6. POTENTIAL FOR HUMAN EXPOSURE

6.1 OVERVIEW

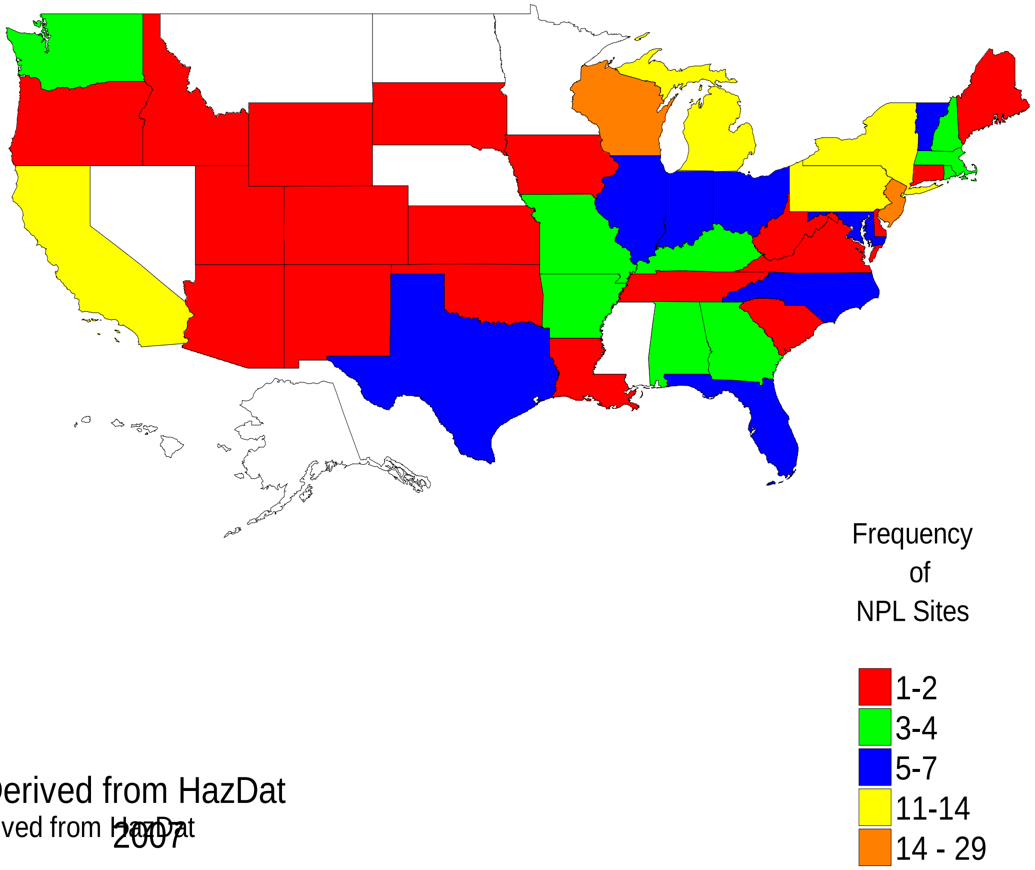
Endosulfan has been identified in at least 176 of the 1,699 hazardous waste sites that have been proposed for inclusion on the EPA National Priorities List (NPL) (HazDat 2007). However, the number of sites evaluated for endosulfan is not known. The frequency of these sites can be seen in Figure 6-1. Of these sites, 174 are located within the United States, 1 is located in Guam, and 1 is located in the Virgin Islands (not shown).

Endosulfan (consisting of 7:3 ratio of α-endosulfan to β-endosulfan) is released to the environment mainly as the result of its use as a restricted-use insecticide. It is not authorized for residential use in the United States. Beginning in July 2012, a voluntary cancellation and phase-out began and is scheduled to be finalized in 2016.

After its release to the environment, endosulfan undergoes a variety of transformation and transport processes. In soil, endosulfan sulfate is the major degradation product from biotic metabolism and is considered to be more persistent than the parent isomers. Neither the α- or β- isomers nor the sulfate are expected to be mobile in soil. Soil erosion, run-off, spray drift, and atmospheric deposition contribute to releases of endosulfan to aquatic ecosystems. In water, hydrolysis to the less toxic endosulfan diol is expected to be the dominant transformation pathway. Volatilization from soil, water, plant surfaces, and transport in dust particles in addition to direct release from spray drift will result in atmospheric levels of endosulfan. Even though monitoring data suggest that most atmospheric endosulfan exists in the vapor-phase rather than the particulate-phase, the relative stability of endosulfan in the atmosphere contributes to its long range transport. Wet deposition of atmospheric endosulfan to remote, high-elevation regions (known as “cold-mountain air trapping”) has been documented in areas of the Sierra Nevada Mountains and the Canadian Rockies. Long-range transport of endosulfan to Arctic regions has been documented and residues have been detected in various Arctic environmental media and biota.

Endosulfan residue concentrations are highest and most prevalent in or nearby regions with intense agricultural activity. Residues have been detected in a variety of media including surface water, sediments, air, aquatic vertebrates, and invertebrates, terrestrial organisms, and in humans. α-Endosulfan, β-endosulfan, and endosulfan sulfate have been detected in a variety of food products during market basket monitoring. Residues are generally higher in fruits and vegetables versus processed foods. As a result, dietary intake is expected to be the major route of exposure to the general adult population and children.

Figure 6-1. Frequency of NPL Sites with Endosulfan Contamination



Derived from HazDat

)erived from HazDat

2007

The presence of endosulfan residues in placenta, cord blood, and breast milk suggests that pre- and post-natal exposure may occur. However, farm workers are expected to have the highest levels of exposures. Estimated risk levels were high for almost all occupation exposure scenarios associated with pesticide handlers. Exposures associated with post-application scenarios (e.g., cutting, weeding) are also expected to be high.

6.2 RELEASES TO THE ENVIRONMENT

The Toxics Release Inventory (TRI) data should be used with caution because only certain types of facilities are required to report (EPA 2005). This is not an exhaustive list. Manufacturing and processing facilities are required to report information to the TRI only if they employ 10 or more full-time employees; if their facility is included in Standard Industrial Classification (SIC) Codes 10 (except 1011, 1081, and 1094), 12 (except 1241), 20–39, 4911 (limited to facilities that combust coal and/or oil for the purpose of generating electricity for distribution in commerce), 4931 (limited to facilities that combust coal and/or oil for the purpose of generating electricity for distribution in commerce), 4939 (limited to facilities that combust coal and/or oil for the purpose of generating electricity for distribution in commerce), 4953 (limited to facilities regulated under RCRA Subtitle C, 42 U.S.C. section 6921 et seq.), 5169, 5171, and 7389 (limited S.C. section 6921 et seq.), 5169, 5171, and 7389 (limited to facilities primarily engaged in solvents recovery services on a contract or fee basis); and if their facility produces, imports, or processes ≥25,000 pounds of any TRI chemical or otherwise uses >10,000 pounds of a TRI chemical in a calendar year (EPA 2005).

Endosulfan has been released to the environment mainly as a result of its use as an insecticide. There are no known natural sources of the compound. Endosulfan and endosulfan sulfate are not contained in the list of chemicals for which releases are required to be reported to EPA for the SARA Section 313 Toxic Release Inventory (TRI) (EPA 1997).

6.2.1 Air

There is no information on releases of endosulfan to the atmosphere from manufacturing and processing facilities because these releases are not required to be reported (EPA 2005).

As a result of its use as an insecticide on fruit trees, vegetables, and other crops, endosulfan is released directly to the atmosphere during application. The compound is applied principally by aerial spray, ground spray, and airblast. The direct release to the atmosphere is commonly a result of spray drift, which immediately contaminates the air surrounding the application area. Volatilization and wind suspension occurring from post-application periods can also be a source of endosulfan in air. Atmospheric endosulfan derived from these sources has the potential to contribute to regional and long-range transport (EPA 2010a).

6.2.2 Water

There is no information on releases of endosulfan to water from manufacturing and processing facilities because these releases are not required to be reported (EPA 2005).

Endosulfan is most commonly released to water by atmospheric deposition, spray drift, runoff, and erosion. Direct release to water bodies is restricted and application restrictions require a buffer distance of 300 feet from surface waters for aerial application and 100 feet for ground application. In California, the buffer is 300 feet for both types of applications (EPA 2010a). Endosulfan is not expected to leach through soil to groundwater based on its low water solubility and its tendency to absorb to soil (EPA 2010a; HSDB 2010).

6.2.3 Soil

There is no information on releases of endosulfan to the soil from manufacturing and processing facilities because these releases are not required to be reported (EPA 2005).

The main routes of release of endosulfan to soils are direct application to crops and atmospheric deposition from spray drift, volatilized material, or from long-range atmospheric transport (EPA 2010a).

6.3 ENVIRONMENTAL FATE

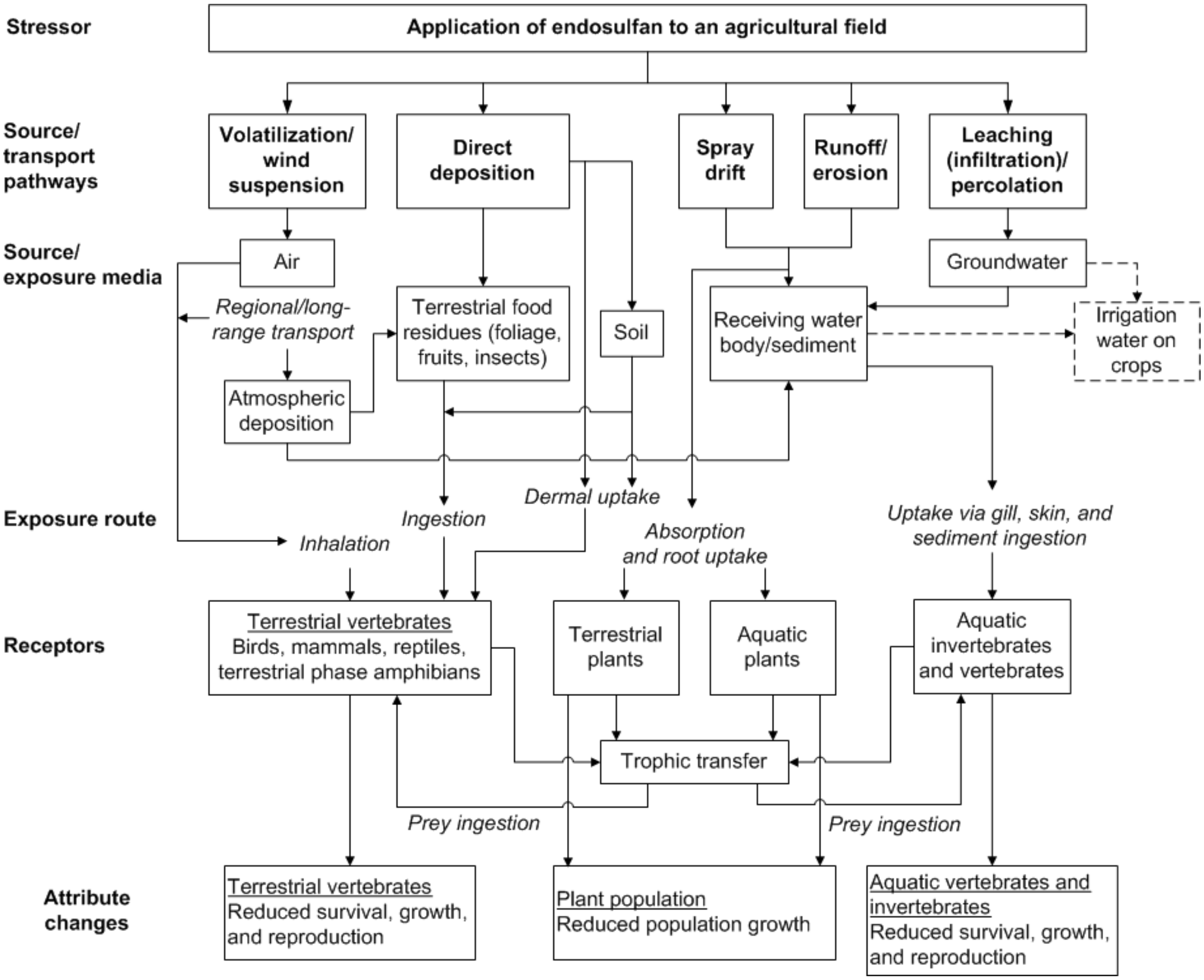
6.3.1 Transport and Partitioning

Endosulfan isomers and endosulfan sulfate are found throughout the environment in various media due to its widespread use, physical properties, and relative persistence. Figure 6-2 contains a conceptual model of how endosulfan moves between environmental compartments starting from field application to potential ecological receptors (EPA 2010a). However, transport and partitioning of endosulfans can be complex and depends greatly on environmental conditions (Weber et al. 2010).

Endosulfan is applied directly to soil through application to crops by aerial, hand, groundboom, airblast, rights-of-way, or backpack sprays, or by chemigation (EPA 2002). Measured K_{oc} values (average for four soils) for α- and β-isomers were 10,600 and 13,500 mL/g, respectively, indicating limited mobility in soil (EPA 2010a). Therefore, leaching to groundwater is not expected to be a concern. This is supported by available groundwater monitoring data, which indicate very low rates of detection in extensive groundwater monitoring programs (USGS 2012b). Model field studies also support this conclusion. Endosulfan did not leach from sandy loam soil following incorporation of 6.7 kg/hectare of the compound (Stewart and Cairns 1974). After sampling periods of 503–828 days, 90% of the residues were found in the top 0–15 cm of soil, 9% at 15–30 cm, and 1% at 30–45 cm. In model soil evaporation beds constructed to test the feasibility of treating pesticide wastes, endosulfan exhibited no movement in loamy sand soil beds up to 54 weeks after the start of the tests (Hodapp and Winterlin 1989). Endosulfan is metabolized in soil to endosulfan sulfate, which is also expected to be immobile based on its estimated K_{oc} of 9,800 (EPA 2010a).

Although it is not applied directly or in the vicinity of water bodies, endosulfan is transported to water via spray drift or atmospheric deposition, or through soil runoff and erosion (EPA 2002, 2010a). Endosulfan has been regularly detected in surface water samples taken from South Florida canals that drain agricultural areas (Harman-Fetcho et al. 2005; Pfeuffer 2011; Scott et al. 2002). Endosulfan transported to water is expected to eventually partition to sediments (EPA 2010a; Weber et al. 2010). However, estimated log air/water partition coefficients for α- and β-endosulfan and endosulfan sulfate ranging from -3.56 to -4.78 indicate that volatilization from water to air is expected to occur, and these chemicals can be considered semi-volatile (EPA 2010a). In a field dissipation study, volatilization was considered to be the dominant route of dissipation for endosulfan and endosulfan sulfate in cotton fields of sub-tropical India. High temperatures and low rainfall were likely influential factors for this behavior (Kathpal et al. 1997). Volatilization of α-endosulfan accounted for 34.5% of total losses from freshly tilled soil during another field study, while volatilization losses of β-endosulfan were much less (14.5%), indicating that the β-isomer is less volatile than the α-isomer (Rice et al. 2002). Volatilization from plant surfaces is also expected to occur, and may be more significant than from soil surfaces. In air sampling studies done in a wind tunnel, 12% of the initial endosulfan application volatilized from a silty sand soil after 24 hours, as compared to 60% from plant surfaces in 24 hours (Rudel 1997). When pure β-endosulfan was allowed to equilibrate in the apparatus, the ratio of the β-isomer to the α-isomer in the gas phase became 8:92 at 20 °C, suggesting a β- to α- conversion (Rice et al. 1997). This conversion would also contribute to total volatilization losses of endosulfans from treated fields.

Figure 6-2. Conceptual Model of the Potential Effects of Endosulfan Application on Ecological Receptors



Note: Endosulfan is expected to absorb strongly to soils and is not likely to leach to groundwater.

Source: EPA 2010a

Because of its semi-volatile nature and relative stability in the atmosphere, endosulfan is susceptible to long-range transport in the environment (Weber et al. 2010). These transport and deposition processes can be localized, regional, or long-range. Atmospheric deposition rates of endosulfans (α-, β-, and sulfate) were estimated in the agricultural intensive Choptank River watershed of the Chesapeake Bay region. Total wet deposition (± combined absolute and relative error) for α-endosulfan was estimated at 0.96 ±0.1 kg/year. Estimated depositions for β-endosulfan and endosulfan sulfate were 2.7±0.3 and 0.5±0.06 kg/year, respectively. Deposition processes can be regional in nature. For example, probabilistic source contribution function (PSCF) modeling performed by Hafner and Hites (2003) suggests that atmospheric endosulfans in the Great Lakes Region are generated in the lower Michigan peninsula, and New York State, and to a lesser extent Pennsylvania.

Regional transport of endosulfans to remote, non-agricultural areas has been documented. Endosulfan residues have been detected in air, snow, rain, lichen, surface water, and sediment samples from Yosemite National Park, California. West to east atmospheric movement of these residues from the agricultural San Joaquin Valley is the likely source. Estimated winter and summer deposition rates of endosulfans (α-, β-, and sulfate) were calculated from data collected during the spring and summer of 2009. Winter deposition was estimated at 1.11 $\mu g/m^2$ with 72% of the total deposition occurring during this season. Summer deposition was estimated at 0.44 $\mu g/m^2$ making the total annual deposition 1.55 $\mu g/m^2$. The dominance of winter deposition is due to the fact most of the precipitation in this region occurs as snow (Mast et al. 2012a, 2012b). Long-range transport to remote regions is evident in the numerous studies that have monitored endosulfan in Arctic environmental media (Hageman et al. 2006a; Hung et al. 2005, 2010; Stern et al. 2005; Weber et al. 2010). Like other organochlorine pesticides in Arctic areas, α-endosulfan has been observed to undergo a “spring maximum event”, where air concentration peaks in the spring (April/May) and again in the fall (October/November). This is different from seasonal trends observed in temperate regions where air concentrations peak in the summer months (Hung et al. 2005).

Organochlorine pesticides like endosulfan undergo a phenomenon known as “cold mountain trapping” where cold temperatures (leading to condensation) and high precipitation rates of high-elevation areas in temperate regions cause increased deposition (mostly through snow) (Daly et al. 2005). This phenomenon for endosulfan has been noted in the results of monitoring activities in National Parks in western United States (Hageman et al. 2006a), in Yosemite National Park (Mast et al. 2012a, 2012b), and in National Parks of western Canada (Daly et al. 2007) (see Section 6.4).

Bioaccumulation and biomagnification potential of endosulfan in organisms varies, but generally suggests that it has the potential to bioaccumulate in organisms and biomagnify in the food webs. This potential has been extensively investigated in aquatic organisms. EPA's Ecological Fate and Risk Assessment for Endosulfan (2010a) extensively summarized experimental bioconcentration factors (BCFs) and bioaccumulation factors (BAFs) for aquatic organisms available in the scientific literature. BCF values ranged from 17.1 to 11,583 in fish species (Hansen and Cripe 1991; Jonsson and Toledo 1993; Schimmel et al. 1977; Toledo and Jonsson 1992; Rajendran and Venugopalan 1991). However, values from Hansen and Cripe (1991) and Schimmel et al. (1977) that met quality screening criteria were 1,146 (in sheepshead minnow for α- and β-isomers) and 2,755 (in striped mullet for α- and β-isomers and endosulfan sulfate). Experimental BCF values in aquatic invertebrates (shrimp, mussel, oyster, clam, and crayfish) ranged from 12 to 600 (EPA 2010a; Ernst 1977; Rajendran and Venugopalan 1991; Roberts 1972; Schimmel et al. 1977). Mesocosm and microcosm bioaccumulation studies reported BAF values similar in range to BCF values ranging from 115 to 1,262 in Bluegill fish, water flea, green algae, oyster, and macrophytes (DeLorenzo et al. 2002; EPA 2010a; Pennington et al. 2004; and unpublished industry studies). BAF is similar to BCF except that BAF takes into account multiple routes of exposure and not just intake from gills. Weber et al. (2010) summarized BAFs based on monitoring data reported for Arctic aquatic fish and mammals. Wet weight BAF values (for α- and β-isomers and endosulfan sulfate) were 1,690 (Arctic char), 7,280 (salmon), and 3,260 (Arctic cod). Lipid weight BAFs (for α- and β-isomers and endosulfan sulfate) were 1.45×10^5 (whole salmon), 3.13×10^5 (whole Arctic cod), 6.76×10^5 (female beluga blubber), 5.65×10^5 (male beluga blubber), 2.21×10^5 (female ringed seal blubber), 2.41×10^5 (male and female ringed seal blubber), and 2.98×10^4–3.52×10^4 (Arctic char muscle). Biomagnification factors (BMFs) were 2.2 for cod to beluga, 1.5 for salmon to beluga, and 0.77 for cod to seals. BMF values represent the ratio of the level of chemical in the predictor versus the concentration in its diet and BMF values greater than 1 indicate a potential to biomagnify up the food web. However, these BMF values may be overestimated since they do not account for metabolism.

Assessing the bioconcentration and biomagnification potential in terrestrial organisms is difficult. Kelly et al. (2007) estimated BMFs for air-breathing organisms ranging from 4.9 to 23 for β-endosulfan. However, this model did account for metabolism. Armitage and Gobas (2007) suggest that application of soil-earthworm-shrew food-chain model illustrates that chemicals with log $K_{oa} \geq 5.25$ and $\log K_{ow}$ values ranging from 1.75 to 12 may biomagnify given that they are not rapidly metabolized (half-life ~2.5 days). Estimated $\log K_{oa}$ values are 6.41 for both isomers and 8.45 for endosulfan sulfate. A measured log K_{oa} value for both isomers is reported at 8.64 (EPA 2010a). Analyses of two cases of intentional endosulfan poisoning reported terminal half-lives in blood serum of 15.2 hours (from 35% endosulfan formulation) and 8.8 hours (endosulfan content unknown) (Eyer et al. 2004). These half-lives suggest that metabolism of endosulfan may significantly attenuate biomagnification in the terrestrial food chain, based on the Armitage and Gobas model. Although there is a lack of comprehensive, standard biological monitoring data in humans, a variety of endosulfan metabolites (-sulfate, -ether, -lactone, -diol) have been detected in adipose tissue, placenta, cord blood, and breast milk of women at nanoscale concentrations. It is unclear the role of continuous, low-dose exposures and metabolism play in these concentrations (Cerrillo et al. 2005). At the very least, these data indicate that human metabolism of endosulfan occurs and may be complex.

6.3.2 Transformation and Degradation

6.3.2.1 Air

The α- and β-isomers of endosulfan are considered to be stable to direct photolysis in the atmosphere because they do not absorb light at wavelengths >300 nm (EPA 2010a). Photolysis of endosulfan isomers has been observed under laboratory conditions using polar solvents and various surface media, but these results are not likely to be relevant in atmospheric conditions. The α-isomer undergoes isomerization to the β-isomer, which is relatively more stable (Dureja and Mukerjee 1982). Both isomers undergo oxidation to endosulfan sulfate via several processes in the environment. Vapor-phase α- and β-endosulfan are expected to be photooxidized by hydroxyl radicals in the atmosphere. The half-life for this reaction has been estimated at about 2 days, assuming a hydroxyl radical concentration of 5×10^5molecules per cm^3 and 12-hour days for both isomers. Reaction of α- and β-endosulfan isomers with atmospheric ozone has been estimated to have a half-life of about 320 days, assuming an ozone concentration of 7×10^{11} molecules per cm^3. Direct photolysis data for endosulfan sulfate are conflicting. Observations from field studies suggest photolysis may play a role in endosulfan sulfate disappearance. A half-life of about 4 days was estimated for reaction with hydroxyl radicals (EPA 2011).

6.3.2.2 Water

Endosulfan undergoes hydrolysis to endosulfan diol in surface water and groundwater. The rate of hydrolysis is influenced by pH, and the values reported in the literature vary somewhat. Under aerobic conditions, both hydrolysis and oxidation of endosulfan can occur, while under anaerobic conditions, only hydrolysis can occur. The hydrolytic half-lives for α- and β-endosulfan under anaerobic conditions at pH 7 were 35.4 and 37.5 days, respectively. At pH 5.5, the half-lives were 151 and 187 days, respectively. In the presence of ferric hydroxide, hydrolysis rates increased at pH 7 and 20 °C (Greve and

Wit 1971). Under aerobic conditions, the half-lives decreased. At pH 7, the half-lives of the chemical degradation (hydrolysis and oxidation) of both α- and β-endosulfan were 23 and 25 days, respectively. At pH 5, the half-lives were 54 and 51 days, respectively. At 20 °C and pHs of 5.5 and 8.0, the half-lives of α-endosulfan in distilled water were 11.3 and 5.3 days, respectively (Kaur et al. 1998). The hydrolysis half-life of α- and β-endosulfan at pH 5 was >200 days, while the half-life at pH 7 was 11 days for α-endosulfan, 19 days for β-endosulfan, and 184 days for endosulfan sulfate. The major degradation product was endosulfan diol, which is considered to be less toxic than the parent compounds or endosulfan sulfate (EPA 2010a).

Endosulfan degradation in a water-sediment system was analyzed in a European study submitted to the EPA by a registrant. The Ohlau system consisted of water at pH 6.8 and sand sediment at pH 6.1 (0.1% organic carbon) and conducted for 120 days at 20ºC. Despite deficiencies concerning test substance purity and stated redox potential, results were considered acceptable in characterizing endosulfan in an aquatic system featuring an aerobic water column and anaerobic sediment. Data indicated relatively rapid transformation of the endosulfan isomers to the endosulfan diol, presumably by hydrolysis (half-lives ranging from 11 to 16 days). Within a month, the endosulfan parents degraded to about 10% of the nominal amount and the diol reached its maximum formation of 35%. Subsequently, the formation of degradates is dominated by the hydroxyl carboxylic acid (44% after 120 days) and endosulfan sulfate (25% after 120 days). Endosulfan sulfate was presumed to be formed by oxidation in the anaerobic sediment, and did not appear to decline after 50 days (EPA 2010a).

Biotic and abiotic transformations of endosulfan in seawater/sediment microcosms have been reported by Cotham and Bidleman (1989). In biotic tests, half-lives for α- and β-endosulfan in seawater-only microcosms (pH≥8) were about 5 and 2 days, respectively. In seawater-only microcosms under sterile conditions at a pH of ≥8, the half-life for α-endosulfan was 2–3 days, whereas the half-life for β-endosulfan was 1–2 days. Half-lives were longer in seawater/sediment microcosms, possibly because of the lower pHs (7.3–7.7) in these test systems; half-lives were 22 and 8.3 days for α- and β-endosulfan, respectively. Endosulfan diol was the main metabolite identified.

6.3.2.3 Sediment and Soil

Biodegradation of endosulfan isomers to endosulfan sulfate is expected to be the dominant fate pathway in soils (EPA 2010a). Endosulfan has been shown to be biodegraded by a wide variety of soil microorganisms in numerous studies. Sixteen of 28 species of fungi, 15 of 49 species of soil bacteria, and

3 of 10 species of actinomycetes metabolized radiolabeled endosulfan in a laboratory study under aerobic conditions (Martens 1976). Endosulfan sulfate was the major product of the fungal metabolism, whereas the bacterial transformation produced endosulfan diol. Degradation of endosulfan by soil fungi and bacteria has also been reported (El Beit et al. 1981). Biotransformation occurs under both aerobic and anaerobic conditions. Aerobic incubation of soil with endosulfan yielded mainly endosulfan sulfate (30–60%), some endosulfan diol (2.6%), and endosulfan lactone (1.2%) (Martens 1977). Flooded (anaerobic) incubation produced mainly endosulfan diol (2–18%), endosulfan sulfate (3–8%), and endosulfan hydroxyether (2–4%). In aqueous nutrient media (20 °C) containing a mixed culture of microorganisms isolated from a sandy loam soil, endosulfan was reported to be transformed to endosulfan diol with half-lives of about 1.1 and 2.2 weeks for α- and β-endosulfan, respectively (Miles and Moy 1979). The predominant formation of endosulfan diol in aquatic systems may indicate hydrolytic degradation rather than biodegradation.

A two-membered bacterial coculture was found to aerobically degrade α- and β-endosulfan efficiently without accumulating any of its metabolites. However, the degradation of soil-bound endosulfan was 4 times slower than in culture media; only 50% of the material (initially at 50 ppm) was degraded in 4 weeks (Awasthi et al. 1997). In an aerobic soil metabolism study using five different soils, half-lives of α-endosulfan ranged from 35 to 67 days and half-lives of β-endosulfan ranged from 104 to 265 days with endosulfan sulfate as the major metabolite. Endosulfan sulfate showed no clear signs of degradation. In a two-phase, two-soil anaerobic study, α-endosulfan anaerobic half-lives were 105 and 124 days and β-endosulfan half-lives were 136 and 161 days (EPA 2010a).

A field study report stated that endosulfan was transformed to endosulfan sulfate following incorporation of 6.7 kg/hectare of the pesticide into sandy loam soil (Stewart and Cairns 1974). The half-lives for α- and β-endosulfan were reported to be 60 and 800 days, respectively. In a field study conducted from 1989 to 1990 in northern India, dissipation (which can include multiple fate pathways) of endosulfan in sandy loam soil was examined (Kathpal et al. 1997). It was found that α-endosulfan could be detected up to 14 and 28 days in two different soil plots, while β-endosulfan could be detected up to 70 and 238 days. The overall half-life for endosulfan degradation ranged from 39.5 to 42.1 days. Endosulfan residues dissipated to an extent of 92–97% in the first 4-week period of application and by about 99% in 238 days. A residue half-life of 15 days for endosulfan (unspecified isomer) has been reported in Australian black soil when incubated at 30 °C at field capacity moisture level (Kathpal et al. 1997). In field dissipation studies submitted to EPA, the half-life of α-endosulfan (encompassing transport and degradation in the soil surface layer) was 46 days in a Georgia tomato field, 70 days in a California cotton field and 6–

11 days in a separate California cotton field. The half-life for β-endosulfan was 90 days in a Georgia tomato field, and 103 and 19–63 days in the respective California cotton fields. Endosulfan sulfate was the dominant degradation product (EPA 2010a).

6.3.2.4 Other Media

Numerous studies have demonstrated that endosulfan is oxidized to endosulfan sulfate on plant surfaces and in soils. Initial residues of endosulfan on treated vegetables generally range from 1 to 100 mg/kg. However, residue levels typically decrease to <20% of initial levels within 1 week after treatment (NRCC 1975). Residues of endosulfan isomers are generally negligible after 2–3 weeks; the α-isomer is much less persistent than the β-isomer. In most plant residue studies, endosulfan sulfate residue levels tend to increase relative to the parent isomers, and other metabolites and appear to be very persistent (Coleman and Dolinger 1982).

α-Endosulfan and β-endosulfan, at concentrations of 1,200–1,400 and 650 μg/kg, respectively, were found to have a half-life of 18.43 days under optimum conditions of temperature, moisture, PH, and enhanced microbial activity during vegetable waste composting using a full-scale continuous rotary drum composter (Ali et al. 2014).

6.4 LEVELS MONITORED OR ESTIMATED IN THE ENVIRONMENT

Reliable evaluation of the potential for human exposure to endosulfan depends in part on the reliability of supporting analytical data from environmental samples and biological specimens. Concentrations of endosulfan in unpolluted atmospheres and in pristine surface waters are often so low as to be near the limits of current analytical methods. In reviewing data on endosulfan levels monitored or estimated 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. The analytical methods available for monitoring endosulfan in a variety of environmental media are detailed in Chapter 7.

6.4.1 Air

Endosulfan has been included in air monitoring programs and in many air monitoring studies conducted within the last 10 years. Results from these studies establish endosulfan as an air contaminant not only in agricultural areas, but in rural, mountainous, and Arctic regions.

Many studies have established α-endosulfan as one of the most prevalent organochlorine pesticides in the Arctic region (Weber et al. 2010). Results from the Arctic Monitoring and Assessment Program (AMAP) analyses of pesticide concentrations sampled from various sites in the Arctic region at various times between 1993 and 2006, indicate endosulfan undergoes long-range atmospheric transport. Sampling stations were located in Canada, Finland, Iceland, Svalbard/Norway, Russia, United States, and Greenland. Endosulfan was measured in Alert, Canada at average air concentrations ranging from 3.3 pg/m^3 in 1993 to 6.5 pg/m^3 in 2003. Concentrations sampled at Nuuk, Greenland between 2004 and 2005 averaged 4.8 pg/m^3. Similar average concentrations were reported at the Russian Arctic stations. The Yukon region in the Canadian Arctic reported the highest average concentrations of endosulfan at 8.3 pg/m^3 at Tagish in 1994 and Little Fox Lake in 2002–2003 (Hung et al. 2010). Like other organochlorine pesticides, α-endosulfan has been observed to undergo a “spring maximum event” at several Arctic sampling stations, where air concentration peaks in the spring (April/May) and again in the fall (October/November). This is different from seasonal trends observed in temperate regions where air concentrations peak in the summer months (Hung et al. 2005).

Shen et al. (2005) mapped air concentrations of α- and β-endosulfan collected from XAD passive air samplers from 2000 to 2001 located across North America, with 31 stations located in Canada, five in the United States, and four in Central America. α-Endosulfan was detected at higher concentrations than β-endosulfan, ranging from 3.1 to 685 pg/m^3, detectable at 39 stations, compared to 0.03–119 pg/m^3, detectable at 30 stations, respectively. The highest concentrations were detected in areas of agriculture.

Endosulfans were detected in outdoor air sampled from three mountainous national park locations in Canada. Passive air samples were taken from sites within Mount Revelstoke National Park in British Columbia, Yoho National Park in British Columbia, and Observation Peak in Banff National Park, Alberta. All three areas lay west of land used for agriculture, but the land in the immediate vicinity of the sampling sites had limited to no agricultural use (Daly et al. 2007).

In 2001, ground level and mid-troposphere (~4,400 m) air samples were collected from the Fraser Valley, British Columbia and analyzed for endosulfan (α- and β-isomers). Ground level samples were taken from rural and urban areas, and mid-troposphere samples were obtained during flight times in aircraft.

Concentrations of the two isomers in rural areas ranged from ~18 to ~82 pg/m^3, with the exception of a period where concentrations exceeded 250 pg/m^3 for several days. This was attributed to high local use of endosulfan products. Concentrations in urban areas were less variable and ranged from ~4 to

~62 pg/m^3. Endosulfans were consistently below the detection limits in the high altitude samples, and therefore, possible trans-pacific movement of atmospheric endosulfans could not be discussed. The authors suggested that the low detection may be attributed to the low temperatures (0 °C) in the troposphere, which would result in partitioning to particulate matter (Harner et al. 2005).

Total endosulfans (α-, β-, and sulfate) were detected in passive air samplers located within the Global Atmospheric Passive Sampling (GAPS) network, which included polar sites (n=4), background sites (n=16), urban sites (n=6), and rural/agricultural sites (n=12) located worldwide. Sampling took place between December 2004 and March 2005. Endosulfan concentrations were highest compared to other organochlorine pesticides tested. Concentrations were highly variable, ranging from tens to hundreds of pg/m^3, the geometric mean was 58 pg/m^3. The α-isomer was the most abundant in the samples, accounting for ~90% of total endosulfans. Results for β-endosulfan and endosulfan sulfate were often below the detection limit. Concentrations were highest in tropical regions where regional applications and greater soil to air exchanges due to temperature may have occurred (Pozo et al. 2006).

Concentrations of endosulfans (α- and β-isomers and endosulfan sulfate) in air were measured by Hoh and Hites (2004) at four sampling sites located in four states in order to model potential sources. Samples were collected from Sleeping Bear Dunes National Lake Shore in Michigan, Bloomington, Indiana, Rohwer, Arkansas, and Cococrie, Louisiana between 2002 and 2003. Concentrations ranged from 0.56 to 1,200 (mean of 142), from 2.7 to 2,000 (mean of 260), from 4.7 to 390 (mean of 100), and from 3.6 to 480 (mean of 100) pg/m^3 for Michigan, Indiana, Arkansas, and Louisiana, respectively. Both Michigan and Indiana sites experienced spikes in endosulfan concentrations during July and August, presumably from local application. Applying the Potential Source Contribution Function (PCSF) model to the results, a large potential source region of endosulfan running from Kentucky, Tennessee, Alabama, and Florida was identified. With the exception of Alabama, these results correlated well with known regional use patterns. Modeling results did not indicate any potential sources west of the sampling sites.

Sun et al. (2006) measured gas phase and particulate phase α- and β-endosulfan in air from seven sampling sites in the Great Lakes region between 1996 and 2003. Samples were taken from sites within the Integrated Atmospheric Deposition Network (IADN), with five of the seven sites located in rural areas. Mean gas phase concentrations from these sites are included in Table 6-1 and mean particulate phase concentrations are included in Table 6-2. Calculated half-lives for gas phase α-endosulfan at these

Table 6-1. Average Concentrations of Gas-Phase α- and β-Endosulfan in Air from the Great Lakes Region

	α-Endosulfan		β-Endosulfan	
Site	Average concentration (pg/m^3; ± standard error)	Number of detects	Average concentration (pg/m^3; ± standard error)	Number of detects
Brule River	23±3.8	177	2.1±0.4	113
Eagle Harbor	27±3.6	250	2.2±0.32	153
Sleeping Bear Dunes	86±14	223	8.9±1.3	193
Sturgeon Point	110±11	272	9.5±1.0	213
Chicago	72±9.2	211	6.0±0.75	143
Burnt Island	21±2.8	312	2.6±0.41	276
Point Petre	110±22	385	24±4.3	347

Source: Sun et al. 2006

Table 6-2. Average Concentrations of α- and β-Endosulfan Particulates in Air from the Great Lakes Region

	α-Endosulfan		β-Endosulfan	
Site	Average concentration (pg/m^3; ± standard error)	Number of detects	Average concentration (pg/m^3; ± standard error)	Number of detects
Brule River	3.6±0.24	171	1.1±0.19	120
Eagle Harbor	4.2±0.22	201	1.1±0.16	134
Sleeping Bear Dunes	11±3.6	201	4.0±1.0	169
Sturgeon Point	7.8±0.83	209	3.7±0.47	195
Chicago	5.6±0.31	199	3.3±0.40	161

Source: Sun et al. 2006

locations ranged from 8.2 to 19 years for four of seven sites (results were not statistically significant for other sites). Particulate phase α-endosulfan half-lives ranged from 3.4 to 8.1 years for five of the seven sites (data were not available for the other sites). Calculated half-lives for gas phase β-endosulfan ranged from 3.2 to 9.7 years and particulate phase half-lives ranged from 3.0 to 8.0 years for five of the seven sites (data were not available for the other sites). These half-lives are based on a regression of the temporal trends of the endosulfan levels at these locations and are not to be confused with the estimated atmospheric photooxidation half-lives of α- and β-endosulfan. High outlier concentrations were observed during summer months, and were likely due to agricultural use. Results indicated that endosulfan concentrations (both isomers) increased from the western Great Lakes Region to the eastern region, possibly due to regional use patterns. Particulate α-endosulfan concentrations declined compared to concentrations recorded in previous years. It was not clear whether usage patterns in the region contributed to this declining trend.

α-Endosulfan, β-endosulfan, and endosulfan sulfate were detected in residential indoor air sampled (n=52) as a part of the Arizona Border Study (NHEXAS-AZ), which collected samples from sites in the Yuma, Nogales/Naco, and Douglas areas. These three testing sites had varied geographies and land use. The Yuma area is highly agricultural with a history of pesticide use. The Douglas area is mountainous with a history of mining and smelting. The Nogales/Naco area is a border town with industrial activity prevalent across the border in Mexico. α-Endosulfan averaged 190 ng per 4 standard semipermeable membrane devices (SMPDs), with a range of 10–1,600 ng per 4 SMPD and an 85% detection rate. β-Endosulfan averaged 87 ng per 4 SMPDs, with a range of 3.7–490 ng per 4 SMPD and an 89% detection rate. Endosulfan sulfate averaged 48 ng per 4 SMPDs, with a range of 19–100 ng per 4 SMPD, but a significantly lower detection rate of 5% (Gale et al. 2009).

6.4.2 Water

Endosulfan is monitored extensively in surface water and groundwater through various state, regional, and national programs. Studies analyzing rainwater, snow, and runoff from across the United States have also been published.

The USGS National Water Quality Assessment (NAWQA) program began in 1991 and obtains water quality data from 51 basins nationwide. These basins include approximately 7,300 surface water sites and 9,800 groundwater wells. Consistent with evidence that endosulfan will adsorb to soil, the available NAWQA groundwater samples obtained between 2002 and 2011 revealed an extremely low detection rate (0.12%) for α-endosulfan and endosulfan sulfate and no detection of β-endosulfan. Only 10 samples collected during this period reported measured or estimated concentrations at levels above detection limits (see Table 6-3). A similarly low rate of detection in surface water and bed sediment was observed for samples obtained between 2006 and 2011. Results from samples containing concentrations above detection limits are summarized in Table 6-4. It is important to note that endosulfan is expected to hydrolyze in aquatic environments to endosulfan diol, which is not analyzed in these samples.

In 2010, EPA published an ecological fate and risk assessment report that summarized the extensive water monitoring data available from NAWQA, EPA's STORET database, California's Department of Pesticide Regulation (CDPR) Surface Water Protection Program Database, and the South Florida Water Management District (SFWMD) DBHydro database, among others. Combining the data from these sources, EPA illustrated regional and national trends of endosulfan presence in U.S. waters over the period of almost 20 years (EPA 2010a).

In south Florida, endosulfan levels in agricultural runoff were analyzed by sampling surface water concentrations in the extensive canals that drain urban and agricultural areas. These canals are managed by the SFWMD. Samples were collected between 1993 and 1997. Average endosulfan concentrations ranged from 9 to 99 ng/L during this period. Using more sensitive analytical methods for samples obtained between 1996 and 1997, endosulfans were detected at a rate of 100% with a peak concentration of 477 ng/L (Scott et al. 2002).

Further analysis of the SFWMD data from 1992 through 2007 by Pfeuffer (2011) revealed several trends concerning endosulfan concentrations in south Florida canals. Surface water concentrations in selected basins for this period are summarized in Table 6-5. Endosulfans were detected in sediment samples taken from South Miami-Dade County (n=142). Endosulfan sulfate had the highest concentration among the three endosulfans in this basin, with an average of 16 μg/kg (25 detections), and a maximum concentration of 120 μg/kg. α-Endosulfan had an average concentration of 6 μg/kg (17 detections) and a maximum of 30 μg/kg. β-Endosulfan had an average concentration of 5 μg/kg (24 detections) and a maximum of 24 μg/kg. Endosulfan has been identified as a chemical of concern in the South Miami-Dade County agricultural area, but the frequency and magnitude of endosulfan detections have decreased since the 1994–1995 growing seasons.

Endosulfans (α- and β-isomers and endosulfan sulfate) were detected in water sampled from 13 sites located in the Biscayne Bay canals of southern Florida (n = 88) obtained between November 2002 and March 2004. These sites were located near ecologically sensitive areas of Everglades National Park and

Table 6-3. α-Endosulfan and Endosulfan Sulfate Detected in Groundwater Sampled for the USGS National Water Quality Assessment Between 2002 and 2011[a,b]

State	County	Well depth (ft)	Land use	Well type	Result date	Chemical	Concentration (µg/L)
Alabama	Mobile	68.4	RC	Urban	06/10/2011	α-Endosulfan	0.0066
New Jersey	Monmouth	32	RC	Urban	08/11/2011	α-Endosulfan	0.0165
New Jersey	Camden	32	RC	Urban	07/25/2011	α-Endosulfan	0.01
New Jersey	Camden	37	RC	Urban	07/13/2011	α-Endosulfan	0.0046
New Jersey	Camden	11	RC	Urban	08/18/2011	α-Endosulfan	0.0064
Virginia	Fairfax	120.43	RC	Urban	08/17/2011	α-Endosulfan	0.0074
Alabama	Mobile	68.4	RC	Urban	06/10/2011	Endosulfan sulfate	0.0042
New Jersey	Cumberland	148.5	NA	Other	05/21/2009	Endosulfan sulfate	0.0068[c]
New York	Suffolk	25	RC	Urban	08/23/2006	Endosulfan sulfate	0.0134[c]
New York	Suffolk	254.5	NA	Other	07/24/2007	Endosulfan sulfate	0.0061[c]

[a] Data represent only samples with concentrations above the detection limits tested during this 10-year period. The rate of detection for α-endosulfan and endosulfan sulfate between 2002 and 2011 was extremely low (0.12%); the rate of detection for β-endosulfan was 0%.
[b] Measured from filtered water
[c] Estimated

NA = not available; RC = residential/commercial

Source: USGS 2012b

Table 6-4. α-Endosulfan and Endosulfan Sulfate Detected in Surface Water and Bed Sediment Sampled for the USGS National Water Quality Assessment Between 2006 and 2011[a,b]

State	County	Land use	Result date	Chemical	Value (μg/L)
Arizona	Maricopa	Agricultural	08/24/2009	Endosulfan sulfate	0.0936
Arizona	Maricopa	Agricultural	09/03/2009	Endosulfan sulfate	0.037
Arizona	Maricopa	Agricultural	08/14/2009	α-Endosulfan	0.0107
Arizona	Maricopa	Agricultural	08/24/2009	α-Endosulfan	0.023
Arizona	Maricopa	Agricultural	08/14/2009	Endosulfan sulfate	0.0492
California	Riverside	Mixed	02/25/2011	Endosulfan sulfate	0.011
California	Riverside	Mixed	07/12/2010	Endosulfan sulfate	0.011[c]
California	Riverside	Mixed	06/21/2006	Endosulfan sulfate	0.0122[c]
Colorado	Mesa	Mixed	08/24/2006	Endosulfan sulfate	0.006[c]
Colorado	Weld	Mixed	09/01/2010	Endosulfan sulfate	0.0082
Colorado	Weld	Mixed	08/18/2010	Endosulfan sulfate	0.0099
Colorado	Weld	Mixed	08/01/2006	Endosulfan sulfate	0.007[c]
Connecticut	Hartford	Mixed	07/31/2008	α-Endosulfan	0.0091
Florida	Palm Beach	Cropland	07/06/2006	Endosulfan sulfate	0.011[c]
Florida	Palm Beach	Cropland	05/10/2007	Endosulfan sulfate	0.023[c]
Florida	Palm Beach	Cropland	05/23/2007	Endosulfan sulfate	0.0091[c]
Georgia	Brooks	Mixed	01/23/2008	Endosulfan sulfate	0.008[c]
Georgia	Brooks	Mixed	11/06/2007	Endosulfan sulfate	0.0091[c]
Georgia	Brooks	Mixed	04/22/2008	Endosulfan sulfate	0.0099[c]
Georgia	Brooks	Mixed	03/11/2008	Endosulfan sulfate	0.0103[c]
North Carolina	Greene	Mixed	08/09/2006	Endosulfan sulfate	0.0154[c]
Nevada	Carson City	Mixed	02/09/2010	α-Endosulfan	0.0038[c]
Oregon	Marion	Agricultural	10/11/2005	Endosulfan sulfate	0.0728
Oregon	Marion	Agricultural	12/08/2005	Endosulfan sulfate	0.0162
Oregon	Marion	Agricultural	04/06/2006	Endosulfan sulfate	0.0316
Oregon	Marion	Agricultural	06/14/2006	Endosulfan sulfate	0.0396
Oregon	Marion	Agricultural	08/10/2006	Endosulfan sulfate	0.0563
Oregon	Marion	Agricultural	10/17/2007	Endosulfan sulfate	0.0285
Oregon	Marion	Agricultural	11/07/2007	Endosulfan sulfate	0.024
Oregon	Marion	Agricultural	12/19/2007	Endosulfan sulfate	0.0118[c]
Oregon	Marion	Agricultural	01/09/2008	Endosulfan sulfate	0.0108[c]
Oregon	Marion	Agricultural	02/06/2008	Endosulfan sulfate	0.0088[c]
Oregon	Marion	Agricultural	02/20/2008	Endosulfan sulfate	0.0096[c]
Oregon	Marion	Agricultural	03/05/2008	Endosulfan sulfate	0.012[c]
Oregon	Marion	Agricultural	03/19/2008	Endosulfan sulfate	0.0105[c]
Oregon	Marion	Agricultural	04/09/2008	Endosulfan sulfate	0.0098[c]
Oregon	Marion	Agricultural	05/07/2008	Endosulfan sulfate	0.0126[c]
Oregon	Marion	Agricultural	05/21/2008	Endosulfan sulfate	0.018[c]

Table 6-4. α-Endosulfan and Endosulfan Sulfate Detected in Surface Water and Bed Sediment Sampled for the USGS National Water Quality Assessment Between 2006 and 2011[a,b]

State	County	Land use	Result date	Chemical	Value (μg/L)
Oregon	Marion	Agricultural	06/04/2008	Endosulfan sulfate	0.012[c]
Oregon	Marion	Agricultural	06/18/2008	Endosulfan sulfate	0.0157[c]
Oregon	Marion	Agricultural	07/09/2008	Endosulfan sulfate	0.0232
Oregon	Marion	Agricultural	07/23/2008	Endosulfan sulfate	0.0222
Oregon	Marion	Agricultural	08/20/2008	Endosulfan sulfate	0.0281
Oregon	Marion	Agricultural	09/03/2008	Endosulfan sulfate	0.0196[c]
Oregon	Marion	Agricultural	09/17/2008	Endosulfan sulfate	0.0194[c]
Oregon	Marion	Agricultural	04/23/2008	Endosulfan sulfate	0.0138[c]
Tennessee	Cocke	Mixed	07/29/2010	Endosulfan sulfate	0.0085
Tennessee	Cocke	Mixed	07/22/2010	Endosulfan sulfate	0.0094[c]
Tennessee	Greene	Agricultural	08/21/2008	α-Endosulfan	0.0042[c]
Tennessee	Greene	Agricultural	08/21/2008	Endosulfan sulfate	0.0039[c]
Tennessee	Cocke	Mixed	08/25/2010	Endosulfan sulfate	0.0081
Tennessee	Cocke	Mixed	08/12/2010	Endosulfan sulfate	0.0123
Tennessee	Cocke	Mixed	09/08/2010	Endosulfan sulfate	0.0063[c]
Washington	Yakima	Agricultural	04/19/2010	Endosulfan sulfate	0.0064[c]
Washington	Benton	Mixed	05/19/2008	Endosulfan sulfate	0.0069[c]
Washington	Benton	Mixed	04/16/2008	Endosulfan sulfate	0.0062[c]
Wisconsin	Kewaunee	Not available	09/04/2007	α-Endosulfan	0.3492[c,d]

[a] Data represent only samples with concentrations above the detection limits tested during this 5-year period. The rate of detection for α-endosulfan and endosulfan sulfate between 2006 and 2011 was extremely low (0.55%); the rate of detection for β-endosulfan was 0%.
[b] Measured from filtered water unless otherwise noted.
[c] Estimated
[d] Estimated in bottom sediments (μg/kg)

Source: USGS 2012c

Table 6-5. Endosulfan Concentrations (μg/L) in Surface Water Measured from the South Florida Water Management District (SFWMD)[a]

Chemical	Basin	Number of detections	Average	Maximum	Geometric mean	Median
α-Endosulfan	Citrus (n=373)	3	0.0036	0.0065	0.0031	0.0022
	South Miami-Dade County (n=311)	63	0.014	0.220	0.011	0.010
	Urban (n=297)	5	0.022	0.076	0.016	0.014
β-Endosulfan	Citrus (n=373)	2	0.004	0.005	0.004	0.004
	South Miami-Dade County (n=311)	32	0.010	0.078	0.008	0.009
	Urban (n=297)	4	0.026	0.077	0.019	0.026
Endosulfan sulfate	Citrus (n=373)	2	0.027	0.048	0.026	0.027
	South Miami-Dade County (n=311)	59	0.039	0.45	0.028	0.025
	Urban (n=297)	8	0.029	0.11	0.018	0.017

[a] Samples collected between April 1992 and December 2007.

Source: Pfeuffer 2011

Biscayne National Park. Many of the canals in these areas drain agricultural or mixed agricultural and urban areas. Concentrations of α-endosulfan ranged from 0.21 to 54 ng/L with an 81% detection rate. β-Endosulfan concentrations ranged from 0.20 to 16 ng/L with a 75% rate of detection. Endosulfan sulfate concentrations ranged from 0.22 to 28 ng/L with a 91% rate of detection. Endosulfan concentrations were highest towards the end of the growing season (Harman-Fetcho et al. 2005).

A combination of discrete water and passive water samples were collected from six stream sites in the Potomac River basin in 2007 and tested for chemical pollutants. β-Endosulfan and endosulfan sulfate were detected in 20 and 40% of passive water samples collected, respectively. No α- and β-endosulfan or endosulfan sulfate was detected in discrete water samples (Kolpin et al. 2013).

Levels of endosulfans were measured in rain water and air from the Choptank River watershed on the Delmarva Peninsula of the Chesapeake Bay. This watershed is located in an agricultural area and is vulnerable to pesticide runoff and atmospheric deposition. Samples were collected from 8 stations in 2000. In rainwater, α-endosulfan had a 13% rate of detection (n=71), an average concentration of 5.1 ng/L, and a range of 1.3–31 ng/L. β-Endosulfan had a rate of detection of 28%, an average concentration of 7.2 ng/L, and a range of 0.27–81 ng/L. Endosulfan sulfate had a rate of detection of 8.5%, an average concentration of 4.1 ng/L, and a range of 0.98–14 ng/L. Endosulfans were generally only detected in rain from June to early August. Total wet deposition rates were estimated as 0.96±0.1 kg/year for α-endosulfan, 2.7±0.3 kg/year for β-endosulfan, and 0.5±0.06 kg/year for endosulfan sulfate. The authors noted that these estimates are probably low compared to actual rates in areas of high pesticide usage (Kuang et al. 2003).

Snow and rain samples collected from 12 sites within Yosemite National Park, California during the spring and summer of 2008 and 2009 and were analyzed for endosulfan. α-Endosulfan and β-endosulfan were detected in 100% of the snow samples with concentrations ranging from 0.15 to 2.1 ng/L. Endosulfan sulfate was detected in 85% of the samples with concentrations ranging from 0.06 to 1.5 ng/L.

They were also among the most frequently detected current-use pesticides in the rainwater samples. Examination of the 2009 rain samples revealed a strong positive correlation between increasing concentrations of endosulfans during the summer months and increasing applications rates in the San Joaquin Valley during the same time. Winter and summer deposition rates were estimated for endosulfans (α- and β-isomers and endosulfan sulfate) and are included in Section 6.3.1, Transport and Partitioning. α-Endosulfan was the only pesticide present above the method quantitation limit in the surface water samples. The authors concluded that estimated water concentrations were sub-parts per trillion and orders of magnitude lower than aquatic benchmarks. Concentrations may be higher during the snowmelt periods of April and May (Mast et al. 2012a, 2012b).

Endosulfan levels in western National Parks were also analyzed by Hageman et al. (2006a, 2006b) as a part of a research project initiated by the U.S. National Parks Service (NPS). Snow pack samples were collected from alpine, sub-Arctic, and Arctic ecosystems from seven National Parks in the spring of 2003. These parks include Sequoia, Rocky, Rainier, Glacier, Denali, Noatak, and Gates of the Arctic. Concentrations of total endosulfans ranged from <0.0040–1.5 ng/L. Calculated deposition rates ranged from <0.19–1,400 ng/m^2. In a follow-up study using data collected from 2003 to 2005, Hageman et al. (2010) estimated the percent concentration due to regional transport for total endosulfans as approximately >90%.

6.4.3 Sediment and Soil

Endosulfans are monitored extensively through national programs such as the USGS NAWQA and National Oceanic and Atmospheric Administration’s (NOAA) National Status and Trends (NS&T) Program. The National Sediment Quality Survey (NSQS) includes sediment monitoring data from January 1990 to December 1998. Data from this source has been summarized in EPA’s ecological fate and risk assessment report for endosulfan, which was published in 2010. Sediment concentrations of endosulfan reported by NAWQA between 2006 and 2011 showed only one sample above the detection limit in Kewaunee, Wisconsin (see Table 6-5).

The NOAA NS&T Program reported a similarly low rate of detection (5.3%) in sediment samples obtained between 2005 and 2009 (NOAA 2012). Approximately 77 samples reported endosulfan (α- and β-isomers and endosulfan sulfate) above detection limits. These concentrations ranged from 0.08 to 12.59 ng/g dry weight. Details are provided in Table 6-6.

Lake sediment samples were collected from 19 lakes within Yosemite National Park, California during the summer of 2008 and 2009 and analyzed for endosulfan. Endosulfan sulfate was the most dominant endosulfan form detected. Total endosulfan concentrations ranged from 1.0 to 5.7 ng/g dry weight. Concentrations in lichen, lake sediments, and surface water (using SPMDs) displayed a positive correlation between increasing concentrations with rising elevation (Mast et al. 2012a, 2012b).

Table 6-6. Sediment Concentrations (ng/g Dry) Obtained by the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends (NS&T) Program Between 2005 and 2009[a]

Study	General location	Latitude	Longitude	Collection date	Chemical	Value
Mussel Watch	Biscayne Bay	25.5333	-80.3232	2/20/2007	Endosulfan sulfate	0.14
Mussel Watch	Commencement Bay	47.2932	-122.433	3/15/2006	β-Endosulfan	0.19
Mussel Watch	Commencement Bay	47.2932	-122.433	3/15/2006	α-Endosulfan	0.62
Mussel Watch	Corpus Christi	27.8522	-97.3598	11/28/2006	Endosulfan sulfate	0.18
Mussel Watch	Eureka	40.8215	-124.171	3/12/2006	Endosulfan sulfate	0.1
Mussel Watch	Everglades	25.9023	-81.5123	2/11/2006	α-Endosulfan	0.34
Mussel Watch	Galveston Bay	29.7045	-94.993	11/17/2006	β-Endosulfan	0.72
Mussel Watch	Galveston Bay	29.7045	-94.993	11/17/2006	Endosulfan sulfate	1.19
Mussel Watch	Green Bay	44.637	-87.8082	9/6/2006	Endosulfan II	0.22
Mussel Watch	Green Bay	44.637	-87.8082	9/6/2006	Endosulfan sulfate	0.43
Kachemak Bay	Homer Harbor	59.6056	-151.426	8/11/2007	Endosulfan sulfate	0.08
Mussel Watch	Hudson River	42.0338	-73.9293	9/24/2009	Endosulfan sulfate	0.83
Mussel Watch	Hudson River	42.0338	-73.9293	9/12/2007	Endosulfan sulfate	1.55
Mussel Watch	Hudson River	41.7089	-73.9406	9/24/2009	Endosulfan sulfate	12.59
Jobos Bay	Jobos Bay-Inner Bay	17.9578	-66.2038	5/28/2008	Endosulfan sulfate	0.15
Jobos Bay	Jobos Bay-Inner Bay	17.9508	-66.2119	5/27/2008	Endosulfan sulfate	0.26
Jobos Bay	Jobos Bay-Inner Bay	17.9508	-66.2119	5/27/2008	β-Endosulfan	0.36
Jobos Bay	Jobos Bay-Inner Bay	17.9391	-66.1852	5/28/2008	β-Endosulfan	2.16
Jobos Bay	Jobos Bay-National Estuarine Research Reserve	17.9268	-66.2408	5/23/2008	β-Endosulfan	0.39
Jobos Bay	Jobos Bay-Outer Bay	17.9537	-66.2892	5/23/2008	α-Endosulfan	0.14
Mussel Watch	Lake Erie	41.9587	-83.233	9/5/2007	α-Endosulfan	0.29
Mussel Watch	Lake Erie	41.6745	-83.2262	9/5/2007	β-Endosulfan	0.33
Mussel Watch	Lake Erie	42.5292	-79.2777	9/17/2009	Endosulfan sulfate	0.72
Mussel Watch	Lake Erie	41.6745	-83.2262	9/10/2009	β-Endosulfan	0.83
Mussel Watch	Lake Erie	42.88	-78.8916	9/20/2009	Endosulfan sulfate	0.94
Mussel Watch	Lake Erie	41.385	-82.5187	9/11/2009	Endosulfan sulfate	1.02
Mussel Watch	Lake Erie	41.6745	-83.2262	9/5/2007	Endosulfan sulfate	1.04
Mussel Watch	Lake Erie	41.6745	-83.2262	9/10/2009	Endosulfan sulfate	1.04
Mussel Watch	Lake Erie	42.5292	-79.2777	9/8/2007	Endosulfan sulfate	1.15
Mussel Watch	Lake Erie	42.1378	-80.0953	9/16/2009	α-Endosulfan	1.17
Mussel Watch	Lake Erie	41.9587	-83.233	9/5/2007	β-Endosulfan	1.26

Table 6-6. Sediment Concentrations (ng/g Dry) Obtained by the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends (NS&T) Program Between 2005 and 2009[a]

Study	General location	Latitude	Longitude	Collection date	Chemical	Value
Mussel Watch	Lake Erie	41.4744	-82.181	9/13/2009	Endosulfan sulfate	1.28
Mussel Watch	Lake Erie	42.88	-78.8916	9/20/2009	β-Endosulfan	1.3
Mussel Watch	Lake Erie	41.7014	-83.4587	9/10/2009	Endosulfan sulfate	1.42
Mussel Watch	Lake Erie	41.6597	-82.825	9/6/2007	Endosulfan sulfate	1.55
Mussel Watch	Lake Erie	41.385	-82.5187	9/11/2009	β-Endosulfan	1.78
Mussel Watch	Lake Erie	41.8933	-83.3248	9/9/2009	Endosulfan sulfate	1.97
Mussel Watch	Lake Erie	41.9112	-80.7877	9/16/2009	Endosulfan sulfate	2.01
Mussel Watch	Lake Erie	41.9247	-80.7183	9/8/2007	Endosulfan sulfate	2.08
Mussel Watch	Lake Erie	41.4994	-81.7188	9/14/2009	Endosulfan sulfate	2.48
Mussel Watch	Lake Erie	41.9247	-80.7183	9/15/2009	Endosulfan sulfate	2.52
Mussel Watch	Lake Erie	41.6597	-82.825	9/12/2009	Endosulfan sulfate	2.53
Mussel Watch	Lake Erie	41.9587	-83.233	9/11/2009	Endosulfan sulfate	2.78
Mussel Watch	Lake Erie	41.9587	-83.233	9/5/2007	Endosulfan sulfate	4.93
Mussel Watch	Lake Michigan	43.2282	-86.3469	9/6/2006	β-Endosulfan	0.19
Mussel Watch	Lake Ontario	44.9799	-74.8916	9/22/2009	Endosulfan sulfate	0.08
Mussel Watch	Lake Ontario	43.3553	-78.6867	9/9/2007	α-Endosulfan	0.17
Mussel Watch	Lake Ontario	44.1442	-76.3247	9/22/2009	α-Endosulfan	0.17
Mussel Watch	Lake Ontario	44.1442	-76.3247	9/11/2007	Endosulfan sulfate	0.25
Mussel Watch	Lake Ontario	43.2578	-77.4953	9/10/2007	Endosulfan sulfate	0.29
Mussel Watch	Lake Ontario	43.4683	-76.5097	9/21/2009	Endosulfan sulfate	0.38
Mussel Watch	Lake Ontario	43.3553	-78.6867	9/9/2007	Endosulfan sulfate	0.47
Mussel Watch	Lake Ontario	44.1442	-76.3247	9/22/2009	Endosulfan sulfate	0.54
Mussel Watch	Lake Ontario	43.2578	-77.4953	9/20/2009	Endosulfan sulfate	0.59
Mussel Watch	Lake Ontario	43.3553	-78.6867	9/19/2009	Endosulfan sulfate	0.67
Mussel Watch	Lake Ontario	43.3387	-78.1878	9/19/2009	Endosulfan sulfate	1.98
Mussel Watch	Lake St. Clair	42.6492	-82.711	9/4/2007	Endosulfan sulfate	0.35
Mussel Watch	Long Beach	33.7232	-118.174	3/7/2006	α-Endosulfan	0.23
Mussel Watch	Marina Del Rey	33.9618	-118.458	3/6/2006	α-Endosulfan	10.85
Mussel Watch	Matagorda Bay	28.6663	-96.2335	12/1/2006	Endosulfan sulfate	0.12
Mussel Watch	Niagara River	43.0468	-78.892	9/18/2009	Endosulfan sulfate	0.28
Mussel Watch	Niagara River	43.0468	-78.892	9/9/2007	Endosulfan sulfate	0.34
Vieques	North Vieques	18.1527	-65.3619	5/28/2007	β-Endosulfan	0.16
Vieques	North Vieques	18.1527	-65.3619	5/28/2007	α-Endosulfan	0.19
Mussel Watch	Pensacola Bay	30.5167	-87.1117	2/1/2006	α-Endosulfan	0.16
Mussel Watch	Puget Sound	47.9727	-122.23	3/17/2006	β-Endosulfan	0.41
Mussel Watch	Puget Sound	47.9727	-122.23	3/17/2006	Endosulfan sulfate	6.25
Mussel Watch	Rappahannock River	37.902	-76.7878	1/6/2007	Endosulfan sulfate	0.08
Mussel Watch	San Diego Bay	32.7247	-117.195	3/5/2006	Endosulfan sulfate	0.11

Table 6-6. Sediment Concentrations (ng/g Dry) Obtained by the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends (NS&T) Program Between 2005 and 2009[a]

Study	General location	Latitude	Longitude	Collection date	Chemical	Value
Mussel Watch	San Diego Bay	32.7247	-117.195	3/5/2006	β-Endosulfan	0.58
Mussel Watch	San Pedro Harbor	33.7067	-118.274	3/7/2006	β-Endosulfan	0.7
Mussel Watch	Sinclair Inlet	47.5852	-122.571	3/15/2006	Endosulfan sulfate	0.23
Mussel Watch	Sinclair Inlet	47.5852	-122.571	3/15/2006	β-Endosulfan	0.39
Vieques	South Southeast Vieques	18.1387	-65.3073	10/24/2007	β-Endosulfan	0.22
Vieques	South Southwest Vieques	18.1055	-65.4413	5/24/2007	β-Endosulfan	0.16
Vieques	South Vieques	18.1057	-65.4391	5/24/2007	β-Endosulfan	0.36
Mussel Watch	Tampa Bay	27.7872	-82.754	2/9/2006	α-Endosulfan	0.39

[a] Data represent only samples with concentrations above detection limits tested during this period. The rate of detection for total endosulfans was 5.3%.

Source: NOAA 2012

Bed sediment samples were collected from six stream sites in the Potomac River basin in 2007 and tested for chemical pollutants. α-Endosulfan was detected in 14% of samples collected at a maximum concentration of 0.01 μg/kg. No β-endosulfan or endosulfan sulfate was detected (Kolpin et al. 2013).

Endosulfan was detected in sediments obtained from 2 major rivers, 11 creeks or sloughs, 8 irrigation canals, and 2 tailwater ponds located in agricultural areas of California's Central Valley. Samples were obtained during "peak use" periods between July and November 2002, and "winter" samples obtained in March 2003. Peak endosulfan concentrations were limited to ponds adjacent to lettuce fields, but a concentration of 17 ng/g was reported in Del Puerto Creek. The authors noted that endosulfan concentrations were generally below acute toxicity thresholds, but may have contributed to toxicity in a few tailwater ponds and irrigation canals with concentrations greater than several hundred ng/g (Weston et al. 2004).

Residential soil samples collected from 11 homes in Atlanta, Georgia in 2006 did not show β-endosulfan in either yard or foundation samples. The method detection limit for this study was 0.60 ng/g (Riederer et al. 2010).

6.4.4 Other Environmental Media

Endosulfan residues have been detected in a variety of the consumer products, as well as aquatic and terrestrial organisms.

The U.S. Department of Agriculture (USDA) monitors levels of endosulfans and endosulfan sulfate in commodity food items for its Pesticide Data Program. Endosulfan (α- and β-isomers) and endosulfan sulfate were detected in samples collected in 2010 from apples, asparagus, cantaloupe, cilantro, cucumbers, hot peppers, lettuce, mangoes, pears, and sweet bell peppers. The results are summarized in Table 6-7. Cucumbers typically had the highest rates of detection (25.3–38.1%), while asparagus and mangoes had very low detection rates (<1% for all three forms of endosulfan). Endosulfan sulfate residues were detected in 44.5% of cantaloupe sampled with a reported concentrations ranging from 0.005 to 0.064 ppm (USDA 2012). Levels of endosulfan and endosulfan sulfate in domestic foodstuffs were determined as part of FDA's Total Diet Studies series (FDA 2005). The results of this monitoring study are summarized in Table 6-8. The highest mean concentrations were reported for endosulfan sulfate in items such as olive oil (0.01363 ppm), fresh/frozen summer squash (0.02050 ppm), peeled

Table 6-7. U.S. Department of Agriculture (USDA) Pesticide Data Program: Distribution of Endosulfan Residues in Fruits and Vegetables (2010)

	α-Endosulfan			β-Endosulfan			Endosulfan sulfate		
Food type	N	Percent detection	Value range (ppm)	N	Percent detection	Value range (ppm)	N	Percent detection	Value range (ppm)
Apple	744	4.3	0.010–0.31	744	8.1	0.010–0.17	744	1.9	0.033–0.087
Asparagus	ND	ND	ND	ND	ND	ND	372	0.3	0.050[a]
Cantaloupe	371	2.4	0.005–0.013	371	0.8	0.005[a]	371	44.5	0.005–0.064
Cilantro	555	1.1	0.010–0.16	325	0.6	0.051–0.19	ND	ND	ND
Cucumber	744	32.8	0.005–0.22	739	25.3	0.007–0.13	734	38.1	0.007–0.16
Hot pepper	186	7.5	0.010–0.083	186	9.1	0.017–0.15	186	15.1	0.005–0.048
Lettuce	743	5.8	0.002–0.051	743	6.2	0.001–0.031	743	6.6	0.004–0.067
Mangoe	372	0.3	0.005[a]	ND	ND	ND	372	0.5	0.005[a]
Pear	743	0.1	0.008[a]	743	2.0	0.008[a]	743	0.3	0.012[a]
Sweet bell pepper	744	11.0	0.004–0.30	744	7.4	0.020–0.38	744	16.9	0.002–0.24

[a] Detected in only one sample.

N = number; ND = not detected

Source: USDA 2012

Table 6-8. Endosulfan Levels in Food Products Sampled for the 2003–2005 Market Basket Survey

	α-Endosulfan				β-Endosulfan				Endosulfan sulfate			
Food type	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)
Apple (red)	8	0.00006	0.0001	0.0002	8	0.00018	0.0002	0.0007	8	0.00018	0.0001	0.0007
Pear, raw	8	0.00001	0.0001	0.0001	8	0.00018	0.0001	0.0004	8	0.00071	0.0003	0.0020
Strawberries, raw/ frozen	8	0.00225	0.0070	0.0110	8	0.00550	0.0100	0.0340	8	0.00293	0.0004	0.0160
Cantaloupe	8	0.00004	0.0003	0.0003	8	0.00015	0.0001	0.0010	8	0.00654	0.0001	0.0230
Raisins	8	0.00003	0.0001	0.0001	8	0.00004	0.0001	0.0002				
Spinach, fresh/ frozen	8	0.00465	0.0001	0.0370	8	0.01080	0.0002	0.0860	8	0.03654	0.0003	0.2850
Collards, fresh/ frozen	8	0.00043	0.0004	0.0030	8	0.00018	0.0001	0.0009	8	0.00091	0.0001	0.0050
Lettuce, iceberg	8	0.00084	0.0003	0.0040	8	0.00046	0.0002	0.0020	8	0.00136	0.0002	0.0050
Broccoli, fresh/ frozen	8	0.00003	0.0002	0.0002	8	0.00003	0.0002	0.0002				
Tomato, raw	8	0.00085	0.0004	0.0030	8	0.00143	0.0004	0.0060	8	0.00103	0.0003	0.0040
Tomato sauce, plain	8	0.00030	0.0001	0.0008	8	0.00049	0.0001	0.0020	8	0.00015	0.0001	0.0007
Green beans, fresh/ frozen	8	0.00146	0.0007	0.0110	8	0.00104	0.0003	0.0080	8	0.00421	0.0007	0.0300
Green beans, canned	8	0.00004	0.0003	0.0003	8	0.00001	0.0001	0.0001	8	0.00004	0.0003	0.0003
Cucumber, peeled	8	0.00171	0.0004	0.0040	8	0.00121	0.0005	0.0020	8	0.01099	0.0009	0.0270
Summer squash, fresh/frozen	8	0.00200	0.0001	0.0060	8	0.00036	0.0001	0.0010	8	0.02050	0.0050	0.0490
Pepper, sweet, green	8	0.00240	0.0002	0.0120	8	0.00556	0.0005	0.0320	8	0.00489	0.0001	0.0280
Squash, winter	8	0.00003	0.0002	0.0002	8	0.00003	0.0002	0.0002	8	0.00499	0.0006	0.0250
Spaghetti w/meat sauce	8	0.00075	0.0002	0.0030	8	0.00085	0.0003	0.0030	8	0.00021	0.0001	0.0005
Dill cucumber pickles	8	0.00315	0.0002	0.0070	8	0.00188	0.0001	0.0050	8	0.00992	0.0004	0.0330
Tomato catsup	8	0.00044	0.0004	0.0010	8	0.00058	0.0001	0.0020	8	0.00024	0.0001	0.0005
Chocolate chip cookies	8	0.00005	0.0002	0.0002	8	0.00003	0.0001	0.0001	8	0.00009	0.0003	0.0004
Candy bar, milk chocolate	8	0.00049	0.0003	0.0020	8	0.00028	0.0001	0.0010	8	0.00145	0.0001	0.0050
BF, macaroni, tomato and beef	8	0.00006	0.0005	0.0005	8	0.00011	0.0009	0.0009	8	0.00011	0.0002	0.0007
BF, applesauce	8	0.00004	0.0001	0.0001	8	0.00010	0.0001	0.0003	8	0.00018	0.0002	0.0005
BF, pears	8	0.00004	0.0001	0.0001	8	0.00009	0.0001	0.0002	8	0.00015	0.0001	0.0003

Table 6-8. Endosulfan Levels in Food Products Sampled for the 2003–2005 Market Basket Survey

	α-Endosulfan				β-Endosulfan				Endosulfan sulfate			
Food type	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)
Yogurt, low-fat, fruit-flavored	8	0.00004	0.0001	0.0002	8	0.00004	0.0001	0.0002	8	0.00003	0.0002	0.0002
Chicken breast, roasted	8	0.00009	0.0007	0.0007	ND	ND	ND	ND				
Brussels sprouts, fresh/frozen	8	0.00001	0.0001	0.0001	8	0.00004	0.0003	0.0003	8	0.00028	0.0002	0.0020
Okra, fresh/frozen	8	0.00062	0.0050	0.0050	8	0.00463	0.0370	0.0370	8	0.00725	0.0580	0.0580
Tuna noodle casserole	8	0.00014	0.0001	0.0008	8	0.00053	0.0001	0.0030	8	0.00039	0.0002	0.0020
Quarter-pound cheeseburger	8	0.00005	0.0002	0.0002	8	0.00005	0.0002	0.0002	8	0.00028	0.0002	0.0020
Taco/tostada with beef and cheese	8	0.00026	0.0003	0.0007	8	0.00026	0.0003	0.0009	8	0.00073	0.0003	0.0020
Pizza, cheese and pepperoni	8	0.00024	0.0002	0.0010	8	0.00050	0.0001	0.0020	8	0.00028	0.0002	0.0009
Black olives	8	0.00006	0.0001	0.0002	ND	ND	ND	ND	8	0.00036	0.0002	0.0009
BF, squash	8	0.00025	0.0020	0.0020	8	0.00004	0.0003	0.0003	8	0.00050	0.0001	0.0030
Breakfast pastry	8	0.00003	0.0002	0.0002	8	0.00005	0.0004	0.0004	8	0.00006	0.0005	0.0005
Macaroni salad	8	0.00004	0.0003	0.0003	8	0.00008	0.0006	0.0006	8	0.00008	0.0001	0.0003
Potato salad	8	0.00016	0.0001	0.0010	8	0.00019	0.0001	0.0010	8	0.00075	0.0002	0.0020
Coleslaw	8	0.00001	0.0001	0.0001	8	0.00003	0.0002	0.0002	8	0.00041	0.0003	0.0020
Lettuce, leaf, raw	8	0.00289	0.0001	0.0050	8	0.00291	0.0001	0.0090	8	0.00607	0.0001	0.0180
Tomato salsa, bottled	8	0.00081	0.0001	0.0030	8	0.00159	0.0003	0.0060	8	0.00079	0.0001	0.0030
Lasagna	8	0.00009	0.0001	0.0003	8	0.00016	0.0002	0.0005	8	0.00010	0.0001	0.0003
Beef with vegetables, Chinese	8	0.00009	0.0003	0.0004	8	0.00008	0.0006	0.0006	8	0.00016	0.0003	0.0007
Chicken with vegetables, Chinese	8	0.00011	0.0001	0.0006	8	0.00005	0.0001	0.0002	8	0.00058	0.0001	0.0030
Chicken filet (broiled) sandwich	8	0.00004	0.0003	0.0003	8	0.00008	0.0002	0.0004	8	0.00016	0.0006	0.0007
Candy, chocolate with nuts	8	0.00005	0.0002	0.0002	8	0.00004	0.0001	0.0002	8	0.00023	0.0002	0.0010
Sweet and sour sauce	8	0.00018	0.0003	0.0006	8	0.00029	0.0004	0.0010	8	0.00008	0.0001	0.0003
Olive oil	8	0.00041	0.0001	0.0010	8	0.00063	0.0002	0.0020	8	0.01363	0.0030	0.0330
BF, plums/prunes with apples/pears	8	0.00001	0.0001	0.0001	8	0.00005	0.0001	0.0003	8	0.00011	0.0001	0.0004
BF, dutch apple betty	8	0.00001	0.0001	0.0001	8	0.00005	0.0002	0.0002	8	0.00008	0.0002	0.0004

Table 6-8. Endosulfan Levels in Food Products Sampled for the 2003–2005 Market Basket Survey

	α-Endosulfan				β-Endosulfan				Endosulfan sulfate			
Food type	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)
BF, chicken with rice	8	0.00001	0.0001	0.0001	8	0.00001	0.0001	0.0001	8	0.00004	0.0003	0.0003
BF, beef and noodles	8	0.00014	0.0001	0.0010	8	0.00026	0.0001	0.0020	8	0.00016	0.0001	0.0010
BF, apples with berries	8	0.00011	0.0001	0.0006	8	0.00021	0.0001	0.0010	8	0.00019	0.0004	0.0006
BF, apples with non-berry fruit	8	0.00010	0.0001	0.0006	8	0.00028	0.0001	0.0010	8	0.00068	0.0002	0.0030
Pork and beans	ND	ND	ND	ND	8	0.00001	0.0001	0.0001	8	0.00001	0.0001	0.0001
Peas	ND	ND	ND	ND	8	0.00004	0.0003	0.0003	8	0.00013	0.0010	0.0010
Peanut butter, smooth	ND	ND	ND	ND	8	0.00003	0.0002	0.0002	8	0.00041	0.0002	0.0009
Peanuts, dry roasted, salted	ND	ND	ND	ND	8	0.00003	0.0002	0.0002	8	0.00036	0.0003	0.0010
Peach, raw/frozen	ND	ND	ND	ND	8	0.00013	0.0010	0.0010	8	0.00009	0.0007	0.0007
Applesauce, bottled	ND	ND	ND	ND	8	0.00005	0.0001	0.0002	8	0.00015	0.0001	0.0005
Cabbage, fresh	ND	ND	ND	ND	8	0.00001	0.0001	0.0001	8	0.00075	0.0020	0.0040
BF, vegetables and beef	ND	ND	ND	ND	8	0.00001	0.0001	0.0001	8	0.00003	0.0002	0.0002
BF, peaches	ND	ND	ND	ND	8	0.00001	0.0001	0.0001	8	0.00003	0.0002	0.0002
Tomato juice, bottled	ND	ND	ND	ND	8	0.00005	0.0001	0.0002	8	0.00001	0.0001	0.0001
Turnip, fresh/frozen	ND	ND	ND	ND	8	0.00019	0.0003	0.0009	8	0.00026	0.0001	0.0009
BF, apricots with mixed fruit	ND	ND	ND	ND	8	0.00001	0.0001	0.0001	8	0.00005	0.0001	0.0003
Cheese, American	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00004	0.0003	0.0003
Beef, ground	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00003	0.0002	0.0002
Pork bacon	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00004	0.0003	0.0003
Bread, whole wheat	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00003	0.0002	0.0002
Crackers, saltine	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00004	0.0003	0.0003
Watermelon	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00005	0.0004	0.0004
Asparagus, fresh/ frozen	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00004	0.0003	0.0003
Potato, boiled	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00031	0.0001	0.0006
Potato, baked	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00066	0.0001	0.0020
Potato chips	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00144	0.0005	0.0060
Quarter-pound hamburger	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00011	0.0001	0.0003
Butter	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00095	0.0001	0.0030

Table 6-8. Endosulfan Levels in Food Products Sampled for the 2003–2005 Market Basket Survey

	α-Endosulfan				β-Endosulfan				Endosulfan sulfate			
Food type	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)	N	Level mean (ppm)	Level min (ppm)	Level max (ppm)
BF, vegetables and chicken	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00001	0.0001	0.0001
BF, chicken noodle dinner	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00004	0.0003	0.0003
BF, turkey and rice	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00001	0.0001	0.0001
BF, fruit dessert/ pudding	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00004	0.0001	0.0002
Cream cheese	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00016	0.0003	0.0007
Pineapple juice, frozen concentrate	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00008	0.0002	0.0004
French fries	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00161	0.0003	0.0070
Eggplant, fresh	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00020	0.0006	0.0010
Fish sandwich	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00004	0.0003	0.0003
Clam chowder, New England	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00004	0.0003	0.0003
Sour cream	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00004	0.0003	0.0003
Salmon	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00003	0.0002	0.0002
Cranberry juice	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00001	0.0001	0.0001
Potatoes, mashed	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00023	0.0004	0.0005
Carrot, baby, raw	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00013	0.0010	0.0010
Ranch dressing, low-calorie	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00013	0.0010	0.0010
Vegetable oil	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00011	0.0009	0.0009
BF, banana/apple	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00003	0.0001	0.0001
BF, macaroni and cheese	ND	ND	ND	ND	ND	ND	ND	ND	8	0.00005	0.0004	0.0004

BF = baby food; max = maximum; min = minimum; N = number of analyses; ND = not detected; RTF = ready to feed

Source: FDA 2005

cucumber (0.01099 ppm), and fresh/frozen spinach (0.03654 ppm). Generally, concentrations were higher in fresh/frozen fruits and vegetables versus processed food products. USDA’s program analyzes a greater number of samples for each food item compared to FDA’s Total Diet Studies. It is important to note that the residue samples in USDA’s Pesticide Data Program tended to be higher when compared to results from FDA.

Studies of carrot and tomato crops sprayed with endosulfan 2–8 days prior to harvest showed that more pesticide remains in the pulp than in the juices of these vegetables. Washing and peeling the vegetables lowered the endosulfan concentration considerably (Burchat et al. 1998).

The NOAA’s Mussel Watch Program monitors contaminant levels in mussels and oysters in over 280 U.S. coastal sites. In EPA’s ecological fate and risk assessment report (EPA 2010a), monitoring levels were summarized from samples analyzed between 1994 and 2009 (see Table 6-9). Endosulfans (α- and β-isomers and endosulfan sulfate) were detected in 64% of samples with an average concentration of 2.0 µg/kg dry weight and a 90th percentile concentration of 4.9 µg/kg dry weight.

Two recent studies monitored endosulfan levels in freshwater fish. In a study monitoring chemical contaminants in bass and carp species in southeastern U.S. rivers, endosulfans were detected at low concentrations (<1.2 ng/g wet weight) and mean concentrations were not calculated due to the large number of samples below the limit of detection. Samples were taken for the Mobile River Basin, Apalachicola-Chattahoochee-Flint River Basin, Savannah River Basin, and Pee Dee River Basin in 2004 (Hinck et al. 2008). Bass, carp, and catfish sampled from the Colorado River basin and tributaries had reported total endosulfan concentrations of <0.02 µg/g, but the largest concentrations (>0.07 µg/g) were found in carp and bass from the Gila River near Arlington, Arizona in August 2003 (Hinck et al. 2007).

USDA’s Pesticide Data Program continued its study of pesticide residues in domestic and imported catfish intended for human consumption. The catfish were mostly farm-raised. In 2010, 384 samples were analyzed for residues of α-endosulfan, β-endosulfan, and endosulfan sulfate. α-Endosulfan was only detected at the limit of detection (0.001 ppm) and β-endosulfan was not detected in any samples. Endosulfan sulfate was detected in 30 samples with a rate of detection of 7.8% and at concentrations ranging from the limit of detection (0.001 ppm) to 0.028 ppm (USDA 2012).

Endosulfan was also detected in the muscle of Pacific cod and Pacific halibut collected from coastal waters of Aleutian Islands, Alaska. Sampling areas were grouped according to their level of military

Table 6-9. Concentration of Total Endosulfans (µg/kg Dry Weight) in Bivalves from the National Oceanic and Atmospheric Administration (NOAA) Mussel Watch Database[a]

Statistic	α-Endosulfan	β-Endosulfan	Endosulfan sulfate	Total endosulfans
Number samples	1,429	1,980	1,277	1,980
Number detected	677	841	379	1,258
Percent detected	47%	42%	30%	64%
Minimum	BDL	BDL	BDL	BDL
Average	1.2	0.9	0.5	2.0
50th Percentile	BDL	BDL	BDL	0.39
90th Percentile	2.4	2.5	0.7	4.9
95th Percentile	4.6	4.4	1.5	7.9
99th Percentile	17.8	11.7	5.3	22.1
Maximum	120	37	192	192

[a] Units of µg/kg dry weight (ppb); BDL = below detection limit (~0.2–0.7 ppb). Averages assume 0.0 ppb for concentrations below detection. Data are from 1994–2008.

Source: EPA 2010a

activity. In the contemporary military group, the geometric mean concentrations of endosulfan in Pacific cod (n=18) and Pacific halibut (n=26) were 2.5 and 3.3 ng/g wet weight, respectively. In the historical military group, the geometric mean concentration in Pacific cod (n=13) was 5.3 ng/g wet weight and 4.6 ng/g wet weight in Pacific halibut (n=23). The reference group concentrations were 3.2 ng/g wet for Pacific cod (n=16) and 3.0 ng/g wet weight for Pacific halibut (n=13). Endosulfan was not detected in any of the fish samples of rock greenling (Miles et al. 2009).

Endosulfan was measured in the major food items of Koeye River grizzly bears in British Columbia, Canada in the fall of 2004. Measured mean concentrations in spawned pink salmon, spawned sockeye salmon, crab, mussels, fall terrestrial vegetation, and spring terrestrial vegetation were 4.21, 2.18, 0.0410, 2.23, 0.926, and 0.833 ng/g lipid weight, respectively (Christensen et al. 2013).

China is the world’s largest producer and exporter of fishery products. Seafood products including 6 species of shrimp, 2 species of crab, and 14 species of shellfish were collected in June and October 2005 and analyzed for various chemical contaminants. Samples were collected from the Guangdong Province, which borders the South China Sea. α-Endosulfan residues had a frequency rate of 3.8%, arithmetic mean of 0.04 ng/g wet weight and a range of 0.02–1.25 ng/g wet weight. β-Endosulfan residues had a frequency rate of 1.4% and a range of 0.02–0.29 ng/g wet weight. Endosulfan sulfate residues had a frequency rate of 1.4% and a range of 0.01–0.35 ng/g wet weight. Residues were found mostly in shellfish species *Perna uiridis*, *Sinonovacula constricta*, and *Crassostrea gigas* (Guo et al. 2007).

Endosulfans were detected in wine corks produced from the bark of the cork oak tree (*Quercus suber*), which are grown widely in regions of western Mediterranean. Wine corks were collected in spring 1999 from wines produced in Greece, wines produced in Cleveland, Ohio, and from a winery in Bloomington, Indiana (these corks were not used as bottle stoppers). The authors did not specify the origin of the corks used in these wine samples. Total endosulfan levels ranged from 3.8 to 29 ng/g lipid. Concentrations were generally higher in the samples obtained from Bloomington, Indiana. Variation of concentrations was high, and the authors suggested that this was due to differences in pesticide usage where cork oak trees are harvested. The production steps, which include the produce, the cork retailers, and the winery, may also contribute to these variations. Wine to cork exchanges may also have contributed to variations (Strandberg and Hites 2001).

Lichen samples collected from Yosemite National Park, California during the spring and summer of 2008 and 2009 were analyzed for endosulfans (α- and β-isomers and endosulfan sulfate). They were detected in 100% of the samples ranging from 2.0 to 24 ng/g dry weight. These concentrations also showed a positive correlation with increasing elevation, suggesting the occurrence of "cold-mountain trapping" (Mast et al. 2012a, 2012b).

Sparling et al. (2001) claimed that heavy pesticide use in the San Joaquin Valley is contributing to population decline of certain amphibians in the Sierra Mountains, which lies downwind from this agricultural area. Residue levels in Pacific tree frog (*Hyla regilla*) tadpoles and adults were analyzed from samples taken from coastal sites (used as controls), Lassen Volcanic National Park, Lake Tahoe, Yosemite National Park, and Sequoia National Park. Generally, endosulfan residues were often zero in the coastal sites and in Lassen Volcanic National Park, which are located west and north, respectively, of the San Joaquin Valley. The maximum concentrations (21.9 ppb) occurred in Sequoia or Yosemite National Park. Endosulfans along with DDx compounds also had the highest frequency of detection (~70–80%) throughout the Lake Tahoe, Sequoia, and Yosemite locations. Although endosulfan was not solely implicated, the authors concluded that there was a mixed but increasing occurrence of endosulfan residues along the west to east gradient, which is consistent with the declining amphibian populations in this area.

Endosulfans were detected in blubber of beluga whales (*Delphinapterus leucas*) collected from 15 sites in the Canadian Arctic at various times between 1993 and 2001. Geometric means for endosulfan (isomers not specified) ranged from 9.7 to 76.3 ng/g wet weight from samples taken from males and females of Resolute Bay (1996, 1999), Grise Fjord (2000), Igloolik (1995, 1997), Coral Harbor (2000), and Arviat (1999). Endosulfan sulfate concentrations (geometric means) ranged from 7.0 to 70.6 ng/g wet weight from sites in Chesterfield Inlet (1997, 1999), Sanikiluaq (1994, 2000), Cape Dorset (1999, 2000), Kimmirut (1994, 1996), Iqaluit (1992, 1996), and Pangnirtung (1996, 1997). Endosulfan along with α-hexachlorocyclohexane, comprised the larger proportion of organochlorine residues in the northern Hudson Bay areas. Concentration variations between the sites were statistically significant but concentrations between male and female samples were not (Stern et al. 2005).

Endosulfan was also detected in other higher trophic aquatic organisms. A study analyzed pesticide concentrations in Bonnethead sharks (*Sphyrna tiburo*) in the Florida estuaries of Apalachicola Bay (9 females, 13 males), Tampa Bay (17 females, 15 males), Charlotte Harbor (5 females, 5 males), and Florida Bay (18 females, 13 males). β-Endosulfan and endosulfan sulfate were not detected in liver samples from any of the sampling locations. β-Endosulfan geometric mean concentrations ranged from 12.77 to 15.13 ng/g in muscle and 1.55–6.60 ng/mL in serum. Endosulfan sulfate was only detected in liver samples from Charlotte Bay (geometric mean 1.00±037 ng/g) and serum samples from Florida Bay (geometric mean 1.93±0.93 ng/mL) (Gelsleichter et al. 2005). Skin and blubber samples (post-mortem) from two blue whales (*Balaenoptera musculus*) stranded off the coast of Baja, California were analyzed for chlorinated hydrocarbons. The two whales were juvenile males, both approximately 18 m long. Endosulfan sulfate was detected at approximately 70 ng/g of lipid in the blubber of whale 1 and at approximately 230 and 380 ng/g of lipid in the skin of whales 1 and 2, respectively (Valdez-Marquez et al. 2004).

6.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

The main route of exposure to endosulfan for the general population is ingestion of food containing residues of endosulfan as a result of application or bioconcentration. A dietary exposure assessment for endosulfan (both isomers and sulfate) was conducted by both the EPA and the CDPR using available monitoring data. Results from the two assessments differed in the data and analytical methods, but both assessments concluded that calculated risks of dietary exposure of endosulfan are below protective benchmarks for all subgroups. The CDPR analysis used residue levels from their own residue monitoring data, as well as data obtained from other sources including consumption data from the USDA Continuing Survey of Food Intakes of Individuals (CFSII), other USDA data, and FDA data. Water residues were not considered in the CDPR dietary exposure assessment since available monitoring data in California did not indicate drinking water or groundwater as a significant source of endosulfan exposure. Results from the CDPR dietary exposure assessment are provided in Table 6-10. The results indicated a chronic dietary exposure (per capita) of 0.19 μg/kg/day and an acute dietary exposure (95th percentile consumption) of 1.85 μg/kg/day for the total U.S. population. Three population subgroups were identified as groups of concern not only from dietary exposure, but also having the highest risk from occupational exposure and/or exposure to the general public. These subgroups were non-nursing infants (<1 year old), children 1–6 years old, and ≥13 year-old nursing females. However, calculated margins of exposure (MOE) were >100 for all subgroups in the CDPR assessment, indicating that these dietary exposures are below levels of concern (Silva and Carr 2010).

The EPA risk dietary exposure assessment used the USDA CFSII data from 1994 to 1996 and 1998 and incorporated drinking water estimated environmental concentrations (EECs) using the DEEM-FCID™ software. Results from the EPA 2007 dietary exposure assessment are provided in Table 6-11. Acute

Table 6-10. California Department of Pesticide Regulation (CDPR) Acute and Chronic Dietary Exposure (µg/kg/day) to Anticipated Endosulfan Residues on Raw Agriculture Commodities (RACs) and the Resulting Dietary Margins of Exposure (MOE) from 1989 to 1992

Population subgroups	Acute exposure[a]	Chronic exposure
U.S. population	1.85	0.19
All Infants <1 year old	3.08	0.22
Infant nursing <1 year old	1.90	0.08
Infant non-nursing, <1 year old	**3.18**	**0.28**
Children 1–6 years old	**3.30**	**0.41**
Children 7–12 years old	2.09	0.29
Female 13–19 years old, not pregnant, not nursing	1.37	0.18
Female ≥20 years old, not pregnant, not nursing	1.51	0.14
Females 13–50 years old	1.39	0.15
Female ≥13 years old, pregnant, not nursing	1.57	0.15
Females ≥13 years old nursing	**2.06**	**0.17**
Males 13–19 years old	1.37	0.21
Males ≥20 years old	1.38	0.15
Seniors ≥55 years old	1.65	0.14

[a] 95th percentile

Source: Silva and Carr 2010

Table 6-11. U.S. Environmental Protection Agency (EPA) Summary of Acute and Chronic Dietary Exposure (μg/kg/day) for Endosulfan[a]

	Drinking water exposure		Food exposure		Food and drinking water exposure	
Population subgroup	Acute[b]	Chronic	Acute[b]	Chronic	Acute[b]	Chronic
U.S. population	0.47	0.003	0.11	0.004	0.48	0.007
All infants <1 year old	1.21	0.010	0.14	0.004	1.21	0.015
Children 1–2 years old	0.47	0.005	0.24	0.013	0.53	0.017
Children 3–5 years old	0.46	0.004	0.18	0.009	0.55	0.014
Children 6–12 years old	0.28	0.003	0.13	0.003	0.30	0.009
Youth 13–19 years old	0.31	0.002	0.09	0.003	0.32	0.006
Adults 20–49 years old	0.35	0.003	0.09	0.003	0.35	0.006
Adults ≥50 years old	0.24	0.003	0.10	0.003	0.26	0.006
Females 13–49 years old	0.33	0.003	0.09	0.003	0.34	0.005

[a] Data for 2007.
[b] The EPA uses the 99.9th percentile of exposure from consumption of food alone (EPA 2000b).

Source: Silva and Carr 2010

dietary exposure from drinking water for the U.S. population was reported at 0.47 µg/kg/day and chronic exposure was reported at 0.003 µg/kg/day. Acute dietary exposure from food was reported at 0.11 µg/kg/day and chronic exposure was reported at 0.004 µg/kg/day for the U.S. population. Acute dietary exposures (99.9th percentile) from both food and drinking water was reported at 0.48 µg/kg/day and chronic exposure was reported at 0.007 µg/kg/day. EPA evaluated risk by calculating the percent population adjusted dose (%PAD), all of which were below levels of concern. The discrepancies between the exposure and risk results between these two studies can be attributed to the type of data used in the analysis, software used (TAS, Inc. EX™ versus DEEM-FCID™, risk characterization values (MOE versus %PAD), and acute toxicity values used to calculate risk (no-observed-effect level [NOEL] of 0.7 versus 1.5 mg/kg/day) (Silva and Carr 2010).

The CDPR estimated public exposure via non-dietary intake of endosulfan. Under a short-term air, bystander exposure scenario, a time-weighted average (TWA) of 1.63 µg/m^3 at the sampler approximately 6.4 m from the eastern edge of the field was calculated. This value was calculated assuming an application rate of 1.5 lb active ingredient (AI)/acre, or 1.7 kg AI/ha, which is below the maximum application rate for apples. This TWA may be an underestimate when compared to fields that have application rates at the maximum allowed level. Adjusting for the maximum allowable rate, a 24-hour concentration estimate of 2.72 µg/m^3 was calculated. Long- and short-term exposure estimates for child and adult swimmers (incorporating incidental ingestion, dermal, and inhalation exposures) were all low, and the associated risks were below health benchmarks (Beauvais et al. 2010a).

Lee et al. (2002) estimated endosulfan inhalation hazard quotients (HQs) using CDPR data from 1990 to 2000. HQs were defined as daily intake/reference dose and were calculated for acute, chronic, and subchronic exposures for both adults and children (<12 years old). All HQs for endosulfan were <1 for both children and adults, indicating levels that are not a concern. However, HQs were generally higher for children than for adults.

The National Occupational Exposure Survey (NOES) conducted by the National Institute for Occupational Safety and Health (NIOSH) from 1980 to 1983 estimated that 3,205 workers in the agricultural services industry were exposed to endosulfan in the workplace in 1980 (NIOSH 1984). The NOES database does not contain information on the frequency, concentration, or duration of exposure of workers to any chemicals; the survey provides only estimates of the number of workers potentially exposed to chemicals in the workplace.

6.6 EXPOSURES OF CHILDREN

This section focuses on exposures from conception to maturity at 18 years in humans. Differences from adults in susceptibility to hazardous substances are discussed in Section 3.7, Children's Susceptibility.

Children are not small adults. A child's exposure may differ from an adult's exposure in many ways. Children drink more fluids, eat more food, breathe more air per kilogram of body weight, and have a larger skin surface in proportion to their body volume. A child's diet often differs from that of adults. The developing human's source of nutrition changes with age: from placental nourishment to breast milk or formula to the diet of older children who eat more of certain types of foods than adults. A child's behavior and lifestyle also influence exposure. Children crawl on the floor, put things in their mouths, sometimes eat inappropriate things (such as dirt or paint chips), and spend more time outdoors. Children also are closer to the ground, and they do not use the judgment of adults to avoid hazards (NRC 1993).

As with the adult general population, the main route of endosulfan exposure for children is through dietary intake. Dietary exposure assessments from the CDPR and EPA-estimated dietary exposure assessments and risks for various child subgroups are provided in Tables 6-10 and 6-11 (Silva and Carr 2010). Calculated risks for all subgroups were below levels of concern, but calculated exposures were generally higher for the child subgroups when compared to the adult sub-groups and the total U.S. population. This is particularly evident in the CDPR results, where calculated exposures (from food only) were 3.18 μg/kg/day for non-nursing infants (<1 year old) and 3.30 μg/kg/day for 1–6-year-old children compared to 1.85 μg/kg/day for the total U.S. population. The EPA dietary exposure assessment results were markedly lower than the CDPR results. The dietary exposure for food only was 0.14 μg/kg/day for all infants (<1 year old) and 0.24 μg/kg/day for children 1–2 years old, compared to 0.11 μg/kg/day for the total U.S. population. The discrepancy between childhood exposures and total population exposures was larger when taking into account intake from both food and drinking water (1.21 μg/kg/day for all infants [<1 year old] and 0.48 μg/kg/day for the U.S. population). Estimated exposures from drinking water were higher for infants and children than from exposures from food intake. It should be noted that the exposure assessments from CDPR and EPA differed in several ways including data sets used, approaches (deterministic vs. probabilistic), populations (users only vs. per capita), and reference points (95th vs. 99th percentiles) (Silva and Carr 2010).

Several studies have been published recently exploring pre-natal and post-natal exposure of endosulfan by analyzing concentrations in breast milk, placenta, and umbilical cord blood. Shen et al. (2007) analyzed α-endosulfan concentrations in placenta and milk from Danish and Finnish mothers (1997–2001). Geometric mean concentrations in Danish and Finnish breast milk samples (n=43 for each group) were 7.41 and 7.3 ng/g lipid, respectively. Geometric mean concentrations in Danish and Finnish placenta samples (n=43 for each group) were 2.28 and 2.52 ng/g lipid, respectively. Placenta concentrations were measured in mothers giving birth to males from southern Spain between 2000 and 2002 (n=220) by Freire et al. (2011). Total endosulfan (sum of α- and β-isomers, endosulfan diol, endosulfan ether, endosulfan sulfate, and endosulfan lactone) geometric mean concentration (n=211) was reported as 4.02 ng/g placenta with a 95% confidence interval of 3.34–4.8 ng/g. The 95th percentile concentration was 27.0 ng/g placenta, and the rate of detection was 95.9%. Cerrillo et al. (2005) also analyzed endosulfan and metabolite levels in mothers and fertile women from Granada and Almeria Provinces in southern Spain. Concentrations were detected in adipose tissue (fertile women), placenta, umbilical cord blood, and breast milk. Results from this study are summarized in Table 6-12. The authors concluded that endosulfan pre- and postnatal exposure between child and mother is a common event, although contributions from environmental and other dietary sources cannot be ruled out.

In a study investigating correlations between prenatal pesticide exposure and cryptorchidism (a male reproductive birth defect) in babies, Damgaard et al. (2006) compared milk samples from 62 mothers of male infants with cryptorchidism (cases) and 68 mothers of healthy male infants (controls). The joint prospective, longitudinal cohort study occurred in Finland and Denmark from 1997 to 2001. α-Endosulfan was detected in all samples with a median concentration of 6.95 ng/g lipid and range of 1.83–17.84 ng/g lipid. Control concentrations were similar with a 6.66 ng/g lipid median and range of 1.19–22.66 ng/g lipid. The authors stated that although singular exposure to α-endosulfan or any other pesticide examined was not significantly correlated with cryptorchidism, the study suggested that exposure to more than one pesticide at low concentrations represents a risk factor for congenital cryptorchidism.

Fernandez et al. (2007) performed a similar study where various organochlorine pesticide concentrations in placenta samples were compared to occurrences of male genital malformations such as cryptorchidism and hypospadias. Samples were obtained from Granada Province in southern Spain from October 2000 to July 2002. Total endosulfan (sum of α- and β-isomers, endosulfan diol, endosulfan ether, endosulfan sulfate, and endosulfan lactone) arithmetic mean concentration (±standard deviation) was 20.8±25.0 ng/g lipid in the case studies (n=48) and 19.7±29.7 ng/g lipid in the control samples (n=114). Maximum

Table 6-12. Endosulfan and Metabolite Concentrations in Adipose Tissue, Placenta, Umbilical Cord Blood, and Breast Milk of Fertile Women and Mothers from Southern Spain

Chemical	α-Endosulfan	β-Endo-sulfan	Endosulfan ether	Endosulfan lactone	Endosulfan diol	Endosulfan sulfated
Adipose tissue (n=149), ng/g fat[a]						
Mean	11.09	6.63	1.72	0.94	5.47	16.16
Standard deviation	86.06	19.63	6.58	1.38	28.02	92.52
Median	0.50	2.50	0.25	0.50	1.25	1.25
Maximum	944.1	219.7	61.83	9.2	338.17	882.98
Percent frequency	26.2	13.4	49.6	16.1	26.8	12.8
Placenta (n=200), ng/g placenta or ng/mL homogenate[b]						
Mean	3.55	4.19	0.33	15.62	12.56	3.57
Standard deviation	5.8	16.02	0.38	19.23	53.25	5.83
Median	0.94	1.00	0.20	2.64	4.35	1.61
Maximum	28.27	150.92	3.08	74.67	527.02	44.45
Percent frequency	55.5	49	50	36	49	67.5
Umbilical cord blood (n=200), ng/mL serum[c]						
Mean	3.34	2.77	1.43	3.88	13.23	2.82
Standard deviation	5.70	1.88	1.61	7.91	11.34	6.09
Median	1.56	2.00	0.81	2.07	9.62	1.20
Maximum	60.25	14.91	8.64	83.89	83.32	36.36
Percent frequency	76.5	62	42.5	60.5	81	33.5
Breast milk (n=23), ng/mL milk[d]						
Mean	0.68	10.70	6.08	4.63	0.60	6.18
Standard deviation	0.35	8.71	14.49	1.14	0.42	4.18
Median	0.87	7.29	0.66	5.00	0.64	5.00
Maximum	1.00	26.89	57.58	5.00	1.00	14.35
Percent frequency	65.2	43.5	100	91.3	43.5	26.1

[a] β-Endosulfan was significantly ($p \leq 0.001$) associated with endosulfan lactone. Endosulfan lactone was also associated with endosulfan ether, endosulfan diol, and endosulfan sulfate.

[b] α-Endosulfan was statistically associated with endosulfan lactone ($p \leq 0.001$) and endosulfan sulfate ($p \leq 0.005$); β-endosulfan was significantly associated with endosulfan diol ($p \leq 0.005$). Among the endosulfan metabolites, endosulfan lactone was associated with endosulfan ether ($p \leq 0.005$) and endosulfan sulfate ($p \leq 0.001$).

[c] α-Endosulfan was associated with endosulfan ether, endosulfan lactone, and endosulfan diol ($p \leq 0.001$), while β-endosulfan was associated with endosulfan diol and endosulfan sulfate ($p \leq 0.001$). There was a significant ($p \leq 0.001$) association among endosulfan diol, endosulfan ether, and endosulfan lactone.

[d] α-Endosulfan was significantly associated with endosulfan sulfate ($p \leq 0.05$), while β-endosulfan was associated with endosulfan ether ($p \leq 0.05$) and endosulfan diol ($p \leq 0.01$). There was a statistically significant association between the two commercial products, α-endosulfan and β-endosulfan ($p \leq 0.05$).

Source: Cerrillo et al. 2005

concentrations were 103 ng/g lipid among the case studies and 189.5 ng/g lipid in the controls. The rate of detection for α-endosulfan was 52.4%. As with Damgaard et al. (2006), positive correlations between male genital malformations were not observed for any singular chemical or metabolite. However, the authors noted a 3.5-fold increase in risk for urogenital malformations when the mother reported taking part in agricultural activities. This trend did not appear in the fathers. However, a 2.98-fold increase risk was found when fathers were asked about specific work tasks and chemical exposures. A seasonal trend of births of males with malformations was also observed, with largest occurrences reported in winter. Data from questionnaires were not sufficient enough to correlate this seasonal trend with increased use of organochlorine pesticides during the previous spring.

Based on a bystander inhalation exposure scenario calculated by Beauvais et al. (2010a), children may be exposed to as much as 2.72 $\mu g/m^3$ of endosulfan (24-hour maximum concentration estimate) if they are within 6.4 m from the edge a field where it is being applied. Endosulfan exposure to children from swimming is expected to be low.

Lee et al. (2002) estimated endosulfan inhalation HQs using CDPR data from 1990 to 2000. HQs were defined as daily intake/reference dose and were calculated for acute, chronic, and subchronic exposures for both adults and children (<12 years old). All HQs for endosulfan were <1 for both children and adults, indicating levels that are not a concern. However, HQs were generally higher for children than for adults.

Since young children spend more time outdoors and have a tendency to ingest soil, it is important to examine child exposure through ingestion. Although no studies have been conducted concerning this subject, exposure through ingestion of soil is not expected to be significant since dietary intake is regarded as the largest source of exposure for endosulfan. However, children may potentially be exposed to endosulfan from oral/dermal exposure if they play in the soil of contaminated areas such as hazardous waste sites. Based on degradation of endosulfan in the environment, child exposures to endosulfan through soil ingestion are not expected to be very significant.

No studies could be located discussing exposure of children to endosulfan after household use by parents. Likewise, no exposure studies could be located concerning the exposure of children whose parent(s) work with endosulfan on a daily basis. However, many studies suggest that pesticides used in the workplace can be brought home through contaminated clothing, shoes, and other materials (NIOSH 1995). Although no documented cases could be located, the possibility exists that endosulfan used in a work setting may be brought home by working parents. It is uncertain what amount of endosulfan exposure a child may encounter under these situations.

6.7 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

Farm workers are expected to be exposed to higher amounts of endosulfan compared to the general population. These exposures may occur through direct handling and application or through latent exposure in fields that were previously sprayed (occupational re-entry). Beauvais et al. (2010b) compared the occupational exposure assessments for endosulfan performed by the CDPR and EPA. Dermal, inhalation, and aggregate exposure estimates short-term are summarized in Table 6-13.

Similar to exposure assessments for the general population, CDPR and EPA results differed greatly due to different assumptions regarding exposure. In general, CDPR calculated significantly higher exposures than EPA. The majority of short-term dermal, inhalation, total, and aggregate MOEs were <100, indicating levels of concern. The exceptions were EPA's low-pressure handwand, mixer/loader handling emulsifiable concentrate (LPHW MLA-EC) short-term exposure scenario and CDPR's seasonal and annual LPHW MLA-EC exposure scenario, which had calculated MOEs >100. Exposures from fields previously sprayed were also assessed by CDPR and EPA. Most short-term (2–4 days) and intermediate-term (10–14 days) MOEs for representative re-entry scenarios were <100 from both studies, indicating levels of concern. Representative exposure scenarios included thinning (almonds, peaches), scouting (broccoli, citrus, cotton, lettuce, potatoes), hand harvesting (broccoli, sweet corn, cucumbers, ornamental cut flowers, ornamental plants, strawberries, and tomatoes), and cane turning (grapes) (Beauvais et al. 2010a).

In one study, the exposure of an individual involved in spraying the compound, while wearing protective overalls, gloves, and breathing mask, was examined (Arrebola et al. 1999). The individual applied 300 L of an endosulfan mixture to plants and later gave 10 urine samples over the course of 3 days. The study found that the highest concentrations occurred 4.3 hours after exposure with concentrations for α- and β-endosulfan reaching 4,289 and 1,079 pg/mL, respectively. The half-lives for the excretion of α- and β-endosulfan were determined to be 23 and 27 hours, respectively.

In addition to individuals who are occupationally exposed to endosulfan, there are several groups within the general population that have potentially high exposures (higher than background levels) to endosulfan.

Table 6-13. Dermal, Inhalation, and Aggregate[a] Short-term Exposure Estimates by CDPR and EPA (mg/kg/day)[b]

Scenario	Dermal		Inhalation		Aggregate	
	CDPR	EPA	CDPR	EPA	CDPR	EPA
GB applicator	0.0439	0.013	0.001	0.00034	0.047	0.017
HPHW MLA-EC	0.501	0.090	0.010	0.0026	0.513	0.096
Aerial flagger	0.371	0.066	0.002	0.00088	0.375	0.070
LPHW MLA-EC	0.013	0.0083	0.0001	0.000026	0.015	0.0044
Aerial applicator	0.786	0.033	0.004	0.00085	0.792	0.037

[a] Aggregate exposure is the combined total of dermal, inhalation, and dietary exposure.
[b] CDPR estimated handler exposures for short-term are defined as acute and up to 1 week; EPA defined short-term durations as 1–30 days and therefore overlaps CDPR's short-term and seasonal estimates.

CDPR = California Department of Pesticide Regulation; EPA = Environmental Protection Agency;
GB = groundbloom; HPHW = high-pressure handwand; LPHW = low-pressure handwand; MLA-EC = mixer/loader handling emulsifiable concentrate; NE = not estimated

Source: Beauvais et al. 2010b

These populations include individuals living in proximity to sites where endosulfan was produced or sites where endosulfan was disposed of.

6.8 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 endosulfan is available. Where adequate information is not available, ATSDR, in conjunction with 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 endosulfan.

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.

6.8.1 Identification of Data Needs

Physical and Chemical Properties. The physical/chemical properties of endosulfan and endosulfan sulfate are sufficiently well characterized to enable assessment of the environmental fate of the compound (HSDB 2009, 2010; NIOSH 2011; O'Neil et al. 2006; Tomlin 2003).

Production, Import/Export, Use, Release, and Disposal. According to the Emergency Planning and Community Right-to-Know Act of 1986, 42 U.S.C. Section 11023, industries are required to submit substance release and off-site transfer information to the EPA. The TRI, which contains this information for 2009, became available in February of 2011. This database is updated yearly and should provide a list of industrial production facilities and emissions.

Although all U.S. producers of endosulfan must be registered under FIFRA, data concerning quantities of endosulfan produced domestically are limited. Annual production can be estimated by analogy to its annual use, for which quantitative data are available. The available use trends over the last two decades indicates use of endosulfan is in decline, based on a reported annual use of 1.38 million pounds between 1990 and 1999 to only 380,000 pounds per year between 2006 and 2008 (EPA 2002, 2010a). As of

March 2012, there were only four active registrants of endosulfan in the United States. Over the 4-year phase-out schedule, these registrants will be cancelling their endosulfan products according to a voluntary agreement (EPA 2012e). Despite this cancellation, production data and continued recordkeeping of endosulfan use would be valuable during the years of cancellation and the years following, so as to understand the impact and its effects on exposure to the public and to workers.

Releases and disposal of endosulfan to the environment are well defined based on the regulatory restrictions and the available monitoring data (CDPR 2011; EPA 2001, 2002, 2010a, 2012c, 2012e; NOAA 2012; USGS 2012c).

Environmental Fate. Overall, the environmental fate mechanisms associated with endosulfan are well documented. Endosulfan partitions to air and is subject to long-range transport (EPA 2010a; Hafner and Hites 2003; Hageman et al. 2006a; Hung et al. 2005, 2010; Kathpal et al. 1997; Mast et al. 2012a, 2012b; Rice et al. 2002; Rudel 1997; Stern et al. 2005; Weber et al. 2010). It is immobile in soils (EPA 2010a; Stewart and Cairns 1974). It is transformed in surface waters and soils via hydrolysis (Greve and Wit 1971; HSDB 2010; Kaur et al.1998) and biodegradation (Cotham and Bidleman 1989; HSDB 2010).

Bioavailability from Environmental Media. Endosulfan can be absorbed following inhalation of contaminated workplace air and ingestion of insecticide-contaminated food (Ely et al. 1967). Dermal contact with or ingestion of endosulfan that is tightly bound to soil particles is an exposure route of concern at hazardous waste sites. No quantitative information is available on the absorption of endosulfan in either adults or children following ingestion or dermal contact with contaminated soils. Therefore, additional information is needed on the uptake of endosulfan from contaminated soil following ingestion or dermal contact. This information would be useful in determining the bioavailability of soil-bound endosulfan.

Food Chain Bioaccumulation. Endosulfan is bioconcentrated by aquatic organisms and supporting data are well established (DeLorenzo et al. 2002; EPA 2010a; Ernst 1977; Pennington et al. 2004; Rajendran and Venugopalan 1991; Roberts 1972; Schimmel et al. 1977; Weber et al. 2010). BMFs from fish to aquatic mammals suggest the potential for biomagnification in aquatic food chains. However, these estimates do not take metabolism into account (Kelly et al. 2007; Weber et al. 2010). Estimating bioconcentration and biomagnification in terrestrial organisms is also difficult due to the lack of biomonitoring data, especially in humans. Recent BMF estimates indicate potential for biomagnification in humans, but understanding of continuous, low-dose metabolism in terrestrial organisms is also lacking

(Kelly et al. 2007). Acute poisoning cases suggest that metabolism in humans may be rapid (Eyer et al. 2004). Methods for estimating biomagnification in terrestrial organisms based on octanol/air and octagonal/water partition coefficients require an understanding of its metabolic potential (Armitage and Gobas 2007; EPA 2010a). Further investigations into the bioaccumulation, biomagnification, and metabolism of endosulfan in humans and terrestrial organisms are needed.

Exposure Levels in Environmental Media. Reliable monitoring data for the levels of endosulfan in contaminated media at hazardous waste sites are needed so that the information obtained on levels of endosulfan in the environment can be used in combination with the known body burden of endosulfan to assess the potential risk of adverse health effects in populations living in the vicinity of hazardous waste sites.

Monitoring data for endosulfan in environmental media are current and extensive. It has been detected in ambient air of temperate and Arctic regions (Daly et al. 2007; Gale et al. 2009; Harner et al. 2005; Hoh and Hites 2004; Hung et al. 2005, 2010; Pozo et al. 2006; Shen et al. 2005; Sun et al. 2006; Weber et al. 2010). Endosulfan is a monitored in groundwater and surface water samples through the USGS NAWQA program, EPA’s STORET database, CDPR Surface Water Protection Database, USDA Pesticide Data Program, and SFWMD DB Hydro database, etc. Altogether, these sources contain over 20 years’ worth of monitoring data for endosulfan in the United States (EPA 2010a). Additional studies have also detected endosulfan in surface water (Harman-Fetcho et al. 2005; Pfeuffer 2011; Scott et al. 2002). Studies have also detected endosulfan in rainwater and snow samples (Hageman et al. 2010; Kuang et al. 2003; Mast et al. 2012a, 2012b). It has been detected in sediment and soil (NAWQA; NOAA NS&T) (Mast et al. 2012a, 2012b; Riederer et al. 2010; Weston et al. 2004). Endosulfan levels in bivalves are monitored as part of the NOAA’s Mussel Watch Program and have been summarized by EPA (2010a). The FDA’s Total Diet Studies (2005) and USDA’s Pesticide Data Program (USDA 2012) detected endosulfan residues in a variety of food products, mostly in fresh and frozen fruits and vegetables. Residues have been detected in fresh and seawater fish and seafood, including catfish caught and raised for human consumption (Guo et al. 2007; Hinck et al. 2007, 2008; Miles et al. 2009; USDA 2012). They have been detected in lichen samples from Yosemite National Park, California (Mast et al. 2012a, 2012b) and wine corks made from cork oak tree (*Q. suber*) (Strandberg and Hites 2001). Endosulfans have been detected in the Pacific tree frog (*H. regilla*) of the Sierra Mountains and California, downwind from the agricultural San Joaquin Valley (Sparling et al. 2001). Endosulfans have been detected in high-trophic aquatic organisms such as beluga whales, blue whales, and Bonnethead sharks (*S. tiburo*) (Gelsleichter et al. 2005; Stern et al. 2005; Valdez-Marquez et al. 2004).

Exposure Levels in Humans. Comprehensive biomonitoring studies for endosulfan are not available. Exposure levels in the general population have been extensively evaluated using available monitoring data. Dietary intake is expected to be the major source of endosulfan exposure to the general public. Dietary exposures were extensively evaluated for various population subgroups by both EPA and CDPR using different data sets and analytical methods. Although the estimated chronic dietary exposures from these studies were significantly different, both values were below protective benchmarks. The major difference between these exposure assessment deals with the incorporation of drinking water into dietary intake. CDPR assumed that drinking water was not a significant source of exposure based on the monitoring data for California. However, EPA incorporated drinking water into their assessment and their model results estimated drinking water exposure to be the same as food intake (Silva and Carr 2010). Comprehensive biomonitoring studies using samples collected from the U.S. population that can be used to better assess endosulfan exposure in the general population would be valuable. CDPR evaluated short-term, bystander inhalation exposure and estimated long- and short-term exposures for child and adult swimmers (Beauvais et al. 2010a). Lee et al. (2002) estimated inhalation HQs using CDPR data from 1990 to 2000.

This information is necessary for assessing the need to conduct health studies on these populations.

Exposures of Children. Dietary exposure assessments were conducted by EPA and CDPR, which included estimates for child subgroups. These assessments used monitoring data in food and drinking water rather than biomonitoring data from urine or blood samples. Results from these assessments indicated that childhood exposures were generally higher compared to exposure estimates for the general adult population (Silva and Carr 2010). Several studies measured endosulfan levels in breast milk, placenta, umbilical cord blood, and maternal adipose tissue, and some explored correlations between elevated endosulfan levels and reproductive malformations in male infants (Cerrillo et al. 2005; Damgaard et al. 2006; Fernandez et al. 2007; Freire et al. 2011; Shen et al. 2007). These studies were conducted in Europe and may not be representative of U.S. exposures. Comprehensive biomonitoring studies using samples collected from the U.S. population would be valuable in assessing endosulfan exposure in children. More data would also be helpful to properly assess endosulfan exposure to children who live, play, or attend school near farmlands that are treated with endosulfan. CDPR estimated inhalation exposure levels under a bystander scenario, but this analysis incorporated several assumptions (Beauvais et al. 2010a). The possibility that farming parents’ work clothes and shoes may carry endosulfan residues into the home also should be studied.

Child health data needs relating to susceptibility are discussed in Section 3.12.2, Identification of Data Needs: Children's Susceptibility.

Exposure Registries. Although endosulfan is monitored extensively in food, surface water, groundwater, air, and media, it is not included in comprehensive biomonitoring studies that analyze tissue, blood, or urine. Endosulfan is not included in the CDC's National Health and Nutrition Examination Survey (NHANES), which monitors chemical concentrations in urine and blood collected from the U.S. population. These data would give the best basis for estimating exposures to the general population, children, and workers. Endosulfan will be gradually phased-out through 2016 and exposures are expected to decrease. As a result, large-scale biomonitoring and environmental monitoring programs will likely be decreased or discontinued. Continuing studies would be valuable in understanding the immediate and long-term impacts on the general population.

6.8.2 Ongoing Studies

No ongoing studies sponsored by NIH or EPA were identified for endosulfan.