Canadian Tissue Residue Guidelines for the Protection of Wildlife Consumers of Aquatic Biota

DDT (TOTAL)

DDT (1,1,1-trichloro-2,2-bis(*p*-chlorophenyl) ethane) is a chlorinated hydrocarbon insecticide that has been used worldwide since the 1940s to control insects that carry diseases (e.g., malaria and typhus), insects that attack agricultural crops (e.g., cotton, peanuts, and soybeans), and biting insects (e.g., blackflies) (ATSDR 1994). The use of DDT in Canada, however, was severely restricted in 1970 and banned in 1985.

Technical grade DDT is a nonflammable, white crystalline or waxy solid at room temperature that is tasteless and almost odourless (Worthing and Hance 1991). It is composed of *p,p'*-DDT (77.1%), *o,p'*-DDT (14.9%), *p,p'*-DDE (1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethylene) (4.0%), *o,p'*-DDE (0.1%), *p,p'*-DDD (1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethane) (0.3%), *o,p'*-DDD (0.1%), and a number of unidentified compounds (3.5%) (USEPA 1980). In the environment, the primary ingredients, *p,p'*- and *o,p'*-DDT, are transformed into a number of breakdown products with similar chemical structures. Of these, *o,p'*-DDE and *p,p'*-DDE tend to be the most persistent in the environment and are, therefore, detected at the highest concentrations.

The highly lipophilic nature of DDT (K_{ow} = 5.5–6.1) (Suntio et al. 1988) presents serious problems for wildlife that feed at high trophic levels in the food chain. Both birds and mammals are capable of accumulating DDTs by ingesting contaminated foods. For example, double-crested cormorants had a whole body BAF (ratio of DDT in bird or mammal to DDT in the diet) of 236 (ww:ww) after being exposed to DDT in their diets for 9 weeks (Greichus and Hannon 1973). High BCFs (ratio of tissue to water concentrations) have also been reported in wildlife exposed to DDT. For example, Tanabe et al. (1994) reported that striped dolphins *(Stenella caeruleoalba)* accumulated DDT in their tissues to levels that were up to 10 million times higher than those in water. It was these properties of DDT that caused Canada to ban its use in 1985 (CCREM 1987).

Currently, the main source of DDT in Canada is atmospheric deposition (Lintott and Waite 1991), arising either from volatilization from soils and aquatic sediments in past use areas (Noble 1990), or from transport from countries where DDT is still in use (Lognathan and Kannan 1994). The high vapour pressures (0.2–1 mPa) and low water solubilities (3–100 $\mu g \cdot L^{-1}$) of DDT and its metabolites (Suntio et al. 1988) cause them to be volatilized, dispersed through the atmosphere (Oehme 1991), and ultimately deposited in cold temperate regions as a result of atmospheric fallout (i.e., “global distillation”) (Lognathan and Kannan 1994). As a result, DDT and its metabolites have been detected in virtually all media across Canada, including remote areas of the Canadian Arctic.

Wildlife in aquatic ecosystems depend on aquatic biota such as fish, shellfish, invertebrates, and plants as their primary source of food. For aquatic-based wildlife species, these food resources provide the main route of exposure to persistent substances, such as DDT, that accumulate in food webs. Table 1 lists the Canadian tissue residue guidelines for the protection of wildlife consumers of aquatic biota. Table 2 summarizes total DDT measurements recently made in Canadian biota (i.e., post-banning of DDT in Canada). The data represent both typically low and high levels of total DDT measured for each organism. Although the data are difficult to compare both temporally and spatially, it is clear that organisms that feed at higher trophic levels (i.e., mammals and birds) have higher levels of total DDT in their tissues.

Toxicity

Exposure to DDT and its metabolites is known to reduce longevity and alter cellular metabolism, neural activity, and liver function (USEPA 1980). In addition, mutagenic and carcinogenic effects, as well as adverse effects on reproduction, growth, and immunocompetence, have been

Table 1. Canadian tissue residue guideline for total DDT[*] for the protection of wildlife consumers of aquatic biota (Environment Canada 1997).

Compound	Guideline value ($\mu g \cdot kg^{-1}$ diet ww)
Total DDT[†]	14.0

[*] Represents a single maximum concentration of DDT in aquatic biota that would not be expected to result in adverse effects on wildlife consumers of aquatic biota.

[†] Total DDT = *o,p'* + *p,p'* DDT; *o,p'* + *p,p'* DDE; *o,p'* + *p,p'* DDD.

Table 2. Recent levels of total DDT in Canadian biota.

Biota		Tissue	Year	Total DDT* ($\mu g \cdot kg^{-1}$ ww)	Reference
Invertebrates:	Freshwater	Whole	1992	0.3–25	Schindler et al. 1993
	Marine	Whole	1993/4; 1987	0.25–180	Muir et al. 1994; Hargave 1994
	Marine	Muscle	1989	0.3–2.4	Swain and Walton 1990
Fish:	Freshwater	Muscle	1991; 1992	0.5–1300	Palmer 1992; Muir et al. 1993
	Freshwater	Liver	1993/94; 1992	14–6310	Muir et al. 1994; Muir et al. 1993
	Marine	Muscle	1993; 1992	0.97–140	Swain and Walton 1990; Bright et al. 1995
	Marine	Liver	1991; 1992	1.9–235	Bright et al. 1995
Amphibians		Whole	1990	16–120	Bright et al. 1995
Reptiles		Muscle	1988/89	0.9–170	Hebert et al. 1993
		Egg	1989; 1990	9.2–392	Bonin et al. 1995
Mammals		Blubber	1991/92; 1986/87	28–101 000	Muir et al. 1992; Muir et al. 1990
		Muscle	1989/90	8.2–40.6	Langois and Langis 1995
Birds		Egg	1992; 1986	12–7425	Braune 1993; Forsyth et al. 1994
		Muscle	1991; 1992	1.5–3044	Braune 1993

*Represents the range of recent values for total DDT found in the literature.

observed in mammalian and avian species exposed to these substances (ATSDR 1994).

Mammalian Toxicity

Acute oral single lethal doses (LD_{50}s) of *p,p′*-DDT range from 113–182 $mg \cdot kg^{-1}$ in rats (Gaines and Linder 1986; Worthing and Hance 1991) to >2100 $mg \cdot kg^{-1}$ in Syrian golden hamsters (Agthe et al. 1970). The available data indicate that *p,p′*-DDE is less toxic to rats than the *p,p′*-DDT isomer, with acute oral LD_{50}s of 380 and 1240 $mg \cdot kg^{-1}$ in male and female rats, respectively (USEPA 1980). The LD_{50}s of *p,p′*-DDD are also relatively high in mice, with values of 1466 and 1507 $mg \cdot kg^{-1}$ reported in females and males, respectively (Tomatis *et al*. 1974).

Several studies have shown that exposure to both the *o,p′* and *p,p′*-DDT isomers can result in adverse reproductive effects. Exposure to 0.53 $mg \cdot kg^{-1}$ bw per day of *p,p′*-DDT for 60 d significantly decreased fertility in female rats (Green 1969), and exposure to 0.7 $mg \cdot kg^{-1}$ bw per day of *o,p′*-DDT for 15 d hastened vaginal patency in young female rats (Wrenn et al. 1970).

The results of animal studies indicate that long-term exposure to sublethal levels of DDTs can result in the formation of tumours and carcinomas in mammals, with most occurring in the liver. Tarjan and Kemeny (1969) determined that exposure to 0.7 $mg \cdot kg^{-1}$ bw per day of *p,p′*-DDT for 180 d increased the incidence of leukemia and malignant tumours in male and female BALBc mice. A study by Tomatis et al. (1974) indicated that exposure to 29 $mg \cdot kg^{-1}$ bw per day of *p,p′*-DDE for 504–518 d resulted in an increased incidence of hepatomas in both male and female mice. Long-term exposure (728–798 d) to the same daily dose of *p,p′*-DDD increased the frequency of lung tumours in both sexes of mice.

Avian Toxicity

DDT and its metabolites, DDE and DDD, generally have moderate to low toxicity to birds when administered as acute oral doses or in the diet (WHO 1989). LD_{50}s of *p,p′*-DDT ranged from 595 $mg \cdot kg^{-1}$ for the California quail *(Callipepla californica)* (Hudson et al. 1984) to >4000 $mg \cdot kg^{-1}$ for pigeons *(Columbia livia)* (Tucker and Crabtree 1970). The single LD_{50}s for *p,p′*-DDD ranged from 386 $mg \cdot kg^{-1}$ for pheasants to >2000 $mg \cdot kg^{-1}$ for mallard ducks *(Anas platyryhnchos)* (Hudson et al. 1984). A 5-d LD_{50} for *p,p′*-DDE of 77.3 $mg \cdot kg^{-1}$ bw per day was reported for 14-d-old Japanese quail *(Coturnix coturnix japonica)* (Hill and Camardese 1986).

Data from several authors indicate that DDT adversely affects the reproduction of avian species, with thin egg shells being one of the most common symptoms. Cecil et al. (1973) determined that eggshell thickness was significantly decreased in white leghorn chickens fed 1.0 mg·kg^{-1} bw per day of *p,p'*-DDT for 60 d. Similarly, Kolaja (1977) determined that eggshell thickness was reduced in mallard ducks administered 0.3 mg·kg^{-1} bw per day of *p,p'*-DDT in their diet for 30 d. Lincer (1975) determined that administering doses as low as 0.50 mg·kg^{-1} bw per day of *p,p'*-DDE to American kestrels *(Falco sparverius)* for 168 d significantly reduced eggshell thickness. In addition, a dose of 0.3 mg·kg^{-1} bw per day of *p,p'*-DDD in the diet of mallard ducks resulted in increased embryo mortality, reduced hatchling survival, and fewer ducklings per hen (Heath et al. 1969).

Tissue Residue Guideline Derivation

The Canadian tissue residue guideline for the protection of wildlife that consume aquatic biota was developed according to the CCME protocol (CCME 1998).

Guideline Derivation for Total DDT

DDT in environmental samples often exists as a mixture of some or all of the metabolites. However, the analytical methods necessary to separate these metabolites are not always available, and the result is, therefore, often reported as total DDT. For this reason a tissue residue guideline for total DDT was deemed appropriate. In addition, a guideline for total DDT is necessary to compare with historical data on DDT levels in environmental samples, also often reported as total DDT. Unfortunately, insufficient information is available to evaluate the toxicity of tissue-associated total DDT directly. Since DDE and DDD are metabolic products of DDT, some or all of the isomers will occur together in the environment. Therefore, a guideline for total DDT was developed using the most sensitive endpoint and the most toxic isomer for mammals and birds using the CCME protocol for the derivation of tissue residue guidelines for the protection of wildlife that consume aquatic biota (CCME 1998). This guideline should be protective for all wildlife irrespective of what isomers are present in the aquatic tissue.

Mammalian Reference Concentration

For mammals, the most sensitive LOAEL was 0.53 mg·kg^{-1} bw per day of *p,p'*-DDT (Green 1969). For the purposes of calculating a TDI of DDT for mammalian species, the NOAEL was calculated by dividing the LOAEL of 0.53 g·kg^{-1} bw per day by 5.6, resulting in a NOAEL of 0.095 g·kg^{-1} bw per day (CCME 1993). The TDI was calculated as follows:

$$\mathrm{TDI} = (\mathrm{LOAEL} \cdot \mathrm{NOAEL})^{0.5} \div \mathrm{UF}$$

where UF = the uncertainty factor. The study by Green (1969) was carried out for 60 d and therefore is considered to be subchronic. While toxicity data are available for several mammalian species, information on wildlife species is lacking. The available data, however, are generally sufficient to evaluate the relative sensitivities of various strains, life stages, and genders of rodent species. In addition, data from long-term studies exist on several sensitive endpoints, such as growth, reproduction, and carcinogenicity. An uncertainty factor of 10 was selected to account for differences in interspecies sensitivites to DDT as well as extrapolation from subchronic to chronic effects. This supports the calculation of a mammalian TDI of 22.4 µg·kg^{-1} bw per day for DDT.

The mammalian TDI was then used in conjunction with the body weights (bw) and daily food intake rates (FI) of the most sensitive wildlife species to calculate reference concentrations (RC) of total DDT, using the following equation:

$$\mathrm{RC} = \mathrm{TDI} \div (\mathrm{FI} \div \mathrm{bw})$$

Among wildlife species, those with the highest FI:bw ratios have the greatest potential exposure to DDT. These species, therefore, are used to calculate the RCs for total DDT. The mammalian RC was calculated to be 94.0 µg·kg^{-1} diet ww of DDT from a TDI of 22.4 µg·kg^{-1} bw per day and assuming a body weight of 0.60 kg and food intake rate of 0.143 kg ww per day for female mink *(Mustela vison)* (CCME 1998).

Avian Reference Concentration

For birds exposed to DDT, the most sensitive endpoint appears to be eggshell thinning and associated reproductive impairment. The most sensitive LOAEL was 0.3 mg·kg^{-1} bw per day (eggshell thinning in mallard ducks) (Kolaja 1977) and the NOAEL was estimated (CCME 1993) to be 0.054 mg·kg^{-1} bw per day. The studies by Heath et al. (1969) and Vangilder and Peterle (1980) were considered to be chronic studies. Although no data were located on the carcinogenic or mutagenic effects of DDT to birds, a large quantity of data exists on the effects of DDT to several avian species, including those known to be sensitive to the reproductive effects of DDT such as raptors. An uncertainty factor of 10, therefore, was used to account for differences in interspecies sensitivities. The LOAEL of 0.30 $mg{\cdot}kg^{-1}$ bw per day was used in conjunction with the NOAEL of 0.054 $mg{\cdot}kg^{-1}$ bw per day to calculate an avian TDI of 13.0 $\mu g{\cdot}kg^{-1}$ bw·per day for DDT.

The avian RC was calculated to be 14.0 $\mu g{\cdot}kg^{-1}$ diet ww of DDT from a TDI of 13.0 $\mu g{\cdot}kg^{-1}$ bw per day, assuming a body weight of 0.032 kg and a food intake rate of 0.03 kg ww per day for Wilson's storm petrel *(Oceanites oceanicus)* (CCME 1998).

Total DDT Tissue Residue Guideline

The lower of the mammalian and avian RCs, 14.0 $\mu g{\cdot}kg^{-1}$ diet on a wet weight basis, was recommended as the Canadian tissue residue guideline for total DDT for the protection of freshwater, marine, and estuarine wildlife that consume aquatic biota.

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