Canadian Water Quality Guidelines for the Protection of Aquatic Life

PERMETHRIN

Permethrin (CAS Registry Number 52645-53-1; IUPAC name 3-phenoxybenzyl(1RS)-cis,trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-carboxylate) is a synthetic active ingredient used in various insecticide products registered for use in Canada. It is an odourless, colourless crystalline solid, or a viscous liquid that is white to pale yellow. Permethrin has the molecular formula $C_{21}H_{20}Cl_2O_3$ and a molecular weight of 391.30 (Kidd and James 1991). Permethrin is practically insoluble in water, with a solubility of 0.006 $mg \cdot L^{-1}$ at 20°C (Tomlin 2000), and is non-volatile with a vapour pressure of 1.5-2.5 µPa at 20°C (Wells et al. 1986). The octanol-water partitioning coefficient (log K_{ow}) is reported to range from 2.9 to 6.5, and the soil organic carbon partition coefficient (log K_{oc}) is reported to range from 1.3 to 2.8 (Schimmel et al. 1983; Montgomery 1993).

Permethrin, first synthesized in 1973, is an ester of the dichloro analogue of chrysanthemic acid and 3-phenoxybenzyl alcohol. Technical products comprise a mixture of four stereoisomers due to the chirality of the cyclopropane ring. The cis-trans isomer ratio is 2:3 and the optical ratio of 1R:1S is 1:1, a racemic mixture (IPCS 1990). Therefore, permethrin contains the [1R, trans], [1R, cis], [1S, trans] and [1S, cis] isomers in the approximate ratio of 3:2:3:2. The [1R, cis] isomer is the most insecticidally active among the isomers, followed by the [1R, trans] isomer.

Permethrin is registered for use in Canada in over 230 products, including technical grade active ingredient and formulated pesticides (PMRA 2004). Trade and other names used for permethrin-based pesticides include, but are not limited to, Ambush, Atroban, Dragnet, Ectiban, Evercide, Permanone, Pounce, Pramex and Raid Fumigator. The various permethrin-based pesticides registered in Canada are used for a wide variety of purposes including: general insecticide products for domestic use; flea and tick control on household pets; insect control on agricultural crops, orchards, nurseries and in greenhouses; biting insect control in livestock (e.g., treated ear tags); as a perimeter application for control of adult mosquitoes around buildings; application to military clothing and mosquito netting; and others. Permethrin is also registered for restricted use on commercial woodlots (PMRA 2004).

For application to agricultural crops, permethrin is available in dusts, emulsifiable concentrates and wettable powder formulations. It is used in Canada to control pests on nut, fruit, vegetable, tobacco, oil seed, ornamental, and cereal crops. Typical application rates for permethrin are 17-70 $g \cdot ha^{-1}$ on nursery trees and shrubs, 35-150 $g \cdot ha^{-1}$ on vegetables, 70-100 $g \cdot ha^{-1}$ on tobacco and cereals, and 100-200 $g \cdot ha^{-1}$ on fruit (PMRA 2004).

Permethrin is not produced in Canada, and information could not be found on quantities of permethrin that are imported. Sales and use data collected by individual jurisdictions can be used to give an indication of the amount of permethrin sold annually in Canada. The combined total annual sales for permethrin in Alberta, Ontario and Prince Edward Island is estimated at 1077.6 kg active ingredient (Environment Canada 2006). In Nova Scotia, approximately 150 litres of pesticide formulation containing permethrin as the active ingredient were sold in 2003 (D. Burns 2004, Nova Scotia Department of Environment and Labour, Halifax, Nova Scotia, pers. com.).

Given the wide variety of practical applications for permethrin in Canada, the sources of permethrin to the environment are multifold. Direct application of permethrin to water bodies is not permitted in Canada. Nonetheless, use of permethrin to control terrestrial pests could potentially result in unintended transport to aquatic habitats and indirect contamination through spray drift, atmospheric deposition, soil erosion and runoff.

Permethrin binds strongly to soil particles and is practically insoluble in water (Carroll et al. 1981; US DASCS 1990). Consequently, leaching rates of permethrin from soil are low (Carroll et al. 1981) and there is little risk of contamination of groundwater

Table 1. Water quality guidelines for permethrin for the protection of aquatic life (Environment Canada 2006).

Aquatic life	Guideline value ($\mu g \cdot L^{-1}$)
Freshwater	0.004*
Marine	0.001*

*Interim guideline.

(Wagenet et al. 1985), as reflected in the low Groundwater Ubiquity Score for permethrin of -1.5 (Vogue et al. 1994). Nevertheless, there have been detections of permethrin in groundwater, but at very low frequency, in both Canada (Briggins and Moerman 1995) and the U.S. (USGS 1998). Permethrin is slightly to non-persistent in soil with half-lives reported between 5-42 days (Kaufmann et al. 1977; Kaneko et al. 1978; Williams and Brown 1979; Carroll et al. 1981; Jordan et al. 1982; Kidd and James 1991; Wauchope et al. 1992). Permethrin is readily broken down in most soils except organic types. The rates of degradation vary depending on the isomer (*cis*- or *trans*-), environmental conditions (e.g., temperature, pH, moisture content, oxidation potential), and microbial community present (Carroll et al. 1981).

Permethrin degrades rapidly in water, primarily through the hydrolysis of the ester bond and oxidation (Lutnicka et al. 1999). Photolysis can also play a role in permethrin degradation (Rawn et al. 1982; Schimmel et al. 1983). Permethrin is more persistent in sediments than in water (Hartley and Kidd 1983; Wagenet et al. 1985). In a laboratory adsorption-desorption study, more than 95% of permethrin in aqueous solution was rapidly adsorbed to sediment, and desorption was minimal even after several water rinses (Sharom and Solomon 1981). Permethrin has been found to bind to suspended solids, dissolved organic matter and sediments (Liu et al. 2004; Lee et al. 2004). Adsorption to coarse solid phases may render permethrin unavailable to microorganisms, thus prolonging its persistence; however, sorption to fine particles, algal cells and bacterial biofilms in sediment may actually enhance the bioavailability of permethrin to benthic invertebrates (Allan et al. 2005). Typical degradation products from ester hydrolysis of permethrin include 3-phenoxybenzyl alcohol, 3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxilic acid, 3-(2,2-dichlorovinyl)-2-methylcyclopropane-1,2-dicarboxilic acid and 3-(4-hydroxyphenoxy)-benzyl-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate, amongst others (Jordan and Kaufman 1986; Kaneko et al. 1978; Leahey and Carpenter 1980; Rawn et al. 1982). Microbial metabolism of 3-phenoxybenzyl alcohol also typically produces 3-phenoxybenzoic acid (Kaufman et al. 1981). Many of these degradation products are further oxidized and degraded and, depending on environmental conditions, can undergo complete mineralization (Jordan et al. 1982; Penick Corporation 1979).

Analytical methods for measuring permethrin in water generally use gas chromatography-mass spectrometry, but may differ in their methods for extraction and detection. Specific methods include those used by Environment Canada's National Laboratory for Environmental Testing (Ed Sverko 2005, National Laboratory for Environmental Testing, Environment Canada, Burlington, Ontario, pers. com.), the Ontario Ministry of the Environment (OMOE 2002) and Bonwick et al. (1995), with detection limits ranging from 0.0005 to 0.02 $\mu g \cdot L^{-1}$.

Only limited data are available on permethrin concentrations in Canadian waters. In a 2003 study from Ontario (John Struger 2004, Environment Canada, Burlington, Ontario, pers. com.) and a 1996 study from Quebec (Giroux 1998), water samples from streams in agricultural areas showed no detection or only trace amounts of permethrin. In British Columbia, 6 sites in the Lower Fraser Valley and Duck Lake were monitored for permethrin. Concentrations were below the detection limit in all but two sites, Cohilickhan Slough (2.70 $ng \cdot L^{-1}$) and Hope Slough (0.61 $ng \cdot L^{-1}$) (Environment Canada 2004). In PEI, water and sediment samples were collected and analyzed for permethrin from three watersheds (Souris, Wilmot and Mill) in 2003. Permethrin was not detected in any of the run-off or stream water samples collected (detection limit of 5 $\mu g \cdot L^{-1}$), but was detected in 4 out of 30 samples of stream sediments, with the highest concentration measured at 10.85 $\mu g \cdot kg^{-1}$ (Environment Canada 2004; Jamie Mutch 2005, PEI Dept. of Environment, Energy and Forestry, Charlottetown, PEI, pers. com.). A study in which surficial sediments from 60 tributaries to Lake Ontario and Lake Erie were analyzed found that *cis*-permethrin was detected in 3% of the sediments, and *trans*-permethrin in 2% (Environment Canada 2004).

Permethrin is a neurotoxin that acts on the axons in the peripheral and central nervous system (IPCS 1990). It prolongs sodium ion permeability of neuron membranes, which results in repetitive activity in the sensory and motor systems (IPCS 1984). Symptoms of intoxication caused by this insecticide include restlessness, incoordination, hyperactivity, prostration and paralysis (Gammon et al. 1981). Permethrin is very highly toxic to aquatic invertebrates and fish (Jarboe and Romaire 1991; Mokry and Hoagland 1990; Holdaway and Dixon 1988), only slightly toxic to some algae (Stratton and Corke 1982), and practically non-toxic to mammals and birds which have the ability to metabolize permethrin rapidly (Hunt and Gilbert 1977; IPCS 1990). Field studies have demonstrated that where permethrin was intentionally introduced to the aquatic environment (e.g., streams, lakes), it has had a major impact on the invertebrate community in that environment. Effects observed include increased invertebrate drift density and invertebrate community changes (Kreutzweiser and Sibley 1991; Werner and Hilgert 1992).

Major metabolites of permethrin are much less toxic than the parent compound to invertebrates and fish (Zitko et al. 1977; Hill 1985). With algae, however, certain metabolites of permethrin appear to be more toxic than permethrin itself (Stratton 1981). Nonetheless, concentrations of metabolites that are toxic to algae are still orders of magnitude higher than concentration of permethrin that are toxic to aquatic invertebrates and fish. Therefore, water quality guidelines for permethrin that are protective of invertebrates will also protect against effects of resulting metabolites on algae.

Environment Canada (2006) provides a review of bioconcentration factors (BCFs) that have been measured for permethrin in freshwater and marine organisms. BCFs reported in the literature range from 44 to 2800, indicating that it is not bioaccumulative.

Water Quality Guideline Derivation

The interim Canadian water quality guidelines for permethrin for the protection of freshwater and marine life were developed based on the CCME protocol (CCME 1991). For more information, see the scientific supporting document (Environment Canada 2006).

Freshwater Life

The most sensitive chronic fish study was conducted by Kumaraguru and Beamish (1986). The authors exposed rainbow trout to permethrin over a 1-6 week period in continuous flow tanks. Small (10 g) and large (100 g) fish were used to determine the impact of permethrin on the growth rate of trout. The authors reported a 21-day LOEC of 0.65 $\mu g \cdot L^{-1}$ for the small trout, while the large trout were more tolerant. Fathead minnows appear to be slightly less sensitive, with a 32-d LOEC of 1.4 µg a.i.$\cdot L^{-1}$ reported for reduced survival and impaired swimming ability (Spehar et al. 1983).

Acute 96-h LC_{50} values for freshwater fish ranged from 0.62 µg a.i.$\cdot L^{-1}$ for juvenile rainbow trout (*Oncorhynchus mykiss*) (Kumaraguru and Beamish 1981) to 540 µg a.i.$\cdot L^{-1}$ for juvenile flagfish (*Jordanella floridae*) (Holdway and Dixon 1988). Other sensitive species include white sucker (*Catostomus commersonii*), Lahontan cutthroat trout (*Oncorhynchus clarkii hensawi*) and Apache trout (*Oncorhynchus gilae apache*) with 96-h LC_{50} values of 1.0, 1.6, and 1.7 µg a.i.$\cdot L^{-1}$, respectively (Holdway and Dixon 1988; Sappington et al. 2001).

A limited number of chronic studies were available for invertebrates. Anderson (1982) investigated the effects of permethrin on the behaviour and survival of stonefly nymphs (*Pteronarcys dorsata*) and caddisfly larvae (*Brachycentrus americanus*) by exposing them in a flow-through system. The 21-d LC_{50} for *B. americanus* was estimated at 0.17 µg a.i.$\cdot L^{-1}$. A 21-d LOEC of 0.042 µg a.i.$\cdot L^{-1}$ was reported for immoblization of *P. dorsata*. McLoughlin et al. (2000) observed similar sensitivity in the amphipod *Gammarus pulex*, with a 6-d LOEC for reduced feeding rate reported at 0.06 µg a.i.$\cdot L^{-1}$.

Acute LC_{50} values for freshwater invertebrates ranged from 0.17 µg a.i.$\cdot L^{-1}$ for the amphipod (*Gammarus pulex*) (McLoughlin et al. 2000) to 940 µg a.i.$\cdot L^{-1}$ for the beaver-tail fairy shrimp (*Thamnocephalus platyurus*) (Sánchez-Fortún and Barahona 2005). A number of studies have also shown *Daphnia magna* to be sensitive to permethrin. Stratton and Corke (1981) reported 48-h LC_{50} values of 0.2 and 0.6 µg a.i.$\cdot L^{-1}$ for juvenile and adult *D. magna*, respectively. Similar 48-h LC_{50} values of 0.43 and 1.06 µg a.i.$\cdot L^{-1}$ were reported in another study with adult *D. magna* (Stratton and Giles 1990), and Thurston et al. (1985) reported 48-h LC_{50} values of <1.4 and <2.5 µg a.i.$\cdot L^{-1}$ for juvenile *D. magna*. Other sensitive invertebrates include the adult crayfish *Orconectes immunis* with a 96-h LC_{50} of <1.2 µg a.i.$\cdot L^{-1}$ (Thurston et al. 1985), larvae of the midge *Tanytarsus dissimilis* with a 48-h LC_{50} of <2.5 µg a.i.$\cdot L^{-1}$ (Thurston et al. 1985), and nymphs of the damselflies *Enellagma* spp. and *Ishnura* spp. with a 24-h LC_{50} of 2.9 µg a.i.$\cdot L^{-1}$ (Siegfried 1993).

Algae are not particularly sensitive to permethrin. Stratton and Corke (1982) examined the toxicity of permethrin and its degradation products towards algae and cyanobacteria. Test cultures included the blue-green algae *Anabaena inaequalis*, *A. cylindrica*, and *A. variabilis*, and the green algae *Chlorella pyrenoidosa* and *Scenedesmus quadricauda*. Permethrin was found to be non-toxic to most of these organisms at the concentrations tested (0 to 1000 µg a.i.$\cdot L^{-1}$). The one exception was the blue-green algae *A. inaequalis* where an EC_{50} of 1600 µg a.i.$\cdot L^{-1}$ for growth was reported.

Acute toxicity values reported for amphibians include a 96-h LC_{50} of 18.2 µg a.i.$\cdot L^{-1}$ for tadpoles of the southern leopard frog (*Rana sphenocephala*) (Bridges et al. 2002) and a 96-h LC_{50} of 115 µg a.i.$\cdot L^{-1}$ for tadpoles of the bullfrog (*Rana catesbeiana*) (Thurston et al. 1985).

The interim water quality guideline for permethrin for the protection of freshwater life is 0.004 µg a.i.$\cdot L^{-1}$. It was derived by multiplying the 21-d LOEC of 0.042 µg a.i.·L^{-1} for the stonefly (*P. dorsata*) (Anderson 1982) by a safety factor of 0.1 (CCME 1991).

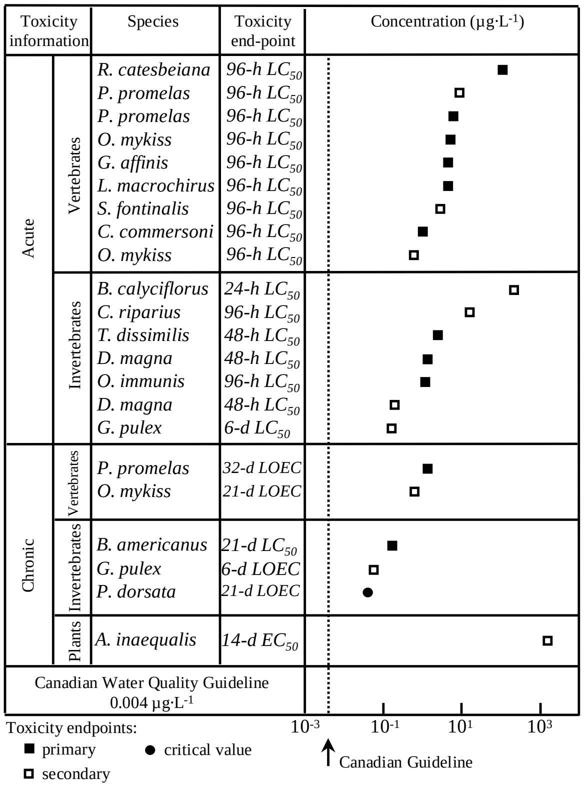

Figure 1. Select freshwater toxicity data for permethrin

Marine Life

Only one chronic study was available for marine fish. Hansen et al. (1983) conducted 28-day embryo-larval toxicity tests using the estuarine species sheepshead minnow (*Cyprinodon variegatus*). Decreased survival was the most sensitive measure of effect in fish exposed to permethrin with NOEC and LOEC values of 10 and 22 µg a.i.·L^{-1}, respectively (Hansen et al. 1983).

Acute 96-h LC_{50} values for marine fish ranged from 2.2 µg a.i.·L^{-1} for Atlantic silversides (*Menidia menidia*) (Schimmel et al. 1983) to 88 µg a.i.·L^{-1} for sheepshead minnow (*C. variegatus*) (Borthwick and Walsh 1981).
Toxicity data for marine invertebrates were only available for acute exposures. Values ranged from a 96-h LC_{50} of 0.018 µg a.i.·L^{-1} for larvae of the stone crab (*Menippe mercenaria*) (Borthwick and Walsh 1981) to a 24-h LC_{50} of 8210 µg a.i.·L^{-1} for larvae of the San Francisco brine shrimp (*Artemia fransciscana*) (Sánchez-Fortún and Barahona 2005). The mysid (*Mysidopsis bahia*) is sensitive to permethrin, with reported 96-h LC_{50} values of 0.02, 0.046 and 0.095 µg a.i.·L^{-1} (Schimmel et al. 1983; Borthwick and Walsh 1981; Cripe 1994). Other sensitive species include the sand shrimp (*Crangon septemspinosa*) with a 96-h LC_{50} of 0.13 µg a.i.·L^{-1} (McLeese et al. 1980) and the pink shrimp (*Penaeus duorarum*) with 96-h LC_{50} values of 0.17 and 0.22 µg a.i.·L^{-1} (Cripe 1994; Schimmel et al. 1983).

Permethrin toxicity data on marine algae were only available for one species, the diatom *Skeletonema costatum*. Walsh and Alexander (1980) reported 96-h EC_{50} values for reductions in diatom cell count and biomass of 68 and 72 µg a.i.·L^{-1}, respectively. Similar results were reported by Borthwick and Walsh (1981) who observed 96-h EC_{50} values for growth reduction in two assays with *S. costatum* of 92 and 124 µg a.i.·L^{-1}.
The interim water quality guideline for permethrin for the protection of marine life is 0.001 µg a.i.·L^{-1}. It was derived by multiplying the 96-h LC_{50} value of 0.02 µg a.i.·L^{-1} for *M. bahia* (Schimmel et al. 1983) by an acute application factor of 0.05 for nonpersistent substances (CCME 1991).

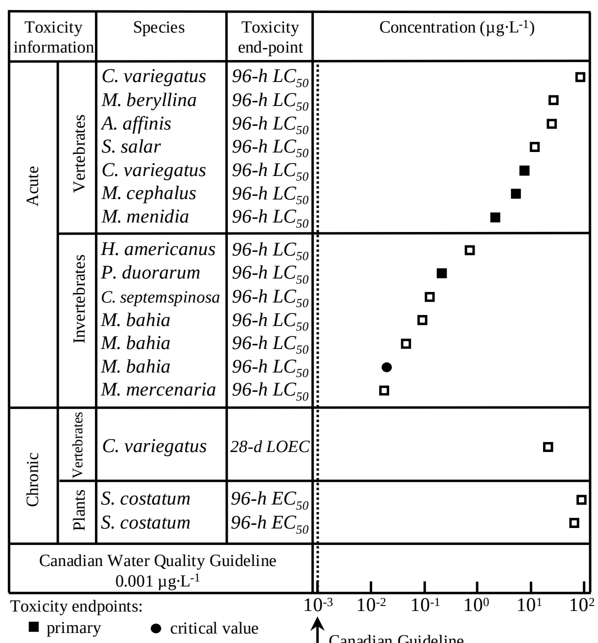

Figure 2. Select marine toxicity data for permethrin

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For further scientific information, contact:

Environment Canada
National Guidelines and Standards Office
351 St. Joseph Blvd.
Gatineau, QC K1A 0H3
Phone: (819) 953-1550
Facsimile:(819) 956-5602
E-mail: ceqg-rcqe@ec.gc.ca
Internet: http://www.ec.gc.ca

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