Canadian Sediment Quality Guidelines for the Protection of Aquatic Life

DDT, DDE, and DDD

Dichlorodiphenyltrichloroethane (DDT) is a chlorinated hydrocarbon compound that exhibits broad-spectrum insecticidal properties (ATSDR 1994). There are several possible configurations of the chlorine atoms on the DDT molecule, resulting in several isomeric forms: *p,p′*-DDT, *o,p′*-DDT, and *m,p′*-DDT (USEPA 1980; WHO 1989). The general term "DDT" is applied to a variety of commercial pesticide formulations that consist predominantly of *p,p′*-DDT and *o,p′*-DDT (WHO 1989), but may also contain minor amounts of dichlorodiphenyldichloroethylene (DDE) and dichlorodiphenyldichloroethane (DDD) (WHO 1989).

In this review, the terms "DDT", "DDE", and "DDD" are used to refer to the sum of isomer concentrations of *p,p′*-DDT and *o,p′*-DDT, *p,p′*-DDE and *o,p′*-DDE, and *p,p′*-DDD and *o,p′*-DDD, respectively. "DDTs" refers to any or all of the six compounds identified above, as well as the metabolites and degradation products of these six compounds. "Total DDT" refers to the sum of the concentrations of *p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDE, *o,p′*-DDE, *p,p′*-DDD, and *o,p′*-DDD.

Because of concerns regarding the bioaccumulative properties of DDTs and evidence linking DDTs to adverse effects in many wildlife species, the use of DDTs in Canada was severely restricted in the early 1970s (CCREM 1987). Under the Pest Control Products Act, registration of the two remaining products containing DDT (both used for rodent control) was discontinued in 1985, with the sale and use of existing stocks permitted until the end of 1990 (Pest Management Regulatory Agency 1997, Ottawa, pers. com.). In addition, DDTs may be present as contaminants in pesticides that are manufactured and used in Canada. DDTs have been identified as Track 1 substances by Environment Canada because they are persistent, bioaccumulative, released primarily as a result of human activities, and considered "CEPA-toxic" under the Canadian Environmental Protection Act (Environment Canada 1997a).

DDTs may enter aquatic ecosystems either directly via pesticide application to surface waters or indirectly via atmospheric deposition. Atmospheric DDTs may arise from spraying operations in countries where this is still permitted (ATSDR 1994) or through the volatilization of DDTs that have been deposited on soils, plants, and surface waters. Airborne DDTs may be transported considerable distances and may be deposited to surface waters. Atmospheric deposition is the most important source of DDTs to Canadian aquatic systems.

In general, DDTs are chemically stable under ambient environmental conditions. The physicochemical properties of these substances, such as low solubility in water and high solubility in lipids (i.e., high K_{ow}) and high K_{oc}, are such that DDT and its metabolites are preferentially incorporated into bed sediments and accumulate in the tissues of aquatic organisms rather than remain in the water column (USEPA 1980). Therefore, sediments represent an important route of exposure for aquatic biota to DDTs.

Canadian interim sediment quality guidelines (ISQGs) and probable effect levels (PELs) for DDT, DDD, and DDE in freshwater and marine sediments were developed according to the procedures described in CCME (1995) (Table 1). These values were developed using a modification of the National Status and Trends Program (NSTP) approach, with the exception of those values for DDT in freshwater sediments. Insufficient toxicity data were available on DDT in freshwater sediments to develop an ISQG using either the modified NSTP approach or the spiked-sediment toxicity test approach. Therefore, the marine ISQG and PEL for DDT were provisionally adopted for freshwater sediments, given that they were the lowest biological effects-based values available. The ISQGs and PELs refer to total

Table 1. Interim sediment quality guidelines (ISQGs) and probable effect levels (PELs) for DDT, DDE, and DDD (μg·kg^{-1} dw).

	Freshwater	Marine/estuarine
DDT		
ISQG	1.19*	1.19
PEL	4.77†	4.77
DDE		
ISQG	1.42	2.07
PEL	6.75	374
DDD		
ISQG	3.54	1.22
PEL	8.51	7.81

* Provisional; adoption of marine ISQG.

† Provisional; adoption of marine PEL.

concentrations of DDT, DDE, and DDD in surficial sediments (i.e., top 5 cm), as quantified by standard analytical procedures.

The majority of the data used to derive ISQGs and PELs for DDT, DDE, and DDD are from studies on field-collected sediments that measured concentrations of DDTs, along with concentrations of other chemicals, and associated biological effects. Data were compiled in the Biological Effects Database for Sediments (BEDS) (Environment Canada 1998). All of the BEDS data sets, with the exception of that for DDT in freshwater, met the minimum data requirements for the development of sediment quality guidelines using the modified NSTP approach. The freshwater guideline derivation tables for DDE and DDD contained 53 and 37 effect entries and 230 and 244 no-effect entries, respectively (Environment Canada 1998). The marine guideline derivation tables for DDT, DDE, and DDD contained 26, 37, and 22 effect entries and 149, 174, and 151 no-effect entries, respectively (Environment Canada 1998). The BEDS represents a wide range of concentrations of DDTs, types of sediment, and mixtures of chemicals. Evaluation of the percentage of effect entries for DDT, DDE, and DDD that are below the ISQGs, between the ISQGs and the PELs, and above the PELs (Figures 1 to 5) indicate that these values define three ranges of concentrations: those that are rarely, occasionally, and frequently associated with adverse biological effects, respectively (Environment Canada 1998).

Toxicity

Adverse biological effects for DDTs in the BEDS include decreased benthic invertebrate diversity and abundance, reduced mortality, and behavioural changes, among others (Environment Canada 1998, Appendixes III to VIII). Mortality is the most common acute toxicological endpoint for sediment bioassays on field-collected sediments and laboratory-spiked sediments. Chronic toxicological endpoints include growth, reproductive effort, and behavioural responses.

For example, in freshwater sediments from Trinity River, Texas, benthic invertebrates were less abundant in sediments with a DDT concentration of 9.28 $\mu g \cdot kg^{-1}$, which is above the ISQG of 1.19 $\mu g \cdot kg^{-1}$ (Dickson et al. 1989). In contrast, sediments having a DDT concentration of 0.885 $\mu g \cdot kg^{-1}$, below the freshwater ISQG, had higher abundances of benthic invertebrates (Dickson et al. 1989). In marine sediments, fertilization of a sea urchin, *Arbacia punctulata*, was reduced by 15.3% in Tampa Bay sediments when concentrations of DDT were 17.4 $\mu g \cdot kg^{-1}$, which is above the marine PEL of 4.77 $\mu g \cdot kg^{-1}$ (Long 1993). A fertilization rate of 79.4% was reported in sediments that contained 0.75 $\mu g \cdot kg^{-1}$ of DDT, which is lower than the marine ISQG of 1.19 $\mu g \cdot kg^{-1}$.

The abundance of several groups of benthic invertebrates, including chironomids, gastropods, and amphipods, was reduced in Midland Bay, Lake Huron, at concentrations of DDE ranging from 2.39 to 2.63 $\mu g \cdot kg^{-1}$, which are above the freshwater ISQG of 1.42 $\mu g \cdot kg^{-1}$ (Jaagumagi 1988; Jaagumagi et al. 1989). Similarly, benthic species richness was low in sediments from Toronto Harbour, Ontario, with 11.5 $\mu g \cdot kg^{-1}$ of DDE, which is above the freshwater PEL of 6.75 $\mu g \cdot kg^{-1}$. However, sediments containing DDE concentrations of 0.625 $\mu g \cdot kg^{-1}$, below the freshwater ISQG, exhibited a high degree of species richness (Jaagumagi 1988; Jaagumagi et al. 1989). In marine sediments, reduction in the abundance of crustaceans, echinoderms, amphipods, polychaetes, sponges, and brittle stars was observed in sediments from the southern California bight, with DDE concentrations ranging from 4 055 to 6 993 $\mu g \cdot kg^{-1}$ (Swartz et al. 1985, 1986; Ferraro et al. 1991). In contrast, abundance of these organisms was higher in sediments that contained DDE concentrations of 56 $\mu g \cdot kg^{-1}$.

For DDD, the freshwater BEDS provided similar examples of adverse biological effects. For example, the abundance of several groups of benthic invertebrates, including chironomids and amphipods, was low in sediments of the St. Clair River, Ontario, containing a mean concentration of 5 $\mu g \cdot kg^{-1}$ DDD, which is above the freshwater ISQG of 3.54 $\mu g \cdot kg^{-1}$ (Jaagumagi 1988; Jaagumagi et al. 1989). In contrast, high abundance of the same species was associated with sediments containing 2.5 $\mu g \cdot kg^{-1}$ of DDD, which is below the freshwater ISQG. Similarly, in marine sediments, a 38% mortality rate was observed in *Ampelisca abdita*, an amphipod, at concentrations of 8.63 $\mu g \cdot kg^{-1}$ of DDD, which exceeds the marine PEL of 7.81 $\mu g \cdot kg^{-1}$ (Bricker et al. 1993). However, no mortality to *A. abdita* occurred in sediments containing 0.62 $\mu g \cdot kg^{-1}$ of DDD, which is half the marine ISQG of 1.22 $\mu g \cdot kg^{-1}$ (EMAP 1991).

Concentrations

DDTs have been detected in sediments from a variety of freshwater locations in Canada, however, data for marine and estuarine sediments are limited. Although present levels of these substances are generally low, elevated concentrations have been measured historically. For example, in 1980, freshwater sediments from the Still Creek basin, Fraser River, British Columbia, contained levels of total DDT ranging from 8 to 189 $\mu g \cdot kg^{-1}$ (Garrett 1980). In 1987, DDT isomers were not detected in this

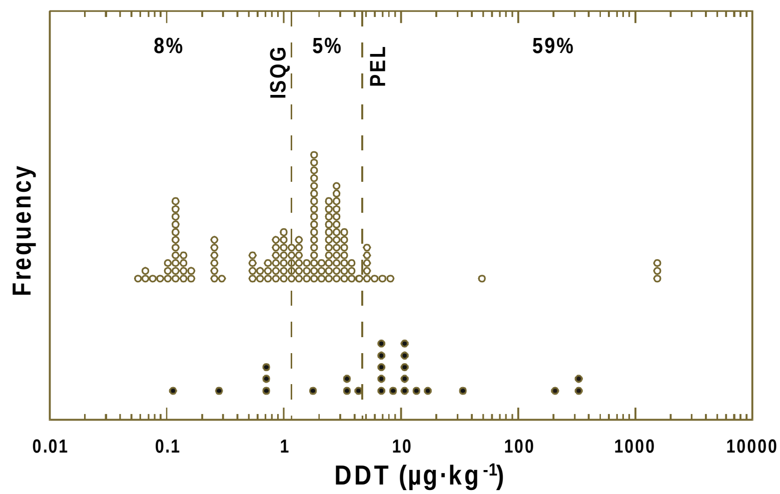

Figure 1. Distribution of DDT concentrations in marine and estuarine sediments that are associated with adverse biological effects (●) and no adverse biological effects (○). Percentages indicate proportions of concentrations associated with effects in ranges below the ISQG, between the ISQG and the PEL, and above the PEL.

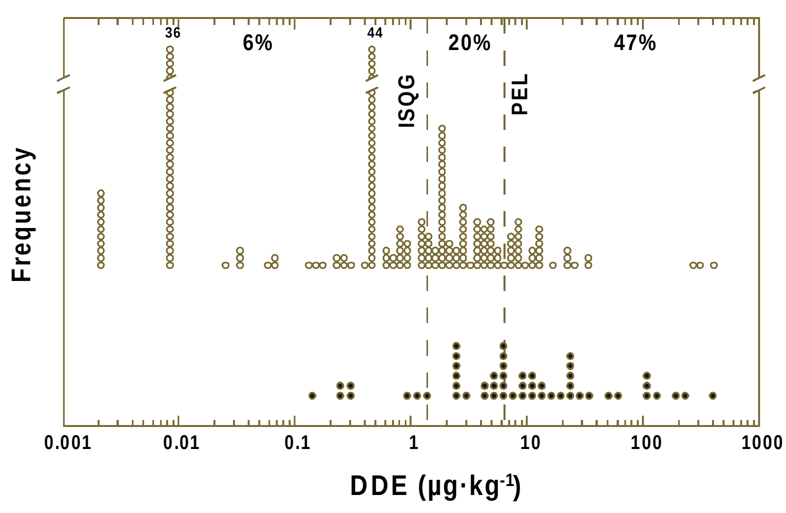

Figure 2. Distribution of DDE concentrations in freshwater sediments that are associated with adverse biological effects (●) and no adverse biological effects (○). Percentages indicate proportions of concentrations associated with effects in ranges below the ISQG, between the ISQG and the PEL, and above the PEL.

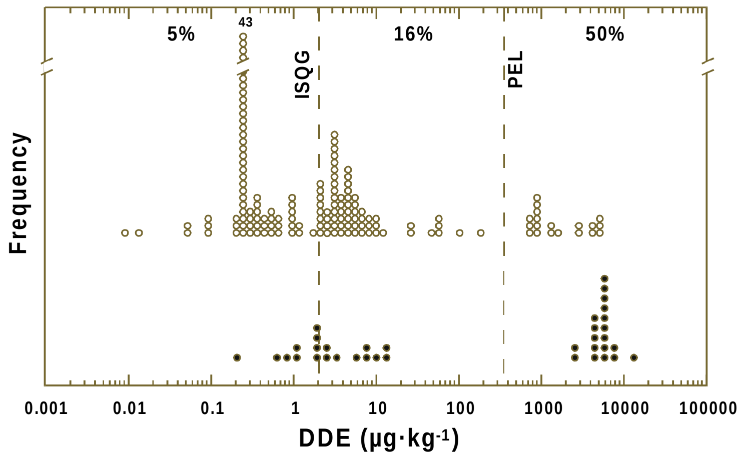

Figure 3. Distribution of DDE concentrations in marine and estuarine sediments that are associated with adverse biological effects (●) and no adverse biological effects (○). Percentages indicate proportions of concentrations associated with effects in ranges below the ISQG, between the ISQG and the PEL, and above the PEL.

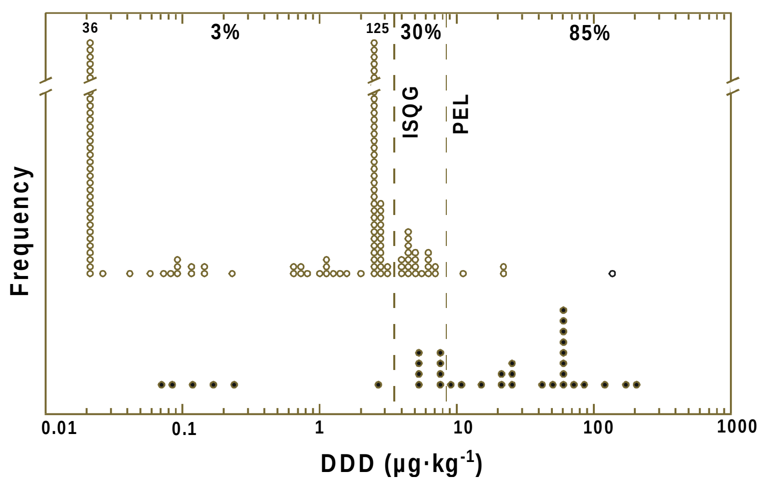

Figure 4. Distribution of DDD concentrations in freshwater sediments that are associated with adverse biological effects (●) and no adverse biological effects (○). Percentages indicate proportions of concentrations associated with effects in ranges below the ISQG, between the ISQG and the PEL, and above the PEL.

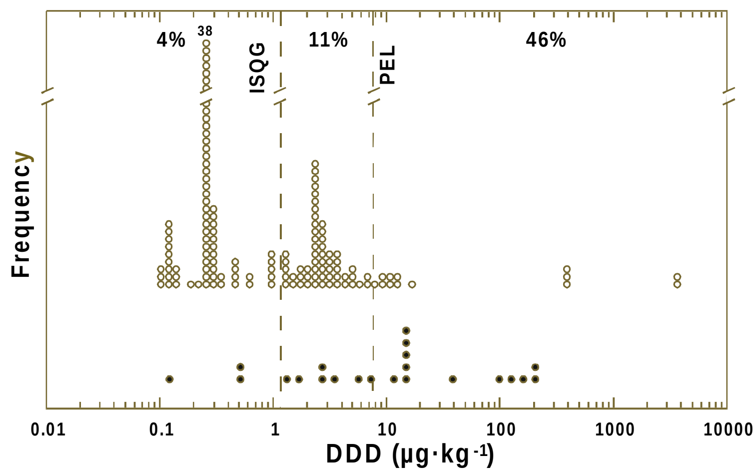

Figure 5. Distribution of DDD concentrations in marine and estuarine sediments that are associated with adverse biological effects (●) and no adverse biological effects (○). Percentages indicate proportions of concentrations associated with effects in ranges below the ISQG, between the ISQG and the PEL, and above the PEL.

drainage basin or any other sites sampled in the lower Fraser River basin (Swain and Walton 1988). In Ontario, historical trends in data show that concentrations of DDTs in freshwater surficial sediments have decreased substantially since the use of DDT-based pesticides was restricted in the early 1970s. For example, mean concentrations of total DDTs in Lake Erie from 1968 to 1971 ranged from 27.9 to 94.8 $\mu g \cdot kg^{-1}$ (Frank et al. 1974, 1977). More recently, samples collected from Lake Erie from 1978 to 1982 had lower levels of total DDTs ranging from <0.5 to 31 $\mu g \cdot kg^{-1}$ (Mudroch 1980; Oliver and Bourbonniere 1985; Bourbonniere et al. 1986). In addition, DDEs and DDDs made up a greater proportion of the latter samples, indicating that the parent compounds, *p,p'*-DDT and *o,p'*-DDT, degrade over time.

Data from Muir et al. (1995) suggest that DDTs are being transported atmospherically to the Arctic. These researchers collected surficial sediments from eight lakes in northern Canada and noted that concentrations of total DDT (ranging from 0.11 to 9.96 $\mu g \cdot kg^{-1}$) declined significantly with increasing latitude. These data support the cold condensation hypothesis, which suggests that the sequential volatilization and condensation of organic pollutants result in 'migration' of these substances towards the poles. However, levels of DDT, DDE, and DDD in freshwater sediments from the Canadian Arctic are generally lower than those reported in other parts of Canada. For example, the concentrations of DDT, DDE, and DDD ranged from 0.01 to 0.34 $\mu g \cdot kg^{-1}$ in Great Slave Lake sediments (Mudroch et al. 1989).

The limited information on historical and present concentrations of DDT in marine and estuarine sediments is primarily from British Columbia. Currently, concentrations of DDT in the Fraser River estuary and Boundary Bay are below the detection limit of <1 $\mu g \cdot kg^{-1}$ (Environment Canada 1998); however, sediments from Boundary Bay in 1994 had concentrations of total DDT as high as 8.1 $\mu g \cdot kg^{-1}$ (Swain and Walton 1994).

Additional Considerations

Regardless of the origin of DDTs in sediments, aquatic organisms may be adversely affected by exposure to elevated levels. The occurrence of adverse biological effects resulting from exposure to DDTs in sediment cannot be precisely predicted from concentration data alone, particularly in the concentration ranges between the

ISQGs and the PELs (Figures 1 to 5). The likelihood of adverse biological effects occurring in response to exposure of aquatic biota to DDTs at a particular site depends on the sensitivity of individual species, the endpoints examined, and a variety of physicochemical (e.g., hydrophobicity and water solubility), geochemical (e.g., TOC and particle size), and biological (e.g., feeding behaviour and physiology) factors that affect the bioavailability of DDTs.

Benthic organisms may be exposed to both particulate and dissolved forms of DDTs in interstitial or overlying waters, as well as to sediment-bound DDTs through surface contact and ingestion of sediment during feeding. Sediments and porewater are believed to represent the primary routes of exposure for infaunal and epibenthic species.

Several properties of DDTs influence their bioavailability to aquatic organisms. Hydrophobicity, as represented by the K_{ow}, and water solubility, have been identified as the two most important factors. Accordingly, substances with high K_{ow}s and low water solubilities are considered to be the most bioavailable to benthic organisms (Landrum et al. 1989). Based on their physicochemical properties, *p,p'*-DDT and *o,p'*-DDT are likely to be the most bioavailable of the six isomers discussed in this fact sheet.

Sediment characteristics, including TOC, particle size distribution, and clay content, may affect the bioavailability of DDTs in sediments (Landrum and Robbins 1990). For example, Nebecker et al. (1989) evaluated the toxicity of DDT to an amphipod, *Hyalella azteca*, in sediments with three levels of TOC (3.0%, 7.2%, and 10.5%) and observed that the toxicity of DDT was inversely proportional to the levels of TOC in the sediment. Physical and chemical characteristics of aquatic systems, such as water temperature, salinity, and nutrients, may also influence the uptake of DDTs by benthic organisms. The physiology and biochemistry of aquatic organisms also play important roles in the uptake and bioaccumulation of DDTs.

It should be noted that, although the ISQGs and PELs recommended for DDTs are developed to be protective of benthic invertebrates, they do not specifically account for the potential for adverse biological effects on higher trophic levels that may result from dietary exposure. Bioaccumulation to high levels in biota, as well as, biomagnification in food chains, are critical aspects of the environmental fate and behaviour of DDTs (WHO 1989). Canadian tissue residue guidelines for the protection of wildlife consumers of aquatic organisms have been developed to consider the effects of bioaccumulative substances, such as DDTs, on higher trophic levels (Environment Canada 1997b). Therefore, Canadian tissue residue guidelines should be used in conjunction with the ISQGs and PELs to evaluate the potential for adverse biological effects on other components of aquatic ecosystems.

Currently, the degree to which DDTs will be bioavailable at a particular site cannot be predicted conclusively from the physicochemical characteristics of sediments or attributes of endemic organisms (Environment Canada 1998). Nonetheless, the incidence of adverse biological effects associated with exposure to DDTs increases as concentrations of DDTs increase in a range of sediment types (Figures 1 to 5). Therefore, the recommended ISQGs and PELs for DDT, DDE, and DDD will be useful in assessing the ecotoxicological significance of DDTs in sediments.

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