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Polychlorinated biphenyl residues in silos in the United States

By

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Contents

I.	Introduction	135
II.	PCB-containing silo coating	137
III.	The contamination process	139
IV.	Cows and PCB residues	140
V.	Regulation of PCB's	142
VI.	Impact of PCB-contaminated silos	143
Summary		145
References		145

I. Introduction

Polychlorinated biphenyls (PCB's) were described in the scientific literature in 1881 by Schmidt and Schultz, and successful commercial production was achieved in 1930 (Hubbard 1965). The manufacturer of these compounds was Swan and Company who recommended their use in commercial applications for protective coatings, waterproofing, flameproofing, electrical insulation, and adhesives along with multiple miscellaneous uses (Penning 1930). PCB's were marketed under the trade name of Aroclors by Swan and Company and then by Monsanto Company. Chemically the Aroclors are relatively inert; thus, they are not hydrolyzed by water and they resist alkalies, acids, and other strong corrosive chemicals. They are not appreciably volatile at normal ambient temperatures and have boiling points which are in excess of 275° C (Monsanto Company). They are stable to long periods of heating and can be distilled at ordinary pressure without any carbonization or decomposition. PCB's are insoluble in aqueous media and very soluble in hydrocarbon solvents. Therefore, these chemical compounds have the chemical

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MONS 086282

and physical characteristics to be persistent in the environment and accumulate up the food chain (PEAKALL and LINCER 1970).

NESBIT and SAROFIM (1972) estimated that sales of PCB's in North America between 1930 and 1970 totaled 5×10^5 tons. This large sales volume is indicative of the industrial versatility of the PCB's, their low cost, and lack of concern for their toxicity. In fact, it was suggested in 1930 (PENNING 1930) that PCB's might be substituted for chicle in chewing gum. TREON *et al.* (1956) investigated the effects of the vapors from heated Aroclor 1242 and 1254 on small animals. The vapors of Aroclor 1242 were not shown to be toxic and the vapors of Aroclor 1254 caused histopathologic injury only when sufficiently concentrated (5.4 μg/L). However, little was known about the biological significance of the Aroclors until after PCB residues were found in fish from Swedish waters in 1966 (JENSEN 1966). Since 1966, PCB's were found to have wide environmental distribution and were found as tissue residues in many animal species. The occurrence of these residues have been reviewed (*Department of Commerce* 1972, EDWARDS 1971, JENSEN *et al.* 1969, PRESBY *et al.* 1970, RISEBROUGH *et al.* 1968, VEITH and LEE 1970).

PCB residues enter the environment primarily through the vaporization of plasticizers, vaporization during open burning, leaks and disposal of industrial fluids, and the intentional spreading such as through the use of pesticide formulations which contained PCB's (NESBIT and SAROFIM 1972). As these uses have led to detectable concentrations of PCB residues in the environment, these sources of residue have contributed only small quantities of the PCB's consumed by human beings. With the exception of fish consumption, the main sources of contamination for human beings have been industrial accidents which directly contaminate food sources and from PCB's in a manufactured food or feed packaging from which the residue has leached into the foodstuff.

The most [illegible] incident of accidental contamination of a food source [illegible] in Japan (KURATSUNE 1972, KURATSUNE *et al.* 1971, KURATSUNE *et al.* 1973). In this incident more than 1,000 persons received a toxic dose of PCB's through rice oil that had been contaminated during its manufacture. It was estimated from this incident that the average patient received approximately two g of PCB's while the [illegible] dose to demonstrate a toxic effect was approximately 0.5 g (KURATSUNE *et al.* 1973). Whereas no significant spill such as the Yusho incident has occurred in the United States, several industrial accidents and/or mistakes have yielded contaminated foodstuffs. The most costly incident which occurred in the United States was in a fishmeal plant on the east coast where fishmeal was contaminated by a leaky heat transfer system as the meal was being prepared for livestock feed (SPAULDING and WESSEL 1972). The meal was subsequently fed to fish and poultry, resulting in extensive losses of fish, meat, and eggs which were impounded and destroyed. Several other episodes occurred where poultry and turkeys were found to have excessively high

MONS 086283

levels of PCB's. These have occurred in Minnesota, Oklahoma, and California; however, the source of the PCB's could not be determined in these incidents (Spaulding and Wesen. 1972). PCB's from transformer fluid were used as a vehicle for herbicides sprayed along the highway right-of-ways in the Martinsburg, West Virginia area. Through this route, PCB's contaminated the dairy cattle grazing area, and PCB residues were detected in the milk (Spaulding and Wesen. 1972).

The use of recycled paper products for packaging of food and feed has also led to the contamination of human foodstuffs with PCB's (Spaulding and Wesen. 1972). The major source of PCB's in recycled paper is carbonless self-duplicating paper (Trout 1972).

In March and April of 1970, the Ohio *State Department of Agriculture* detected a chlorinated hydrocarbon residue, presumed to be a PCB, in the milk from several dairy farms. Initially, the milk from one of these farms was excluded from the market by the Ohio State *Department of Agriculture* for containing a DDT residue. Upon further investigation it was determined that this residue was a PCB. The source of the PCB residue was determined to be an ingredient of the coating materials inside the silos. Since that time it has been determined from regulatory experience and information supplied by silo companies that PCB's had been used in the coating materials in silos located in Ohio, Indiana, Kentucky, Tennessee, Michigan, Illinois, Alabama, Georgia, North Carolina, South Carolina, Pennsylvania, Maryland, West Virginia, Virginia, and New York (Willett 1973).

II. PCB-containing silo coating

The organic acids produced by the ensiled material in farm concrete silos cause considerable decomposition and erosion of inside walls. Therefore, coating materials have been used for many years to resist the corrosive action of the silage juices and thus extend the useful life of the silo. Through the years, many products have been used to coat the inside of silos including wax, mineral oil, epoxy resins, and many other resinous coating. One such coating, Cumar[1], was the coating responsible for most, if not all, of the PCB-contaminated silos.

Cumar was formulated in the summer of 1941 for the Concrete Silo Company of Bloomfield, Indiana (Egan 1971). The formula for the Cumar silo coating is shown in Table I. It was based on formulations developed by Seymour (1941) for the protection of concrete silos from the organic acids produced during the ensiling process. Aroclor 1254[2] was incorporated because of low cost, good ability to plasticize, and excellent

[1] The silo coating called Cumar which contains Aroclor 1254 (*Monsanto Company*) should not be confused with Neville R-16-A resin which has the same name.

[2] Aroclor 1254 is a mixture of chlorinated biphenyls containing about 54% chlorine (*Monsanto Company*).

MONS 086284

Table 1. *Formulation of Cumar silo coating* (Egan 1971).

Ingredient	Amount
Paraplex No. 2 (dark flaked)	40 lb
R-16-A Neville resin (flaked)	40 lb
Clay	20 lb
Perlite	45 lb
Aroclor 5460	20 lb
Aroclor 1254	70 lb
Xylol	13 gal
S.C. Solvent-100	5 gal
S.C. Solvent-3	8 gal

chemical resistance. Aroclor 5460[a], a polychlorinated terphenyl (PCT), was a resin fortifier. The application of Cumar varied with various silo companies which were involved with its use. The wide distribution of this coating material on silos in the eastern half of the United States was the result of various mergers and disbursements of the silo companies involved.

Michigan Silo Company, founded in 1910 and incorporated in 1916, had initial production facilities located in Charlotte, Michigan, White Marsh, Maryland, and Massillon, Ohio (Egan 1971). Concrete Silo Company, with silo production facilities in Bloomfield, Indiana and Monteagle, Tennessee, was later acquired and was operated as a subsidiary of Michigan Silo Company. After acquisition of Concrete Silo Company, the headquarters for the entire operation were in Bloomfield, Indiana. In 1958, a corporate reorganization split the company's assets; thus, the Massillon plant became the Michigan Silo Co., Inc., and likewise the Bloomfield Silo Co., Inc. was formed in Bloomfield, Indiana and the Monteagle Silo Company, Inc. was formed in Monteagle, Tennessee.

Personnel of the Michigan Silo Co., Inc. have stated that the purpose of Cumar on new construction was to provide a sealant to retain moisture in the joint mortar during curing and to protect the mortar from silage acid (Egan 1971). On recoating, the material was applied to the joint mortar and in some instances on the entire surface of the silo. Cumar had been used for recoating of silo interiors since 1948 throughout the entire territories served by the old Michigan Silo Company. Silos constructed by the Monteagle Silo Company, Inc. and the southern division of Michigan Silo were primarily of a waxed stave construction. Therefore, the use of Cumar by these companies was limited to the mortar between the stave blocks as it would not bond to the waxed surface.

It is difficult to estimate the exact number of silos which have been treated with the Cumar silo coating. Records of the company which used this material prior to 1966 are either nonexistent or unavailable. To fur-

[a] Aroclor 5460 is a mixture of chlorinated terphenyls containing about 60% chlorine (*Monsanto Company*).

MONS 086285

ther complicate number estimates, many of the silos which were coated with Cumar have been torn down or are no longer in use.

The Michigan Silo Co., Inc. reported 108 sales of Cumar during the period 1969 through the end of 1969 (EGAN 1971). Records for Montcalm or Bloomfield Silo Companies are not complete for the Cumar sales or use. The best estimate to date as to the number of silos constructed by the present or predecessor companies and to which the PCB-containing Cumar coating was applied is from 5,000 to 6,000 silos (*U. S. Department of Agriculture* 1973).

III. The contamination process

Cumar was just one of many coating materials which have been applied to silos in order to fill the pores of concrete and reduce the surface vulnerable to acid attack. When Cumar was applied to a silo wall, solvents in the coating material penetrated into the concrete, thus carrying the PCB's and PCT's into the silo wall. After the carrier solvents evaporated, a firm coating material was left on and in the inside walls of the silo. The mechanisms by which the PCB's from this Aroclor-containing coating were transferred to silage was originally thought to be only mechanical, as by flaking off of chips of the coating. It is not uncommon for these chips to contain 5,000 to 20,000 ppm of Aroclor 1254. However, recent work has shown that this Aroclor is soluble in silage juices (SCHMITZ *et al.* 1971). The solutes may well be lactic and acetic acid, which are important organic acids formed when silage ferments (PRATT *et al.* 1958). Aroclor 1254 has been shown to be soluble in acetic acid (*Monsanto Company*). After PCB's are dissolved by the silage juices, they diffuse into the silage; therefore, the residue concentrations are significantly higher in the silage adjacent to the silo wall as compared to that in the central portion of the silage. SCHMITZ *et al.* (1971), WILLETT and CONRAD (1973), and WILLETT (1974) have shown that most of the Aroclor residue was within the first 15 cm from the silo wall. Residues were not detected beyond 120 cm from the silo wall. One might expect this to be a typical distribution in most silos; however, the distribution will probably be influenced by time and temperature. Superimposed on the diffusion distribution of the Aroclor residue in the silage was the confounding effect of precipitation and the downward migration of the silage juices. This effect tends to cause an accumulation of the Aroclor residue near the base of the silo (WILLETT 1974).

Failure of the Cumar coating occurs with time, and flakes of the material fall into the silage. Theoretically, when animals consume these contaminated chips of coating they could receive substantial doses of the Aroclor residue. Through continued use of the silo all of the coating material will eventually flake off of the silo walls. At this point, many dairymen are unaware that they have a PCB residue problem, as there is no visible coating left on the silo wall. However, organic acids from

MONS 086286

the silage dissolved PCB's which had penetrated into the concrete. These residues subsequently diffused into the silage. Silos last coated with PCB compounds 16 years ago still caused significant contamination of the silage long after traces of the coating had disappeared (Willett 1973 a).

Reported levels of Aroclor 1254 from scrapings of contaminated silos have a range between 100 to greater than 20,000 ppm (Skrentny 1971, Willett 1974). Aroclor 1254 in silage adjacent to these contaminated surfaces has ranged from less than one to greater than 7,000 ppm. Most frequently these residues are detected at levels between ten and 60 ppm.

Although the PCT's are extracted by the same procedure used for Aroclor 1254, PCT's are not eluted at the maximum temperatures of the gas liquid chromatography (GLC) column used for routine analyses of Aroclor 1254 (Fries and Marrow 1972). Fries and Marrow (1972) were able to detect PCT residues in silage and silo wall scrapings; however, methodology problems prevented their quantitation. The relative GLC peak areas for standard Aroclor 1254 and those extracted from silages were essentially the same (Fries 1972 b, Skrentny *et al.* 1971, Willett 1974). These results indicated that microorganisms and/or juices in the silage do not selectively alter or extract the Aroclor components (Fries 1972 b). *Figures 1 A and B show typical chromatograms of Aroclor 1254* standard and silage extract (Willett 1973 c).

IV. Cows and PCB residues

When cows were fed PCB's orally or consumed PCB-contaminated silage, residues were detected in the milk within six hours (Willett 1973 c). When constant levels of PCB's were continually fed to cows the concentration in milk fat increased rapidly for the first five to ten days. The concentration in milk fat continued to increase and approached a steady state in 40 to 60 days (Fries 1972 a). Platonow *et al.* (1971) showed that the PCB's fed to cows were deposited in the fat portion of the milk. They also showed a strong predilection of the PCB residues for the lipid phase of various products produced from milk.

Upon GLC analysis, PCB residues found in milk after cows had consumed Aroclor 1254 were quite different in appearance from the standard Aroclor 1254 (Fig. 1 C). Fries *et al.* (1973) observed that the peaks with shorter retention times did not occur in the milk samples, whereas the later peaks were more predominant. Based on the work of Simons and Welti (1971), Fries *et al.* (1973) determined that the major components of the PCB residues which occur in milk were the pentachlorobiphenyls and the hexachlorobiphenyls. Those components of the standard which did not appear in the milk as residues were theorized to be tetrachlorobiphenyls and some of the pentachlorobiphenyls.

Fries *et al.* (1973) estimated that cows that were continuously fed Aroclor 1254 excreted about 21% of the daily intake in the milk. However, these data were obtained when cows were fed 200 mg of Aroclor

MONS 086287

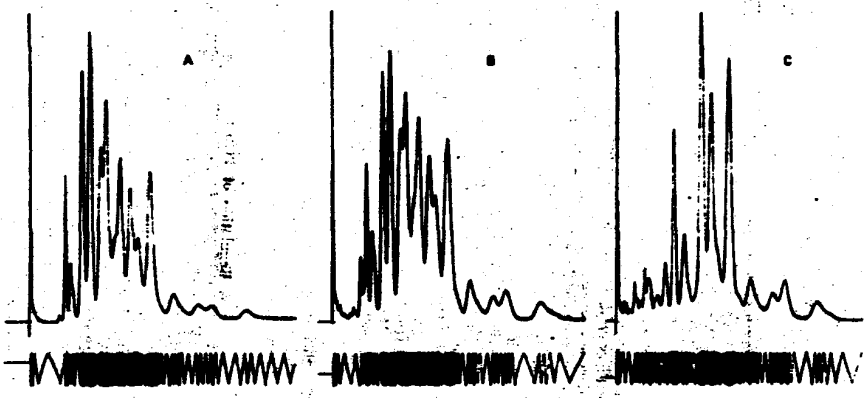

Fig. 1. Typical gle chromatograms: *A* = commercial grade Aroclor 1254 standard, *B* = Aroclor 1254 extracted from silage, and *C* = the Aroclor 1254 residue extracted from the milk of cows. These chromatograms were from an F & M model 402 with a ^{63}Ni detector. A 2.44-m glass column of 4% DC-200 and 6% QF-1 (1:1) on Gas Chrom Q (80/100 mesh) with argon-methane (95:5) as carrier and purge. Operating conditions were: flash heater 230° C, column oven 210° C, detector 300° C, and gas flow 40 to 50 ml/min (Wellett 1973 c).

MONS 088288

1254/day for 60 days. This quantity of Aroclor was at least four times higher than would be expected from an average PCB-contaminated silo. Based on the results of Willett (1974), one can calculate that a more reasonable figure for daily intake of PCB's might be in the range of one to 20 mg/day. Currently, research is underway to determine if similar excretion rates occur when cows are fed the lower levels of PCB's (Willett 1973 c).

Upon withdrawal of PCB residues from dairy cattle, there was a rapid decline in concentration of the residues in milk fat (Fuhs *et al.* 1971, Fuhs 1972 a). Fuhs *et al.* (1973) demonstrated with cows fed Aroclor 1254 that the decline in concentration of PCB's in milk fat is adequately described by a two-component, first-order system equation. The first or fast component in this system was about 15 days in length and the residue concentration declined about 50%. The second component or slow-rate of decline was much slower, representing a daily decline of 1%. This slow-component remained linear throughout the duration of the aforementioned experiments. The rates of decline for Aroclor 1254 concentrations perform very similarly to those of DDE which is the most persistent degradation product of DDT when fed to cows (Fuhs 1972 a, Fuhs *et al.* 1972 and 1973). In contrast, DDD and *p,p'*-DDT and *o,p'*-DDT were transferred to body fat and milk fat at much lower rates than DDE and PCB (Fuhs 1972). Activated charcoal (Fuhs *et al.* 1970, Wilson *et al.* 1968) and certain barbiturates (Fuhs *et al.* 1969) have been used to influence the rate of excretion of various organochlorine compounds. However, the use of activated carbon and phenobarbital were ineffective in reducing the final concentration of PCB's in milk fat (Fuhs 1972 a). As yet there is no known treatment which can increase the rate of excretion of PCB residues from lactating dairy cows.

V. Regulation of PCB's

At least ten Federal laws are potentially relevant for regulation of PCB's (Davies 1972). The Federal Food, Drug, and Cosmetic Act (21 U.S.C. 301 *et seq.*) is applicable for regulating the movement in commerce of dairy and livestock products containing PCB residues from farm silos, and action has been taken by the *U. S. Food and Drug Administration* (FDA) under its rule-making authority provided in this Act.

The FDA in 1969 first established an "internal guideline" or "working level" of 5.2 ppm in whole milk and declared that whole milk with PCB residues exceeding that level would be excluded from commerce (Kolarz 1972). Regulations pursuant to the Food, Drug, and Cosmetic Act [21 U.S.C. 342(a), 346, 348, 371] were published on July 6, 1973 (*Department of Health, Education, and Welfare* 1973) and became effective after September 4, 1973, amending 21 Code of Federal Regulations Parts 3, 128, and 135 and adding new Part 128.

These regulations are intended to limit "the sources by which PCB's

MONS 086289

1254/day for 60 days. This quantity of Aroclor was at least four times higher than would be expected from an average PCB-contaminated silo. Based on the results of Willett (1974), one can calculate that a more reasonable figure for daily intake of PCB's might be in the range of one to 20 mg/day. Currently, research is underway to determine if similar excretion rates occur when cows are fed the lower levels of PCB's (Willett 1973 c).

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MONS 086289

1941 and farm families generally consume the milk produced on their farm. There have not been reports of serious illnesses among midwestern and mideastern United States farm families such as those documented by the Japanese Yusho incident (KURATSUNE 1972, KURATSUNE *et al.* 1972).

The greatest hazard from a silo that has residues of PCB's is a financial one to the dairyman involved. If the milk market is determined to contain PCB's by a regulatory agency, the producer suffers a loss as the product must be impounded and destroyed. The producer must also replace the feedstuff stored in the contaminated silo. These losses may well exceed $14,000 in a single year (WILLETT 1973). FRIES (1973) found that when cows were producing unmarketable milk due to PCB residues, the PCB concentrations in their body were also high. Therefore, a dairyman cannot minimize his losses by selling contaminated cows and replacing them until sufficient time has elapsed for decontamination. An additional cost is that of decontaminating or replacing the silo.

WILLETT and CONRAD (1973) and WILLETT (1974) have demonstrated that silos with PCB residues can be decontaminated and returned to use by carefully cleaning and recoating the silo surfaces with a protective barrier coating. Whereas these barrier coatings have not been 100% effective, the small quantities of residue that can be detected in the silage have not been sufficient to raise the concentration of PCB's in milk of animals fed this silage to the actionable level prescribed by the FDA. On silos that are in good repair the procedure of carefully cleaning the silo surfaces and recoating proved to be a practical approach as the cost was only ten to 18% of the cost of replacing the silo. WILLETT (1973 b) has outlined recommendations for procedures to be followed before recoating silos and has described the important properties of coating materials if they are to be effective barriers. Some of the properties of a good barrier coating include adequate adherence to the concrete surface, acid resistance, temperature resistance, durability, waterproof characteristics, and no content of organochlorine compounds or toxic chemicals which can contaminate silage. Other considerations for coating selection include environmental quality and Occupational Safety and Health Act (*Department of Labor* 1972) standards for the protection of the environment and workers who may be applying or handling the materials. With careful management dairymen should be able to get many additional years of service from a silo that was once contaminated with PCB's through careful use of a barrier coating and routine maintenance.

The number of silos contributing PCB residues to human food is declining. Unfortunately, the rate of decline is unknown. Several factors are responsible for fewer of these problem silos. Companies previously formulating and using Cumar discontinued the use of this coating as soon as silos were identified as sources of PCB residues in milk. Similarly, *Monsanto Company* will no longer sell Aroclors for use in surface coatings. Dairymen have been advised by regulatory agencies, the *U. S. Department of Agriculture*, the Agricultural Extension Service, and field repre-

MONS 086291

tentatives from cooperatives to have their silage analyzed for residues if they suspect a PCB problem. As a result, many of the contaminated silos have been dismantled, others have been abandoned, and many have been effectively decontaminated with a barrier coating. Efforts to locate and eliminate contaminated structures are continuing. Such activities should reduce further the number of farm silos responsible for the entry of PCB residues into the human food chain.

Summary

Polychlorinated biphenyls (PCB's) have been applied commercially to a wide range of products for over 40 years. Since PCB's are chemically inert they resist degradation and are, therefore, found throughout the global ecosystem and can accumulate up the food chain. Many of the PCB's which are widely spread in our environment have little importance when considering the accumulation of PCB's in man. The direct contamination of food products with PCB residues is the main contamination source for human beings. One use of PCB's that led to the contamination of dairy products was a coating used on the inside of farm silos.

The coating responsible for most, if not all, of the PCB-contaminated silos was called Cumar and was used in the eastern half of the United States between 1941 and 1970. The PCB (Aroclor 1254) that was a component of this coating was dissolved by organic acids produced during the ensiling process. The PCB residues then diffused into the silage. Detected residue concentrations in silage adjacent to silo walls have been greater than 7,000 ppm.

When PCB-contaminated silage was fed to livestock the residues accumulated in the body fat and deposited in the fat portion of milk. When the source of PCB's was withdrawn from dairy cattle the concentration in milk fat declined rapidly and can be described by a two-compartment first-order system equation.

It has been estimated that 5,000 to 6,000 silos have been treated with coatings containing PCB's. However, the number of contaminated silos is declining as they are being dismantled, torn down, or decontaminated with barrier coatings. Surveys have indicated that PCB residues could be detected in milk from about 7% of the farms investigated. Many of the detectable residues were below the FDA temporary tolerance of 2.5 ppm on a fat basis.

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MONS 086294

Subject Index

Aatram and Aatrex, see Atrazine
Abate, automated analyses 87
——— LC 51
Absorption of vapors 117 ff.
Adsorption of vapors 115 ff.
Aerosols, centrifuging 114
——— electrostatic precipitation 112
——— filtration 106
——— impingement 107 ff.
——— sampling 105 ff.
——— sedimentation 112
——— thermal precipitation 114
Afalon, see Linuron
Air samples, analytical determination 127
——— sampling, alterations of constituents 101
——— sampling, analytical limitations 101
——— sampling, collection limitations 100
——— sampling, combination systems 125
——— sampling-detection systems 121
——— sampling duration 48
——— sampling, effectiveness 100
——— sampling, experimental design 94 ff.
——— sampling, freezeout 121
——— sampling, grab samplers 121
——— sampling, metering devices 103
——— sampling, Nylon nets 123
——— sampling, packed absorption columns 119 ff.
——— sampling, paper strips 124
——— sampling, pesticides in (see also specific compounds) 91 ff.
——— sampling, quantity measurement devices 101
——— sampling, rates and rate measurement devices 97, 104
——— sampling, samplers vs. detectors 102
——— sampling, sequential samplers 100–102
——— sampling, size of sample 96
——— sampling trains 125
——— sampling, units of measure 97
——— sampling, vacuum sources 102
Aldrin in air 107, 119, 120, 124, 125
Alfalfa 51
Alvit, see Dieldrin
Ammonia analyzers and automated analyses 19, 65
Amosone, see 2,4-D
Anderson samplers 110, 111
Anilines, automated analyses 11
Anofex, see DDT
Anti-ChE analyzers 3
Antimony, automated analyses 30
Aphoxide, automated analyses 28
Aphoxide, see Tepa
Apples 13, 26
Aroclors, see PCB
Aromatic amines, automated analyses 11
Arsenic acid, automated analyses 38
Arsenic analyzers 30
——— automated analyses 30
Asparagus 26
Ativan, see Lorazepam
Atrazine, automated analyses 63, 64, 66
——— in soil, automated sample preparation 38
Automated ammonia analyzers 19
——— analyses, advantages 2, 69
——— analyses, future 69 ff.
——— analytical systems (see also specific compounds) 19 ff.
——— analyzers, recognition 71 ff.
——— anti-ChE analyzers 3
——— arsenic analyzers 30
——— azinphosmethyl analyzers 19
——— aziridine analyzers 22
——— biphenyl analyzer 25
——— carbaryl analyzer 25
——— carbon disulfide analyzer 26
——— cyanide analyzers 30
——— data handling 57 ff.
——— diazotization and coupling analyzers 11
——— diquat analyzer 28
——— dithiocarbamate analyzer 27
——— element analyzers 30 ff.
——— gas chromatography 15, 40, 43 ff.
——— IR detection 56
——— lead analyzers 32
——— liquid chromatography 47 ff.
——— malathion analyzer 26
——— mercury analyzers 34
——— naphthol analyzer 25
——— nicotine analyzer 28
——— organobromine analyzers 17
——— organochlorine analyzers 13
——— organonitrogen analyzers 17
——— organophosphorus analyzers 15
——— organosulfur analyzers 18
——— paraquat analyzer 28
——— pesticide analyzers (see also specific compounds) 1 ff.
——— phenols analyzer 30
——— polarography 56
——— sample preparation 38 ff.

MONS 086295