



Environmental Fate of Propylene Oxide

April 2016

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1 Introduction

Propylene oxide (PPO) is used in agriculture as a fumigant for the post-harvest fumigation of fruits, nuts, and spice crops (US EPA 2006). PPO can also be used in air-tight sterilization chambers for sterilizing packaged foods, health care materials, and other heat-sensitive products (US EPA 2006). PPO was first discovered in 1860 and has been commercially available for the treatment of food products since the early 1900s (IARC 1985; IARC 1994; Kahlich *et al* 2005). Major U.S. PPO manufacturers are Dow Agrosiences, ABERCO INC. and ARCO Chemical Company for industrial use. PPO was first registered with the United States Environmental Protection Agency (US EPA) in June 1986 as a postharvest fumigant for tree nuts in crop group 14 (this group includes almond, beech nut, Brazil nut, butternut, cashew, chestnut, chinquapin, hazelnut, hickory nut, macadamia nut, pecan, and walnut) and subgroup 19 for herbs and spices, with a general residue tolerance of 300 ppm (US EPA 1987). The only product registered in California is distributed by ABERCO INC. (CDPR 2016).

No data indicate that PPO is a naturally occurring product; instead it enters the environment mainly through evaporation from PPO's production, handling, storage, transport, and use. It is estimated that major contribution to total PPO exposure occurs through atmospheric releases (Alberta 2002; EC 2008). Primarily PPO is produced by one of two processes: from direct oxidation of propylene with air or oxygen or via the intermediate propylene chlorohydrin (NRC 2010; Gardiner *et al.* 1993). This report reviews the relevant literature to address the environmental fate, major agricultural uses, and chemistry of PPO in California with special attention to atmospheric, terrestrial and aquatic fates.

2 Physical and Chemical Properties

PPO is a clear, colorless, highly flammable and volatile liquid at ambient temperature and pressure (IPCS 1985; Alberta 2002; NRC 2010). Its odor has been described as ethereal (IPCS 1988; IPCS 1985; Alberta 2002; NCBI 2016), alcoholic, sweet (Lewis 1997; NCBI 2016), and neutral to pleasant (Verschueren, 2001). PPO is easily ignited; the vapors are heavier than air and under prolonged exposure to fire or intense heat can cause explosion (IPCS 1988; 1985; NCBI 2016). Decomposition of PPO upon heating results in emission of irritating vapors and acrid smoke (NCBI 2016). Interaction of PPO with active catalytic surfaces and hydroxides of alkali metal; aqueous acids,

amines, and acids may cause hazardous polymerization (EPA 1994). The physical and chemical properties of PPO are presented in Table 1.

Table 1 Physical and chemical properties of propylene oxide

Property	Value	Reference
Chemical Name	Propylene oxide	
Synonyms	1,2-Epoxypropane, methyloxidrane,	
Empirical Formula:	C ₃ H ₆ O	
CAS Registry Number	75-76-9	
Physical State	Liquid	
Color	Colorless	
Molecular Weight (g/mol)	58.08	
Melting Point	-112.13 °C	<u>Budavari 1996</u>
Boiling Point	35 °C	<u>Budavari, 1996; Lewis 2000</u>
Density	0.821 g/cm ³ (6.861 lb./gal)	<u>Dow Chemical 2015</u>
Specific gravity (liquid)	0.859 at 0 °C	<u>Verschueren 2001; Alberta 2002;</u>
Specific gravity (gas) (air =1)	2	<u>IPCS, 1988; IPCS 1985</u>
Log Kow	0.05	<u>Shell Chemical 2011</u>
Log Koc	25	<u>Hansch et al. 1995</u>
Henry's Law constant (HLC)	6.96 X 10 ⁻⁵ atm- m ³ /mole)	<u>Grosjean1991</u>
Vapor pressure	538 mmHg at 25 °C	<u>Ontario 2001; Dow Chemical 2015</u>
Solubility in water	40.5 wt % (400–590 g/L)	<u>Dow Chemical 2015; Bogyo et al.</u>
Solubility	Soluble in alcohol and ether Miscible	<u>Budavari 1996; Alberta 2002</u>

Source: Alberta Environment. 2002. Assessment report on propylene oxide; Shell Chemicals. 2011. Propylene oxide Product Stewardship Summary; NCBI. 2016. Compound summary propylene oxide

3 Use Profile

The only agricultural use of PPO is the disinfestation and sterilization of agricultural products (including spices and nuts). PPO is registered by the US EPA as a fumigant to prevent microbial spoilage caused by fungi, yeasts, molds, and bacteria, and for the control of insects in food commodities including herbs and spices, tree nuts, and cocoa powder and to treat several dried foods (US EPA 2006; APVMA 2012).

PPO fumigation has been used by the California nut industry for insect and microbial control for decades (ABC 2007; ABC 2008; CDPR 2009). Several research projects demonstrated that PPO fumigation is an effective treatment for almond pasteurization (Harris et al., 2015). In 2007, the United States Department of Agriculture (USDA) issued a Letter of Determination that raw almonds in the United States must be pasteurized to mitigate risk of salmonella either by steam pasteurization or by treatment with propylene oxide gas (USEPA 2007).

In California, applicators are required by law to report use of any registered pesticide to the California Department of Pesticide Regulation (DPR); a Pesticide Use Report (PUR) is then generated for each agricultural pesticide application. A PUR record includes information on the product used, type of crop, application date, amount applied, application method, and the location of the application. The PUR database provides location information per one-mile square section as defined by the Public Land Survey System (PLSS). Using the PUR database, we queried for PPO pesticide applications from 2010 to 2014. As displayed in Figure 1, highest use of PPO occurred in Stanislaus County followed by Yolo, Kern and Tehama counties.

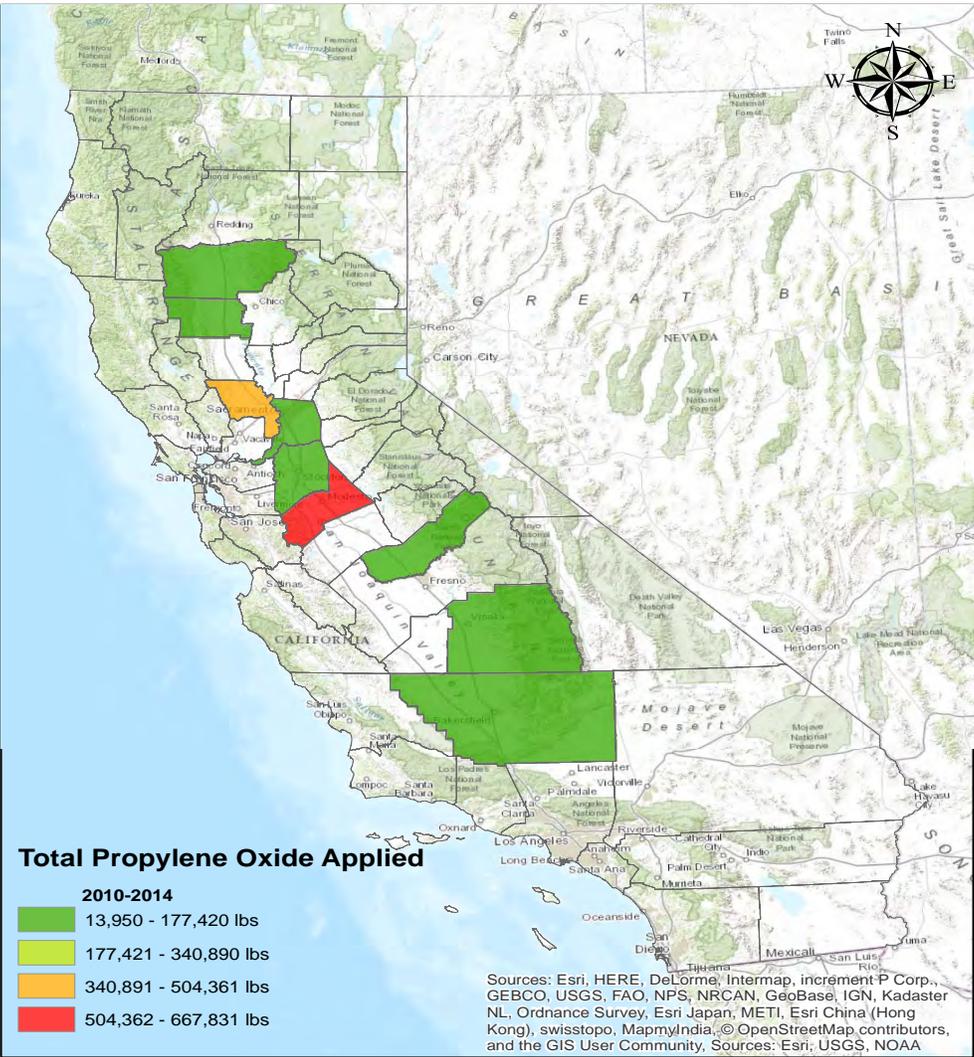


Figure 1 Map displaying cumulative applications of propylene oxide in California. Data covers the period from January 1, 2010, to December 31, 2014.

Based on 681 records queried from the PUR database, a total of 1,765,141 pounds of PPO were applied in California from 2010 – 2014 (Figure 2). For all the years queried, 2011 was the year with the highest total amount of PPO applied (418,210 lbs) followed by 2013 (410,360 lbs). All applications of PPO from 2010 – 2014 were commodity fumigations (100 % of the total reported use). Additionally, commodity fumigation use of PPO in 2013 was 1.3 times higher than in 2012 or 2014, while a significant decrease in agricultural application of PPO was being observed in 2014 (324,083 lbs).

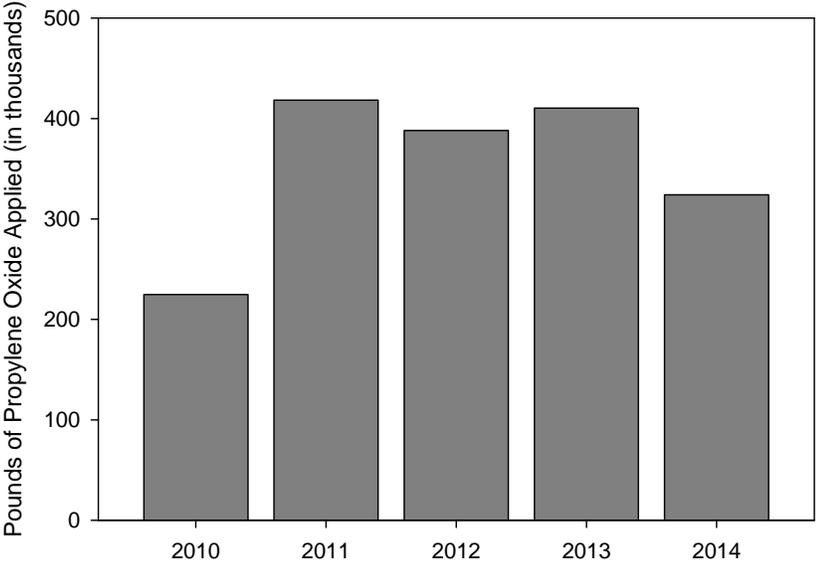


Figure 2 Application of propylene oxide as commodity fumigation on California crops during 2010-2014

Table 2 represents PPO use by county from 2010-2014. Stanislaus County was the county with the highest reported use of PPO from 2010-2014 (521,168 lbs.), followed by Yolo County (374,847 lbs.). Additionally, with the exception of Madera County, 2011 was the year with the highest reported PPO use for all counties in which PPO was applied. Figure 3 indicates that among the listed counties, Stanislaus (30%) and Yolo Counties (21%) consistently had the highest amount of PPO applied for all years queried. On the contrary, Glenn County had the lowest PPO use reported (1%) for all four years.

Table 2 Total pounds of propylene oxide used in each California county from 2010 – 2014

Counties	2010	2011	2012	2013	2014	Grand Total
Glenn	2,040	2,730	3,720	2,640	2,820	13,950
Kern	69,840	26,190	25,020	31,140	24,210	176,400
Madera			44,351	48,800	43,568	136,718
Sacramento	28,519	27,291	28,785	33,003	23,625	141,223
San Joaquin	9,171	29,089	49,258	42,720	9,424	139,663
Stanislaus	59,356	118,588	143,046	145,569	54,609	521,168
Tehama	21,832	24,195	24,409	53,816	41,861	166,113
Tulare	5,051	21,600	17,414	19,245	31,750	95,060
Yolo	28,766	168,527	51,911	33,427	92,216	374,847
Grand Total	224,574	418,210	387,913	410,360	324,084	1,765,141

Source: PUR database

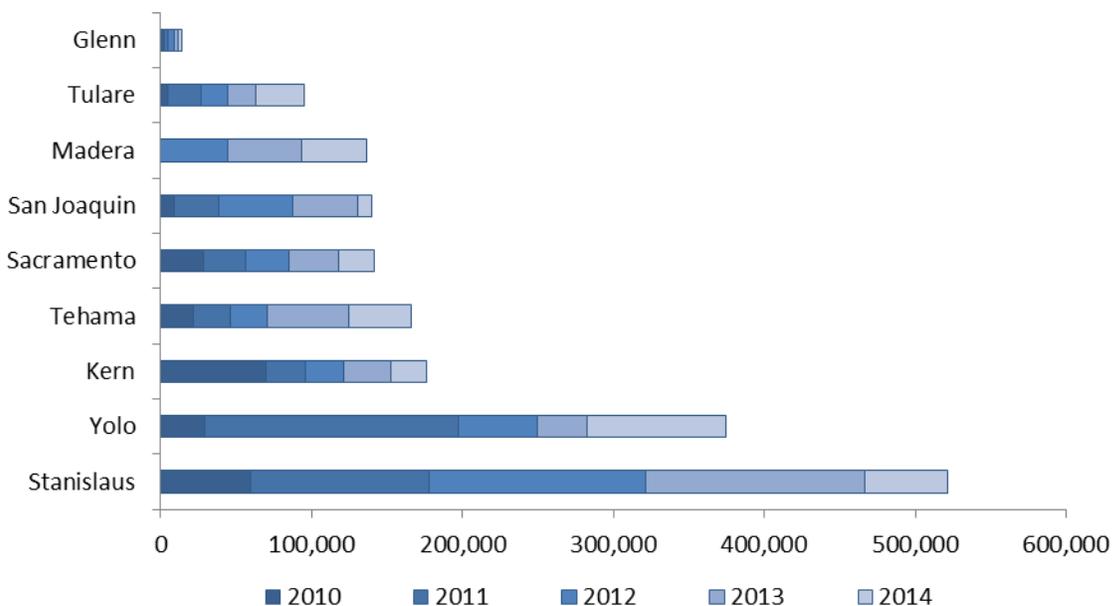


Figure 3 Pounds of propylene oxide used in each California county from 2010 – 2014

4 Environmental Fate and Degradation

PPO is not expected to persist in the environment due to its significant mobility in water, air, and soil (US EPA 2006). The environmental fate and degradation of PPO with special attention provided to air, soil and water are discussed below.

4.1 Environmental Fate and Degradation in Air

PPO, with a vapor pressure of 538 mmHg, is expected to exist solely as a vapor in the atmosphere (Dow 2015). Once in air, PPO reacts with hydroxyl radicals to create formanhydride, formaldehyde, acetyl formyl oxide and methyl glyoxal, which themselves are expected to degrade further (Alberta 2002; APVMA 2012). PPO is not expected to react with ozone. It is expected to eventually transform into carbon dioxide and water, and it is not considered an important cause of photochemical air pollution (APVMA 2012). The rate of this reaction has been determined by a number of investigators. Table 4 summarizes the published results.

Table 3 Rate of reaction of propylene oxide with hydroxyl radicals in the atmosphere

Rate Constant ($10^{-12} \text{ cm}^3 \cdot \text{Molecule}^{-1} \cdot \text{s}^{-1}$)	Half-life (days)*	Method	Reference
1.11±0.75	14.4	Relative rate (n-butane as reference compound)	<u>Edney et al. (1986)</u>
0.46±0.052	32.3	Absolute rate (n-butane as reference compound)	<u>Wallington et al. (1988)</u>
1.30±0.8	12.3	Relative method	<u>Winer et al. (1979)</u> (in RIVM1988)
0.53±0.07	30.1	Absolute method	<u>Zetsch and Stahi (1981)</u> (in RIVM 1988)
2.40	6.6	Smog chamber	<u>Pitts (1979)</u> (in IUCLID)

*Using OH radical concentration of $5 \times 10^5 \text{ molecule cm}^{-3}$

Source: EU 2002, Risk assessment report: Methyloxirane (propylene oxide)

PPO reactions with O_3 and NO_3 are negligible, though daytime removal by reaction with hydroxyl radicals is expected to occur at a slow rate (Grosjean, 1990). Grosjean (1991) proposed a tentative mechanism for the reaction of PPO with hydroxyl radicals as shown in Figure 4. Hydrogen atom abstraction is expected to involve the tertiary C_2 hydrogen atom, and unimolecular decomposition for each alkoxy radical may involve C-C as well as C-O bond scission. This process leads to formaldehyde, CH_3CO , acetaldehyde, and HCO, which reacts with O_2 to give carbon monoxide and HO_2 .

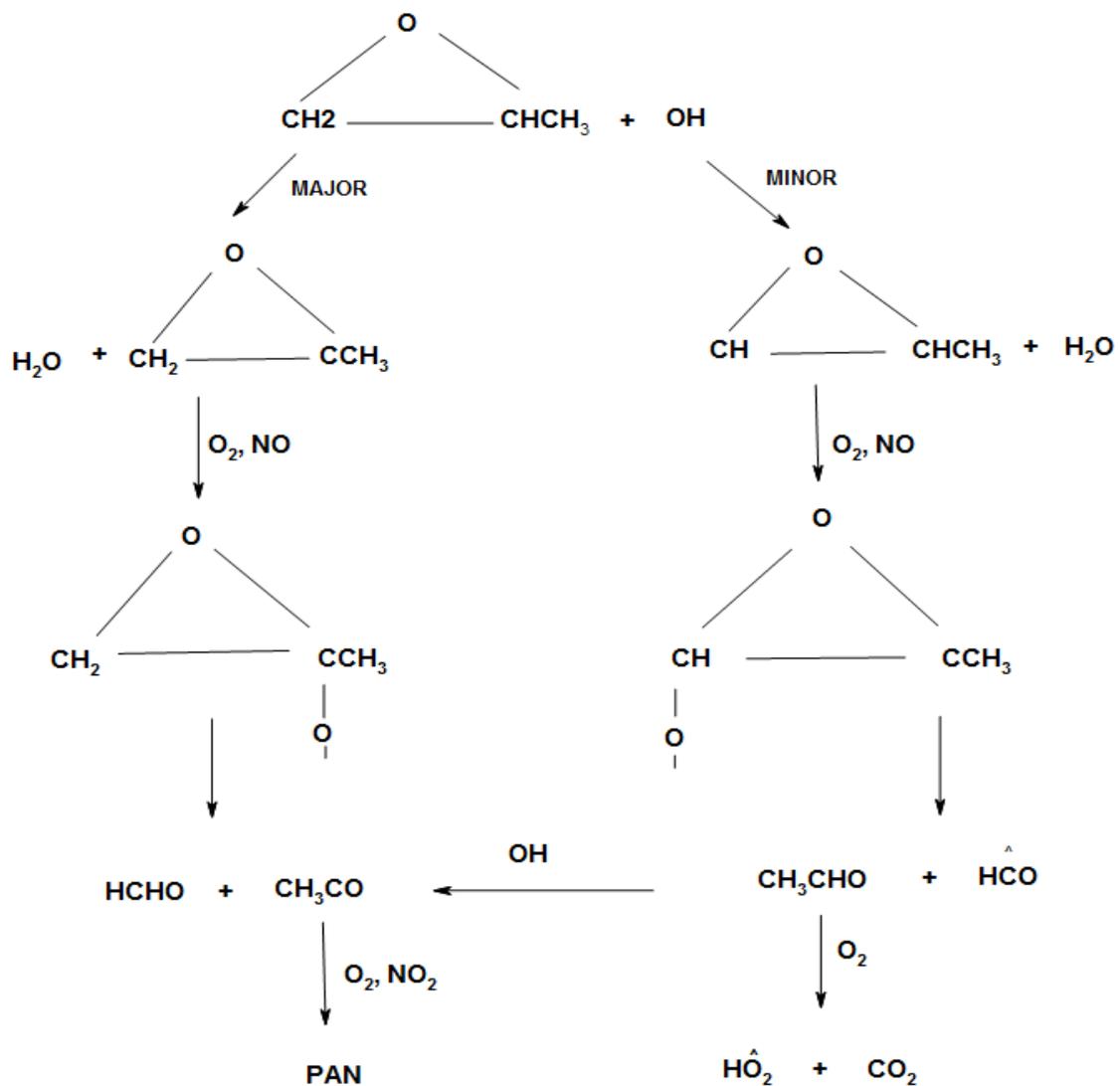


Figure 4 Proposed mechanism of the reaction of propylene oxide with hydroxyl radicals

Source: Grosjean, 1991. Atmospheric Chemistry of Toxic Contaminants: Journal of the Air & Waste Management Association

4.1.1 Photolysis

PPO does not absorb solar radiation appreciably at wavelengths greater than 300 nm (it has a maximum absorption at 199.5 nm) (APVMA 2012). Thus, direct photolysis does not occur. Its main degradation pathway is expected to be through reaction with photochemically produced hydroxyl radicals.

4.1.2 Other Pathways

Due to the high water solubility of PPO, it can also be removed by precipitation. PPO is not expected to persist in air, as it has a half-life of up to 20 days (IARC 1994; EU 2002). However, this half-life is long enough to subject PPO to transboundary transport and to affect ecosystems following dry and wet deposition.

4.2 Environmental Fate and Degradation in Soil

There are no direct application routes that have been found for PPO to the soil compartment, and the only route for soil exposure is through deposition from air (EU 2002). Due to its high K_{oc} (25) and low $\log K_{ow}$ (0.05) values, PPO is not expected to adsorb to suspended solid and sediment and will have a high mobility in this environmental compartment (Shell 2011; Canada 2008). The most important PPO removal process from soil is through volatilization (Henry's Law constant of 6.96×10^{-5} atm·m³/mole) (Grosjean, 1991). Hence hydrolysis and adsorption are not expected to be important fate processes in soil ecosystems (EU 2002). It is indicated in various studies that degradation in wet soil occurs through hydrolysis, while in dry soil PPO will evaporate into the air (ECIC 2009).

4.2.1 Biodegradation

Studies indicate that PPO is a likely candidate for biodegradation by microorganisms (APVMA 2012). It appears that microbial degradation of PPO is favored in the presence of acclimated microorganisms. Therefore, PPO is assumed to be inherently biodegradable (APVMA 2008). Microorganisms from the effluent of a biological sanitary waste treatment plant were found to degrade PPO very slowly (IPCS 1985). However, biodegradation is unlikely to contribute to the

overall degradation of PPO due to PPO's high volatility at ambient temperatures, which leads to volatilization and thus less likely to be available to degrading microorganisms (EU 2002). Evidence for the biodegradation of PPO is variable. One study was reported where the bacterium, *Nocardia* A60, was able to utilize PPO as a carbon source for growth (Danyluk *et al.*, 2005). *Pseudomonas graveoleus* and *P. fluorescens* are also reported to be capable of slowly biodegrading PPO at concentrations of <700 ppm in industrial wastewaters using aeration tanks (APVMA 2012).

4.3 Environmental Fate and Degradation in Water

PPO is highly soluble in water (590 g/L). If released to water, PPO is expected to hydrolyse to form 1, 2-propylene glycol, which relatively rapidly degrades in water (APVMA 2012). The length of time for PPO degradation will vary based on the conditions of the aquatic medium. In fresh water, aqueous solutions of PPO degrade to propylene glycol with an average half-life of 12 days, and in seawater, PPO will degrade to chloropropanol with an average half-life of 2-4 days (EU 2002).

4.3.1 Hydrolysis

PPO can degrade in water through hydrolysis and related ionic reactions involving the cleavage of a carbon-oxygen bond with an estimated half-life of 4-12 days (at pH's 7-9) and 6.6 days (at pH 5) at 25 °C (Alberta 2002). The hydrolysis reaction can be spontaneous (under neutral conditions), or acid or base catalyzed (Bogyo *et al.*, 1980). PPO reaction with halogen ions in water is fairly common. For example, 1-chloro-2-propanol (90 %) and 2-chloro-1-propanol (10 %) are formed by the reaction of PPO with chlorine in water under neutral pH; hence, PPO is classified as readily hydrolysable (APVMA 2012). It has been estimated that 70 % of PPO biodegrades in 30 days in water (US EPA 2006).

4.3.2 Volatilization

PPO is a highly volatile compound. Based upon this compound's estimated Henry's Law constant (6.96×10^{-5} atm-cu m/mole) and water solubility ($5.9 \times 10^{+5}$ mg/l), it is expected to volatilize moderately from water surfaces (NCBI 2016; EC 2008). The volatilization half-life for a modelled river (1 m deep, flowing 1 m/sec, wind velocity of 3 m/sec) has been estimated at 12 hours and volatilization from a modelled Lake was determined to be 6 days (NCBI 2016). Due to its

chemical-physical properties, PPO is not expected to be partitioned from water to sediment (APVMA 2012; NCBI 2016).

4.3.3 Photolysis

Although PPO's water solubility is considered moderate ($5.9 \times 10^{+5}$ mg/l), wet deposition of category constituents is not likely to play a significant role in their atmospheric fate because they rapidly photodegrade (EU 2002). Therefore, in aquatic environments, the aqueous photolysis and hydrolysis will not play a major role in the transformation of category constituents because they are not susceptible to these reactions (EU 2002).

4.3.4 Aquatic Organisms

The available aquatic toxicity data indicate that PPO is slightly toxic to fish, with the rainbow trout being reported as the most sensitive species having a 96-h LC₅₀ of 52 mg/L (Shell Research 1986; EU 2002). Table 5 represents the acute toxicity effects of PPO to aquatic organism. It is reported that the 3 common salt-water mullets show a similar sensitivity to the freshwater species, having a 96-h LC₅₀ of 89 mg/l (Crews 1974). Deneer *et al.* (1988) carried out long-term toxicity test with the guppy (*Poecilia reticulata*). They reported a 14-day LC₅₀ of 31.9 mg/l for this species. The lowest no-observable effect concentration (NOEC) was reported for *Oncorhynchus mykiss* (96-h NOEC of 20 mg/l) (Shell Research 1986). No toxicity data for non-aquatic non-mammalian organisms were identified. Due to the limited potential for exposure to aquatic organisms and to the low toxicity observed, PPO poses a negligible risk to the aquatic environment from its intended use.

Table 4 Acute toxicity tests of exposure of propylene oxide to aquatic organisms

Species	Size (mm)	pH	Dissolved oxygen (mg/l)	Water hardness (mg/l CaCO ₃)	Water temp (°C)	Flow/static	Effect conc	Data validity	Reference
							* n - nominal conc * m -measured conc		
Freshwater species									
Bluegill <i>Lepomis macrochirus</i>	30-40	--	--	--	23	Static	96-hr NOEC 150 mg/l (n) 96-hr LC ₅₀ 215 mg/l (n) 24- hr LC ₁₀₀ 240 mg/l (n)	Use with care ²⁾	Crews (1974)
Mosquito fish <i>Gambusia affinis</i>	20-30	--	--	--	23	Static	96-hr NOEC 130 mg/l (n) 96-hr LC50 141mg/l (n) 48- hr LC100 240 mg/l (n)	Use with care ²⁾	Crews (1974)
Goldfish <i>Carrasius auratus</i>	62±7	6 - 8	>4	--	20±1	Static	24-hr LC50 170 mg/l (m)	Use with care ²⁾	Bridié et al. (1979)
Rainbow trout <i>Oncorhynchus mykiss</i>	44-51	8-8.4	8-10.2	218-228	18.5	Static renewal	96-hr NOEC 20 mg/l (n) 96-hr LC50 52 mg/l (n) 72-hr LC100 100 mg/l (n)	Valid ¹⁾	Shell Research (1986)
Guppy <i>Poecilia reticulata</i>	--	6.8-7.1	>4.6	--	23	Semi-static	14-day LC50 549.5 µmol/l =31.9 mg/l (n)	Not Valid ¹⁾	Deneer et al. (1988)
Saltwater species									
Common mullet <i>Mugil cephalus</i>	70-80	--	--	--	23	Static	96-hr NOEC 80 mg/l (n) 96-hr LC50 89 mg/l (n) 48-hr LC100 100 mg/l (n)	Use with care ²⁾	Crews (1974)

NOEC and LC50 based on survival

1) EPA Test; 2) APHA Guideline; 3) Insufficient experimental detail

Source: European Union Risk Assessment Report Methyloxirane (Propylene Oxide). EU 2002

5 Persistence

Different studies report that PPO meets the persistence criterion in air (half-life of ≥ 2 days) (EC 2008). As PPO is hydrolysable with a half-life of 10.7–21.7 days and is up to 100% biodegradable, it is not expected to persist in water. A model-predicted biodegradation half-life of 15 days in water (BIOWIN 2000) was used to estimate the PPO half-life in soil and sediment by applying Boethling's extrapolation factors ($t_{1/2 \text{ water}}: t_{1/2 \text{ soil}}: t_{1/2 \text{ sediment}} = 1:1:4$) (Boethling, 1995). The resultant values indicate that PPO with half-life values of 182 days is not persistent in water, soil, and sediments (EC 2008).

6 Human Health

The main identified route of human exposure is through inhalation, but no relevant data have been published listing ambient PPO levels at a distance from point sources. Also there is insufficient data to estimate the levels of exposure to (PPO) use as fumigant through residues in food. An analysis revealed that the main PPO degradation products found in fumigated food products are

chloropropanols and 1, 2-propanediol, which are more persistent than the parent compound (IPCS 1988).

Based on human exposure and observed data in rodents, PPO is identified as “possibly carcinogenic to humans” (IARC 1994), and a “probable human carcinogen” (US EPA 1994). US EPA review of data submitted by industry indicated that a threshold mode of action for the carcinogenic effects is highly plausible (US EPA 2006). While this information is being reviewed further, a potential human health risk of concern associated with the current registered uses of PPO exposures has been identified (US EPA 2006). Csandy and Filser (2007) conducted a study on rodent species in order to evaluate the effects of PPO exposure on physiological toxicokinetics. It has been identified that the toxicokinetics of PPO are similar in humans and rats, and the data suggest that both genotoxic and non-genotoxic mechanisms may play a role. Based on the significant uncertainties of exposure of the general population to PPO, it can be expected that actual exposure levels do not exceed these estimates (US EPA 2012).

7 Regulation

All pesticides sold or distributed in the United States are required to be registered with the US EPA. Additionally in California, all pesticides must also be registered with DPR. PPO is a registered fumigant in the United States nut industry, used for insect and microbial control for decades. PPO has a tolerance of 300 ppm for processed spices, cocoa beans or cocoa powder, processed nutmeats (except peanuts), and edible gums (US EPA 2006). Different recommended permissible exposure limits (PEL) for PPO have been recommended by different agencies: OSHA (Occupational Safety and Health Administration) recommended a PEL for PPO as 100 ppm at 8-h time weighted average (8-h TWA); the California Division of Occupational Safety and Health, better known as Cal/OSHA, recommended a PEL for PPO as 20 ppm at 8-h TWA.

Based on the Risk Assessment Initiation Report, PPO is ranked as the fifth active ingredient of top 10 active ingredients DPR has prioritized for risk assessment initiation (CDPR 2014). The US EPA has classified PPO as a “probable human carcinogen” and has listed this fumigant as a toxic air contaminant and “hazardous air pollutant” (US EPA 2006). In 1996, PPO was classified as a restricted use product (RUP) by US EPA (US EPA 2006). RUPs are not available for purchase or use by the general public and they are deemed to have “the potential to cause unreasonable adverse

effects to the environment and injury to applicators or bystanders without added restrictions.” Use by a certified applicator is required under the RUP classification. In California the use of a RUP requires a permit by a County Agricultural Commissioner’s office.

8 Summary

PPO is a product registered for control of stored pests in spices, in-shell nuts, nutmeats, and other dried foods. Due to its high vapor pressure and its tendency to hydrolyze, PPO does not persist in soil or water; bioaccumulation and adsorption to soil or sediment are negligible. When released to soil, it quickly evaporates into the atmosphere and, when released in air, PPO is photochemically degraded by hydroxyl radicals to form acetylformyl oxide, formaldehyde, and methylglyoxan. Once PPO is introduced into water, it reacts to form propylene glycol (hydrolysis). Due to its relatively high water solubility, PPO removal from air by rainfall is expected. Although the primary route of human exposure to propylene oxide is by inhalation, foods treated with PPO may contain residues of this fumigant unless adequate ventilation/aeration of the treated commodity is provided (CFIA 2011).

The major crops to which PPO is applied in California are almonds, walnuts, cashews and other different nut crops. The only use of PPO is commodity fumigation. During 2010-2014, a total of 1,765,141 pounds of PPO were applied in Glenn, Kern, Madera, Sacramento, San Joaquin, Stanislaus, Tehama, Tulare and Yolo counties. A noticeable upward trend in PPO use from 2004-2014 has been observed, and 2011 was the year of highest PPO use reported.

PPO is identified as a high priority chemical active ingredient as it is considered to pose a great potential for exposure to individuals in California (CDPR 2014). PPO has been classified as a RUP by USEPA on the basis of its carcinogenicity and genotoxicity. In California the use of a RUP requires a permit by a County Agricultural Commissioner’s office. Requiring a permit allows Commissioners to make sure restricted pesticide users follow appropriate procedure to prevent harmful effects.

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