

Monitoring of Pesticide & Chemical Residues in Air, Soil, and Non-Target Vegetation

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Introduction

Pesticide sprays can directly hit non-target vegetation, or can drift or volatilize from the treated area and contaminate air, soil, and non-target plants. Some pesticide drift occurs during every application, even from ground equipment (Glottfelty and Schomburg, 1989). Drift can account for a loss of 2 to 25% of the chemical being applied, which can spread over a distance of a few yards to several hundred miles. As much as 80–90% of an applied pesticide can be volatilised within a few days of application (Majewski, 1995). Despite the fact that only limited research has been done on the topic, studies consistently find pesticide residues in air. According to the USGS, pesticides have been detected in the atmosphere in all sampled areas of the USA (Savonen, 1997). Nearly every pesticide investigated has been detected in rain, air, fog, or snow across the nation at different times of the year (U.S. Geological Survey, 1999). Many pesticides have been detected in air at more than half the sites sampled nationwide. Herbicides are designed to kill plants, so it is not surprising that they can injure or kill desirable species if they are applied directly to such plants, or if they drift or volatilise onto them. Many ester-formulation herbicides have been shown to volatilise off treated plants with vapors sufficient to cause severe damage to other plants (Straathoff, 1986). In addition to killing non-target plants outright, pesticide exposure can cause sublethal effects on plants. Phenoxy herbicides, including 2,4-D, can injure nearby trees and shrubs if they drift or volatilise onto leaves (Dreistadt *et al.*, 1994). Exposure to the herbicide glyphosate can severely reduce seed quality (Locke *et al.*, 1995). It can also increase the susceptibility of certain plants to disease (Brammall and Higgins, 1998). This poses a special threat to endangered plant species. The U.S. Fish and Wildlife Service has recognized 74 endangered plants that may be threatened by glyphosate alone (U.S. EPA Office of Pesticides and Toxic Substances, 1986). Exposure to the herbicide clopyralid can reduce yields in potato plants (Lucas and Lobb, 1987). EPA calculated that volatilisation of only 1% of applied clopyralid is enough to damage non-target plants (US EPA, 1990). Some insecticides and fungicides can also damage plants (Dreistadt *et al.*, 1994). Pesticide damage to plants is commonly reported to state agencies in the Northwest. (Oregon Dept. of Agriculture, 1999; Washington Dept. of Health, 1999). Plants can also suffer indirect consequences of pesticide applications when harm is done to soil microorganisms and beneficial insects. Pesticides including those of new the generation, e.g., dacthal, chlorothalonil, chlorpyrifos, metolachlor, terbufos and trifluralin have been detected in Arctic environmental samples (air, fog, water, snow) (Rice and Cherniak, 1997), and (Garbarino *et al.*, 2002). Other studies have identified the ability of some of these compounds to undergo short-range atmospheric transport (Muir *et al.*, 2004) to ecologically sensitive regions such as the Chesapeake Bay and the Sierra Nevada mountains (LeNoir *et al.*, 1999; McConnell *et al.*, 1997; Harman-Fetcho *et al.*, 2000,

Thurman and Cromwell, 2000). One long-term study that investigated pesticides in the atmosphere of British Columbia (BC), dating from 1996 (Belzer *et al.*, 1998) showed that 57 chemicals were investigated at two sampling sites (Agassiz and Abbotsford) in the Fraser Valley, from February 1996 until March 1997. Atrazine, malathion, and diazinon, highly toxic chemicals identified as high-priority pesticides by Verrin *et al.* (2004), were detected as early as the end of February (72 pg/m^3) until mid-October (253 pg/m^3), with a peak concentration in mid-June of 42.7 ngm^{-3} . Dichlorvos is a decomposition product of another pesticide, Naled (Dibrom) (Hall *et al.*, 1997). Captan and 2,4-D showed the highest concentrations and deposition rates at these two sites, followed by dichlorvos and diazinon (Dosman and Cockcraft, 1989). Air concentrations of currently used pesticides in Alberta were investigated in 1999 at four sampling sites that were chosen according to geography and pesticide sales data (Kumar, 2001). Triallate and trifluralin were the two mostly detected pesticides at the four sites. Insecticides (malathion, chlorpyrifos, diazinon and endosulfan) were detected intermittently with concentrations in the range $20\text{--}780 \text{ pg/m}^3$. South of Regina, Saskatchewan, in 1989 and 1990, 2,4-D reached 3.9 and 3.6 ng/m^3 at the end of June (Waite *et al.*, 2002a). Triallate, dicamba, bromoxynil concentrations were also higher in 1989 (peak concentration of 4.2 ng/m^3 in mid-June) compared with 1990 ($600\text{--}700 \text{ pg/m}^3$ in mid-June). In a more recent study, Waite *et al.* (2005) studied spatial variations of selected herbicides on a threesite, 500km transect that included two agricultural sites—Bratt's Lake, located 35 km southwest of Regina and Hafford to the North—and a background site at Waskesiu. Some acid herbicides were also investigated in South Tobacco Creek, Manitoba during 1993–1996. Once again, maximum concentrations occurred during periods of local use (Rawn *et al.*, 1999a). A neutral herbicide, atrazine, was also investigated in 1995 (Rawn *et al.*, 1998). It was first detected in mid-April, peaked mid-June at about 300 pg/m^3 , and was detected until the end of October. The insecticide dacthal was identified throughout the sampling periods in 1994, 1995 and 1996 (Rawn and Muir, 1999) even though it was not used in this area ($<20\text{--}300 \text{ pg/m}^3$).

Non-target organisms

Pesticides are found as common contaminants in soil, air, water and on non-target organisms in our urban landscapes. Once there, they can harm plants and animals ranging from beneficial soil microorganisms and insects, non-target plants, fish, birds, and other wildlife. Chlorpyrifos, a common contaminant of urban streams (U.S. Geological Survey, 1999), is highly toxic to fish, and has caused fish kills in waterways near treated fields or buildings (US EPA, 2000). Herbicides can also be toxic to fish. According to the EPA, studies show that trifluralin, an active ingredient in the weed-killer Snapshot, “is highly to very highly toxic to both cold and warm water fish” (U.S. EPA, 1996). In a series of different tests it was also shown to cause vertebral deformities in fish (Koyama, 1996). The weed-killers Ronstar and Roundup are also acutely toxic to fish (Folmar *et al.*, 1979; Shafiei and Costa, 1990). The toxicity of Roundup is likely due to the high toxicity of one of the inert ingredients of the product (Folmar *et al.*, 1979). In addition to direct acute toxicity, some herbicides may produce sublethal effects on fish that lessen their chances for survival and threaten the population as a whole. Glyphosate or glyphosate-containing products can cause sublethal effects such as erratic swimming and labored breathing, which increase the fish's chance of being eaten (Liong *et al.*, 1988). 2,4-D herbicides caused physiological stress responses in sockeye salmon (McBride *et al.*, 1981) and reduced the food-gathering abilities of rainbow trout (Little, 1990). Several cases of pesticide poisoning of dolphins have been reported worldwide. Because of their high trophic level in the food chain and relatively low activities of drug-metabolising enzymes, aquatic mammals such as dolphins accumulate increased concentrations of persistent organic pollutants (Tanabe *et al.*, 1988) and are thereby vulnerable to toxic effects from

contaminant exposures. Dolphins inhabiting riverine and estuarine ecosystems are particularly vulnerable to the activities of humans because of the restricted confines of their habitat, which is in close proximity to point sources of pollution. River dolphins are among the world's most seriously endangered species. Populations of river dolphins have been dwindling and face the threat of extinction; the Yangtze river dolphin (*Lipotesvexillifer*) in China and the Indus river dolphin (*Platanista minor*) in Pakistan are already close to extinction (Renjun, 1990; Perrin *et al.*, 1989; Reeves *et al.*, 1991; Reeves and Chaudhry, 1998). In addition to habitat degradation (such as construction of dams) (Reeves and Leatherwood, 1994), boat traffic, fishing, incidental and intentional killings, and chemical pollution have been threats to the health of river dolphins (Kannan *et al.*, 1993b, 1994, 1997; Senthilkumar *et al.*, 1999). Earlier studies reported concentrations of heavy metals (Kannan *et al.*, 1993), organochlorine pesticides and polychlorinated biphenyls (PCBs) (Kannan *et al.*, 1994), and butyltin compounds (Kannan *et al.*, 1997) in Ganges river dolphins and their prey. The continuing use of organochlorine pesticides and PCBs in India is of concern (Kannan *et al.*, 1992; Kannan *et al.*, 1997a; Kannan *et al.*, 1997b; Tanabe *et al.*, 1998). The Ganges river basin is densely populated and heavily polluted by fertilizers, pesticides, and industrial and domestic effluents (Mohan, 1989). In addition to fish, other marine or freshwater animals are endangered by pesticide contamination. Exposure to great concentrations of persistent, bioaccumulative, and toxic contaminants such as DDT (1,1,1-trichloro-2,2-bis[*p*-chlorophenyl]ethane) and PCBs has been shown to elicit adverse effects on reproductive and immunological functions in captive or wild aquatic mammals (Helle *et al.*, 1976; Reijnders, 1986; Ross *et al.*, 1995; Martineau *et al.*, 1987; Kannan *et al.*, 1993; Colborn and Smolen, 1996). Aquatic mammals inhabiting freshwater systems, such as otters and mink, have been reported to be sensitive to chemical contamination (Leonards *et al.*, 1995; Leonards *et al.*, 1997). 2,4-D or 2,4-D containing products have been shown to be harmful to shellfish (Cheney *et al.*, 1997) and other aquatic species (U.S. EPA, 1989; Sanders, 1989). The weed-killer trifluralin is moderately to highly toxic to aquatic invertebrates, and highly toxic to estuarine and marine organisms like shrimp and mussels (U.S. EPA, 1996). Since herbicides are designed to kill plants, it makes sense that herbicide contamination of water could have devastating effects on aquatic plants. In one study, oxadiazon was found to severely reduce algae growth (Ambrosi *et al.*, 1978). Algae is a staple organism in the food chain of aquatic ecosystems. Studies looking at the impacts of the herbicides atrazine and alachlor on algae and diatoms in streams showed that even at fairly low levels, the chemicals damaged cells, blocked photosynthesis, and stunted growth in varying ways (U.S. Water News Online, 2000). The herbicide oxadiazon is also toxic to bees, which are pollinators (Washington State Department of Transportation, 1993). Herbicides may hurt insects or spiders also indirectly when they destroy the foliage that these animals need for food and shelter. For example spider and carabid beetle populations declined when 2,4-D applications destroyed their natural habitat (Asteraki *et al.*, 1992). Non-target birds may also be killed if they ingest poisoned grains set out as bait for pigeons and rodents (US EPA, 1998). Avitrol, a commonly used pigeon bait, poses a large potential for ingestion by non target grain feeding birds. It can be lethal to small seed-eating birds (Extoxnet, 1996). Brodifacoum, a common rodenticide, is highly toxic to birds. It also poses a secondary poisoning hazard to birds that may feed on poisoned rodents (US EPA, 1998). Herbicides can also be toxic to birds. Although trifluralin was considered "practically nontoxic to birds" in studies of acute toxicity, birds exposed multiple times to the herbicide experienced diminished reproductive success in the form of cracked eggs (U.S. EPA, 1996). Exposure of eggs to 2,4-D reduced successful hatching of chicken eggs (Duffard *et al.*, 1981) and caused feminisation or sterility in pheasant chicks (Lutz *et al.*, 1972). Herbicides can also adversely affect birds by destroying their habitat. Glyphosate treatment in clear cuts caused dramatic decreases in the populations of birds

that lived there (MacKinnon *et al.*, 1993) Effects of some organochlorines (OCs) on fish-eating water birds and marine mammals have been documented in North America and Europe (Barron *et al.*, 1995; Cooke, 1979; Kubiak *et al.*, 1989). Despite the continuing usage, little is known about the impacts of OCs in bird populations in developing countries. Among the countries that continue to use OCs, India has been one of the major producers and consumers in recent years. As a consequence, wild birds in India are exposed to great amounts of OC pesticides (Tanabe *et al.*, 1998). Use of OCs in tropical countries may not only result in exposure of resident birds but also of migratory birds when they visit tropical regions in winter. The Indian sub-continent is a host to a multitude of birds from western Asia, Europe and Arctic Russia in winter (Woodcock, 1980). Hundreds of species of waterfowl, including wading birds such as plovers, terns and sandpipers, migrate each winter to India covering long distances (Grewal, 1990). While concentrations of OC pesticides in wholebody homogenates of birds have been reported elsewhere (Tanabe *et al.*, 1998), concentrations of OCs in prey items and in eggs of Indian birds have not been reported.

A few studies related to the decline in the populations of bats in various parts of the world to OC exposure were also being conducted (Altenbach *et al.*, 1979; Clark, 1976; Clark, 1983; Clark, 1981; Geluso *et al.*, 1976; Jefferies, 1976; Thies and Mc Bee, 1994). The world population of bats was estimated to be 8.7 million during 1936 and it declined to approximately 200,000 in 1973 (Geluso *et al.*, 1976) It has recovered slightly to an estimated number of 700,000 in 1991 (Geluso *et al.*, 1976; Thies and Mc Bee, 1994). High tissue concentrations of *p,p'*-dichlorodiphenyldichloroethene (*p,p'*-DDE) have been found in bats in Carlsbad Caverns in Mexico and in New Mexico in the USA (Geluso *et al.*, 1976; Thies and Mc Bee, 1994). Occurrence of stillbirths in little brown bats exposed to high concentrations of PCBs, *p,p'*-DDE, and/or oxychlordanes was documented (Clark, 1976; Jefferies, 1976). These observations indicate that bats can accumulate high concentrations of OCs and may be affected by their potential toxic effects. The flying fox or the new world fruit bat, short-nosed fruit bat and Indian pipistrelle bat are resident species and are very common in South India. Their habitat is mainly agricultural areas, rock caves, and abandoned houses in domesticated areas. Insects constitute an important diet for many bats, allowing the passage of OCs in their body (Mc Bee *et al.*, 1992). Several studies found OC pesticides and PCBs in livers and eggs of birds in developed countries (Becker, 1989; Bernardz *et al.*, 1990; Cade *et al.*, 1989; Castillo *et al.*, 1994; Mora, 1996; Mora, 1997). Similarly, several studies reported OCs in a variety of biota including humans and wildlife from India (Senthilkumar *et al.*, 2000). However, no study has used whole body homogenates of birds, which is important to evaluate biomagnification features and body burdens of OCs (Mc Bee *et al.*, 1992). Earlier studies used specific body tissues to estimate biomagnification of OCs. However theoretically, estimation of biomagnification factors requires whole body concentrations rather than specific tissue concentrations.

Impact through food commodities

For determining the extent of pesticide contamination in the food stuffs, programs entitled 'Monitoring of Pesticide Residues in Products of Plant Origin in the European Union' started to be established in the European Union since 1996. In 1996, seven pesticides (acephate, chlopyrifos, chlopyrifos-methyl, methamidophos, iprodione, procymidone and chlorothalonil) and two groups of pesticides (benomyl group and maneb group, i.e. dithiocarbamates) were analysed in apples, tomatoes, lettuce, strawberries and grapes. An average of about 9 700 samples has been analysed for each pesticide or pesticide group.

For each pesticide or pesticide group, 5.2% of the samples were found to contain residues and 0.31% had residues higher than the respective MRL for that specific pesticide. Lettuce was the crop with the highest number of positive results, with residue levels exceeding the MRLs more frequently than in any of the other crops investigated. The highest value found in 1996 was for a compound of the maneb group in lettuce which corresponded to a mancozeb residue of 118 mg/kg. In 1997, 13 pesticides (acephate, carbendazin, chlorothalonil, chlopyriphos, DDT, diazinon, endosulfan, methamidophos, iprodione, metalaxyl, methidathion, thiabendazole, triazophos) were assessed in five commodities (mandarins, pears, bananas, beans, and potatoes). Some 6 000 samples were analysed. Residues of chlorpyriphos exceeded MRLs most often (0.24%), followed by methamidophos (0.18%), and iprodione (0.13%). With regard to the commodities investigated, around 34% contained pesticide residues at or below the MRL, and 1% contained residues at levels above the MRL. In mandarins, pesticide residues were most frequently found at levels at or below the MRL (69%), followed by bananas (51%), pears (28%), beans (21%) and potatoes (9%). MRLs were exceeded most often in beans (1.9%), followed by mandarins (1.8%), pears (1.3%), and bananas and potatoes (0.5%). Estimation of the dietary intake of pesticide residues (based on the 90th percentile) from the above-mentioned commodities, where the highest residue levels of the respective pesticides were found, shows that there is no exceeding of the ADI with all the pesticides and commodities studied (European Commission, 1999). In 1998, four commodities (oranges, peaches, carrots, spinach) were analysed for 20 pesticides (acephate, benomyl group, chlopyriphos, chlopyriphos-methyl, deltamethrin, maneb group, diazinon, endosulfan, methamidophos, iprodione, metalaxyl, methidathion, thiabendazole, triazophos, permethrin, vinclozolin, lambdacyalothrin, pirimiphos-methyl, mercabam). With regard to all four commodities investigated in 1998 (oranges, peaches, carrots, spinach), about 32% contained residues of pesticides at or below MRL, and 2% above the MRL (1.8% for EU-MRLs, 0.4% for national MRLs). Residues at or below the MRL were found most often in oranges (67%), followed by peaches (21%), carrots (11%) and spinach (5%). MRL values were exceeded most often in spinach (7.3%), followed by peaches (1.6%), carrots (1.2%) and oranges (0.7%). The intake of pesticide residues has not exceeded the ADI in any case. It was found to be below 10% of the ADI for all pesticides. The exposure ranges from 0.35% of the ADI for the benomyl group to 9.9% of the ADI for the methidathion group. In 1999, four commodities (cauliflower, peppers, wheat grains, and melon) were analysed for the same 20 pesticides as in the 1998 study (European Commission, 2001). Overall, around 4700 samples were analysed. Residues of methamidophos exceeded MRLs most often (8.7%), followed by the maneb group (1.1%), thiabendazole (0.57%), acephate (0.41%) and the benomyl group (0.35%). The MRL for methamidophos was exceeded most often in peppers and melons (18.7 and 3.7%, respectively). The residues of the maneb group exceeded the MRL most often in cauliflower (3.9%); residues of thiabendazole exceeded the MRL most often in melons (2.8% of the melon samples). With regard to all the commodities investigated, around 22% of samples contained residues of pesticides at or below the MRL and 8.7% above the MRL. Residues at or below MRL were found most often in melons (32%), followed by peppers (24%), wheat grains (21%) and cauliflower (17%). MRL values were exceeded most often in peppers (19%), followed by melons (6.1%), cauliflower (3%) and wheat grains (0.5%). The intake of pesticide residues did not exceed the ADI in any case. It was below 1.5% of the ADI for all pesticides. The exposure ranged between 0.43% of the ADI for methamidophos and 1.4% of the ADI for endosulfan. The intakes for the highest residue levels in a composite sample for chlorpyriphos, deltamethrin, endosulfan and methidathion were below the ARfD for adults. They range between 1.5% of the ARfD for deltamethrin and 67% of the ARfD for endosulfan (Nasreddine and Parent-Massin, 2002). In spite of food contamination, most pesticide deaths recorded in hospital surveys are the result of self-poisoning

(Eddleston, 2000). The Global Burden of Disease Study 6 estimated that 798 000 people died from deliberate self-harm in 1990, over 75% of whom were from developing countries (Murray and Lopez, 1996). More recent WHO estimates showed that over 500 000 people died from self-harm in Southeast Asia and the Western Pacific during 2000 alone (WHO, 2001). Suicide is the commonest cause of death in young Chinese women and Sri Lankan men and women (Murray and Lopez, 1996; Sri Lankan Ministry of Health, 1995; WHO, 2001).

Conclusion

In India the first report of poisoning due to pesticides was from Kerala in 1958, where over 100 people died after consuming wheat flour contaminated with parathion (Karunakaran, 1958). This prompted the Special Committee on Harmful Effects of Pesticides constituted by the ICAR to focus attention on the problem (Report of the Special Committee of ICAR, 1972). In a multi-centric study to assess the pesticide residues in selected food commodities collected from different states of the country (Surveillance of Food Contaminants in India, 1993), DDT residues were found in about 82% of the 2205 samples of bovine milk collected from 12 states. About 37% of the samples contained DDT residues above the tolerance limit of 0.05 mg/kg (whole milk basis). The highest level of DDT residues found was 2.2 mg/kg. The proportion of the samples with residues above the tolerance limit was highest in Maharashtra (74%), followed by Gujarat (70%), Andhra Pradesh (57%), Himachal Pradesh (56%), and Punjab (51%). In the remaining states, this proportion was less than 10%. Data on 186 samples of 20 commercial brands of infants formulae showed the presence of residues of DDT and HCH isomers in about 70 and 94% of the samples with their maximum level of 4.3 and 5.7 mg/kg (fat basis) respectively. Measurement of chemicals in the total diet provides the best estimates of human exposure and of the potential risk. The risk of consumers may then be evaluated by comparison with toxicologically acceptable intake levels. The average total DDT and BHC consumed by an adult were 19.24 mg/day and 77.15 mg/day respectively (Kashyap *et al.*, 1994). Fatty food was the main source of these contaminants. In another study, the average daily intake of HCH and DDT by Indians was reported to be 115 and 48 mg per person respectively, which were higher than those observed in most of the developed countries (Kannan *et al.*, 1992).

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