ENVIRONMENTAL ISSUES REGARDING PENTACHLOROPHENOL

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INTRODUCTION

The focus of my talk today is on the environmental issues relating to pentachlorophenol. We cannot, however, intelligently discuss the environmental issues regarding pentachlorophenol (penta) unless we first outline its various uses. Penta's main use, as we are well aware, is as a wood preservative. In fact, it has been used as a wood preservative since its introduction into the market in the 1930's. It is still used extensively in a wide variety of agricultural and industrial applications as a fungicide, bactericide, herbicide, molluscicide, algicide, insecticide, and slimicide (1). Although the level of use has declined somewhat in recent years, worldwide production is estimated to be about 50,000 metric tons (110,231,130 lbs.) (2). Penta's environmental issues today have been created by its past uses and misuses for both industrial and agricultural applications. I will list these issues and then expand upon them later:

The First Issue Is:	The misuse of penta; using it in applications for which it was not intended.
The Second Issue Is:	The omnipresence of penta residues in the environment and the source of those residues.
The Third Issue Is:	The neutral impurities contained in commercial technical penta sold today.
The Fourth Issue Is:	Penta's long-term (chronic) and short-term (acute) toxicity to animals.
The Fifth Issue Is:	The fate of penta and its neutral impurities in treated wood.
The Sixth Issue Is:	The proper disposal of penta wastes produced by various manufacturing processes.

First Issue: The misuse of penta; using it in applications for which it was not intended.

The use of penta within log homes has created an issue in the United States which needs some explanation. Prefabricated houses and, in particular, log homes have become more popular because of the lower cost of their construction. The log home industry had a problem, however, with sapstain's occurring on the green logs before construction of the log home. The logs could be sitting in storage or on site prior to construction for several months before the actual building of the log home would commence, thus making it necessary to treat the logs with an antisapstain agent after fabrication of the logs. Usually a 0.5% solution of penta was sprayed onto the log or the log was dipped in the solution. Consequently, after the log was incorporated into the log house, approximately half of the log had an interior exposure and half had an exterior exposure. The part of the log which formed the interior walls exposed the residents of the those log homes to penta vapors. Technically, this is a misapplication of penta for penta is not recommended for interior use where vapors can concentrate. To this day, however, nobody has proven any ill health effects from those exposures (3). Recently, the penta industry has tested and recommended certain coatings which can be applied on the wood in order to minimize further interior exposures of penta (4).

The dairy herd incident which became an issue in Michigan in 1977 resulted from another misapplicaiton of penta. Penta residues, ranging from the low ppb level to well over 1 ppm in concentration, were found in the blood of some dairy cows in 12 herds throughout Michigan. Due to the fact that the presence of penta was established in the cow, many dairy health problems were thought to be caused by penta and its contaminants. As a result, the Michigan Department of Agriculture placed an emmergency ban on the sale of penta until an investigation could be completed. I might add that this was during the time when Michigan was still having its

Polybrominated Biphenyl (PBB) problem with other dairy herds in the area. The results of the investigations by the veterinarian experts indicated that penta residues most likely entered the cow via penta treated wood used to construct feed troughs, silos, and bunker silos. It appears that the penta would slowly migrate into the food which eventually was eaten by the cow.

After a series of hearings were held in Michigan during which results of dairy herd studies were presented, it was the conclusion of the hearing judge that neither penta nor its contaminants were the cause of the dairy herd health problems. The health problems of the cows were due to a variety of common problems including poor dairy management. Neither penta residues nor its neutral impurities were found in the cow's milk. As a result, the ban was rescinded in 1978. It was futhermore concluded, however, that penta treated wood as well as any other chemically preserved wood should not be used in the construction of feed containers for animals, unless a barrier were included in the construction to prevent direct contact with the food (5).

Another misuse of penta can result from the fact that penta can be absorbed through the skin when it is dissolved in an organic solvent. Therefore, proper protective clothing and equipment should be worn in these use applications. Five cases of penta toxicosis resulting from neglect of the above precautions were reported by Bergner *et al.* in 1965 (6). Workers at a wood treating plant in Winnipeg, Canada were dipping wood with bare hands in a vat of 4% penta dissolved in a petroleum solvent. One of the workers died from an overexposure to the penta/solvent system. This is clear misuse of penta. At the present time, all nonpressure wood treating operations in the United States dip wood mechanically.

A last example of a misuse of penta occurred in 1969, in St. Louis (7). Two infant deaths occurred in a hospital where the sodium salt of penta was conventionally used to control mildewing of diapers, but an accident occurred where it was used at 10 times the permissible concentration. The liver is the major detoxifying organ in the body. The infants' underdeveloped livers were overburdened and, therefore, were unable to detoxify the penta. Penta or its sodium salt is no longer used as a diaper mildew control agent.

In my opinion, the misuse of penta has been one of the major causes of heightened public concern and increased regulatory restrictions on pentachlorophenol. More education of the users of penta is needed in order to help prevent these problems in the future.

Second Issue: The omnipresence of penta residues in the environment and the source of those residues.

A. Residues of penta are found in the atmosphere

One of the major concerns about penta residues in the environment is how do they get there? Aerial transportation seems to be one significant cause. Despite a seemingly low vapor pressure, penta is quite volatile. It evaporates from the surface of water and from treated wood in microgram (μg) amounts (8, 9, 10). Snow pack samples collected during the winter of 1977-78 from 19 locations in Ontario were analyzed for the presence of toxic substances, including penta (11). Trace amounts of penta, i.e., ($<$0.001 μg/l to 0.03 μg/l) 1 to 3 ppt of snow melt, were detected in samples from those 19 sites. Those sites which were positive for penta came from several national parks and two provincial parks which are located in the Hudson Bay watershed. It was concluded that the presence of penta in the snow demonstrated that aerial transport of penta occurs year round. Also, penta vapors have been detected in log homes ranging from 0.2 to 0.38 $\mu g/m^3$ (12).

Wood treating plants using penta have an obvious potential exposure to air-borne penta particulates as well as vapors. In one plant up to 1.7 $\mu g/m^3$of penta has been detected (13). In the U.S.A. the OSHA allowable threshold limit value (TLV) for penta is 500 $\mu g/m^3$ of penta for workers exposed to penta in the working environment. This value is approximately 300 times higher than the value I have previously discussed. No ill health effects have been reported from any of the above instances of atmospheric exposures.

B. Penta residues are also detected in water

Penta in ppm concentrations (25 - 150 ppm) has been detected in raw effluent from a series of wood treatment plants (14). Also, domestic and industrial sewage as well as some river waters throughout the United States and Canada contained ppm concentrations of penta (15). In 1977, 85 water samples from stream mouths, nearshore areas adjacent to stream mouths and interconnecting rivers and channels on the Canadian shores of the Great Lakes were analyzed for penta. Levels of penta ranging from $<$5 ppt to 1.4 ppb were observed. Only 8 sites produced samples with no detectable pentachlorophenol. The source of this penta remains uncertain. Thirteen sewage effluent samples from 7 treatment plants in southern Ontario were also analyzed for penta which was observed in all samples, ranging from 65 ppt to 1.3 ppb (16). Penta ranging from 100 ppt to 700 ppt has been detected in domestic well-water in Northern California as well as in Oregon drinking water (15, 17).

C. Penta residues are found in food

Penta was one of a number of pesticides routinely monitored in raw food by the USFDA market basket survey. In 1973 - 1974, 10 out of 360 composite food samples contained penta at 10 to 30 ppb. Penta residues were also found in chicken and fish at low levels (18).

D. Furthermore, there are penta residues in humans

Penta has been widely detected in human urine, as well as in blood and fat. Low levels (mean value 6.3 ppb) were detected in 85% of urine samples taken from the general U.S. population. Based on human feeding studies, it has been determined that the half-life of penta (i.e., the length of time 1/2 the penta concentration is excreted) in the plasma of exposed individuals was 33 hours or 1.3 days. Eighty-six percent (86%) of an administered oral dose of penta was excreted in the urine (19). Consequently, penta is excreted relatively rapidly by humans. In general, penta's neutral impurities have not been detected in humans.

E. Alternate sources of penta residues in the environment

Residues of penta and associated chlorophenols do not necessarily arise only from the production or uses of pentachlorophenol. A number of other chemicals, including some common pesticides, are metabolized to penta by plants, animals, and microorganisms. In particular, metabolism of γ-BHC orLindane® has been shown to form penta in rats (20), humans (21), and higher plants (22). Also, hexachlorobenzene (HCB) is converted to penta in rodents, chickens, fish, monkeys, and other organisms (23). HCB is a major contaminant of all chlorinated solvents sold today. Moreover, background residues of HCB and γ-BHC can be found everywhere in the environment. The extent, however, of these contributions to penta's environmental residues remains under study.

It has been reported that chlorination of a 1 ppm aqueous solution of phenol produced detectable penta and other lower chlorinated phenols (24). The suggestion that penta might be naturally generated remains unsupported, although several closely related compounds, including 1,4-dimethoxy and 1-hydroxy - 4-methoxy - 2,3,5,6-tetrachlorobenzene, and 2,3,5,6-tetrachloro-anisole reportedly are naturally biosynthesised by micro-organisms (25, 26 and 27).

The higher chlorinated dioxins and dibenzofurans present as residues in the environment do not seem to be naturally occurring, although traces of these compounds have been reported as pyrolysis products from many common sources, e.g., municipal incinerators and other combustion processes (28).

While there is no denying the fact that penta can be detected in a majority of environmental samples, there are two questions which need to be asked and answered in order to gain a clearer perspective of the present information:

1. The first is the accuracy of the determinations for penta. When concentrations of penta are reported in the ppm, ppb, and ppt ranges, verification needs to made. At these very low levels of detection, verification by using known standards as well as mass spectrometry is

necessary. Also, analytical interferences can easily occur and the use of different analytical procedures may affect the outcome of the results (29, 30). Therefore, verification must become a part of the routine analytical procedure. Just stop and think for a moment what these very low concentrations mean.

1 ppm = 1 drop of vermouth/80 fifths of gin
1 ppb = 1 drop of vermouth/80,000 fifths of gin
1 ppt = 1 drop of vermouth/80,000,000 fifths of gin

These are pretty dry martinis by anybody's standards!

2. Second, the detection of penta concentrations of ppm or less raises issues of penta's chronic toxicity, i.e., whether or not long-term health effects occur. Penta is not a carcinogen (31, 32, 33). No long-term health effects due to low levels of penta exposures have ever been reported.

Third Issue: The neutral impurities contained in commercial technical penta sold today.

Current manufacturing practices in the production of technical penta also produce additonal chlorinated compounds in ppm concentrations which are known as neutral impurities. Presently, technical penta is manufactured by the direct chlorination of phenol in the U.S. and Canada. Typically, the penta content is approximately 86% to 92% and the tetrachlorophenol content is approximately 4% to 7% (34). Modern analytical methods show that most penta samples contain contaminants called chlorinated dibenzo-p-dioxins and chlorinated dibenzofurans in concentrations totalling approximately 1,500 ppm (34). The chlorodioxin usually present in the highest concentration is the comparatively nontoxic octachlorodibenzo-p-dioxin (OCDD). Analysts have not detected any 2,3,7,8-tetrachlorodibenzo-p-dioxin (the most toxic dioxin) in penta, and, indeed, none has been found in penta made in the U.S.A. or Canada (35). Two isomers of heptachlorodibenzo-p-dioxin and three of the ten possible isomers of hexachlorodibenzo-p-dioxin have also been detected in penta. One isomer of hexachlorodibenzofuran and one isomer of heptachlorodibenzofuran were also detected. Much smaller amounts of other chlorinated compounds were reported, e.g, polychlorinated diphenyl ethers and polychlorinated phenoxy phenols (29, 36). It is extremely difficult to prevent the formation of these neutral impurities in technical commercial penta. This is due to the fact that the same chemical laws apply to the formation of the neutral impurities as they do to the formation of penta during its manufacture (37).

Technical tetrachlorophenol (tetra) which contains 20% penta and 80% tetra is gaining more of a market share for wood treating applications, mainly in the antistain uses and it may be useful to discuss the neutral impurity concentrations in tetra as well. Generally speaking, the chlorodibenzo-p-dioxin and chlorodibenzofuran concentrations are about one order of magnitude or less in total concentration when compared to technical penta (38). Those concentrations exist in tetra sold in the solid form and not in the final antistain solutions. The chlorodibenzo-p-dioxin concentrations in typical antistain solutions sold in the U.S.A. are approximately 2 ppm total, whereas the hexachlorobenzo-p-dioxin concentrations are in the range of 20 ppb (39).

Fourth Issue: Penta's long-term (chronic) and short-term (acute) toxicity to animals.

Penta has an acute toxicity which is well defined; however, its chronic toxicity, including that due to the presence of neutral impurities, is still being studied. Penta is used because it is toxic to micro-organisms (i.e., as a bactericide and fungicide), lower and higher plants (i.e., as an algicide and herbicide) and invertebrate and vertebrate animals (i.e., as an insecticide and molluscicide). It is not surprising to note that it is also toxic to man. Penta, if one ingests enough, can kill man. It is a simple dose response relationship. Penta if dissolved in a solvent is absorbed by and corrosive to skin and causes irritation; it is highly irritating to the nose and throat. In mammals, acute exposure (i.e., short-term) leads to elevated body temperature, increased respiratory rate, elevated blood pressure, hyperglycemia, and cardiovascular distress (40).

Long-term dietary exposure of rats to purified penta produced, at the highest dose (30 $\mu g/m^3$), only mild biochemical and physical effects, which did not alter life span or tumor incidence (41). Penta was not teratogenic or mutagenic in the standard tests but was embryotoxic (42, 43).

In contrast, several studies comparing technical grade penta with purified grade of penta showed the former to produce greater liver damage and altered biochemistries consistent with the presence of dibenzo-p-dioxins and dibenzofurans (44, 45). Some, but not all, of these neutral impurities are toxic. The toxic symptoms are delayed; degeneration occurs in liver, thymus, and often in skin, and a variety of clinical chemistry changes are observed which are different symptoms from those produced by pure penta. Penta contains hexachloro-, heptachloro-, and octachlorodibenzo-p-dioxins. Heptachlorodibenzo-p-dioxin and octachlorodibenzo-p-dioxin make up more than 99% of the total chlorodioxin content in penta. The hexachlorodibenzo-p-dioxin (HxCDD) constituents are generally thought to be the toxic chlorodioxin components in penta. A mixture of the two HxCDD isomers contained in technical penta are oral carcinogens as was recently determined by the National Cancer Institute (46).

Despite its obvious toxicity and the occasional reports of accidental acute intoxication, there has been remarkably little evidence of human damage due to long-term exposure (47). The U.S. wood treating industry claims a good health and safety record, and a six-year epidemiological study concluded that no long-term health effects could be observed in occupationally exposed workers (48). Presently, this six-year study is in the process of being extended to approximately 15 years. The results should be available next year. The total cost of this study approaches 1/3 million dollars.

Fifth Issue: The fate of penta and its neutral impurities in treated wood.

A concern exists today regarding whether or not penta and its neutral impurities remain in the treated wood. The treatment of wood with penta dissolved in light petroleum or liquefied petroleum gas causes deep penetration into the cells of the wood. While some of the compound inevitably "bleeds" out and may vaporize or be washed away, with time the biological activity decreases and extractable penta declines (49). This decrease is gradual (greater than 25 years) depending primarily upon environmental conditions.

The concentrations of neutral penta impurities forced into the wood may remain relatively constant throughout the impregnated depth, but penta migrating to the surface may be converted to OCDD, HpCDD, and HxCDD by sunlight. These concentrations, however, remain very low due to the fact that the chlorodioxins also degrade in the presence of sunlight (50). The environmental and health significance of these occurrences are thought to be negligible.

Sixth Issue: Proper disposal of penta wastes produced by various manufacturing processes.

The disposal of hazardous wastes has become a regulatory concern today. Penta wastes are classified as hazardous and, therefore, need special attention from the industry. Penta is biodegradable; for example, 96% is removed by an activated sludge process (51). When sludge containing penta from commercial wood treating operations was composted in permeable soil with penta concentrations of 200 ppm or less, at least 98% of that penta was destroyed in 205 days (36). The use of ozone or other oxidants in the presence of UV light has proved to be effective in degrading both penta and its neutral impurities (52). High temperature combustion at 800C is also effective (53). Other common methods of waste disposal, such as open pits, landfill sites, on-site burning, deep-sea burial, or deep-well injection are not recommended because of mobility and toxicity of penta.

In conclusion, while there actually is little evidence that low level environmental exposures to penta present an imminent hazard to man or animals, a history of actute intoxication associated with the use of technical penta, often by untrained people, reflects a clear hazard. On the other hand, penta has long proven its usefulness as an inexpensive and effective preservative and pesticide, especially in the developing nations. No truly satisfactory alternatives to penta have been found and improved safety will depend on better formulation, user education, and restriction of certain low-priority, high-contact uses, such as, indoor uses, laundry products, and cosmetics. For the future, the chemical mechanism by which the neutral impurities are synthesized must be better elucidated. If this is achieved, neutral impurity reduction in technical penta can be achieved.

Very few chemicals have been studied as much as penta. The scientific literature on penta has expanded rapidly, e.g., some 200 chemical abstracts citations appeared in 1978 alone—and several valuable reviews already exist (54, 55). The more we study penta, the more we will know how to use it safely. There is no doubt there are existing environmental problems with penta, however, all of these are manageable. One main challenge to the wood treating industry and the wood treating chemical producers is the education of the users of penta in how to handle various penta solutions and penta treated products safely.

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