Fate of Polychlorinated Biphenyls, Metals, and Other Elements in Papers Fed to Lactating Cows

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Lactating cows were fed 30% of either newspaper, brown or grey cardboard, or computer paper as a cellulose substitute in their ration for periods up to 38 days. Polychlorinated biphenyls (PCBs) present perhaps as ink dye carriers in the paper were excreted in milk up to levels of 76 ppb and stored in tissues such as renal fat up to levels of 1540 ppb. Differential metabolism and storage of certain of the PCBs were indicated. Analysis for 47 metals and other elements did not indicate significant transfer of these from the diet to milk or tissues. Milk production declined 20–33% during the paper feeding experiment but the animals were ending their lactations and were probably more susceptible to ration changes. *In vivo* dry matter digestibilities were 59.8, 51.6, 52.7, and 77.9% for newspaper, brown or grey cardboard, and computer paper diets, respectively. Corresponding *in vitro* dry matter digestibilities for the paper-containing pellets were 60.2, 68.5, 59.5, and 75.1%.

Waste paper has been incorporated into farm animal feeds as a means of disposal and to serve as a substitute form of cellulose in the dairy ration. Mertens *et al.* (1971a) incorporated up to 20% newsprint in diets of lactating cows. Office paper has been fed to sheep (Nishimuta *et al.*, 1969). More recently diets containing up to 30% newspaper have been experimentally fed to cows (Sherrod and Hansen, 1973). Several other investigators have also experimented with paper as a farm animal feed supplement (Daniels *et al.*, 1970; Thomas *et al.*, 1970; Williams *et al.*, 1958; Millett *et al.*, 1973; Rook and Campling, 1959; Kesler *et al.*, 1967; Dinius and Oltjen, 1971, 1972). In most of these experiments only one paper source was used. *In vitro* studies indicate that there are large differences in digestibility between paper types (Mertens *et al.*, 1971b; Mertens and Van Soest, 1971) suggesting that research with various paper sources is needed.

In a recent analytical survey of papers for the possible presence of toxic constituents appreciable concentrations of polychlorinated biphenyl (PCB) type compounds and various heavy metals were found in certain newspapers, cardboards, and computer paper (Serum *et al.*, 1973). PCBs have also been reported in paper board (Thomas and Reynolds, 1973) and carbonless copying paper (Masuda and Kagawa, 1972) which may be included in paper products recycled into paperboard. PCBs have also been reported in a wide variety of paper products by others (Shahied *et al.*, 1973; Stanovick *et al.*, 1973; Food Chemical News, 1971). The toxicologic significance of PCBs in the environment has been amply reviewed (Peakall, 1972; Edwards, 1971; Environmental Health Perspectives, 1972; Peakall and Lincer, 1970). In the work reported, papers known by analysis to contain PCBs (Aroclors) and metals have been fed to lactating cows with subsequent analysis of milk and tissues for these toxicants.

EXPERIMENTAL SECTION

Cattle Feeding. Newspaper, brown cardboard, grey cardboard, and computer paper comprise four major classes of paper which may contain PCBs and these were chosen for study. The newspaper was the Buffalo Evening News collected between December, 1970, and April, 1971. The brown cardboard consisted of various used boxes collected locally. The grey cardboard consisted of rolls of grey paperboard (Bogus Paper, FCF Paper Co., Rochester, N.Y.). Used computer paper was obtained from the Cornell University Dairy Records Processing Laboratory.

About 300 lb of each paper was ground in a hammer mill. The pulverized material was mixed with other feed constituents so as to comprise a complete dairy ration. The level of 30% paper in the pellets was selected to obtain a maximum intake of paper and PCB contamination. The feed composition is shown in Table I. The feed constituents were thoroughly mixed and then pressed into 1.27-cm pellets. Four cows (three Holsteins and one Ayrshire) were each fed pellets containing one of these papers. The pellets were fed throughout periods up to 38 days beginning with an 8-day conditioning period. During the conditioning period the normal daily amounts of hay and grain were progressively reduced while the quantity of pellets was increased. It was necessary to continue feeding 1.14–2.28 kg of hay and 0.91–1.82 kg of grain daily after the conditioning period in order that the cows would accept the paper-containing pellets. Morning and evening samples of the total mixed milk were taken prior to feeding paper (control) and daily throughout the feeding period. The morning and evening milk samples were combined each day prior to analysis. Feces was collected using specially designed gutter trays and urine was collected *via* catheter for 7 days at the end of each feeding period. Dry matter and Kjeldahl nitrogen analyses were performed by standard methods. Cell wall determinations and *in vitro* digestibilities of the paper-containing pellets were determined by the procedures of Goering and Van Soest (1970). At the end of the feeding period the animals were sacrificed and tissue samples taken for analysis. Table II lists pertinent data concerning the feeding trial. The total intake of PCBs by each cow was calculated from the quantities of the particular paper and alfalfa consumed and the concentrations of PCBs found initially in these feed components (see Table III).

Methods of Analysis. The analysis for PCBs was done using the published method (Pesticide Analytical Manual, 1971) involving fat extraction, isolation using acetonitrile partitioning, and column chromatography on Florisil. Feces samples were processed using procedure 212.13a in the above reference. Final determination was performed by electron affinity gas chromatography as previously reported (Serum *et al.*, 1973) except that the gas chromatographic column contained 5% OV-17 on 80–100 mesh Chromosorb AW and was 6 ft long. Quantitation was made by measuring the heights of the three peaks emerg-

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Table I. Feed Composition

Constituent	Wt, %
Paper	30
Ground alfalfa	30
Corn meal	30
Soybean meal	3
Sodium chloride	1
Monosodium phosphate	1
Molasses	5

ing at retention times of 30.4, 34.3, and 44.8 min which corresponded to those in gas chromatograms of Aroclor 1254. The method of isolation and determination also permitted analysis of Aroclor 1242 which may be commonly present in waste paper.

Analysis for 44 elements (Al, Ar, As, Au, Ba, Br, Ca, Ce, Cl, Co, Cr, Cs, Cu, Eu, F, Fe, Hf, I, In, K, La, Lu, Mg, Mn, Na, Nd, Ni, Rb, Ru, S, Sb, Sc, Se, Sm, Su, Sr, Ta, Th, Ti, U, V, W, Y, and Zn) was performed using nondestructive neutron activation analysis. Milk samples were blended and tissue samples were ground and mixed. The samples were dried and 1 g of each was subsampled and weighed into a polyethylene vial (1.5 cm i.d. × 2 cm high). The samples were irradiated twice, once for a short period (which varied depending on sodium and chlorine content but on the order of 1 min) and again for a period of approximately 4 hr. The neutron flux to which the samples were exposed was about 10^{12} neutrons/cm^2 per sec (1.2×10^{12} for the short irradiation and 1.3×10^{12} for the long irradiation). After the short irradiations the samples were counted within a few minutes on a Ge(Li) counting system for a period of 8 min. Data were acquired using a nuclear data No. 4420 multichannel analyzer and stored on magnetic tape for later processing. For the long irradiations, the samples were counted as soon as practical, considering the level of the activity due to sodium. This period of time varied from 2 to 5 days after the end of the irradiation.

After acquisition, the data were processed using an IBM 370 computer and a developed program. The main function of this program is to find the areas under preselected peaks in the γ spectra. The program fits the peaks to a modified gaussian distribution, assuming a small asymmetry for the peaks, on a background represented by a quadratic polynomial. The program is capable of subtracting interference due to peaks close to the peak of interest. The computation of parts per million was based on comparison of the peak areas determined as above with peak areas determined for standards run under known circumstances.

Table III. Concentration of PCBs Initially in the Feed Components

Sample	PCBs, ppb
Newspaper	120
Brown cardboard	47
Grey cardboard	610
Computer paper	320
Ground alfalfa	6
Corn meal	<5
Soybean meal	15
Sodium chloride	<4
Monosodium phosphate	<4
Molasses	<2

The determination of cadmium and lead was performed by dry ashing 5 g of the samples (up to 475°) by the respective AOAC methods (1965) followed by analysis using conventional flame atomic absorption spectrophotometry with a Perkin-Elmer Model 303 instrument. The determination of mercury was accomplished by oxygen flask combustion (Gutenmann and Lisk, 1960) of 1-g samples dry weight) and flameless atomic absorption analysis (Hatch and Ott, 1968).

RESULTS AND DISCUSSION

The concentrations of PCBs in the feed components are listed in Table III. Paper was the main contributor to PCBs in the cattle diets. Grey cardboard and some brown cardboards are made from recycled paper. PCBs serving, for instance, as dye carriers in the ink apparently survive the ink removal and regeneration process to contaminate the cardboard products. Ink used again on the cardboard can also contribute to the PCB content. Newspaper is usually prepared from virgin wood cellulose materials and does not contain recycled constituents. Ink used on it, however, may again contain PCBs. The presence of traces of PCBs in other of the feed constituents may be due to the ubiquitous presence of these compounds. As pointed out by Price *et al.* (1972) PCBs have been identified in a range of samples including sediments, river water, sewage,

Table II. Data Relevant to the Feeding Trial

Paper	Cattle breed	Wt of cow, kg	Total pellets[a] consumed during trial, kg	Total PCB intake, mg/kg body wt	Duration of feeding, days	Total milk produced during trial, kg	Total milk produced during a similar period immediately before trial, kg	% decrease in milk production
Newspaper	Holstein	682	194	0.011	36	790	1111	20.2
Brown cardboard	Holstein	705	670	0.015	38	878	1130	23.0
Grey cardboard	Ayrshire	500	418	0.154	37	667	975	31.6
Computer paper	Holstein	512	333	0.064	39	527	790	33.3

[a] Containing 30% paper.

Table IV. Recovery of PCBs from Samples

Sample	Added, ppm	Recovery, %	Estimated sensitivity, ppb
Milk	0.04	83	1
	0.1	77	
Urine	0.2	94	10
Feces	0.2	105	14
Brain	0.2	58	5
	0.3	57	5
Heart	0.2	68	5
	0.3	57	5
Kidney	0.2	80	5
	0.3	89	5
Liver	0.2	66	5
	0.3	85	5
Spleen	0.2	67	1
	0.3	77	1
Brisket fat	0.2	67	5
	0.5	93	5
Omental fat	0.6	66	5
	1.66	83	5
Renal fat	0.6	68	5
	1.66	64	5
Chuck	0.2	73	5
	0.3	72	5
Round	0.2	64	5
	0.3	67	5
Ground alfalfa	0.2	75, 101	6
Corn meal	0.2	81	5
Soybean meal	0.2	76	5
Sodium chloride	0.2	75	4
Monosodium phosphate	0.2	99	4
Molasses	0.2	93	2

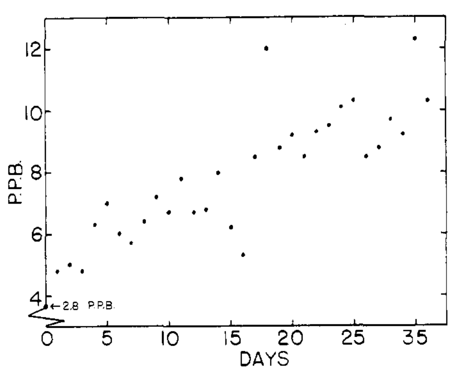

Figure 1. Graph showing the concentrations of PCBs (parts per million, fresh weight) in milk as a function of length of time of feeding newspaper in the dairy ration.

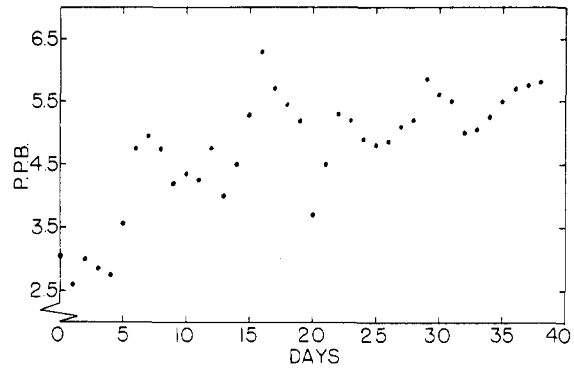

Figure 2. Graph showing the concentrations of PCBs (parts per million, fresh weight) in milk as a function of length of time of feeding brown cardboard in the dairy ration.

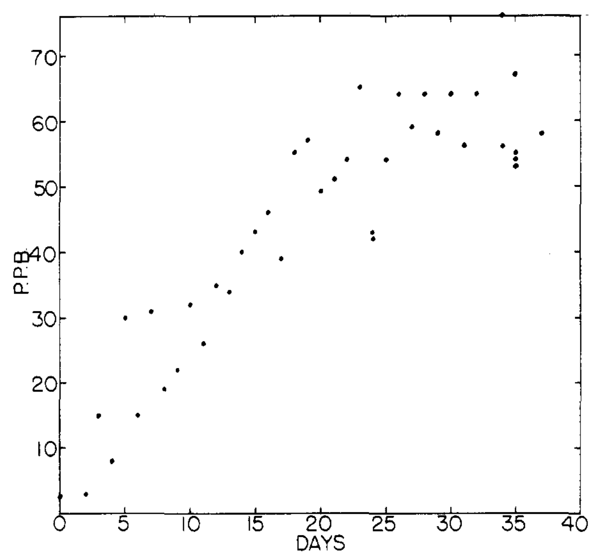

Figure 3. Graph showing the concentrations of PCBs (parts per million, fresh weight) in milk as a function of length of time of feeding grey cardboard in the dairy ration.

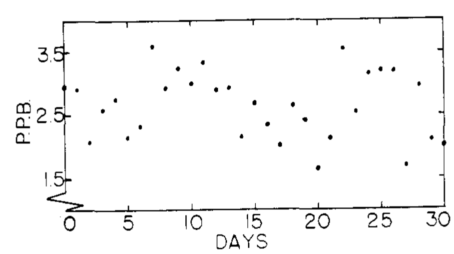

Figure 4. Graph showing the concentrations of PCBs (parts per million, fresh weight) in milk as a function of length of time of feeding computer paper in the dairy ration.

ink, newsprint, plastic bags, and plastic products. It should be noted that the sales of PCBs by one manufacturer (Monsanto Co.) recently have been restricted to use in sealed sources. Their use in inks may therefore decline. Halogenated flame retardants used on fabrics which may survive recycling back into certain types of paper (Erasable typing paper has a 25% rag content) are another possible source of contamination. Spurious gas chromatographic peaks have also been observed in grains stored in cloth bags (Levi and Nowicki, 1972). They may also occur in the coatings used in silos (Skrentny *et al.*, 1971).

The recoveries and estimated sensitivities of the method for PCBs are shown in Table IV. The residues of PCBs found in milk as a function of the lengths of time of feeding are illustrated in Figures 1-4. The PCB concentration shown for day zero corresponded to the prefeeding control level. Replicate analyses were made of certain samples (the 24- and 35-day milk samples in Figure 3). The concentrations of PCBs found in the various tissues and excreta are listed in Table V. The concentrations of PCBs in milk and tissues as shown in Figures 1-4 and in Table V were not corrected for per cent recovery.

The increase in the concentration of PCBs in the milk of the animals fed newspaper and brown and grey cardboards as feeding progressed is evident in Figures 1-3. Based on the level of PCB intake the cow fed grey cardboard is expectedly highest. The highest PCB concentrations were also found in the fat samples of this cow (Table V). The animal fed computer paper became sick and developed diarrhea which continued throughout the experi-

Table V. Residues of PCBs in Cattle Tissues and Excreta

	PCB concn (ppb fresh wt) in cow fed			
Tissue	News-paper	Brown card-board	Grey card-board	Com-puter paper
Brain	22	6.0	34	4.8
Heart	10	9.7	83	25
Kidney	33	12.7	80	17
Liver	24	18.3	210	5.1
Spleen	6.7	5.0	<1.5	5.4
Brisket fat	110	36.0	320	67
Omental fat	175	77.3	1480	96
Renal fat	194	76.0	1540	110
Chuck	17	16.8	48	11
Round	7	25.7	30	5.5
Urine[a]	<10	<10	<10	<10
Feces[a]	<14	<14	29.0	<14

[a] Urine and feces collected on the last day milk was sampled.

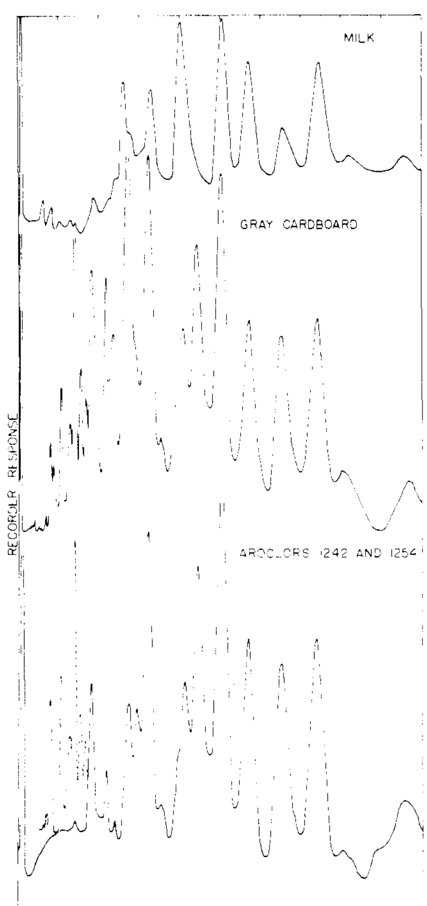

Figure 5. Chromatograms of: (top) milk samples on the last (37th) day of the cow fed grey cardboard; (middle) grey cardboard; and (bottom) Aroclor standards 1242 (dashed line) and 1254.

ment. The absence of elevated concentrations of PCBs in her milk and fatty tissue might appear to reflect their diminished absorption owing to a shorter residence time of food materials in the gut. This may cause the measured *in vivo* digestibility of computer paper to be somewhat lower than normal. A number of subsamples of each of the ground papers were taken and thoroughly mixed prior to analysis to minimize the possibility that a biased sample of paper was analyzed. Thus the values listed in Table III for the initial concentrations of PCBs in the papers fed are believed to be representative and valid.

Figure 5 shows chromatograms of the milk sampled on the last day (37th day) from the cow fed grey cardboard (top chromatogram), grey cardboard (middle chromatogram), and Aroclors 1242 (dashed line) and 1254 (bottom chromatogram). The corresponding peak retention times in the chromotograms are evident. Most of the peaks in the chromatograms of Aroclors 1242 and 1254 also appear in the chromatograms of grey cardboard. Certain peaks such as those of Aroclor 1242 and notably those in the chromatogram of grey cardboard having retention times of 10.6, 13, 13.9, and 26.5 min were absent in the chromatogram of milk. Consistently, certain peaks such as those emerging at 15.5, 19.7, and 39.2 min were proportionally smaller in the milk chromatograms than the corresponding peaks in the grey cardboard chromatogram, thus indicating lack of absorption or possible metabolism after absorption. The same spectrum of chromatographic peaks, with the specific ones mentioned above reduced in size, also appeared in the chromatograms of tissues of the cow fed grey cardboard. Each of the other paper types (newspaper, brown cardboard, and computer paper) had peaks corresponding to Aroclor 1242 as well as 1254. The milk and tissues from these cows also showed approximately the same spectrum of peaks, *i.e.* mainly those of Aroclor 1254 and only small or negligible levels of those in Aroclor 1242. The same peaks that appeared smaller in milk and tissues of the cow fed grey cardboard again were reduced in size in the milk and tissues of the other cows. This would therefore further indicate differential absorption or metabolism of certain of the components of Aroclor 1254 and 1242. It should be noted, however, that the electron affinity detector response of Aroclor 1242 is only about one-third that of Aroclor 1254. The absence or near absence of Aroclor 1242 may at least in part therefore have been due to a lack of detector sensitivity. Differential metabolism (Grant *et al.*, 1971) and storage (Curley *et al.*, 1971) of PCBs in rats has also been indicated. The excretion and persistence of PCBs in cows milk, its storage in tissues (Fries *et al.*, 1972, 1973; Platonow *et al.*, 1971; Saschenbrecker *et al.*, 1972) and its transplacental transfer in the bovine (Platonow and Chen, 1973) have been studied. Excretion of PCBs in the urine and feces was minor or undectable (Table IV) but some elimination of PCBs was observed in the urine and feces of preadolescent girls (Price *et al.*, 1972).

Apparent digestibility coefficients, the consumed input minus the fecal output divided by the consumed input, were determined for dry matter, cell wall, and crude protein. Since there was only one animal per treatment, an *in vitro* technique was also used to determine dry matter and cell wall digestibility of the paper-containing pellets. The chemical composition and digestibilities of the paper-containing pellets and rations are given in Tables VI and VII. Table VI shows that the addition of the small amount of hay and grain to the paper-containing pellets did not significantly change the chemical composition between the complete ration and the pellets. The reason for this is that pellets were the greatest amount of the total ration with the cows consuming 2.3, 2.0, 2.0, and 2.0% of their body weight daily as pellets.

The digestibilities given in Table VII confirm the variation in digestibility among different types of paper observed by Mertens and Van Soest (1971). The *in vitro* dry matter digestibilities agree with those of Mertens and Van Soest (1971) in that computer paper was most digestible followed by brown cardboard, grey cardboard, and newspaper. However, the *in vivo* digestibilities of the brown and grey cardboards were much lower than those predicted by the *in vitro* observations. Crude protein digestibili-

Table VI. Chemical Composition of Paper-Containing Pellets and the Complete Rations Fed to the Cows

	Brown cardboard		Computer paper		Newspaper		Gray cardboard	
	Pellets	Ration	Pellets	Ration	Pellets	Ration	Pellets	Ration
Crude protein, %	11.9	12.6	12.9	13.6	13.0	13.8	11.4	12.4
Cell wall, %	44.8	43.7	45.7	43.7	41.9	40.7	50.5	48.0
Cellulose, %	28.6	27.4	29.5	26.5	23.9	22.9	31.0	28.5
Lignin, %	8.3	7.9	5.5	5.3	7.7	6.9	9.1	8.4

Table VII. *In Vitro*[a] and *in Vivo*[b] Digestibilities of the Paper-Containing Pellets and the Complete Rations Fed to the Cows

	Brown cardboard, %		Computer paper,[c] %		Newspaper, %		Gray cardboard, %	
	Pellet	Ration	Pellet	Ration	Pellet	Ration	Pellet	Ration
Dry matter	68.5	51.6	75.1	77.9	60.2	59.8	59.5	52.7
Cell wall	58.9	26.0	76.0	73.1	36.8	31.2	46.7	33.4
Protein		47.1		72.7		64.4		49.3

[a] Pellet digestibilities were determined *in vitro*. [b] Ration digestibilities were determined *in vivo*. [c] The animal fed computer paper was sick during the first 2 days of the digestion trial.

ties for these two rations were also lower than expected. The protein digestibilities of all four rations should have been similar because the same protein sources were used to supplement each diet. Since there was only one animal per treatment, it is probable that the differences between *in vivo* and *in vitro* digestibilities could be attributed to animal variation.

The results of analysis of the papers fed, milk (sampled on the last day of feeding paper to each cow), liver, kidney, and muscle (round) (taken at the time of slaughter) for metals and other elements by neutron activation and atomic absorption analysis are listed in Tables VIII and IX (see paragraph at end of paper regarding supplementary material). There was no correlation between the concentration of particular elements in the various papers and their levels in the respective tissues or milk samples. As would be expected, certain elements such as lead and cadmium tended to be higher in liver and kidney tissue than in muscle or milk since these organs are known to concentrate heavy metals.

Considering toxic elements, a high concentration of lead (80 pm) found in newspaper may have derived largely from the inks used in the colored pages. Hankin *et al.* (1973) reported a range from 8 ppm of lead in black and white pages to 3600 ppm in colored pages. The inks ranged from 275 ppm of lead in black ink to 29,000 ppm in yellow ink. Only a small (less than 0.02%) proportion of lead fed as lead nitrate to cows was found to be excreted in milk (Stanley *et al.*, 1971). The concentrations of lead in the milk (2.14–3.85 ppm) and tissue samples (0.9–7.5 ppm) are in about the same range as those reported as typical for lead in dairy products and meats (Schroeder *et al.*, 1961). The concentrations of barium found in the various milk (3.2–34 ppm) and tissue (0.01–21.4 ppm) samples appeared high and of concern. These concentrations, however, are in the same range as those found in human tissues (Schroeder, 1970). Barium administered subcutaneously as barium chloride to guinea pigs was shown to accumulate in the following tissues in decreasing order: bone, muscle, bone marrow, digestive tract, lungs, kidneys (Truhaut and Berrod, 1962). Daniels *et al.* (1970) found no accumulation of minerals by beef liver in cattle fed newspaper.

In summary, cattle consuming a diet containing 30% (by weight) paper excreted up to 76 ppb of PCBs (in grey cardboard) in the milk. Residues of PCBs in milk appear to increase asymptotically after several days of consuming paper (see Figures 1–3). Residues of PCBs in tissues attained their highest level (1540 ppb) in renal fat of the cow fed grey cardboard. The present tolerance for PCBs in milk is 0.1 ppm. The tolerance in poultry is 5 ppm on a fat basis. Fat-soluble compounds such as the polychlorinated terphenyls can contaminate certain paper products (Villeneuve *et al.*, 1973) and may be potential contaminants of animal products (Fries and Marrow, 1973). PCBs and related compounds are now no longer permitted for use in paper products.

Certain PCB isomers appeared to show differential absorption, metabolism, and tissue storage. Contamination of milk and tissues by toxic elements in paper products did not appear to be significant. *In vivo* dry matter digestibilities were 59.8, 51.6, 52.7, and 77.9% for newspaper, brown or grey cardboard, and computer paper diets, respectively. Corresponding *in vitro* dry matter digestibilities for the paper-containing pellets were 60.2, 68.5, 59.5, and 75.1%.

The decrease (20.2–33.3%) in milk production found (see Table II) in this experiment was also noted in the work of Mertens *et al.* (1971a,b). The animals were ending their lactations and were probably more susceptible to ration changes.

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Supplementary Material Available. Tables VIII and IX giving results of analysis for metals and other elements will appear following these pages in the microfilm edition of this volume of the journal. Photocopies of the supplementary material from this paper only or microfiche (105 × 148 mm, 24× reduction, negatives) containing all of the supplementary material for the papers in this issue may be obtained from the Journals Department, American Chemical Society, 1155 16th St., N. W., Washington, D. C. 20036. Remit check or money order for $3.00 for photocopy or $2.00 for microfiche, referring to code number JAFC-74-954.

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Photodecomposition of Methidathion

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Irradiation of methidathion was carried out with ultraviolet light of a wavelength of 254 mμ. The photoproducts were isolated and purified with different systems of tlc and by comparison of their infrared and mass spectra with the spectra of synthetized samples. The following products were identified: the P=O analog; *O,O,S*-trimethyldithiophosphonic acid; *O,O,S*-trimethylthiophosphonic acid; *O,O,O',O'*-tetramethyl-*S,S*-methylbis(dithiophosphonic) acid; 2-methoxy-4-methylthiomethyl-Δ^2-1,3,4-thiadiazolin-5-one; 2-methoxy-Δ^2-1,3,4-thiadiazolin-5-one; 2-methoxy-4-methyldithiomethyl-Δ^2-1,3,4-thiadiazolin-5-one; bis(2-methoxy-Δ^2-1,3,4-thiadiazolin-5-on-4-yl) disulfide; 1,3,4-oxodiazolidine-2,5-dione.

The fate of methidathion (Supracide, GS 13005, *O,O*-dimethyl 5-[(2-methoxy-Δ^2-1,3,4-thiadiazolin-5-on-4-yl)-methyl] phosphorodithioate), a Ciba-Geigy insecticide, has been studied in several biological systems. Esser and Müller (1966) and Esser *et al.* (1968) following the metabolic pathway in the rat and in plants showed an appreciable breakdown of the applied [5-*carbonyl*-^{14}C]methidathion to $^{14}CO_2$ (27.4% CO_2 after 14 days on beans, 31.6% CO_2 after 56 days on apples, and 80.5% after 4 days in rats).

Bull (1968), using labeled compounds on cotton, alfalfa, rats, and fifth instar tobacco budworms, concluded that in insects and plants CO_2 was not a major end product of the metabolism of the heterocyclic ring and reported the presence of several water-soluble metabolites. Esser *et al.* (1968) identified three main metabolites in the rat: the 4-methylthiomethyl derivative of the intact heterocyclic ring and its oxidation products, the sulfoxide and the sulfone. Cassidy *et al.* (1969a,b), after treating alfalfa with methidathion, reported the presence of the oxygen analog and noncholinesterase inhibiting water-soluble ^{14}C residues. Polan *et al.* (1969), studying the physiological effects and the metabolism in the bovine, found very low levels of the parent compound in the tissues and no evidence for the presence of the oxygen analog. Rüfenacht (1968) described the synthesis of methidathion and the possible metabolites. Analytical methods for routine determinations of residues in plants, fruits, and soils have been published by Eberle *et al.* (1967) and Mattson *et al.* (1969). Eberle *et al.* reported, besides methidathion,

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