with SE-20/OV-210 on Gas Chrom Q (66%). The oven temperature was programmed from 160° to 220°C. at 2° per minute. The recovery of all chemicals from spiked samples was greater than 92% with a relative standard deviation ($N = 12$) of 6% for HCB and DDE and 13% for PCB and DDT. The detection limits were 0.01 ppm for DDE and DDT, 0.1 ppm for PCBs, and 0.005 for HCB.

Statistical Analysis

Prior to statistical analyses, the data were examined for the assumptions of normality and for homogeneity of variances; using logarithmic transformations when suitable. We tested for differences in residue concentrations (fresh weight in eggs and both fresh and lipid-basis in others) between species for each sample type using (1) one way analysis of variance, or (2) Kruskal-Wallis tests when sample sizes for any group were <5 or when the assumptions were not satisfied (Sokal and Rohlf 1981). Principal component analysis (PCA) was used to express the covariation in residue levels of PCBs, DDT, DDE, and HCB among the three species. Briefly, principal component analysis is a multivariate statistical technique that is useful for reducing the dimensionality of data. Here there are n individuals of 3 species and 4 organochlorine residue concentrations, an $n \times 4$ data matrix. It is impossible to illustrate the relationships of the residue concentrations for the n individuals in 4-dimensional space, but principal component analysis calculates a reduced number of variables (principal components) that explains a high proportion of the variation in the original variables. Principal component (PC 1) corresponds to the axis that explains the highest proportion of the variance in the original data. Principal component 2 (PC 2) and subsequent principal components explain proportionately less percentages of the variation with the constraint that each pair of principal components are uncorrelated ($r = 0$) (Tatsuoka 1971). We primarily used PCA to graphically display the covariation among the chemicals and the differences in concentrations between individuals of the three species.

Results

Organochlorine Chemical Residue Levels

Of the four chemicals analyzed, PCB concentrations were highest and HCB concentrations lowest in the

Table 1. Mean and range (in parentheses) of concentrations for PCBs, HCB, DDE, and DDT in lipid basis and fresh weight for A) egg, B) pre-fledge, C) post-fledge muscle, and D) pre-fledge blood in the herring gull, the ring-billed gull, and the common tern. ND represents not detectable

Sample type and species	Sample size	Lipid[a] content %	Lipid basis (ppm)				Fresh weight (ppm)			
			PCBs	HCB	DDE	DDT	PCBs	HCB	DDE	DDT
A. **Egg**										
Herring Gull	10	—					11.3	0.05	7.6	2.8
							(3.9–17.6)	(.03–.07)	(3.4–12.4)	(1.2–4.9)
Ring-billed Gull	10	—					5.5	0.03	2.1	1.4
							(ND–17.4)	(.01–.09)	(0.9–3.2)	(0.2–3.5)
Common Tern	10	—					5.2	0.06	2.5	0.9
							(2.5–8.2)	(.02–.37)	(0.8–6.8)	(0.6–1.6)
B. **Pre-fledge Muscle**										
Herring Gull	10	5.5	34	0.16	7.2	3.5	1.9	.01	0.4	0.2
		(4.6–7.2)	(4–68)	(ND–.32)	(1.0–18.7)	(ND–9.8)	(0.2–3.3)	(ND–.02)	(0.1–1.1)	(ND–0.6)
Ring-billed Gull	10	5.7	11	0.08	2.2	1.4	0.6	0.00	0.1	0.1
		(4.5–9.0)	(7–15)	(ND–.13)	(ND–3.0)	(ND–2.0)	(0.4–1.0)	(ND–.01)	(ND–0.2)	(ND–0.1)
Common Tern	5	8.9	30	0.09	3.6	3.9	2.5	0.01	0.3	0.3
		(6.8–11.1)	(11–47)	(.08–.12)	(1.7–5.2)	(1.5–6.4)	(1.3–3.4)	(.01–.01)	(0.2–0.4)	(0.2–0.4)
C. **Post-fledge Muscle**										
Herring Gull	4	5.8	62	0.21	17.0	8.1	3.1	0.01	0.8	0.4
		(4.2–7.1)	(8–167)	(ND–.58)	(2.2–49.4)	(1.2–22.7)	(0.5–6.9)	(ND–.02)	(0.1–2.1)	(0.1–0.9)
Ring-billed Gull	2	6.6	46	0.23	5.2	3.4	3.0	0.02	0.3	0.2
		(6.3–6.9)	(43–49)	(.21–.25)	(4.3–6.1)	(3.2–3.7)	(2.7–3.4)	(.02–.02)	(0.3–0.4)	(0.2–0.2)
Common Tern	7	8.1	63	0.17	6.3	6.5	5.1	0.01	0.5	0.5
		(7.0–9.9)	(51–86)	(ND–.25)	(5.1–7.7)	(5.5–8.8)	(4.2–7.5)	(ND–.02)	(0.4–0.7)	(0.4–0.8)
D. **Pre-fledge Blood**										
Herring Gull	6	0.8	52	1.90	8.1	5.8	0.4	0.01	0.06	0.04
		(0.6–1.2)	(16–154)	(.74–2.80)	(ND–28.7)	(ND–20.8)	(0.2–1.1)	(.01–.02)	(ND–.20)	(ND–.15)
Ring-billed Gull	10	0.8	18[b]	0.35	ND	1.7	0.1	0.00	ND	0.01
		(0.5–1.2)	(ND–36)	(ND–1.72)	(ND–ND)	(ND–2.2)	(ND–0.2)	(ND–.01)	(ND–ND)	(ND–.02)
Common Tern	5	1.7	14	0.20	1.1	1.5	0.2	0.00	0.02	0.03
		(1.2–2.8)	(11–19)	(ND–.29)	(0.8–1.6)	(1.1–2.4)	(0.2–0.3)	(ND–.01)	(.02–.02)	(.02–.03)

[a] Lipid content was not measured in egg samples
[b] One missing value for ring-billed gull PCBs, based on n = 9

four sample types and for all three species (Table 1). DDE residues were higher than DDT residue concentrations except in the pre-fledge muscle samples for the common tern and the pre-fledge blood samples for the ring-billed gull and common tern. All residue concentrations for all three species increased from the pre-fledge to the post-fledge stage even though this is a period of dilution due to growth. One high value was observed for the HCB concentration in an egg sample of the common tern. The HCB concentration in this sample was six times greater than any other HCB concentration value (0.37 ppm vs the next highest value of 0.06 ppm). A statistical test for outliers (Sokal and Rohlf 1981) indicated that this value was an outlier (Dixon's test, $p < 0.001$). Therefore, subsequent analyses were performed with and without this value.

Differences between species for egg and pre-fledge muscle samples were tested by oneway analysis of variance, and Duncan's Multiple Range Test was used to identify which species were different. Kruskal-Wallis tests were used for the post-fledge samples. The results of the pre-fledge blood samples are not presented because of inconsistent results with the pre-fledge muscle samples. There were similar results using fresh weight or lipid-basis data and using the original or logarithmic-transformed data for the muscle samples. The results are presented for logarithmic-transformed lipid-basis values in pre- and post-fledge muscle samples.

In the egg stage, concentrations of PCBs, DDE, and DDT were higher for herring gulls than for ring-billed gulls and common terns ($P < 0.01$), but ring-billed gulls and common terns had similar concentrations for all three residues. There were no differences between the three species in HCB egg residues with or without the common tern HCB outlier in the analysis. Concentrations of PCBs and DDT in prefledge muscle samples were similar in the common tern and herring gull and both were higher ($P < 0.1$) than in the ring-billed gull. Concentrations of HCB and DDE in pre-fledge muscle samples were higher ($P < 0.05$) in the herring gull than in the ring-billed gull, but concentrations in the common tern were not different from either the herring or ring-billed gull. No significant differences in residue levels in the post-fledge samples were found between any of the three species. However, there was a distinct pattern of increasing residue concentrations of PCBs, DDE, and DDT in the common tern relative to the gull species from the egg to the post-fledge stage.

Covariation Among Residues

The distribution of residue concentrations for each stage are shown for the results of the PCA in Figure 1. Generally, PCBs, DDE, and DDT concentrations were positively correlated at each stage ($.77 < r < .93$), but HCB concentrations were more independent from PCB, DDE, and DDT concentrations ($.03 < r < .76$). The interpretations of the PCA for each stage is similar. Principal component 1 (PC 1 in Figure 1) was positively correlated with PCB, DDE, and DDT concentrations (all $r > .89$), while PC 2 was positively correlated with HCB concentrations (all $r > .52$). Because of these patterns of covariation among the residue concentrations, PC 1 explained a high proportion of the variance (65 to 83%) in all three stages. PC 1 and PC 2 together accounted for >91% of the variation in all three stages.

Figure 1 also expresses the relative distribution of residues in individual birds for the three species for each sample type. In general, the herring gull showed the most variation in residue concentrations in all three sample types, while the residues present in the ring-billed gull were relatively consistent in the pre-fledge stage. Again, this illustrates that the common tern showed a distinct pattern of increasing residue concentration from the egg stage to the post-fledge stage.

Blood and pre-fledge Muscle Residues

There were no consistent patterns of association between residue concentrations in the pre-fledge

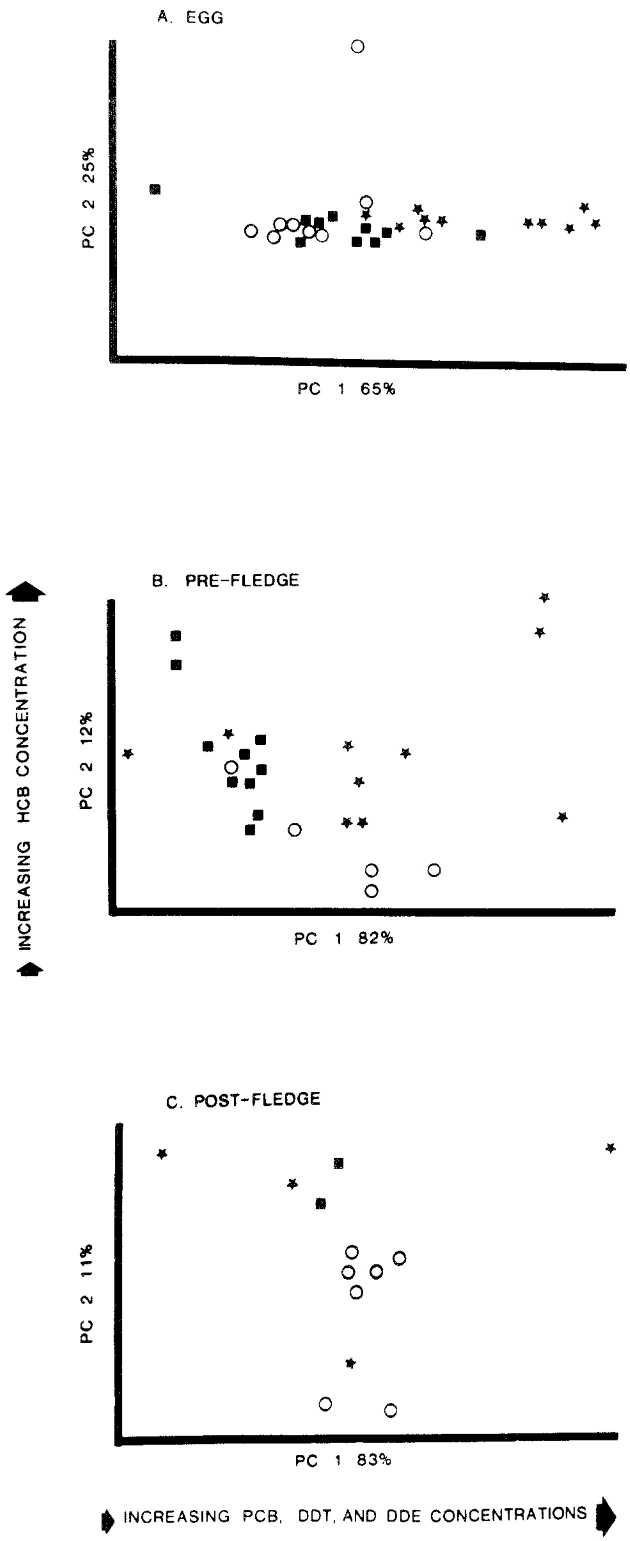

Fig. 1. Distribution of samples for larid species, herring gull (★), ring-billed gull (■), and common tern (○), according to principal component 1 (PC 1) and principal component 2 (PC 2) from principal components analysis of four organochlorine chemical residue concentrations (PCBs, DDE, DDT, and HCB) during each of three life history stages. The egg stage is based on fresh weight values and the pre and-post-fledge stages on lipid base values

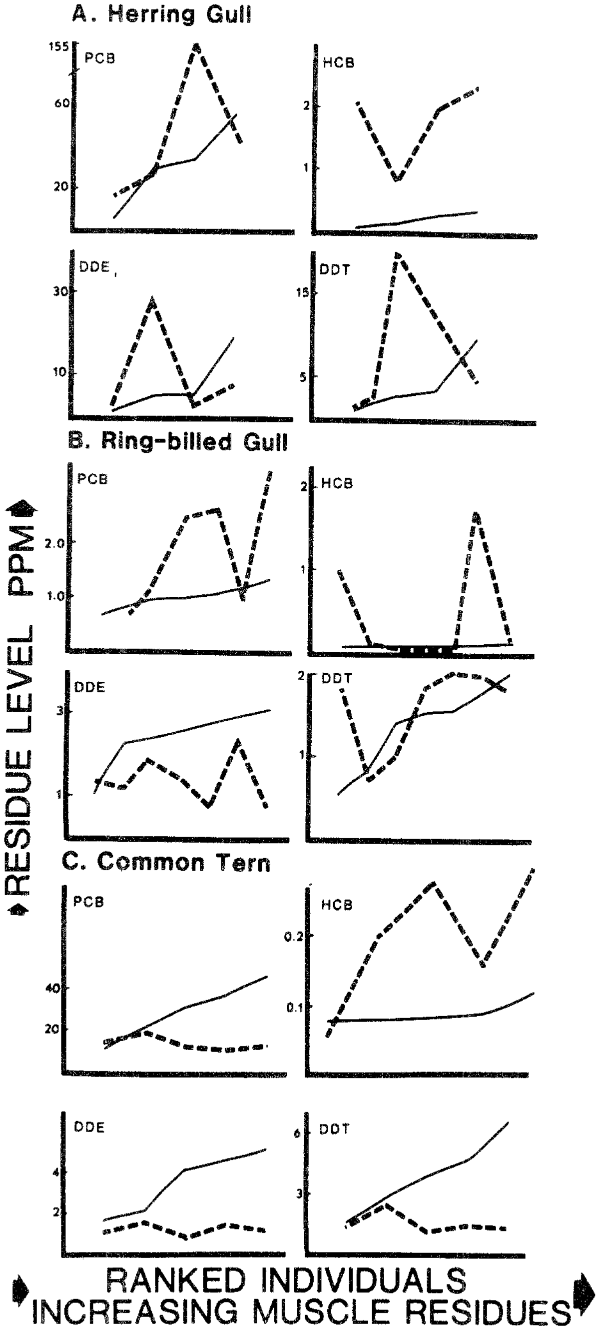

Fig. 2. Relationship between concentrations (lipid basis) in muscle residue (solid line) and blood (dashed line) for four organochlorine compounds in pre-fledgling young of three species of larids. Individual samples of pre-fledge muscle were ranked in order of increasing residue concentrations

muscle and the blood for any combination of the four residues and three species (Figure 2). In each case, residue concentrations in the blood were not similar to those in the muscle even though they were taken from the same individual. There were also no consistent patterns in the differences between species in comparisons of pre-fledge muscle and blood data.

Discussion

Comparisons With Previous Studies

The PCB residue concentrations in eggs of all three species are 2 to a factor of 10 less than values reported in earlier years for herring gulls in the Great Lakes (Gilman *et al.* 1977) and for herring and ring-billed gulls in the northern part of Lake Superior (Ryder 1974). In contrast, the PCB and DDE concentrations in eggs are about two times greater than for the arctic tern (*Sterna paradisaea*) nesting in sparsely-populated areas such as southwestern and northern Finland (Lemmetyinen *et al.* 1977; Lemmetyinen and Rantamaki 1980). Comparisons with common tern eggs collected on the East Coast of the United States in 1980 indicate the PCB concentrations are similar, but DDE concentrations were generally higher (Custer *et al.* 1983).

In comparison, DDT residue concentrations in the herring gull eggs were more than 20 times greater than those found by Gilman *et al.* (1977) in all the Great Lakes for data gathered in 1974 and 1975. These concentrations are still more than a factor of 5 less than the concentrations observed by Keith (1966) in 1964 and 1965 for a herring gull colony that was contaminated with DDT.

Few comparative data are available for DDT in the common tern and ring-billed gull. The concentrations of PCBs and DDE observed here are substantially lower than those observed for the common tern in the lower Great Lakes in 1971 (Frank *et al.* 1975) and in 1972 (Morris *et al.* 1976). In contrast, these concentrations are higher than those reported in eggs in the 1970's for the Arctic tern in Finland (Lemmetyinen *et al.* 1977; Lemmetyinen and Rantamaki 1980) and for the double-crested cormorant (*Phalacrocorax auritus*) or white pelican (*Pelecanus erythrorhynchos*) in the prairie provinces of the United States (Anderson *et al.* 1969).

Data are even more limited for comparisons of HCB residues, but the concentrations in herring gull eggs are similar to those reported for herring gull eggs collected in the mid-1970's from Lake Michigan and about one-half those reported from the northern and eastern portions of Lake Superior (Gilman *et al.* 1977).

Generally, lower residue levels were observed in the ring-billed gull in comparison with the herring gull for all sample types. This was also observed by Ryder (1974) in egg samples for populations of these species in northern, Lake Superior. Although generally higher concentrations of all residues were observed in the eggs of the herring gull in this study, Frank *et al.* (1975) observed higher concentrations of DDT and lower concentrations of PCB in the eggs of common terns as compared with the herring gull. Few additional data are available for comparisons in this regard or for comparisons with the later life history stages.

Residue Concentrations and Reproduction

Various factors have been associated with population changes in these species and reductions in reproductive success due to organochlorine chemical contamination is one possibility. Field and experimental evidence suggests DDE and other compounds or metabolites of DDT are related to eggshell thinning in several species of North America and England (Blus *et al.* 1972a, 1972b; Ohlendorf *et al.* 1979). The herring gull has been relatively insensitive to DDE-induced egg shell thinning (Hickey and Anderson 1968; Gilman *et al.* 1977), and the PCB and DDE concentrations observed here are much lower than other studies that concluded there were no deleterious effects of these residues on reproductive performance in herring or ring-billed gulls (Vermeer and Reynolds 1970; Ryder 1974).

In common terns, mean DDE concentrations of 6.67 and 7.72 ppm wet weight were associated with abnormal eggs (Fox 1976). The DDE concentrations of some common tern eggs fall within this range of values. In comparison, Switzer *et al.* (1971, 1973) found no correlation between egg shell thinning and DDE residues in common terns for a similar range of concentrations. They suggest that nest predation and nest abandonment as well as other factors were probably responsible for lower reproductive success in this species. However, they did not eliminate the possibility of other interactions between DDE and the physiology of reproduction. Common terns may be more sensitive to chemical contamination than the gull species and the residue levels cannot be eliminated as a potential contributing factor to the reproductive success of this species in the Duluth-Superior Harbor.

Residues and Species' Life History

The observed differences in residue levels between species are probably a function of differences in each species life history. The common tern primarily feeds on small fish, crustaceans, and aquatic invertebrates, while the gull species are more omnivorous and feed on fish, insects, worms, and a variety of plant and animal remains (Martin *et al.* 1951). The common tern migrates the longest distance, because it breeds in the northern hemisphere and winters in the southern portion of the northern hemisphere and in the southern hemisphere (Godfrey 1976). In contrast, the ring-billed gull breeds in the northern United States and southern Canada and winters along coastal areas and inland lakes of the United States and Central America. The herring gull is the least migratory because it overwinters throughout the Great Lakes.

Data presented here showed that residue levels for the common tern were lowest in comparison with gulls in the egg stage and that those levels gradually increased relative to the gulls in the prefledge and post-fledge stage. This suggests common terns accumulated organochlorine chemical residues within western Lake Superior and at a faster rate than the gull species. Because the common tern feeds exclusively from food sources in the water, the contaminant concentrations are likely higher in food sources found in the water than in the surrounding areas where gulls are feeding, such as in open fields, streets, lawns, and garbage dumps. At least two complicating factors make these generalizations difficult. (1) The percentage of lipid in the pectoral muscle is highest for the common tern in comparison to the gulls. Therefore, if the residues are lipophilic, then residue concentrations should be highest in the species with the highest fat content. If it is assumed that the pectoral muscle is representative of the overall distribution of fat in the entire body of gulls and terns, then it is possible to conclude that concentrations are indeed higher in the common tern. However, examination of the relative distribution of fat within these species would be necessary. Fat content may be high in the pectoral muscle of the common tern, because it is the most-migratory and high in the skin of the herring gull because it overwinters in cold climates. (2) Females excrete contaminants into their eggs, which reduces the body load in females (Särkkä *et al.* 1978; Lemmetyinen and Rantamaki 1980). It is likely that the excretion rates are not the same among different species (Peakall *et al.* 1973) and, therefore, higher excretion rates by gulls compared with the common tern could explain the higher concentrations in the eggs of the gull species.

Population Dynamics of Larids

Population levels of the ring-billed gull have increased substantially in Lake Superior during the

1970's (Davis and Niemi 1980; ARDC 1984), while populations of the herring gull have increased by about 25% from 1978 to 1984 along a portion of lake Superior (Goodermote 1984). Populations of the common tern have remained relatively stable in the Duluth-Superior Harbor between 1977 and 1983, but reproductive success was observed to be relatively low in 1982 and 1983 (Cuthbert *et al.* 1984). From 1935 to the mid-1970's major declines in common tern populations were reported from much of the northeastern United States (Kress *et al.* 1983).

Regarding these population changes, it is interesting to note that the lowest concentrations of DDE were found in the ring-billed gull whose population has increased the most over the past ten years. Conversely, the common tern (the species most likely to be affected by organochlorine chemicals) population has decreased substantially. Neither of these conclusions can be considered confirmatory because of the confounding influence of many additional factors involved in the population dynamics of these two species. The results do suggest, however, that attention should be paid to the potential effects of organochlorine chemicals on population changes in common terns found in the western end of Lake Superior.

Muscle and Blood Residues

If residue concentrations in muscle could be predicted from residue concentrations in the blood, then samples of blood taken from the same individuals over time could provide a measure of bioaccumulation of residues within a defined area and without sacrificing the individual. Unfortunately there were no consistent patterns between these two sample types. Furthermore, the relative ranks in residue concentrations between species were not consistent. For example, the pre-fledge muscle samples indicated that the common tern had mean PCB concentrations that were more than three times the ring-billed gull, but blood samples indicated that the common tern and the ring-billed gull had similar concentrations of PCBs. If only the blood values were used in the interpretations of residue concentrations among the species, the conclusions would have been different in comparison with the muscle data. This raises the question, "Which analysis is correct?" To be consistent and comparable with previous studies, the results were interpreted with the muscle data. These results, however, indicate that further study of the relative distribution of residue concentrations in birds may be needed.

Larids as Biological Monitors

The concentrations of PCBs and DDE in eggs were similar and up to three times greater than the highest concentrations observed in any fish species studied in the western end of Lake Superior in the early 1970's (Veith *et al.* 1977). In addition, contaminants such as HCB were detected in concentrations that were twice the concentrations observed in lake trout (*Salvelinus namaycush*) in Lake Superior (Veith *et al.* 1977). Detecting concentrations of compounds present in low concentrations were not possible in most fish samples (Veith op cit.), but were detected in most of the bird samples. Unfortunately, the migratory behavior of many bird species precludes their use in identifying the source of such pollutants. However, the results indicate that the uptake of residues by young-of-the-year seasonal residents of birds could be detected within a watershed system. The ability to detect those chemicals that occur in low concentrations in the environment, coupled with the ability to identify specific individuals, is encouraging for the use of birds in monitoring the presence or contamination levels of a variety of compounds in aquatic communities.

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