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Relation of DDE and PBB Serum Levels in Farm Residents, Consumers, and Michigan Chemical Corporation Employees

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Results of serum PBB determinations on 524 Michigan diary farm residents and consumers of products from the farms, 55 chemical workers, and 56 Wisconsin farm residents are reported. Mean and median values were highest for the chemical workers, followed by consumers from and residents of quarantined and nonquarantined farms. Serum DDE was higher among chemical workers, but was similar for all other groups.

Statistical analysis of serum PBB and serum DDE levels was done with respect to quarantine status, age, sex, and obesity. The most significant correlate with PBB was quarantine status. Serum DDE, age, sex, or obesity were not consistently correlated with serum PBB. For DDE, age was invariably the most significant correlate.

Both serum PBB and DDE were higher in males than females in husband-wife pairs in most cases, although the differences occurred less frequently among older age groups.

These results support the hypothesis that PBB exposure was a recent interim exposure whereas DDE exposure has been cumulative throughout a person's lifetime. Higher PBB and DDE mean concentrations in serum of Michigan Chemical workers suggests an occupational exposure to these chemicals.

During November 1976, a group of 984 Michigan diary farm residents were examined for possible clinical effects of polybrominated biphenyls (PBBs). Serum PBB determinations were part of this study and were carried out according to methodology protocols set forth by the Center for Disease Control, Atlanta (*1*). Serum DDE [1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethylene] is also readily estimated with this procedure, and it has been recorded along with PBB for each survey participant.

At this time, serum PBB and DDE are available for 524 farm family members and 55 Michigan Chemical Corporation employees, who were also examined at the time of the survey. These data have been analyzed statistically with respect to several variables, including farm quarantine status, age, family group, obesity, and sex.

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Materials and Methods

Serum was stored frozen (−15°C) in glass vials, with Teflon-lined caps, which had been prewashed with detergent and water and rinsed in succession with tap water, distilled water, acetone, and hexane. Sera were prepared and analyzed according to the method of Liddle, Price, and Bayse (*1*). Briefly, a hexane–ether extract (3 × 5 ml) of methanol (2 ml)-treated serum (4 ml), was eluted through a Florisil (1.6 g) column topped with sodium sulfate. The first 13 ml hexane eluate was analyzed by electron-capture gas chromatography (Ni^{63} detector) by use of a 3 ft × ¼ in. glass column with 1% OV101 on 80/100 Supelcoport.

Recoveries for DDE and PBB from hexane and of PBB from spiked serum (10–100 ppb in DMSO) were 88 ± 3%, 98 ± 2%, 92 ± 2% (SE), respectively for 47 batch runs. PBB was recovered from spiked serum as low as 0.2 ppb. Levels below this or below

the daily system blank were reported as nondetectable.

Gas chromatographic conditions (Perkin-Elmer 900 or 3920) were such that the major PBB peak used for all calculations had a retention time of 8 min; injector and interface 275°C, column temperature 235°C, Ni^{63} detector 265°C. The limit of detection was 1–2 pg total injection. Calibration curves were determined daily, from at least four standard injections of 10–400 pg/μl, by using a computer-generated exponential curve (log–log plot; slope ca. 0.9). PBB was calculated as the major peak by using electronically integrated area. DDE was calculated from the response factor of two to three injections of a standard solution of 100 pg/μl.

Results

Summary values for PBB and DDE are presented in Table 1 for three groups. Wisconsin farm residents (229) were examined as a control population in March 1977, and of 56 sera examined, two had detectable PBB levels of 0.5 and 1.1 ppb. The latter was a man who had recently changed residence from a Michigan farm.

For Michigan farm residents, mean and median serum PBB values were lower than for 55 Michigan Chemical Company employees, who included ten former PBB production workers and 45 production employees in other departments in the plant. Serum PBB for the chemical workers was also higher than for male farm residents older than 18 years, who corresponded more closely by sex and age. Michigan and Wisconsin farm residents had similar serum DDE levels.

Mean and median serum PBB values for quarantined farm families and consumers from quarantined farms were each significantly higher than either comparable nonquarantined group (Table 2). On the other hand, serum DDE was similar. Persons less than one year old or residents on farms for less than one year had lower serum PBB and DDE than other groups.

Serum PBB values were not detectable ($\leq$ 0.2 ppb) in four of 524 farm family members, two in the nonquarantined group and two among residents for less than one year or infants less than one year old (Table 3). Serum PBB values at or near the limit of detection (0.2 ppb) numbered 18, of which nine occurred in nonquarantined groups (n = 181) and nine among quarantined farm residents (n = 283). All serum PBB among Michigan Chemical Company employees were above 1 ppb. Thus the distribution of serum PBB for chemical workers and quarantined groups showed a marked trend toward higher values.

Serum DDE correlated significantly with age in all groups and sub-groups (various quarantine categories), a trend attributable to accumulated dietary intake of DDT-related environmental residues (2). Serum PBB correlated negatively with age in most quarantine subgroups of farmers, but this trend rarely achieved statistical significance (Table 4). Serum PBB was positively correlated with

Table 1. Serum PBB and DDE summary values for Michigan farmers, Michigan Chemical Corporation employees, and Wisconsin farmers.

		PBB, ppb		DDE, ppb	
	n	Median	Mean ± S. D.	Median	Mean ± S. D.
Michigan farmers	524	2.6	23.7 ± 119.3	9.3	12.9 ± 11.9
Michigan Chemical Co. employees	55	9.3	123.0 ± 367.0	14.5	21.4 ± 18.1
Wisconsin farmers	56	—	—[a]	9.7	11.4 ± 7.4

[a] PBB not detected in 54/56 persons. PBB observed at 1.1 ppb in one person, identified as recently moved from a Michigan farm, and at 0.5 ppb in another person.

Table 2. Frequency distribution of PBB among farming groups and Michigan Chemical Corporation employees.

		Farmers		Consumers		
PBB in serum, ppb	Michigan Chemical Co. employees	Quarantined	Not quarantined	Quarantined	Not quarantined	Less than 1 year old or resident less than 1 yr.
$\leq$0.2	—	9	7	—	4	2
>0.2–10.0	28	206	139	28	21	18
10.1 – 50.0	17	45	7	6	3	—
50.1 – 500.9	6	19	—	4	—	—
> 500.0	4	4	—	2	—	—
n	55	283	153	40	28	20

Table 3. Serum PBB and DDE in Michigan farm family members classified by quarantine status.

		PPB, ppb		DDE, ppb	
	n	Median	Mean ± S.D.	Median	Mean ± S.D.
Resident on					
Quarantined farm	283	3.9	33.9 ± 150.6[a]	9.2	12.9 ± 13.3
Nonquarantined farm	153	1.4	2.9 ± 5.4[b]	10.1	13.0 ± 9.8
Consumer from					
Quarantined farm	40	4.2	56.6 ± 150.1[a]	11.0	16.1 ± 11.9[c]
Nonquarantined farm	28	2.2	3.4 ± 3.7[b]	8.1	9.9 ± 7.1[d]
Farm resident for less than one year or infant less than one year old[e]	20	1.3	1.9 ± 1.7	5.3	8.5 ± 8.9

[a, b] Groups with different superscripts had significantly different mean PBB values; i.e., both nonquarantined consumers and farmers had lower PBB than both analogous quarantined groups.

[c, d] The means of DDE for these two groups were significantly different (*t* test). This difference is attributed to a comparable significant difference in mean ages of these two groups (37.7 ± 18.2 vs. 19.6 ± 15.6).

[e] Not included in *t* test calculations.

Table 4. Correlations of PBB, DDE, and Age[a]

	DDE vs. age	PBB vs. age	PBB vs. DDE	*n*
All farmers	0.53 (0.001)	−0.05 (NS)	−0.05 (NS)	523
Quarantined group	0.54 (0.001)	−0.09 (NS)	−0.07 (NS)	322
Nonquarantined group	0.49 (0.001)	−0.15 (0.02)	−0.07 (NS)	181
Michigan Chemical Corp. employees	0.60 (0.001) 0.58 (0.001)[b]	−0.17 (NS) −0.07 (NS)[b]	0.03 (NS) 0.10 (NS)[b, c]	55
Without PBB production workers	0.62 (0.001) 0.60 (0.001)[b]	0.10 (NS) 0.30 (0.20)[b]	0.54 (0.001) 0.60 (0.001)[c]	45

[a] Level of significance (*p*) indicated in parentheses. Nonsignificant correlations indicated by NS.

[b] Vs. years of employment.

[c] Controlling for age by multiple partial correlation.

serum DDE levels among Michigan Chemical Company employees who had not been involved in PBB production. Serum DDE and PBB correlated with another bromine-containing chemical residue determined in these workers as well, and these trends are attributable to nondirect (casual) exposure within the occupational environment.

Correlations of serum PBB and DDE have been examined with respect to quarantine status, age, and sex. Typically, within a family, serum PBB and DDE are higher for males than females, and serum DDE is higher for older persons. Serum PBB correlated negatively with age in several subgroups of this population of 523 dairy farmers, but the trend was not statistically significant. These findings suggested two hypotheses: that serum DDE levels would follow a pattern such that young females < young males < older females < older males; or that serum PBB would follow a pattern such that young males > young females > older males > older females. The data in Table 5 were used to test this hypothesis. For serum DDE, the trend was observed for both mean and median values for the quarantined group, and the means were signifi-

Table 5. Relation of serum PBB and serum DDE to age and sex.[a]

Population			PBB values, ppb			DDE values, ppb		
Consumers and residents	Sex	Age, yr	Mean ± SD	Median	*n*	Mean ± SD	Median	*n*
Quarantined farms	F	< 18	28.0 ± 83.5	2.3	40	4.2 ± 2.2[b]	3.4	40
	M	< 18	67.7 ± 178.4[c]	7.3	51	6.3 ± 5.1[b, d]	5.3	51
	F	> 18	18.2 ± 58.6[c]	2.5	102	14.9 ± 14.3[d]	11.0	102
	M	> 18	28.2 ± 123.6	4.4	129	16.9 ± 10.3	15.7	129
Nonquarantined farms	F	< 18	3.1 ± 5.4	1.3	37	8.4 ± 7.5	5.8	37
	M	< 18	4.8 ± 9.2[e]	1.7	35	6.5 ± 3.8[f]	5.9	35
	F	> 18	1.7 ± 1.7[e, g]	0.9	57	14.5 ± 8.1[f, h]	13.1	57
	M	> 18	3.1 ± 3.0[g]	2.2	51	17.6 ± 11.4[h]	16.9	51

[a] Values bearing same superscripts were significantly different (*t* test).

cantly different for two of three comparisons. For residents of nonquarantined farms and consumers of their products the median values followed the hypothetical trend, but the mean value for young females was greater than for young males, although not statistically significant.

For serum PBB, median values for males were consistently higher than for females. However, no other consistent trends with respect to age and sex were observed.

Male-female differences in PBB adipose tissue residue concentrations have been noted by Meester and McCoy (3), attributable in part to higher body fat content in females. Among farming families examined so far, we have observed that serum PBB and DDE follow this trend in adult male–female pairs of similar age within a family. In less than 20 of 108 pairs was the PBB in the male value less than that in the female (Fig. 1). Even fewer cases existed, mostly among older pairs, in which both serum PBB and DDE were lower for the male. These differences were not obviously explicable by greater male obesity, etc.

Discussion

Serum PBB was not significantly correlated with serum DDE among the group of 523 dairy farmers for which these variables have been determined. Both PBB and DDE exposures are probably derived mainly from a common dietary source, but the onset, duration, and degree of exposure have been different for the two chemicals.

Morgan and Roan (4) concluded that "elimination of DDE would require the better part of man's natural life-span." DDE is a DDT-derived environmental contaminant, the results of cumulative, low-level dietary exposure. Thus, a consistently significant correlation of serum DDE with age has been observed, even in subgroups of this population as small as single families. The age–DDE association had been reported by Hayes as early as 1958 (2), and other DDE studies have shown lower levels among young children (5). Serum DDE occurred at similar levels among Michigan farmers, regardless of quarantine status, and Wisconsin farmers.

PBB exposure among dairy farmers has been of shorter duration, perhaps more intense, than to DDE–DDT. Therefore, no consistent correlation was observed for PBB with age. Very young children or farm residents for less than one year had lower serum PBB than other groups, suggesting that either intensity of exposure was lower than in preceeding years or that one year was insufficient time to accumulate high levels of PBB under such circumstances. Consumers and residents of nonquarantined farms had significantly lower PBB levels, suggesting a dose–response relationship

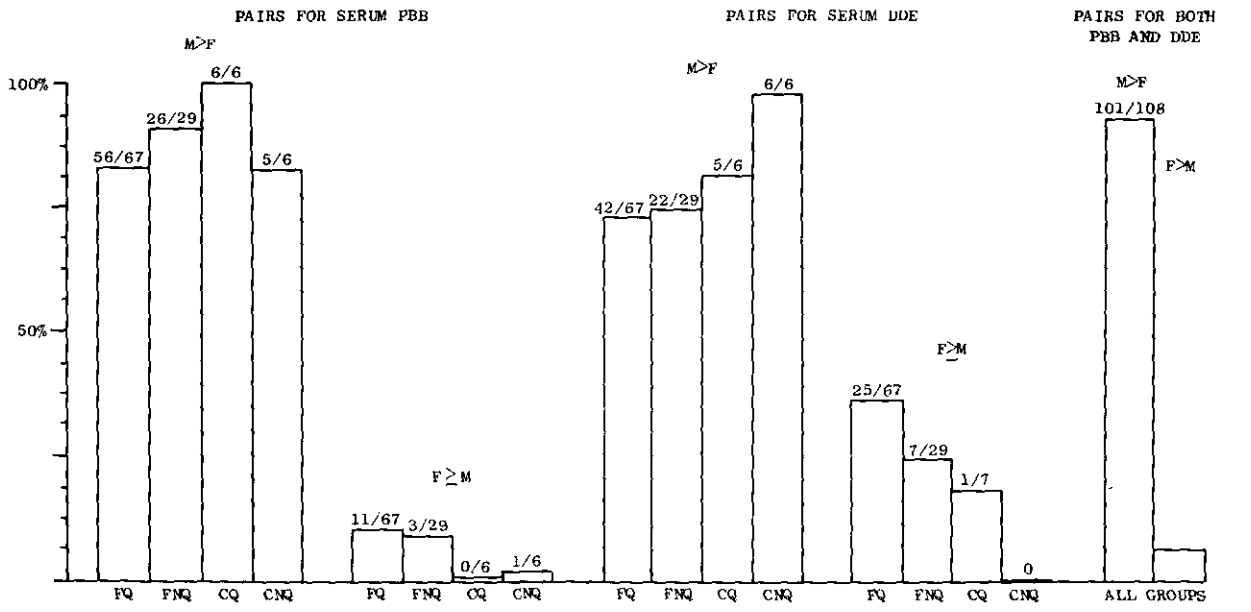

FIGURE 1. Relative serum PBB and DDE levels in male-female (husband-wife) pairs: (FQ) farmers, farms quarantined; (FNQ) farmers, farms not quarantined; (CQ) consumers, farms quarantined; (CNQ) consumers, farms not quarantined.

between consumption of contaminated food and serum PBB.

Both PBB and DDE are higher for males than females in several group comparisons. It has been suggested that the greater proportional body fat in women may account for this difference, but exposure and other factors may also be important. For example, males may have consumed more contaminated meat or may have had more direct contact with PBB.

For Michigan Chemical Corporation employees, a significant correlation was observed for serum PBB with DDE, which is attributed to contact exposure to both within the work environment. PBB and DDT have at different times been manufactured by this company. Both PBB and DDE were correlated with years of employment, and mean and median serum levels of both chemicals were higher than for farm groups.

Summary

Evaluation of serum PBB and DDE concentrations among persons in Michigan suggests different intensities and durations of exposure, and for chemical workers, a different route of exposure. Serum PBB represents a relatively recent exposure, with concentration related to intensity of exposure as designated by relevant quarantined–nonquarantined groupings or by occupational exposure.

Serum DDE represents a cumulative exposure to environmental DDT-related pollutants since 1940, so that age reflects the maximum number of years of exposure. For chemical workers, both serum DDE and PBB levels suggest occupational exposure.

REFERENCES

1. Liddle, J. A., Price, H. A., and Bayse, D. D. Human health consequences of polybrominated biphenyls (PBBs) contamination of farms in Michigan. Protocol for Specimen Analysis and Quality Assurance Program, July 1976. Preprint courtesy Dr. Liddle, CDC, Atlanta, Ga.
2. Hayes, W. J., Jr., et al. Storage of DDT and DDE in people with different degrees of exposure to DDT. Arch. Ind. Health 18: 398 (1958).
3. Meester, W. D., and McCoy, D. J., Sr. Human toxicology of polybrominated biphenyls. Paper presented at the Symposium on Environmental Toxicology, Seattle, Wash., Aug. 4, 1976.
4. Morgan, D. P., and Roan, C. C. The metabolism of DDT in man. Essays Toxicol. 5: 39 (1974).
5. Davies, J. E., et al. Pesticides in people. Pestic. Monit. J. 2: 80 (1968).