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POLYCHLORINATED BIPHENYL CONTAMINATED SILOS: ARE THEY A HAZARD?

L.B. Willett

Polychlorinated biphenyls (PCBs) are commercially used mixtures that have been the subject of unprecedented public interest, regulatory action, and research during the past 10 years. Since these compounds have been utilized commercially for more than 40 years, they have become widespread in our ecosystem and frequently turn up as undesirable contaminants of livestock and ultimately human food. Since these lipophilic compounds are persistent, accumulative, toxic, and identified as potential carcinogens, there have been considerable efforts to eliminate them from our food chain. As part of this effort to prevent human exposure to PCBs, extensive monitoring programs have been implemented and strict tolerances imposed.

One source of PCBs in the human food chain has been certain concrete stave farm silos that were treated with a coating material that was called Cumar. This coating was formulated in 1941 for the Concrete Silo Company of Bloomfield, Indiana, to protect the inside concrete surfaces of the silo from the erosive actions of the organic acids which are produced during fermentation of silage. Cumar was also used as a sealant to retard the rate of curing of mortar used to seal the joints between staves.

Cumar received broad distribution across the Eastern one-half of the United States as a result of various mergers of silo companies. Figure 1 indicates those states known to have PCB-contaminated silos as identified by regulatory surveys or silo company records. Principally, the Concrete Silo Company (with production facilities in Bloomfield, Indiana, and Monteagle, Tennessee) and Michigan Silo Company (with facilities at Charlotte, Michigan; Massillon, Ohio; White Marsh, Maryland; and Peoria, Illinois) utilized and distributed this coating. Thus, the greatest concentration of Cumar-treated silos appears to be in the Upper Midwest and generally in the vicinity of silo manufacturing plants. The

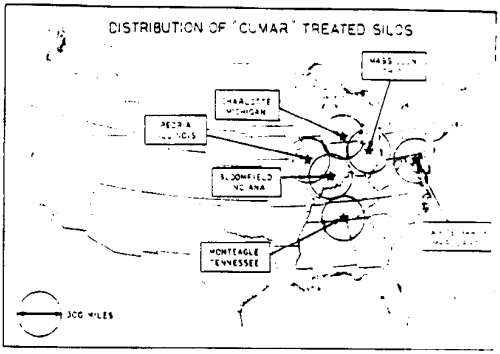

Figure 1. Shaded states have been identified by silo company or regulatory agency having Cumar-treated silos. The greatest concentration of these silos should be within 150 miles of the Michigan-Concrete Silo Company, plants.

shaded areas on Figure 1 represent areas within a 150-mile radius from various Michigan or Concrete Silo Company facilities.

The reported formulation for Cumar (Willett and Hess, 1975) indicated that the mixture contained approximately 19% Aroclor 1254, a polychlorinated biphenyl mixture, and 5.4% Aroclor 5460, a mixture of polychlorinated terphenyls. Once this mixture was applied to the silo wall, and the carrier solvents evaporated, the cured coating contained approximately 33% PCB. Typically, 100 ft.^{2} of silo wall were covered per gallon. Thus, a silo with a diameter of 16 feet and height of 50 feet would require 25 gallons of material, if completely coated, and 35 pounds of PCBs would be deposited on the walls.

Although silos had been treated with Cumar since 1941, they were not identified as a source of PCB in milk until 1970 when the Ohio Department of Agriculture recognized that residues in cows' milk, previously thought to be metabolites of DDT, were in fact PCBs. Silos were subsequently identified as the source of residue, despite distinct differences in gas-liquid-chromatogram peak profiles of PCBs found in milk when compared to profiles in silage or pure Aroclor 1254 standards (Willett and Hess, 1975). Apparently, congeners of the mixtures of PCBs are selectively absorbed or excreted by cattle while the residues that migrate into silage maintained a composition nearly identical to the original formulation. Originally it was thought that contamination of cows occurred when chips

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of the coating entered the feed and were consumed. Subsequently studies revealed that the PCBs were soluble in the organic acids, including lactic and acetic, that were produced during the fermentation process. Once dissolved in these acids, the residues diffused toward the center and leached toward the bottom of the silo (Willett, 1974). Traces of PCB have been detected in the center of the silo; however, the majority of the residues were found in the silage within one foot of the silo wall.

The rate at which the PCB content of silos declines is probably very dependent on use and management practices on individual farms. Apparently, the most important losses of PCB content occur as the Cumar coating begins to disintegrate and flake off the silo wall. However, sufficient PCB can remain in the silo to cause objectionable residues in milk and tissues of animals that consume feed from the structure for more than 20 years after all traces of the coating have peeled away.

The foremost problem created by PCB-contaminated silos during the early 1970's was the economic losses incurred by livestock producers when milk or animals could not be marketed when the PCB content exceeded the "internal guidelines" established by the U.S. Food and Drug Administration. The first of the regulations was established in the fall of 1969, which excluded the sale of milk containing more than 5 ppm of PCB in fat. Tolerances were to be lowered twice in subsequent years. In September 1973, a "temporary tolerance" of 2.5 ppm was established and the current "tolerance" of 1.5 ppm was established in August 1979.

Studies conducted by the U.S. Department of Agriculture (Fries, 1972) and the Ohio Agricultural Research and Development Center (Willett and Liu, 1982) showed that polychlorinated biphenyls, like other halogenated hydrocarbon pesticides and industrial chemicals, have a strong predilection for fat, therefore accumulating in fatty tissues, and are found associated with milk fat. When consumed, they are absorbed from the gastrointestinal tract, carried by the blood, and stored in the adipose tissue. When lactating cows are actually consuming feed which contains PCB, approximately 20% of the amount consumed is excreted in the milk. Some of the PCBs are passed directly though the G.I. tract and are excreted in the feces. Very little PCB finds its way into urine. When the PCB residues are withdrawn from the diet, concentrations in milk fat will decline following first-order kinetics. The initial decline in milk fat concentration will be about half in the first 15 days. Thereafter, the half-life of the decline in PCB concentration will be 6 to 8 weeks. Thus, if cows were consuming feed that contained PCB, and the concentration in the milk fat was 12 ppm, and if all PCBs were removed from the diet, it would require approximately 22 weeks for the residues to decline to the 1.5 ppm tolerance. Keep in mind that lactation is one of the more efficient means for animals to excrete PCB. Therefore, nonlactating animals will excrete PCB much more slowly. Some estimates have suggested that the half-life of PCBs in nonlactating animals is more than one year.

"When lactating cows are actually consuming feed which contains PCB, approximately 20% of the amount consumed is excreted in the milk."

The amounts of PCB routinely consumed and accumulated by cattle, when fed from a Cumar-treated silo, are not sufficient to result in a toxic syndrome. In fact, toxicity was not induced when cattle were experimentally fed much higher doses of PCB during distribution and clearance studies (Fries, 1972; Willett and Liu, 1982; L.B. Willett, unpublished studies).

Aside from concern for contaminated products and financial losses, there were questions of the potential hazards to consumers. It was apparent there were two distinct consumer groups: the general public; and farm families that ate and drank meat and milk products that they produced.

In 1973, we conducted a survey of all bulk milk received by a single processor in order to assess the amount of PCB that might be consumed by persons who purchased milk regularly from one source (Willett, 1980). This milkshed was in the immediate vicinity of one of the Michigan Brand silo manufacturing plants. The results of this study indicated that approximately 5% of the bulk shipments contained some PCB but the concentration in the whole milk supply was only 0.005 ppm. It was estimated that a consumer of 1 kg of milk per day would consume 5 ng daily or 1.8 mg of PCBs per year. This was considerably below the FDA suggested maximum intake.

This milkshed was again sampled for PCB one year later, after efforts to get dairymen to voluntarily abandon or decontaminate their silos. The concentrations of PCB in the milk supply had decreased suggesting the program was having a successful impact.

Obviously, exposure of farm families to a silo contaminated with PCB would likely be much greater than that of the general public which may obtain food from more diverse sources. Currently, the Michigan Department of Public Health has a cohort study underway which includes farm families that have PCB-contaminated silos. Fortunately, PCBs do not appear to be uniquely toxic to humans (Drill et al, 1982) and the symptoms of industrial exposure to high concentrations of PCB have not been generally reported among the farm families.

"One successful approach to eliminate further objectionable contamination of food-producing animals with PCB from this source was developed at the Ohio Agricultural Research and Development Center."

Initially, it was recommended that these contaminated structures be abandoned or torn down. Although this was the most positive way to assure that food would not continue to be contaminated, destruction of silos was not popular with the livestock industry and feasible alternatives were sought. One successful approach to eliminate further objectionable contamination of food-producing animals with PCB from this source was developed at the Ohio Agricultural Research and Development Center. The procedure required thorough cleaning of the silo wall and application of an approved barrier coating.

When properly recoated and subsequently managed, silos will not result in actionable residues in food or feed products. However, silos which were

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not properly cleaned prior to recoating are destined to again be a residue problem. Cleaning procedures that cut away all loose surface materials (high pressure water, 500-5000 psi, and sand blasting have been the most successful) will remove most of the PCBs from the silo wall plus will leave a sound surface to which barriers can bond. Without a sound surface, failure of some coatings has occurred within one year where many years without loss of integrity can be expected when the job is done properly.

Unfortunately, during the middle 1970s many silos were recoated by "fly-by-night" operators who did not use the recommended procedures and/or coatings. As a result, some livestock producers were left with the false security that their PCB problems were over. In recent years some low concentrations of PCBs have been detected in milk supplies which had been relatively free of the contaminant. Seldom have these concentrations approached or exceeded the federal tolerances of 1.5 ppm in fat.

If silos are not currently responsible for significant contamination of human food, then what is the problem?

The Congress of the United States passed a "landmark" piece of legislation in 1979. Called the "Toxic Substances Control Act," this was an effort to protect consumers from various chemical hazards in food, the workplace, and the environment. In that legislation, PCBs were specifically regulated and some tolerances established. One section of this law requires that materials which contain more than 50 ppm of PCBs be disposed of in a regulated hazardous waste site approved for PCBs. When scrapings are taken from the surface of silos that were once treated with Cumar, the PCB concentration generally exceeds the 50 ppm limit. For example, a survey of 12,097 concrete stave silos in Michigan revealed 298 that were contaminated between 0.7 ppm and 140,000 ppm of PCB.

It is important to remember that scrapings represent only the concentration of PCBs directly on the inner surface of the silo and not in the entire structure, unless the PCBs had penetrated deeply into the concrete. Without significant penetration into the silo, the concentration of PCBs, based on the weight of the entire silo, would be less than 50 ppm for most silos.

In order to determine if the polychlorinated biphenyls from Cumar had penetrated into the silo wall, staves from silos in Ohio and Michigan that were known to be contaminated with PCBs were sectioned. The concentrations of PCBs at various depths were determined. The results revealed that insignificant amounts of PCB actually penetrated into the stave, with nearly all the PCBs on or within 10 mm of the inner silo wall surface. Therefore, scrapings can be used to estimate the amount of PCB in the entire silo using the following equation:

$$Ce = Cs \cdot Ds/Dw$$

where:

C_e = concentration in entire silo
C_s = concentration in scrapings
D_s = depth of scrapings
D_w = thickness of silo wall

In some of the Michigan Brand silos only the joint mortar was coated with Cumar, thus less PCB is present. If the scrapings were collected from the joint mortar of one of these silos, the concentration estimate for the entire silo can be adjusted for the portion not coated by multiplying by 0.23.

"It is now apparent that most of the polychlorinated biphenyls that entered human food from farm silos did so before we ever knew them as an environmental contaminant."

Figure 2 shows the relationship between concentrations of PCBs in scrapings versus PCB concentration of the entire silo when scrapings of various thicknesses are taken. This figure

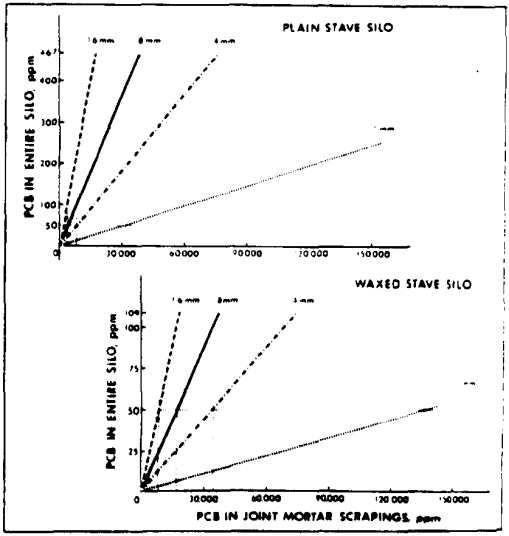

Figure 2. The relationship between the concentration of PCBs in scrapings from the joint mortar of silos and the PCB concentration in the whole silo. The top graph represents plain stave silos which were completely coated while the bottom graph is for waxed stave silos which only had the Cumar applied to the joint mortar. If scraping thickness was 0.8 mm, the concentration in the entire silo would be less than 50 ppm as long as the PCB concentration in scrapings was less than 4,000 ppm in plain steve silos and less than 17,000 ppm in waxed steve silos.

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represents both silos that were completely coated (plain stave silo) and those generally coated only on the joint mortar (waxed stave silo). Scrapings collected by personnel from our laboratory generally are between 0.1 and 1.5 mm thick. We have on occasion been sent samples of scrapings which were much thicker, particularly when entire sections of joint mortar were peeled away.

Based on these estimates, the majority of PCB contaminated silos would not exceed 50 ppm in the total structure and thus would not require disposal in a hazardous waste facility. This is fortunate because such disposal would be tremendously expensive and would occupy a great deal of valuable space that could be utilized for more hazardous materials.

Presently, there are no uniform regulations defining the ultimate fate of silos which have been coated with Cumar. In some areas they may be allowed to remain in use as long as no residue problems result, while in others they may need to be completely abandoned. I'm sure that new policies will be forthcoming within the next few years.

Cumar-treated silos are now at least 13 years old, since the last reported uses of this coating were in 1970. Some date back to the early 1950s and late 1940s and have reached the end of their usefulness. It is time for these obsolete structures to be removed. Unless these older structures were recoated with Cumar, it is highly unlikely that they would exceed the federal 50 ppm regulation. Prior to destruction of the structures it would be advisable to determine if state or local regulations would regulate reuse of structural materials, on-farm burial, or landfill disposal. Since these staves contain only small amounts of PCBs, which are bound to the surface, they seem to represent only a minimal future hazard once removed from direct contact with humans, food, feed or livestock.

Demolition of silos on or near active livestock operations will require special care as dust and debris from the structure can represent a significant source of PCB contamination to livestock. Unlike the situation depicted in Figure 3, feeding, watering, and housing areas must be protected and the exercise lots thoroughly cleaned before animals are given access.

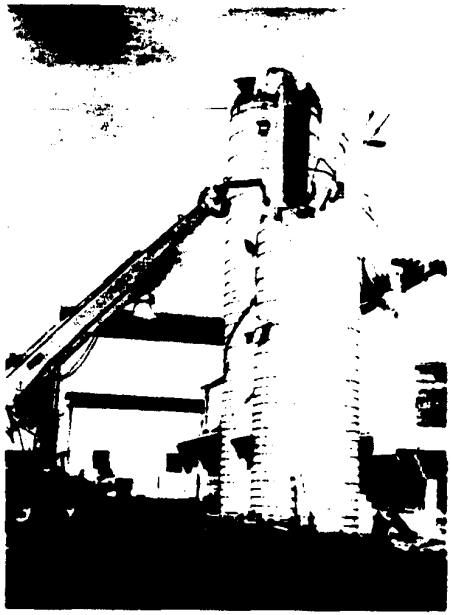

Figure 3. Care must be taken to prevent contamination of livestock, feed, water, housing and exercise areas with PCB-laden dust when silos are being demolished.

The results of an extensive survey of silos in Michigan have revealed that a few silos do exist that exceed the 50 ppm limit. Since the PCBs are maintained on the inner surface, available cleaning procedures (such as high pressure water at 5,000 psi) can remove sufficient amounts of PCB so that only the small amount of material removed would require special handling, and the remaining silo could be disposed of on site or in accordance with state or local regulations.

Soon after the initial discovery of PCBs in silos, two potential hazards were identified. One was concern for human health, including the general public and farm families; the second hazard was financial loss resulting from embargoed food products and replacement or loss of the silo. It is now apparent that most of the polychlorinated biphenyls that entered human food from farm silos did so before we ever knew them as an environmental contaminant. (This certainly was before PCB became a household word that has become apparently synonymous with toxic and carcinogenic.) After the residues of PCB were identified in silos great strides were made to warn farm families that PCBs were present and to eliminate the silo as a source of PCB in human food. These efforts have generally been successful. Residues are only infrequently discovered and rarely

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do they exceed tolerances. Obviously, the PCB from these silos does not pose a health threat to the general public.

The threat of a financial hazard still lingers. There are still, spread over at least 18 states, between 1,000 and 6,000 aging structures that were once treated with a relatively small quantity of PCB (Willett and Hess, 1975). The PCB content has declined through the years via erosion and extraction by silage juices. Many silos have undergone cleaning and recoating while others have simply been abandoned. However, sufficient residue may remain to result in objectionable residues in food if they were put back in service, thus representing a hazard to an unwary purchaser of a farm. Fortunately, most of these structures have less than the 50 ppm of PCBs that would require special hazardous waste disposal.

The current financial hazard to owners with contaminated structures remains unclear, and hinges on whether regulatory agencies will require demolition of silos, and if special and expensive procedures will be required. ■

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