5. POTENTIAL FOR HUMAN EXPOSURE

5.1 OVERVIEW

Chlorpyrifos enters the environment as the result of its use as a broad spectrum insecticide/acaricide for treatment of crops, lawns, ornamental plants, domestic animals, and a variety of building structures. Unintentional releases to the environment include improper indoor application, redeposition of air residues, spills, and the disposal of chlorpyrifos wastes. Indoor use by unlicensed or untrained applicators has occasionally resulted in excessive human exposure. EPA (1997) reported that most of the more serious chlorpyrifos poisonings appear to involve either the misuse or inappropriate use of the pesticide by pest control operators.

The important physical and chemical characteristics which influence the fate and transport of chlorpyrifos in the environment are its low solubility, volatility, and strong affinity for colloidal matter. Abiotic hydrolysis, photodegradation, and biodegradation are all important processes for the transformation and degradation of chlorpyrifos. Chlorpyrifos bioconcentrates to only a limited extent, and has little mobility in most soils. Chlorpyrifos exists in the atmosphere primarily in the vapor phase, but can partition to particulates. Chlorpyrifos is not persistent in water, due to volatilization and strong adsorption to particulate matter.

Indoor air, food, and soil are the environmental media with the highest degree of chlorpyrifos contamination; ambient air, groundwater, and surface water have lesser degrees of contamination. Although a large amount of chlorpyrifos is used in various environments (see Chapter 4), levels of general exposure are mediated by its limited mobility and persistence, and by environmental degradation processes.

Several subpopulations are at higher risk of exposure: workers in industries that manufacture and formulate chlorpyrifos, those who apply the insecticide, and farm workers who enter treated fields after the insecticide has been applied. Among the general population, people who use the insecticide in homes and gardens and people who ingest food exposed to chlorpyrifos are at higher risks of exposure.

5. POTENTIAL FOR HUMAN EXPOSURE

Chlorpyrifos has been identified in at least 7 of the 1,428 current or former EPA National Priorities List (NPL) hazardous waste sites (HazDat 1996). However, the number of sites evaluated for chlorpyrifos is not known. The frequency of these sites within the United States can be seen in Figure 5- 1.
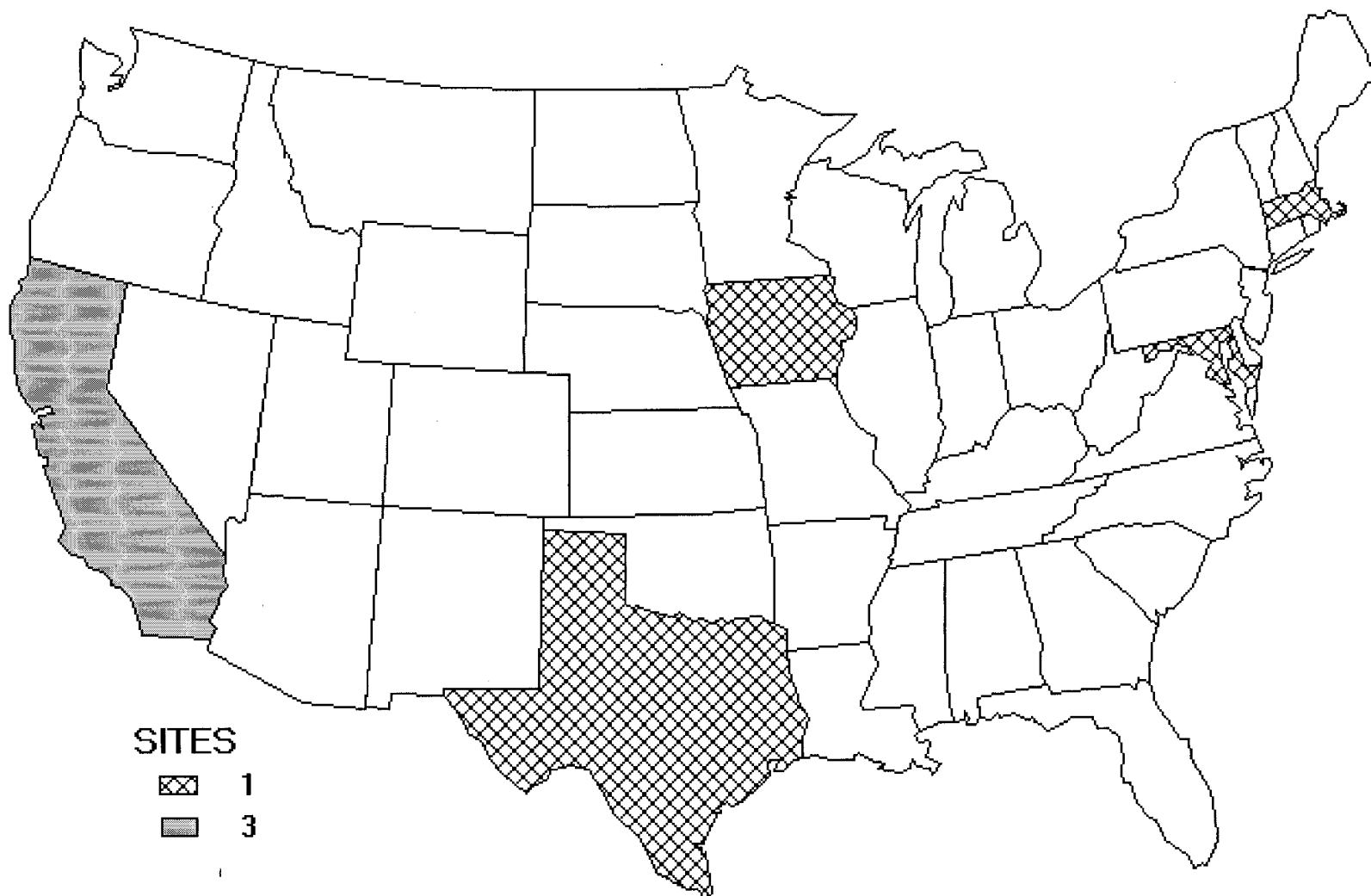
5.2 RELEASES TO THE ENVIRONMENT

5.2.1 Air

Chlorpyrifos enters the atmosphere as a result of its use as an insecticide/acaricide. Chlorpyrifos is released to the atmosphere by volatilization during foliage or soil application by ground or air broadcast equipment (Racke 1993). Air emissions from chlorpyrifos production have been reported to be 0.5 kg per 1,000 kg (one metric ton) produced (Sittig 1980). The Toxics Release Inventory in 1992 did not require reporting of chlorpyrifos releases to air (EPA 1993c). No information was found on detections of chlorpyrifos in air at NPL hazardous waste sites (HazDat 199).

5.2.2 Water

Chlorpyrifos is released to water during foliage or soil application as an insecticide/acaricide by ground or air broadcast equipment and during subsequent runoff or leaching (Racke 1993). Leaching and runoff from treated fields, pesticide disposal pits, or hazardous waste sites may inadvertently contaminate both groundwater and surface water with chlorpyrifos. Entry into water can also occur from accidental spills, redeposition of atmospheric chlorpyrifos, and discharge of waste water from chlorpyrifos manufacturing, formulation, and packaging facilities (HSDB 1996; Racke 1993). In the past, chlorpyrifos was aerially applied to water over swamps for mosquito abatement; however, it is no longer registered for this use. No other uses are known which result in direct application to water (EPA 1986). The Toxics Release Inventory in 1992 did not require reporting of chlorpyrifos releases to water (EPA 1993c). There is also a potential for release of chlorpyrifos to water from hazardous waste sites. Chlorpyrifos has been detected in surface water samples collected at 4 of the 7 NPL sites and in groundwater samples collected at 1 of the 7 NPL sites where chlorpyrifos has been detected in some environmental medium (HazDat 1996). The HazDat information used includes data from NPL sites only.

Figure 5-1. Frequency of NPL Sites with Chlorpyrifos Contamination

SITES

▨ 1

▩ 3

Derived from HazDat 1996

5. POTENTIAL FOR HUMAN EXPOSURE

5.2.3 Soil

Chlorpyrifos is released in agricultural, home, and garden soil during direct soil or foliar treatment, and from disposal of chlorpyrifos-containing wastes in hazardous waste sites (HSDB 1994). Much of the chlorpyrifos (or its metabolites) applied to foliage eventually reaches soil (Racke 1993). Soil in waste disposal sites may include manufacturing wastes containing chlorpyrifos. A primary method for disposing of liquid pesticide wastes has been the dumping of liquid materials into soil evaporation pits, ditches, and ponds. Topsoil from such discharge areas is expected to be contaminated with pesticides; the soil from one discharge pit contained chlorpyrifos at concentrations of 1,012-3,193 mg/L in the top 7.5 cm (Winterlin et al. 1989). Soil from tail water pits used for collecting irrigation runoff may also be a source of chlorpyrifos if the soil is treated with this insecticide (Kadoum and Mock 1978). Chlorpyrifos may also enter soil by redeposition of atmospheric chlorpyrifos (Racke 1992). Entry may also occur from spills during storage, transport, or equipment loading and cleaning, although the sophistication of contemporary management practices limits this amount. The Toxics Release Inventory in 1992 did not require reporting of chlorpyrifos releases to soils (EPA 1993c). Chlorpyrifos also can be released to soils and sediments from hazardous waste sites. Chlorpyrifos has been detected in soil samples collected at 3 of the 7 NPL sites and in sediment samples collected at 1 of the 7 NPL sites where chlorpyrifos has been detected in some environmental medium (HazDat 1996). The HazDat information used includes data from NPL sites only.

5.3 ENVIRONMENTAL FATE

5.3.1 Transport and Partitioning

The vapor pressure of chlorpyrifos is 1.9×10^{-5} mm Hg at 25 °C (2.5×10^{-8} atm) (Racke 1993). This suggests that while chlorpyrifos is in the atmosphere, it will exist primarily in the vapor phase but will also partition to available airborne particulate (Eisenreich et al. 1981). Experimental evidence during fog events (Glotfelty et al. 1990) supports this hypothesis. The removal rate by dry deposition is low for such compounds (Schroeder and Lane 1988); therefore, depending on its reactivity characteristics and the amount of available airborne particulate, chlorpyrifos may travel long distances in the air. The low solubility of chlorpyrifos at 1.12 mg/L at 24 °C (Felsot and Dahm 1979) indicates that dry deposition is a more important process than wet deposition.

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The transport of chlorpyrifos from water to air can occur due to volatilization. Compounds with a Henry's law constant (H) of $<10^{-5}$ atm-m^3/mol may volatilize slowly from water (Lyman et al. 1990). Therefore, chlorpyrifos, with an H value of 6.6×10^{-6} atm-m3/mol at 25 °C (Downey 1987) may volatilize slowly from water. The dimensionless Henry's law constant (H′) or air/water partition coefficient for chlorpyrifos, as calculated from vapor pressure and solubility data, has been reported to be 5×10^{-4} (Glotfelty et al. 1987), 7.3×10^{-4} (Suntio et al. 1987), and $1.7 \pm 0.3\times10^{-4}$ (Fendinger and Glotfelty 1990). Using these data, the estimated volatilization half-life from a river 1 meter deep flowing 1 m/set with a wind velocity of 3 m/sec is estimated to be 9 days (Lyman et al. 1982).

The amount of chlorpyrifos available to be volatilized from surface water is reduced by sediment adsorption. Chlorpyrifos has a strong affinity for soil colloids, as evidenced by its measured range of organic carbon-adjusted soil sorption coefficient (K_{oc}) of 973-31,000 (Felsot and Dahm 1979; Kenaga 1980; McCall et al. 1980; Racke 1993). This suggests that chlorpyrifos in natural water ecosystems adsorbs strongly to suspended solids and sediments, and that this process may transport considerable amounts of chlorpyrifos from water to particulate matter. Several studies have reported very low concentrations of chlorpyrifos in surface waters (see Section 5.4.2).

In macrophyte-dominated freshwater model ecosystems, *Elodea nutalli* vegetation adsorbed a large proportion of the dose of chlorpyrifos applied and hampered mixing of the insecticide in the water column (Brock et al. 1992). Only a relatively small proportion of the applied dose became incorporated in the sediment. In open-water model ecosystems, however, mixing was rapid and the sediment compartment, particularly its upper layer, was a sink for chlorpyrifos.

Aquatic bioconcentration factors (BCF) ranging from 1 to 5,100 for chlorpyrifos and metabolites have been determined extensively from laboratory and field studies (ASTER 1996; Cid Montafies et al. 1995; Macek et al. 1972; Mulla et al. 1973; Odenkirchen and Eisler 1988; Racke 1993). These studies suggest that chlorpyrifos bioconcentrates to varying degrees in different organisms, and with different doses and durations of exposure. It has been suggested that the BCF values determined-during short duration single-dose exposure studies may not be indicative of long-term exposure due to nonattainment of equilibrium conditions (Racke 1993). It has been observed that 5-9 days are necessary for steady-state conditions (Hedlund 1973; Welling and deVries 1992).

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The transport processes that may move chlorpyrifos from soil to other media are volatilization, leaching, runoff, and biotransfer by plants. Post-application volatilization of chlorpyrifos applied as an agricultural insecticide and subsequent atmospheric transport is thought to be a primary means by which chlorpyrifos is dispersed throughout the environment. Volatilization is affected by soil cultivation practices. Cumulative losses of chlorpyrifos by volatilization from no-till (NT) and from conventionally tilled (CT) plots were measured by Whang et al. (1993). The NT/CT flux ratio increased from a factor of about 3 on days 1 and 2 to a factor of 12 by day 26. Soil dryness did not often limit volatilization, and differences in soil moisture resulting from different tillage practices were not usually a major reason for differences between fluxes.

Volatilization rates, which result from the complex interplay between chlorpyrifos sorbed to soil, dissolved in the soil pore water, and present in the soil air spaces, can be quite variable. Chlorpyrifos (applied to the soil at 11 $\mu g/cm^2$) was captured from 3 moist soils (0.3 bar soil moisture tension, 25 °C) by blowing an airstream of 1 km/hour over the soils. The calculated flux rate ranged from 80-290 g/hectare/day during the first 3 days, with 62-89% of applied chlorpyrifos remaining after 36 hours (McCall et al. 1985). Racke et al. (1991) observed significantly less volatility over a longer exposure period with ranges of 3-39 g/hectare/day, and >90% of the applied chlorpyrifos remaining after 30 days. When applied as a foliar spray, chlorpyrifos volatilized from corn leaves rapidly. In the laboratory, 80% volatilized within 48 hours at 30 °C with a simulated wind speed of 0.8 km/hour (McCall et al. 1985). A field study confirmed the fairly rapid rate of volatilization, with an observed half-life of about 1.5 days on corn and soybean foliage (McCall et al. 1984).

Leaching studies have shown chlorpyrifos to have little mobility in soil. Laboratory leaching studies revealed that all the surface-applied residues of chlorpyrifos were confined to the upper 5 cm of several soils after elution with 20 cm of water (Harris et al. 1988; McCall et al. 1985). Field studies have confirmed this lack of mobility, with chlorpyrifos residues being confined to the upper 12 inches of soils in several trials (Fontaine and Teeter 1987; Oliver et al. 1987). The leaching and dissipation of the applied ^{14}C-chlorpyrifos in sandy soil under simulated field precipitation, drainage and temperature was less than 0.2% (Fermanich and Daniel 1991). Amounts of chlorpyrifos lethal to termites moved to a depth of at least 30 cm in decomposed granite soil from the Santa Ana River bed in Colton, California, after it was applied at 500 ppm to the top 7.62 cm of soil in a long column of 34 mm diameter and ≈130 mL of water was dripped through (Smith and Rust 1992).

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Studies indicate that runoff of chlorpyrifos is of minor environmental significance. In a study conducted in an Iowa cornfield, approximately 0.003% of 3 applications of chlorpyrifos was transported via runoff to a pond within the watershed (McCall et al. 1984). Information from irrigated environments (e.g., turf) indicates that because of the lack of erosion of soil particles, strongly sorbed chlorpyrifos is not transported via runoff (Watschke and Mumma 1989). Even during a simulated l00-year rainfall event (13.6 cm) occurring less than a week after application, only between 0.10 and 0.29% of the applied chlorpyrifos was present in runoff. In another study of runoff from turfgrass treated at 1.12 kg/hectare with irrigation applied at 150 mm/hour, no residue of chlorpyrifos was detected at 5 μg/L (minimum detection level) (Harrison et al. 1992). The movement of chlorpyrifos was studied from 1985 to 1987 in a small agricultural Saskatchewan watershed (Waite et al. 1992). In 1985-86, 3-4 million hectares of farmland were treated with insecticides at application rates as high as 1 kg/hectare to control grasshopper infestations. The frequency of occurrence and concentrations of chlorpyrifos in groundwater, surface water and runoff from spring snow melt were measured. No chlorpyrifos was found in any of the samples at detection limits of 1 ppb in 1985 and 0.1 ppb in 1986.

Spills are an important way that chlorpyrifos enters surface waters. A spill of chlorpyrifos into a marine bay resulted in initial water concentrations of up to 300 μg/L, but because of sediment sorption, dissipation, and dilution, the concentration had dropped to below detectable levels within 17 days (Cowgill et al. 1991).

Some research has shown that only very small levels of chlorpyrifos are taken up by plant roots, translocated, or metabolized by plant tissues (Kenaga et al. 1965; Smith et al. 1967). Cranberry bean plants were hydroponically grown in nutrient solutions containing 50 ppm of chlorpyrifos emulsifiable concentration. After 72 hours, only 0.07-0.1% of the radioactivity present, composed of TCP and other degradation products, had been translocated to the plant tops. In another experiment (Smith et al. 1967), one leaf of the cranberry bean plant was treated foliarly with 1 mg of chlorpyrifos. After 7 days, <1% of the chlorpyrifos applied was found in nontreated areas of the plant.

However, other researchers have found that soil-applied doses of chlorpyrifos are transported to foliage (Rouchaud et al. 1991). Cauliflower and brussels sprouts were treated with chlorpyrifos by pouring it onto soil around the stem of the plant for protection against the root fly. During plant growth, chlorpyrifos and its soil metabolites were transported from soil into the plant foliage, where it could give a secondary plant protection against the foliage insects. The foliage concentrations of the

nonsystemic chlorpyrifos was ≥1 mg/kg fresh weight during a period of about 44 days after soil treatment in brussels sprouts crops and a period of 3.5 days in cauliflower crops.

5.3.2 Transformation and Degradation

Chlorpyrifos undergoes a number of different transformation and degradation reactions in the environment as discussed in the following sections. The resulting environmental transformation products are shown in Figure 5-2.
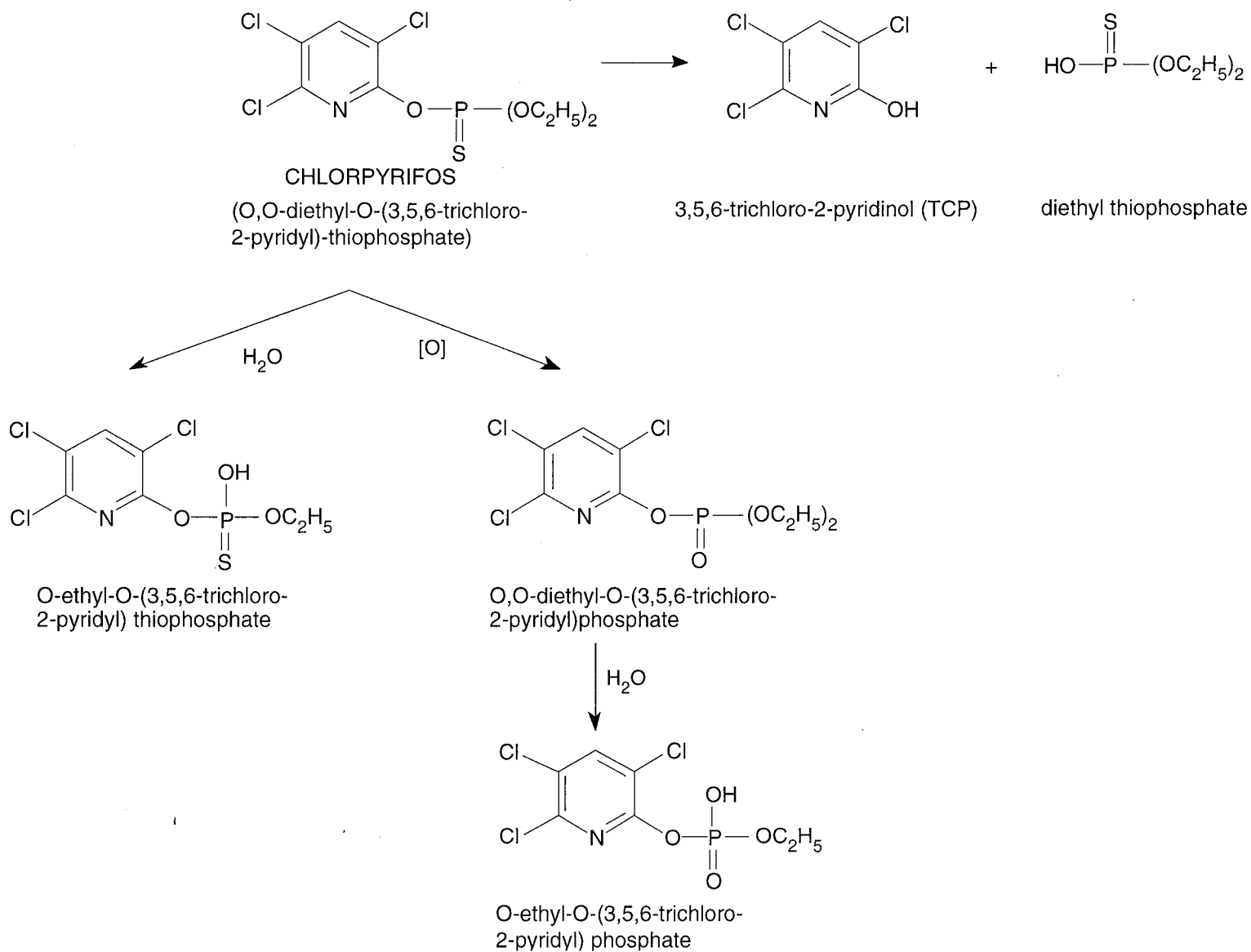
5.3.2. 1 Air

Both chlorpyrifos and its degradation product, TCP, have ultraviolet (UV) absorbencies above 295 nm, indicating their susceptibility to photodegradation by sunlight. The photodegradation half-life of chlorpyrifos in the laboratory is approximately 2.6 days (Fontaine and Teeter 1987). While in the atmosphere, chlorpyrifos will react with photochemically induced hydroxyl radicals. Its estimated halflife is 6.34 hours (Atkinson 1987).

5.3.2.2 Water

The processes primarily responsible for the transformation and degradation of chlorpyrifos in water are abiotic hydrolysis and photosensitized oxidation. Neutral hydrolysis is favored below pH 9, whereas alkaline hydrolysis dominates above pH 9 (Macalady and Wolfe 1983). Thus, both the disappearance half-life and the products are pH-dependent. Neutral hydrolysis yields O-ethyl-O-3,5,6-trichloro-2-pyridyl phosphorothioate, while alkaline hydrolysis occurs by base-catalyzed cleavage at the phosphate ester linkage to produce TCP and phosphorthioic acid. Neutral hydrolysis is pseudo-first-order kinetics, while alkaline hydrolysis is second-order kinetics (Wolfe 1988). Keeping the temperature at 25 °C, the half-life of chlorpyrifos in distilled water was 89.14 days at pH 1, and 0.01 days at pH 12.9 (Macalady and Wolfe 1983). At 20 °C, it has a half-life of 120 days at pH 6.1 and 53 days at pH 7.4 (Freed et al. 1979). The activation energy for the hydrolysis of chlorpyrifos at pH 7.4 is 14 kcal/mol, indicating its sensitivity to temperature change. Laboratory studies on the interaction of chlorpyrifos with Cu^{2+} have demonstrated metal-catalyzed hydrolysis and have provided rate constants for this pathway (Blanchet and St. George 1982).

Figure 5-2. Environmental Degradation Pathways of Chlorpyrifos

CHLORPYRIFOS
(O,O-diethyl-O-(3,5,6-trichloro-
2-pyridyl)-thiophosphate)

3,5,6-trichloro-2-pyridinol (TCP)

diethyl thiophosphate

O-ethyl-O-(3,5,6-trichloro-
2-pyridyl) thiophosphate

O,O-diethyl-O-(3,5,6-trichloro-
2-pyridyl)phosphate

O-ethyl-O-(3,5,6-trichloro-
2-pyridyl) phosphate

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Photodegradation in water is possible since chlorpyrifos absorbs in the UV region at >295 nm; however, its relative importance as a dissipative force in the environment is unclear. Laboratory studies from artificial light sources may not be very useful for predicting environmental photodegradation kinetics (Miller and Zepp 1983). For example, chlorpyrifos in natural waters is usually very strongly sorbed to suspended particulate and bottom sediment, and thus less readily available to photolytic forces than chlorpyrifos in clear distilled water in the laboratory.

Brock et al. (1992) performed experiments using macrophyte-dominated freshwater ecosystems and open-water model ecosystems. In both systems, 50% of the chlorpyrifos dose applied had disappeared on day 8 post-treatment. In the long run, loss of chlorpyrifos was more rapid in the macrophytedominated ecosystems than in the open-water ecosystems.

Under field conditions, chlorpyrifos exhibits very short persistence in the water compartment of aquatic ecosystems, and half-lives as short as several hours have been observed. This is due to its considerable volatility from water (arising from low solubility and moderate vapor pressure) and its high association with sediment. The rate of disappearance of chlorpyrifos from river and well waters in a pH range of 8-8.5 was studied in the laboratory at a range of temperatures and under conditions of light and dark (Frank et al. 1991). The half-life for the disappearance of chlorpyrifos was 4.8 days at 21 °C and 27 days at 4 °C, indicating that temperature plays a major role in the degradation of chlorpyrifos in water. The half-life for disappearance of chlorpyrifos was 56 days in the dark and 46 days in the light at 21 °C, indicating that sunlight photolysis is not a major route of chlorpyrifos degradation in water.

The persistence of chlorpyrifos in surface water was studied (Hughes et al. 1980) by application of 10 ppb chlorpyrifos to polyethylene-lined ponds and a single natural pond inoculated with leaf litter. In early post-treatment, there was rapid partitioning to adsorption on bottom sediments and polyethylene; 30-60% disappeared from the water within 24 hours. The time for the concentration of chlorpyrifos to decline to 0.01 ppb in the polyethylene-lined pond was estimated to be 40 to >200 days compared to 18 days for the natural pond. The desorption from sediments was considerably slower from organic matter than from polyethylene. Desorption from the polyethylene contributed to residual concentrations in the water of artificial ponds for up to 18 months. Similar results were noted in an artificial lake treated with chlorpyrifos: lake water concentrations peaked 1 day after treatment at 0.9 μg/L and leveled near 0.2 μg/L after 3 weeks (Mulla et al. 1973).

First-order degradation rate constants of chlorpyrifos were determined in estuarine water and sediment/water slurry systems (Walker et al. 1988). Half-lives of chlorpyrifos in sediment/slurry systems calculated from these rate constants ranged from 12 to 30 days for the non-sterile system, and 16-51 days for the sterile system. Half-lives for the seawater-only systems ranged from 13 to 41 days for the non-sterile systems and 3.5-24 days for the sterile systems. The half-life of chlorpyrifos in seawater was 24 days in a sediment-seawater slurry (Schimmel et al. 1983). These data indicate that abiotic processes predominate in estuarine systems.

5.3.2.3 Sediment and Soil

Chlorpyrifos may undergo degradation on the surface of soils by photo-induced reactions. Laboratory photodegradation of chlorpyrifos on soil surfaces with UV light (254 nm from mercury lamps) demonstrated that three different photochemical processes (hydrolysis, dechlorination, and oxidation) take place simultaneously (Walia et al. 1988). The oxidative and dehalogenated products formed during photo-irradiation of soil undergo further photolysis to form chloropyridinols and O,O-diethyl phosphorothioic acid. The oxon is unstable; it tends to hydrolyze more rapidly than chlorpyrifos and does not accumulate in the soil. With the passage of time, the percentage of chlorinated pyridinols also decreased, suggesting that these products are mineralized in the soil under UV-photo-irradiation conditions. Under simulated sunlight conditions, the rate of photodegradation of chlorpyrifos on a leaf surface was slow. Chlorpyrifos was stable up to 10 days; then the oxon (1.5%) and the hydrolytic product, TCP (2.5%), were detected. Dehalogenated analogs of chlorpyrifos could be detected only after 15 days of constant irradiation. Under these conditions, the photo-oxidation process was more predominant than the photohydrolytic or dehalogenation process. Formation of such photoproducts on an irradiated soil surface was very fast, but the rates in the laboratory will differ from those found under environmental conditions.

Chlorpyrifos undergoes transformation in soil by the processes of abiotic hydrolysis and microbial degradation. A few studies have attempted to separate abiotic chemical hydrolysis from-microbial processes and to determine their relative importance (Miles et al. 1979, 1983). The half-lives of chlorpyrifos in muck (48% organic matter [OM]) and loam (2.7% OM) were determined in sterilized and natural soils at 3 temperatures (3, 15, and 28 °C). The results indicate that in sterile soils, chlorpyrifos is progressively more degraded by abiotic hydrolysis as the temperature increases, and that it degrades faster in sandy loam than in muck (after 24 weeks, 38 versus 68% remaining at the

highest temperature). An explanation for the soil difference may lie in the pH. The sterile loam had a pH of 6.5, whereas the sterile muck had a pH of 5.9, indicating that increasing pH increases degradation. The degradation study of chlorpyrifos in natural soil gave the same progression for increasing temperature, and it continued to degrade faster in the loam than in the muck (half-lives of 16, 6, and 2.5 weeks versus >24, 15, and 6 weeks at the respective temperatures). All half-lives were shorter in the natural soils as opposed to the sterile soils, however, indicating microbial degradation in addition to abiotic chemical hydrolysis.

Some researchers have concluded that chlorpyrifos is not catabolized (Racke and Coats 1988, 1990) because it is resistant to enhanced degradation by microbes. When chlorpyrifos is applied to fields with a soil history of chlorpyrifos use, the breakdown of chlorpyrifos is not enhanced, and is often delayed (Racke and Coats 1988; Somasundaram et al. 1989). The biotic process at work is probably co-metabolism. Patterns of persistence were observed in a variety of agricultural soils after treatment with ^{14}C-chlorpyrifos and its hydrolysis product, TCP (Racke et al. 1988). In soils with no previous history of chlorpyrifos use, significant quantities of TCP and soil-bound residues were produced, but little $^{14}CO_2$. In soils with a history of chlorpyrifos use, neither TCP nor soil-bound residues accumulated, but large quantities of $^{14}CO_2$ were produced. Direct treatment of fresh samples of each of these soils with ^{14}C-TCP resulted in rapid mineralization of TCP to $^{14}CO_2$ only in those soils with a history of prior chlorpyrifos use. The rapid mineralization of TCP in these soils was microbially mediated. It is unclear if catabolic or co-metabolic processes are predominant (Racke and Robbins 1990) in the degradation of TCP. TCP exhibited sorption (K_d) coefficients of between 0.3 and 20.3 mL/g (mean of 3.1) and calculated mean Koc coefficients for the neutral and anionic forms of 3,344 and 54 mL/g, respectively.

In a study of persistence of chlorpyrifos in a silt loam soil, the disappearance rate was fast in the first 15 days, but slowed after that. The pseudo-first-order rate constants were 0.041 day^{-1} and 0.044 day^{-1}, for the band treatment at seeding, and 0.04 day^{-1} for the drench at seeding. The calculated half-lives ranged from 15.8 to 17.3 days (Szeto et al. 1988).

5.4 LEVELS MONITORED OR ESTIMATED IN THE ENVIRONMENT

Reliable evaluation of the potential for human exposure to chlorpyrifos depends, in part, on the reliability of supporting analytical data from environmental samples and biological specimens. In

reviewing data on chlorpyrifos levels monitored in the environment, it should also be noted that the amount of chemical identified analytically is not necessarily equivalent to the amount that is bioavailable.

5.4.1 Air

Chlorpyrifos has been detected in both outdoor and indoor air; of special concern are levels in fogwater and environments receiving broadcast pesticide application, and in selected indoor environments such as poorly ventilated and artificially lit environments and the infant breathing zone (25 cm above the carpet). Selected studies documenting chlorpyrifos concentration and persistence in these environments include Anderson and Hites (1988); Fenske et al. (1990); Jackson and Lewis (1981); Leidy et al. (1992); Lewis et al. (1988); Moye and Malagodi (1987); Vaccarro (1993); and Wright et al. (1991, 1994). Special issues in these environments are discussed below.

Substantially higher chlorpyrifos concentrations were measured in the infant breathing zone than in the adult breathing zone, implying a vertical gradient with the treated carpet serving as a source of volatilized chlorpyrifos (Fenske et al. 1990). All concentrations in the infant breathing zone exceeded the National Academy of Sciences interim guideline of 10 $\mu g/m^3$. This study also indicated that broadcast applications appear to produce average levels 5-10 times higher and peak levels l-2 orders of magnitude greater than other application procedures, with peak concentrations occurring 3-7 hours after application. Following treatment in the crawl spaces, significantly more chlorpyrifos was present in the air of houses built over sand than in the air of houses built over clay soils. However, no differences were found between rooms or construction types (slab, crawl, crawl-slab) (Wright et al.1988). The air of storage rooms in commercial pest control buildings was found to have a higher concentration (220 ng/m^3) of chlorpyrifos than office rooms (126 ng/m^3). The same study detected levels of chlorpyrifos from 20 to 1,488 ng/m^3 in the air of 6 food preparation serving areas following application of a 0.5% emulsion spray into cracks and crevices, although concentrations dropped considerably over 24 hours in all areas. Chlorpyrifos was detected in homes and pest control offices and vehicles, with residues ranging from 0.1 to 5 $\mu g/m^3$ (Leidy et al. 1992). Air concentrations in commercial pest control vehicles ranged from 9 to 221 ng/m^3 (Wright and Leidy 1980).

High fogwater concentrations (320-6,500 ng/L) were reported at Parlier, Corcoran, and Lodi, California, relative to air concentrations (0.6-14.7 ng/L), with enrichment factors of 160-260 (Plimmer

1992). Other researchers have found similar enrichment factors (Glotfelty et al. 1987, 1990). Enrichment was attributed to the effect of temperature correction, colloidal organic matter, and adsorption. The enrichment factor has also been correlated to hydrophobicity, as indicated by K_{ow} (Valsaraj et al. 1993).

There is less evidence of general contamination of ambient air, although residues have been detected. Ambient air monitoring at 10 U.S. locations in 1980 resulted in 14 detections from 123 samples, with a maximum of 100 ng/m^3 and an arithmetic mean of 2.1 ng/m^3 (Carey and Kutz 1985). This same study reported 2 detections of chlorpyrifos from 11 air samples in Pekin, Illinois, in 1980. Ambient air and wet-deposition monitoring of chlorpyrifos in California indicated that atmospheric transport is occurring from the Central Valley, where chlorpyrifos is used agronomically, to the Sierra Nevada Mountains; concentrations decrease with distance from the source area and elevation (Zabik and Seiber 1993). A maximum concentration of 6.5 ng/m^3 was recorded in the valley; the maximum value midslope was 0.083 ng/m^3. A loading rate of 0.8 μg/m^2 to Sierra National Park was calculated.

5.4.2 Water

Chlorpyrifos has been detected in groundwater and surface water, but only rarely, and generally well below levels of concern. Hallberg (1989) reported that chlorpyrifos was detected (concentrations unspecified) in 0.2% of 334 samples from groundwater used for public drinking water supply in Illinois, but was not detected in 15 Iowa samples. This same study detected chlorpyrifos in 45% of the wells in the vicinity of agrochemical dealers, and in 1.4% of farm water supply wells. In a survey of surface waters in southern Ontario from 1975-77, chlorpyrifos was detected in 3 of 949 samples from 11 agricultural watersheds (Braun and Frank 1980). Krill and Sonzogni (1986) reported no detections of chlorpyrifos in groundwater sampling of 358 wells in Wisconsin. In a study of 54 wells in California, Maddy et al. (1982) found no detectable levels of chlorpyrifos. Pionke et al. (1988) and Pionke and Glotfelty (1989) found no detectable levels of chlorpyrifos in a study of 21 wells and 2 springs (detection limit of 4 ng/L) in Pennsylvania. Maddy et al. (1982) found no detectable levels of chlorpyrifos in a study of 53 wells in California. In an intensive monitoring effort, Richards and Baker (1993) detected chlorpyrifos in 0-l .06% of 750 samples for each of 7 tributaries to Lake Erie from 1983 to 1991. A maximum chlorpyrifos concentration of 480 μg/kg in runoff from irrigated cropland in California was reported by Leonard (1990). Total seasonal losses as a percentage of application were 0.02-0.24, and were attributed to aerial application during irrigation. Chlorpyrifos

detections were not reported as part of the national surface water monitoring program for 1976-80 (Carey and Kutz 1985).

5.4.3 Sediment and Soil

Limited data on chlorpyrifos residues in soils or sediments were located. At a detection limit of 0.01 mg/kg, chlorpyrifos was not detected in sediment samples collected from Lakes Superior and Huron, including Georgian Bay, in 1974 (Gloschenko et al. 1976). Chlorpyrifos detections were not reported in sediments as part of the national surface water monitoring program for 1976-80 (Carey and Kutz 1985). Soil evaporation pits, ditches, and ponds have been used to dispose of liquid pesticide wastes in California (Winterlin et al. 1989). A core soil sample taken from one such pit in northern California contained detectable levels of chlorpyrifos to a depth of 67.5 cm (Winterlin et al.1989).

5.4.4 Other Environmental Media

The Food and Drug Administration (FDA) identified chlorpyrifos in four grain samples and in four samples of animal feed in 1975 (Duggan et al. 1983). The FDA's pesticide residue monitoring program for domestic and imported food commodities detected chlorpyrifos 33 times in 1,044 samples in unspecified foods at unspecified concentrations during fiscal years 1978-82 and 295 times from 3,744 samples during fiscal years 1982-86 (Yess et al. 1991a, 1991b). From October 1, 1981, to September 30, 1986, the FDA Los Angeles District Laboratory detected chlorpyrifos in 1,969 of 19,851 samples of domestic and imported food and feed commodities (Luke et al. 1988). Chlorpyrifos was detected in 440 of 4,916 samples analyzed as part of the FDA Total Diet Study between 1986 and 1991. As part of the FDA's Pesticide Monitoring Program for domestic and imported foods, chlorpyrifos residues have been detected during 1988-89, 1989-90, 1990-91, and 1991-92 (FDA 1990, 1991, 1992, 1993). Chlorpyrifos was detected in domestic feed, lavender, lettuce, cantaloupe, peanuts, bell peppers, summer squash, and cherry tomatoes; and in imported apples, green beans, cabbage, coriander, cucumbers, eggplant, feijoa, kiwi, green leaf lettuce, cantaloupe, honeydew, nectarine, Chinese peas, peaches, peppers, spinach, squash, tomatillos, and tomatoes (Hundley et al. 1988). In the FDA's Revised Market Basket Study (FDA 1995), ready-to-eat foods were analyzed for pesticides and industrial chemicals repetitively for 10 years (1982-91). During that period, 37 market baskets, each containing 234 food items, were collected. Chlorpyrifos was detected in 121 of the food

items a total of 718 times; the average concentration found was 0.0036 μg/g (ppm). Gartrell et al.(1986) found chlorpyrifos in meat, fish and poultry, grain and cereal products, garden fruits, oils and fats, and sugar. Chlorpyrifos was detected in 121 different domestic foods (0.9% of samples) in 1988 and 128 domestic foods (1% of samples) in 1989 by state regulatory monitoring (Minyard et al. 1991). In a study of processed foods imported into Hawaii from western Pacific rim countries, chlorpyrifos was detected in oriental-style noodle soup and roasted peas at concentrations of 4.7 ppb and 10.95 ppb, respectively (Gans et al. 1994). In a pesticide residue screening program conducted in 1989-91 in San Antonio, Texas, on 6,970 produce samples, chlorpyrifos was detected in 41 produce samples (lemons, oranges, peppers, turnips), with a detection limit of 0.25 ppm (Schattenburg and Hsu 1992). In a study of pesticide residue contamination of processed milk-based and soy-based infant formula, chlorpyrifos was not detected (Gelardi and Mountford 1993). However, in a study of pesticide residues in cornposited milk, chlorpyrifos was found in 23 of 806 composite samples (Trotter and Dickerson 1992).

The EPA Office of Water has recommended that chlorpyrifos residues be monitored by states in their fish and shellfish contaminant monitoring programs in watersheds where this pesticide has been or is currently used extensively in agriculture (EPA 1993c. While no fish or shellfish consumption advisories are currently in effect for chlorpyrifos, this contaminant has not been widely monitored in state fish contaminant monitoring programs or the U.S. Fish and Wildlife Service National Contaminant Biomonitoring Program (EPA 1993c). In a national study, EPA (1992a) did detect chlorpyrifos in fish in 26% of 362 sites, with mean and maximum concentrations of 4.09 ng/g and 344 ng/g, respectively.

5.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

The general population is exposed to chlorpyrifos primarily by inhaling indoor air and ingesting food containing chlorpyrifos, and through skin contact during or after pesticide application. Chlorpyrifos has been very infrequently detected in ambient air, and only at very low concentrations (see Section 5.4.1). It is not anticipated that the general population would experience substantial levels of exposure by inhaling ambient air. Chlorpyrifos has rarely been detected in drinking water (see Section 5.4.2), and consumption of chlorpyrifos-contaminated drinking water is not considered a significant exposure route for the general population.

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Chlorpyrifos has been detected in some foods (see Section 5.4.4), so ingestion may be a route of exposure for the general population. The FDA has estimated daily food intakes of chlorpyrifos for different age/sex groups in the United States. The FDA estimated the dietary intake of chlorpyrifos for a 14-16-year-old male in the United States to be 3.4 ng/kg body weight/day, which is much lower than the Food and Agricultural Organization of the United Nations/World Health Organization's (FAO/WHO) acceptable daily intake (ADI) of 10 μg/kg body weight/day and ATSDR's intermediate oral MRL and EPA's RfD of 3 μg/kg body weight/day (FDA 1992; IRIS 1994).

Other than during home and garden insecticide application, exposure of the general public to chlorpyrifos through skin contact is not anticipated.

The Non-Occupational Exposure Survey (NOES) conducted by NIOSH from 1981 to 1983 on the number of workers and the number of facilities where workers could be potentially exposed to chlorpyrifos in the United States estimated that 911 janitors and cleaners in meat packing plants, and bread, cake, and related product industries; 10,452 pest control workers; and 41 groundskeepers and gardeners in the medical industry were potentially exposed (NOES 1994). The American Conference of Governmental Industrial Hygienists (ACGIH) (1993-94) recommends that workplace air levels of chlorpyrifos not exceed 0.2 mg/m^3 as a time-weighted average (TWA) for an 8-hour workday, 40-hour workweek and not exceed a 0.6 mg/m^3 short-term exposure limit (STEL). The STEL is a 15-minute TWA exposure which should not be exceeded during a workday, even if the 8-hour TWA is within the threshold limit value (TLV)-TWA; also exposure should not be >15 minutes and should occur not more than 4 times per day.

Workers involved in the manufacture, formulation, handling, or application of chlorpyrifos, or those involved in the disposal of chlorpyrifos-contaminated wastes are likely to be exposed to higher concentrations by dermal contact and inhalation than the general population. Persons working with plants that have been previously treated with these compounds also can be exposed by absorption through the respiratory system or skin (Aprea et al. 1994). A study of pet handlers responsible for flea control in California in 1987 indicated that chlorpyrifos was associated with increased frequency of blurred vision, flushing of skin, and a decrease in urination (Ames et al. 1989). In a study of airborne and surface concentrations of chlorpyrifos after application in offices, Currie et al. (1990) found airborne concentrations peaked 4 hours after application at 27 μg/m^3, and surface residue concentrations peaked at 5.9 ng/cm^2 48 hours after application. Airborne levels were found to be

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lower in furnished offices than unfurnished offices. When granular chlorpyrifos at 0.75 active ingredient per acre was applied to a field by air, the estimated inhalation exposure to chlorpyrifos was 0.02 mg per 8-hour day for the pilot and 0.03 mg per 8-hour day for the ground staff (Myram and Forrest 1969). The estimated inhalation exposure to chlorpyrifos for workers using ground machines was 0.33 mg per 8-hour day (Myram and Forrest 1969).

Hodgson et al. (1986) reported symptoms of organophosphate intoxication among five office workers after chlorpyrifos treatment for termites. The duration of symptoms and erythrocyte cholinesterase levels over time suggested redistribution of the active ingredient after absorption to a second body compartment, with subsequent slow release into the bloodstream. Estimated potential dermal exposure (i.e., unprotected by clothing) of three greenhouse workers in Florida ranged from 17,500 to 24,000 μg/hour (Stamper et al. 1989), with highest exposure to applicators' legs. Tyvek[®] protective clothing afforded 89%, ±5% protection.

In a study of termiticide applicator exposure in eight North Carolina homes, exposures of 0.1-98 $\mu g/m^3$ were reported. Exposure levels were higher in houses constructed over a crawl-space (Wright et al. 1988). The NOES reported detectable levels of chlorpyrifos in indoor, outdoor, and personal air in Jacksonville, Florida, and in Springfield/Chicopee, Massachusetts (Whitmore et al.1994). Concentrations tended to be highest in summer, lower in spring, and lowest in winter. Indoor and personal air concentrations were generally higher than outdoor concentrations. Of 11 carpets sampled in the study, all had detectable levels of chlorpyrifos in carpet dust, (mean concentration of 5.8 μg/g), suggesting that infants and toddlers may be at higher risk of exposure. NAS/NRC (1982) recommends that air levels in houses not exceed 10 μg chlorpyrifos/m^3. Measurements of pesticides or their metabolites in human biological specimens, such as urine, are considered an appropriate way of approximating total pesticide exposure through all routes of entry into the body. As part of the National Health and Nutrition Examination Survey III (NHANES III), urine samples were collected from approximately 1,000 adults ranging in age from 20 to 59 years. These individuals represented a relatively board spectrum of the U. S. population, including individuals from both sexes-and different age groups, races/ethnicities, urban/rural residences, and regions of the country (Needham et al. 1995). Hill et al. (1995) examined the ranges of pesticide residues found in the urine of approximately 1,000 U.S. adults and found that 3,5,6-trichloro-2-pyridinol (TCP), considered a fairly specific metabolite and indicator of exposure to chlorpyrifos or chlorpyrifos-methyl, was present at detectable levels in 82% of the (993) individuals examined. Further, 31% of those subjects had urinary TCP concentrations of

5 μg/L or greater. This was consistent with the report of Bartele and Kastl (1992) that TCP was present in the pooled urine of unexposed control subjects at a concentration of 5 μg/L.

5.6 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

Workers in industries that manufacture and formulate chlorpyrifos and applicators of the insecticide are at higher risk than the general population for chlorpyrifos exposure. Farm workers who enter treated fields after insecticide application may also be exposed to chlorpyrifos at higher levels than the general population. Those who use the insecticide for homes and gardens are also at higher risk of exposure to chlorpyiifos. Although no investigative evidence from the hazardous waste sites was located, it is likely that people who live near hazardous waste sites containing chlorpyrifos wastes are at higher risk of exposure to chlorpyrifos.

5.7 ADEQUACY OF THE DATABASE

Section 104(i)(5) of CERCLA, as amended, directs the Administrator of ATSDR (in consultation with the Administrator of EPA and agencies and programs of the Public Health Service) to assess whether adequate information on the health effects of chlorpyrifos is available. Where adequate information is not available, ATSDR, in conjunction with the NTP, is required to assure the initiation of a program of research designed to determine the health effects (and techniques for developing methods to determine such health effects) of chlorpyrifos.

The following categories of possible data needs have been identified by a joint team of scientists from ATSDR, NTP, and EPA. They are defined as substance-specific informational needs that if met would reduce the uncertainties of human health assessment. This definition should not be interpreted to mean that all data needs discussed in this section must be filled. In the future, the identified data needs will be evaluated and prioritized, and a substance-specific research agenda will be proposed.

5.7.1 Identification of Data Needs

Physidal and Chemical Properties. As seen in Table 3-2, the relevant physical and chemical properties of chlorpyrifos are known (HSDB 1994; Sanbom et al. 1977), and it is possible to predict

the environmental fate and transport of chlorpyrifos based on K_{ow} K_{oc} and H. Therefore, further data acquisition and research are not recommended as a high-priority activity.

Production, Import/Export, Use, Release, and Disposal. Knowledge of production and use data for a chemical is important in predicting its potential for environmental contamination and human exposure. Since chlorpyrifos is produced by two manufacturers (SRI 1994), to maintain confidentiality, its recent production volume is not known. Similarly, data concerning the import and export volumes for chlorpyrifos in recent years have not been located. There is currently no federal requirement to report the use of chlorpyrifos. The most recent estimates of its yearly use in the United States were published in 1986 (Gianessi 1986). Therefore, more current estimates of use and projected trends are needed. No information in the available literature was located that indicates the use of chlorpyrifos in any consumer products other than edible crops and vegetables during and after their planting. Although some information regarding the disposal of wastes containing chlorpyrifos is available, more detailed and recent information would be helpful. The standards promulgated by the EPA for the disposal of wastes containing chlorpyrifos are available (Berlow and Cunningham 1989).

According to the Emergency Planning and Community Right-to-Know Act of 1986, 42 U.S.C. Section 11023, industries are required to submit chemical release and off-site transfer information to the EPA. The Toxics Release Inventory (TRI), which contains this information for 1992, became available in May of 1994. This database will be updated yearly and should provide a list of industrial facilities and emissions. However, no TRI data were located for chlorpyrifos because this chemical is not required to be reported. As with most pesticide agents, it is virtually impossible to make decent quantitative estimates of the amounts of chlorpyrifos produced, used, disposed, imported and exported. This presents some fundamental problems in making more than the most general sorts of risk assessments. Improved information for any of these categories is considered a major data need.

Environmental Fate. Information regarding the fate of chlorpyrifos in air was limited in the literature. Although the available data indicate that the concentration of chlorpyrifos in air will be low (Carey and Kutz 1985), more information would help predict the residence time and distance of its aerial transport. Knowledge about the fate of chlorpyrifos in water is also limited. Although it has been estimated that sorption onto particulates and settling into the sediment are important for chlorpyrifos in water, more information regarding the relative importance of sorption for removal of chlorpyrifos from water to sediment would be helpful. There is some evidence in the literature

regarding the mobility of chlorpyrifos in soil. Additional information on the degradation of chlorpyrifos in water and air and the fate of the degradation products in soil would be helpful.

Bioavailability from Environmental Media. Available information regarding the rate of chlorpyrifos absorption following inhalation, oral, or dermal contact has been discussed in Section 2.3, Toxicokinetics. Although no data on the bioavailability of chlorpyrifos from contaminated air are available, the bioavailability from inhalation exposure is expected to be high because chlorpyrifos is likely to be present in the vapor phase and not in the particulate phase in the adsorbed state. Similarly, no data on the bioavailability of chlorpyrifos from water, soil, or plant material are available; however, chlorpyrifos is adsorbed rather strongly to soil. Since the part that remains adsorbed to soil or sediments may be, at most, partially bioavailable, chlorpyrifos is expected to have reduced bioavailability from soil and water. Data on the bioavailability of chlorpyrifos from actual environmental media and the difference in bioavailability for different media need further development.

Food Chain Bioaccumulation. Measured BCF values for chlorpyrifos are available for a large number of aquatic invertebrate and fish species (Odenkirchen and Eisler 1988; Racke 1993). Research on accumulation of chlorpyrifos applied to soils in the roots, stems, and leaves of plants has also been undertaken (Rouchaud et al. 1991).

Exposure Levels in Environmental Media. A number of studies have been conducted dealing with chlorpyrifos concentrations in indoor air. Although some data on the levels of chlorpyrifos in ambient air are available (Carey and Kutz 1985), these data are neither current nor general enough to estimate inhalation exposure to chlorpyrifos for the general population in the United States. Limited data on the level of chlorpyrifos in drinking water were located in the literature. More recent data regarding the levels of chlorpyrifos in ambient air, drinking water, and soil are needed. Data on chlorpyrifos levels in food and recent estimates of the human intake of chlorpyrifos from foods are available (Duggan et al. 1983; FDA 1990, 1991, 1992, 1993; Gelardi and Mountford 1993; Gunderson 1988; Luke et al. 1988; Schattenburg and Hsu 1992; Yess et al. 1991a, 1991b).

Reliable monitoring data for the levels of chlorpyrifos in contaminated media at hazardous waste sites are needed so that the information obtained on levels of chlorpyrifos in the environment can be used

in combination with the known body burden of chlorpyrifos to assess the potential risk of adverse health effects in populations living in the vicinity of hazardous waste sites.

Exposure Levels in Humans. Aside from the NHANES III and Hill et al. (1995) data, no other quantitative information on chlorpyrifos levels in human tissues and body fluids for a control population, populations near hazardous waste sites, or occupationally exposed groups were located. Additionally, data on the levels of chlorpyrifos and its metabolites in body tissues and fluids in symptomatic, exposed individuals, as well as RBC and plasma ChE activity levels in these persons, are needed to correlate exposure levels with adverse symptoms and to identify levels of ChE inhibition associated with the onset of toxic manifestations. One potential source of this information is the American Association of National Poison Control Centers.

Exposure Registries. No exposure registries for chlorpyrifos were located. This substance is not currently one of the compounds for which a subregistry has been established in the National Exposure Registry. The substance will be considered in the future when chemical selection is made for subregistries establishment. The information that is amassed in the National Exposure Registry facilitates the epidemiological research needed to assess adverse health outcomes that may be related to exposure to a substance.

5.7.2 Ongoing Studies

As part of the National Pesticide Impact Assessment Program, research is in progress at North Carolina State University (Leidy) to study the movement of herbicides into poorly drained soils of the Tidewater region of North Carolina and to determine the dislodgeable residue of chlorpyrifos from carpet samples.

Research is in progress at the University of Florida, Belle Glade (Snyder) to quantify organophosphate losses in percolate, retention in soil and thatch, and removal in grass clippings.

Research is in progress at the University of Florida, Gainesville (Moye and Wheeler), Texas A&M (Plapp), and Clemson (Camper) to determine the metabolic fate of chlorpyrifos in different media.

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Researchers at the University of Puerto Rico (Singmaster and Acin-Diaz) are determining the dissipation and persistence of chlorpyrifos in surface and vadose-zone soils and water.

The USDA Agricultural Research Service (ARS) (Wauchope) is determining chlorpyrifos residues for 22 minor food crops at Tifton, Georgia, 7 crops at Yakima, Washington (Toba), and chlorpyrifos residues in coffee in Puerto Rico (Acin-Diaz, Liu, Armstrong).

The USDA-ARS in Riverside, California (Spencer and Yates) is studying water and pesticide management systems for minimizing groundwater and air contamination, as well as the persistence (fate and transport) of chlorpyrifos (Gaston).

The USDA-ARS in Beltsville, Maryland (Wright and Hapeman) are quantifying chlorpyrifos volatilization, transport, partitioning, and deposition.

The University of Nevada at Reno (Seiber), with funding from the U.S. Department of Agriculture, is studying the aerial transport and deposition of organophosphate pesticides, including chlorpyrifos, in Sierra Nevada forests.

The National Taiwan University (Hsu and Epstein), with funding from the USDA, is investigating the effects of different processing/cooking variables on chlorpyrifos residues in meat and poultry products.

Research is in progress at the University of Nebraska (Shea) to determine the mobility and bioavailability of chlorpyrifos in soil and at Iowa State University to compare degradation kinetics at high as opposed to low concentrations, persistence of TCP, and effect of temperature and moisture on degradation of chlorpyrifos (coats).

The U.S. Department of Energy (DOE) is funding a cooperative research and development agreement (CRADA) between Argonne National Laboratory (Kakar), the University of Notre Dame, and COGNIS, Inc., to study the biodegradability of pesticides by direct enzyme treatment.

The University of California at Davis (Kilgore), with funding from the U.S. Department of Agriculture, is developing methods to measure exposure, ab,sorption, and toxicity of pesticides to

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workers and is preparing guidelines for best management practices to reduce worker exposure to pesticides.