Use Information and Air Monitoring Recommendation for the Active Ingredient Chloropicrin in California.

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By

Edgar Vidrio, Pam Wofford, and Fabio Sartori

Air Program
Environmental Monitoring Branch
Department of Pesticide Regulation
1001 I Street
Sacramento, CA, 95814

INTRODUCTION

The California Department of Pesticide Regulation (DPR) provides this recommendation to assist the California Air Resources Board (CARB) with the selection of appropriate locations to perform seasonal ambient air monitoring in Santa Barbara County for the pesticide active ingredient chloropicrin as part of the proposed monitoring included in the 2016 Budget Act. This recommendation also includes general information regarding the physical and chemical properties of chloropicrin as well as its reported use in the state.

Chloropicrin is a clear, colorless, and nonflammable liquid with a moderate vapor pressure and boiling point (Ajwa et al. 2010). Following a field fumigation, chloropicrin rapidly diffuses through the soil in all directions and dissipates following a first-order kinetics decay function, with estimated half-lives ranging from 1.5–5.8 days (Wang et al. 2000). Volatilization is the major pathway through which chloropicrin dissipates from soil, although tarps can significantly reduce volatilization (Gao et al. 2008). Table 1 lists selected physical and chemical properties of chloropicrin.

Chloropicrin was first registered in the U.S. in 1975 and is used as a broad spectrum, non-selective fumigant to primarily eliminate the soil fauna, acting as antimicrobial, fungicide, herbicide, insecticide, and nematicide. It is used for (1) pre-plant soil fumigations, (2) residential uses (warning agent for sulfuryl fluoride in structural fumigations), and (3) other specialized fumigations, such as spot tree replant sites and remedial wood treatments. Because of its high volatility, chloropicrin is not expected to be found as a residue in/or any agricultural commodities that are subject to its application (USEPA 2008). It is often used in combination with 1,3-D as a fungicide to achieve a broad spectrum control (Ajwa et al. 2010).

Major exposure risks are primarily for handlers, bystanders, and workers, because of the possible movement off-site following chloropicrin use as a fumigant. In December 2010, DPR issued a Risk Management Directive (RMD) on pesticide products containing the active ingredient chloropicrin to mitigate unacceptable acute exposures to residents and bystanders. Improved risk reduction was achieved primarily by focusing on calculating buffer zones for site-specific meteorological conditions and specific tarp types, as well as focusing on selecting appropriate time intervals prior to tarp cutting (CDPR 2015).

Since methyl bromide is being phased out as a fumigant in most recent years, growers have been using alternative fumigants, particularly 1,3-dichloropropene (1,3-D) and chloropicrin (Ware and Whitacre 2004). The latter is sufficiently reactive that it may contribute to the formation of tropospheric ozone, a major component of photochemical smog in urban and rural areas (Carter et al. 1997; Gan et al. 2000; Ajwa et al. 2013).

CHLOROPICRIN USE IN CALIFORNIA

Pesticide use information was obtained for the calendar years 2012–2014 from the Pesticide Use Report (PUR) database maintained by DPR (CDPR 2011). The most commonly used products containing chloropicrin were Pic-Clor 60 (32 %), Tricon50/50 (18 %), Tri-Clor (9 %), Tri-Clor Ec (8 %), Terr-O-Gas (7 %), Inline (6 %), and Pic-Brom 25 (5 %) (Table 2). These were used in 86 % of all agricultural chloropicrin applications or 6,103 applications.

Table 1. Selected physical and chemical properties of chloropicrin.

Property	Value	Description	Reference
Common name	Chloropicrin		
Chemical names	trichloro(nitro)methane, nitrochloroform		Kidd and James 1991; Ware and Whitacre 2004
CAS registry number	76-06-2		
Molecular formula	Cl_3CNO_2		
Molecular weight (g/mol)	164.4		Kidd and James 1991
Boiling point (°C)	112.4 (at 757 mm Hg)		Kidd and James 1991
Density (g/mL)	1.66 (20 °C)		Kidd and James 1991
Water solubility (g/L)	1.62 (25 °C)	Limited solubility in water. Miscible with most organic solvents, such as acetone, benzene, ethanol, and methanol.	Kidd and James 1991;Wilhelm et al. 1996
Vapor pressure (kPa)	3.2 (25 °C)	Moderate vapor pressure	Kidd and James 1991
K_d[a]	1.2–5.9	Freundlich equation as $Q = K_d \cdot C_w^{1/n}$, where Q (µg/g) is adsorbed amount per gram of soil, K_d adsorption constant per gram of soil, and C_w the equilibrium concentrations in water (µg/g).	Kawamoto and Urano 1989
K_{oc} (L/kg)[b]	36.05	Low soil adsorption	USEPA 2008
log(K_{ow})[c]	1.8–4.7		Kawamoto and Urano 1989
K_H (atm m^3/mole)	$2.51 \cdot 10^{-3}$	High Henry's Law Constant	CDFA 1989
$t_{1/2}$[d]	4.5 days	Based on laboratory incubations of sandy loam soils at 25 °C	Wilhelm et al. 1996
	1.3 hr	Flooded soils (treated at 500 lbs/ac) incubated at 25 °C under dark conditions.	Wilhelm et al. 1996
	1.5, 4.3, and 0.2 days for Arlington, Carsitas, and Waukegen soil, respectively	Investigation on the effects of soil temperature and moisture on chloropicrin degradation in Arlington sandy loam (coarse loamy, mixed, thermic Haplic Durixeralf; Riverside, CA), Casitas loamy sand (mixed, hyperthermic Typic Torripsament; Coachella Valley, CA), and a Waukegen silt loam (fine silty over sandy or sandy-skelatal, mixed, mesic Typic Hapludoll; Rosemont, MN).	Gan et al. 2000
	6.3, 13.9, and 2.7 days (same as previous soils)	Same as previous but for the corresponding sterilized soils.	Gan et al. 2000

aK_d, soil-water partition coefficient; bK_{oc}, soil-water partition coefficient normalized for soil organic carbon; cK_{ow}, octanol-water partition coefficient; d$t_{1/2}$, half-life value estimated using a first-order kinetics regression model.

Table 2. Summary of commercial products containing chloropicrin used in California for agricultural applications during 2012–2014.

Product name	% as active ingredient[1]	Number of applications[2]	
		n	%
Pic-Clor 60	56.6, 59.6	2628	32.4
Tri-Con 50/50	49.7, 50	1431	17.7
Tri-Clor	99	770	9.5
Tri-Clor Ec	94	672	8.3
Terr-O-Gas	20, 25, 33, 41.5, 50	602	7.4
Inline	33.3	477	5.9
Pic-Brom 25	24.9, 25	383	4.7
Tri-Con 57/	42.7, 43	271	3.3
Telone C-35	34.7, 35	268	3.3
98-2 Contai	2	155	1.9
Mbc-33 Soil	33	127	1.6
Mbc-33	32.8	122	1.5
Chloropicri	99	109	1.3
Brom-O-Gas	2	51	0.6
Tri-Con 45/	54.7	10	0.1
Pic Plus Fu	85.5	3	0.0
Metapicrin	100	2	0.0
Brom-76	1	1	0.0
Methyl Brom	2	1	0.0
Telone C-17	16.5	1	0.0

[1] "n" = number of applications; "%" = number of applications as percentage of total number (8107).
[2] Percentage of chloropicrin on a mass basis contained in a product. Certain products under the same name were reported in the PUR with different percentages.

Top counties and sites/crops

The cumulative use of chloropicrin in California in the top three counties during 2012–2014 was highest in 2012 (6,297,400 lbs or 75 % of statewide total applied mass during the same period) and lowest in 2013 (5,464,600 lbs or 71 % of the total yearly mass applied in the California) (CDPR 2016, Table 3). The overall highest uses (2012–2014) were in Ventura (28% of total statewide use), Monterey (25 %), and Santa Barbara Counties (16 %). The other counties with lower uses over the same period were Santa Cruz (8 %), Siskiyou (6 %), and San Luis Obispo Counties (5 %) (Table 3). These data indicate that the use of chloropicrin is greatest in coastal areas in the southern coastal regions of Ventura and Santa Barbara counties and in the northern coastal regions of Monterey and Santa Cruz counties where berry-type crops are typically cultivated.

Chloropicrin use was highest on strawberry and raspberry in Ventura County representing 24 % and 3 %, respectively, of total chloropicrin mass applied during 2012–2014 in California (Table 4). Similarly, in Monterey County, use on these two crops represented 23 % and 1 %, respectively, of total chloropicrin mass applied during 2012–2014 in California (Table 4).

Table 3. Number of applications (n) and reported use of chloropicrin by year in the top ten California counties (ordered from top to bottom) during 2012–2014[1] (CDPR 2016).

County/Year	2012	2013	2014	2012	2013	2014	2012-2014			
	n			lbs			Total number of applications	CA[2] (%)	Total pounds applied	CA[1] (%)
Ventura	536	389	407	2,742,600	2,174,500	2,283,200	1332	16	7,200,300	28
Monterey	717	605	717	2,157,000	2,015,000	2,263,300	2039	25	6,435,300	25
Santa Barbara	287	216	207	1,397,800	1,275,100	1,416,700	710	9	4,089,600	16
Santa Cruz	267	251	268	655,300	659,000	784,500	786	10	2,098,800	8
Siskiyou	181	195	177	592,600	508,000	452,000	553	7	1,552,600	6
San Luis Obispo	147	141	139	417,600	446,800	480,700	427	5	1,345,100	5
San Joaquin	123	189	140	166,700	184,800	227,200	452	6	578,700	2
Merced	115	141	104	95,200	198,200	192,400	360	4	485,800	2
Fresno	92	119	119	92,600	105,100	150,600	330	4	348,300	1
San Diego	29	38	76	56,600	114,700	124,800	143	2	296,100	1
Combined Total	2,494	2,284	2,354	8,374,200	7,681,300	8,375,300	7,132		24,430,800	
% of CA Total	87	88	89	94	94	93	88		94	

[1]Numbers are rounded to hundreds.

[2]Percentage of total chloropicrin mass applied in California during 2012-2014.

Table 4. Reported chloropicrin use by crop/site in the top five California counties (from left to right) based on total use during 2012–2014[1].

Ventura		Monterey		Santa Barbara		Santa Cruz		Siskiyou	
Crop/Site	Total Pounds	Crop/Site	Total Pounds	Crop/Site	Total Pounds	Crop/Site	Total Pounds	Crop/Site	Total Pounds
Strawberry	6,209,400	Strawberry	6,013,400	Strawberry	3,838,500	Strawberry	1,330,600	Soil application	1,547,400
Raspberry	701,400	Raspberry	248,300	Raspberry	189,300	Raspberry	696,000	Barley	2,900
Tomato	142,500	Lettuce	61,800	NOCFG[a]	33,200	Blackberry	49,000	Strawberry	2,400
Peppers	79,800	Spinach	42,300	Blackberry	28,500	NOCFGP[b]	7,000		
NOCFG	48,300	Blackberry	30,100	NGCFG[d]	150	NOCFG	5,800		
NOCFGP	8,500	NOCFG	24,300			NOGTPM[c]	4,600		
Blueberry	5,600	Lettuce	9,400			Soil Application	4,100		
Peppers	2,600	Asparagus	4,000			Research commodity	900		
Lemon	1,300	Peppers	1,700			Blueberry	800		
Blackberry	1,100								

[1]Numbers are rounded to hundreds

[a]NOCFG, Nursery, Outdoor Cut Flowers or Greens; [b]NOCFGP, Nursery, Outdoor Container/Fld Grown Plants; [c]NOGTPM, Nursery, Outdoor Grown Trnsplnt/Prpgtv Mtrl; [d]NGCFG, Nursery, Greenhouse Cut Flowers or Greens.

Top townships and sections

The PUR database provides location information as a one mile square section defined by the Public Land Survey System (PLSS). The top two use sections during 2012–2014 in California were located in Ventura County and Santa Barbara Counties, with chloropicrin use totals of 352,000 lbs (1 % of the total chloropicrin mass) and 315,000 lbs (1 % of statewide total mass), respectively (data not shown). The third (S10N34W25), fourth (S10N34W5), and fifth (S10N33W18) overall highest sections were all located in Santa Barbara County near the city of Santa Maria and highway 101 and each corresponded to roughly 1 % of the 2012–2014 statewide total mass applied in California.

Use information can also be summarized on a township level (6x6 mile area or 36 square miles). The top three townships were located in Ventura and Santa Barbara counties (Table 5): the chloropicrin mass applied during 2012–2014 in the two Ventura County townships corresponds to 7 % (S01N21W) and 5 % (S02N22W) of the total chloropicrin mass applied in California during the same period, whereas that applied in Santa Barbara County corresponds to 7 % (S10N34W).

The mass applied in township S10N34W, which includes the city of Santa Maria, was ranked the highest overall for 2014 (Table 6). Additionally, CARB at the request of DPR has conducted seasonal air monitoring in both Ventura and Santa Barbara Counties (S01N21W and S10N34W) in 2014 and 2015 during times of high chloropicrin use. Although 2012-2014 use was higher in township S01N21W, 2014 and 2015 monitoring results indicate that chloropicrin air concentrations were higher in the S10N34W township located in Santa Barbara County.

Table 5. Reported chloropicrin use in the top 10 townships in the state of California during 2012–2014[1].

County	Township	Sum (lbs)	CA[2] (%)
Ventura	S01N21W	1,925,400	7
Santa Barba	S10N34W	1,876,500	7
Ventura	S02N22W	1,415,800	5
Monterey	M14S03E	1,368,300	5
Ventura	S01N22W	1,256,600	5
Monterey	M12S02E	1,230,500	5
Monterey	M14S02E	1,166,500	5
Ventura	S02N21W	1,129,900	4
Santa Barba	S10N33W	1,086,100	4
San Luis Obispo	S11N35W	933,400	4

[1]Numbers are rounded to hundreds

[2]Percentage of total chloropicrin mass applied in California during 2012-2014.

Table 6. Chloropicrin mass applied in the top 10 townships based on use (from top to bottom) by year in the state of California during 2012–2014[1].

2012				2013				2014			
County	Township	Sum (lbs)	CA[2] (%)	County	Township	Sum (lbs)	CA[2] (%)	County	Township	Sum (lbs)	CA[2] (%)
Ventura	S01N21W	783,500	8.8	Ventura	S01N21W	602,600	7.3	Santa Barbara	S10N34W	641,400	7.1
Santa Barbara	S10N34W	663,700	7.4	Santa Barbara	S10N34W	571,400	7.0	Monterey	M14S03E	550,800	6.1
Ventura	S02N22W	525,700	5.9	Ventura	S02N22W	423,100	5.2	Ventura	S01N21W	539,300	6.0
Ventura	S01N22W	475,500	5.3	Monterey	M14S03E	415,400	5.1	Ventura	S02N22W	467,000	5.2
Ventura	S02N21W	447,000	5.0	Santa Barbara	S10N33W	407,500	5.0	Monterey	M12S02E	458,100	5.1
Monterey	M14S03E	402,100	4.5	Monterey	M12S02E	399,300	4.9	Monterey	M14S02E	422,300	4.7
Monterey	M12S02E	373,100	4.2	Ventura	S01N22W	375,300	4.6	Ventura	S01N22W	405,800	4.5
Monterey	M14S02E	372,000	4.2	Monterey	M14S02E	372,100	4.5	Ventura	S02N21W	369,200	4.1
Santa Barbara	S10N33W	354,900	4.0	Ventura	S02N21W	313,600	3.8	San Luis Ob	S11N35W	335,000	3.7
San Luis Obispo	S11N35W	294,500	3.3	San Luis Obispo	S11N35W	303,900	3.7	Santa Barbara	S10N33W	323,700	3.6

[1]Numbers are rounded to hundreds

[1]Percentage of total chloropicrin mass applied in California during 2012-2014.

Seasonal patterns

When considering the time series of the most recent years (2012–2014), maximum monthly use within a calendar year for the combined top five counties occurred always in September (Figure 1). Similarly, when considering the same time series but split by county, the month of highest use was September in Monterey County (2,743,000 lbs), Santa Barbara County (1,740,000 lbs), and Ventura County (2,062,000 lbs).

When considering the temporal series for the 2014 overall highest use township (S10N34W, Santa Barbara County), the 12-week period with overall highest use (for the cumulative use during 2012–2014) was during weeks 33–44, i.e., the 12-week period starting the last two weeks of August (Figure 2, top plot). This trend remained approximately the same when considering the use split by years (Figure 2, bottom plot). In 2012, 2013, and 2014, the first application of week 33 occurred on August 12, August 18, and August 17, respectively. Instead the last application of week 44 in 2012, 2013, and 2014 occurred on November 12, November 13, and November 8, respectively.

Figure 1. Total chloropicrin mass applied in a specific month in the top five California counties (based on use) during 2012–2014.

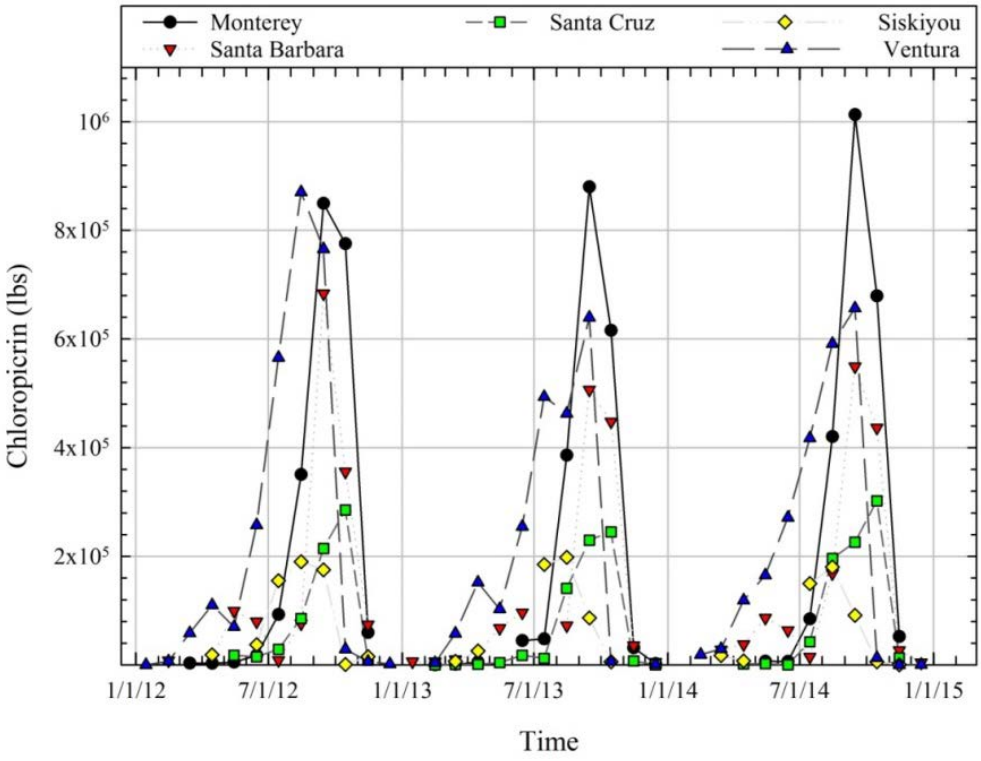
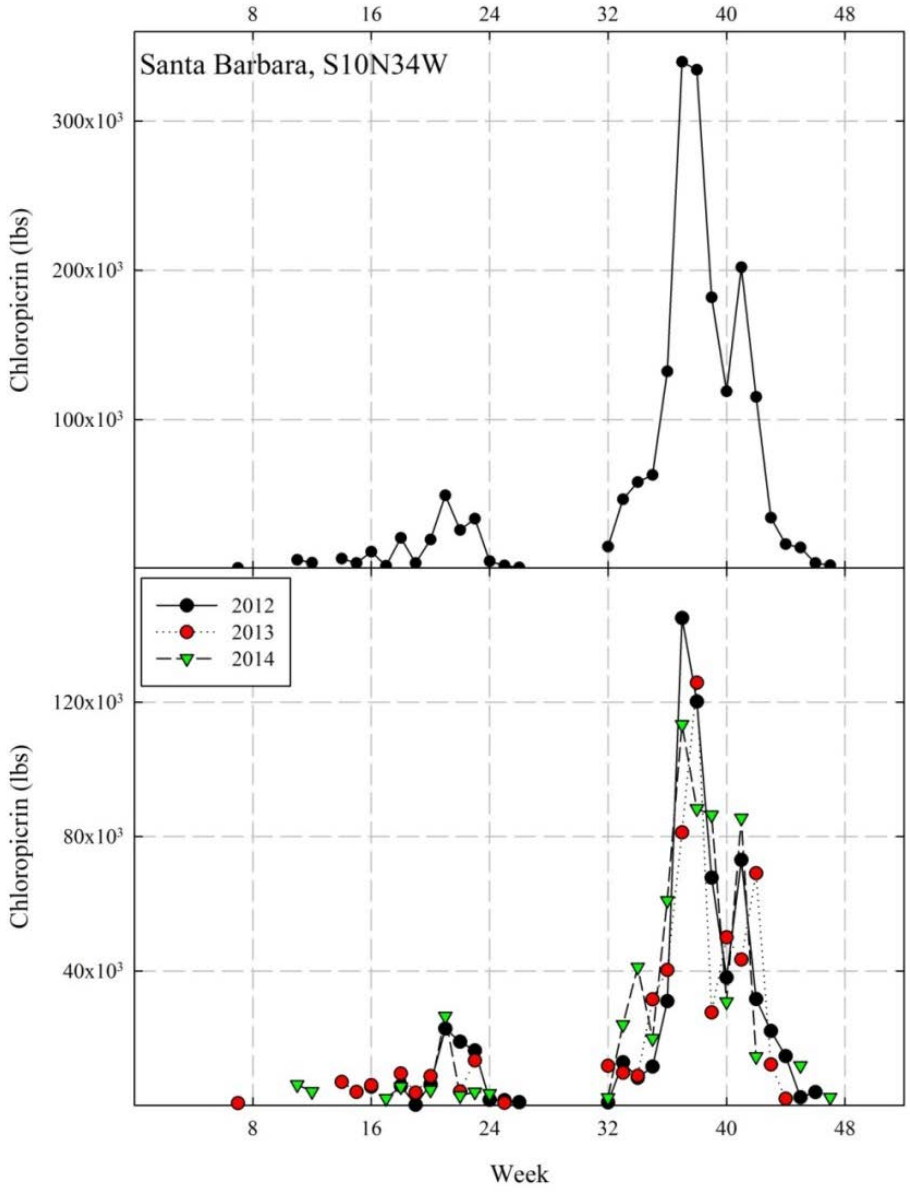


Figure 2. Weekly chloropicrin mass applied in the top township (2014 6 mile x 6 mile area) of Santa Barbara County (top plot) during 2012–2014 and split by year (bottom plot).



C. RECOMMENDATION

Ambient Air Monitoring

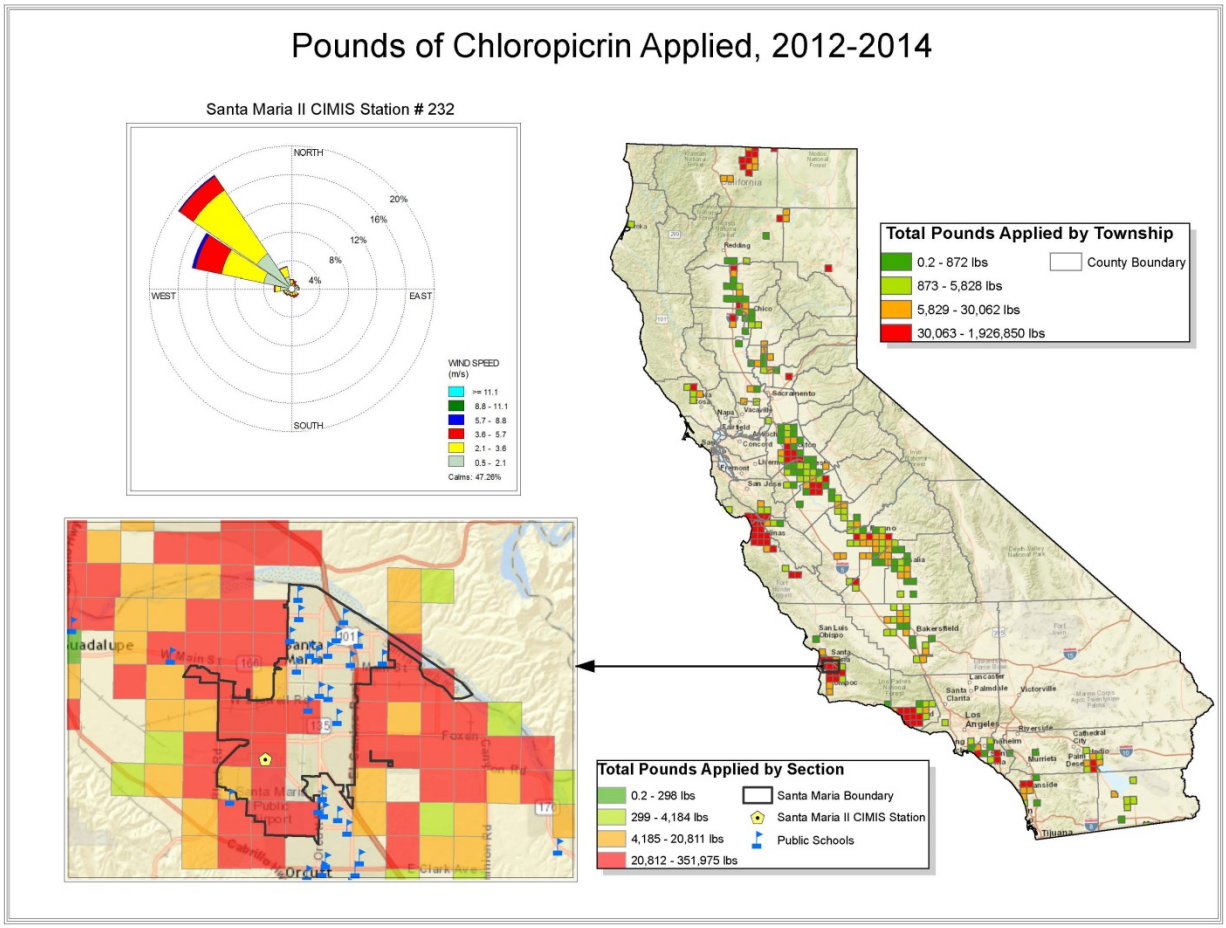
Township S10N34W, which is located in Santa Barbara County and makes up most of the Santa Maria City area, was the California township with the highest mass of chloropicrin applied in 2014 (641,400 lbs; 7.1% of total state chloropicrin use). At the request of DPR, CARB conducted ambient air monitoring for chloropicrin in 2014 and 2015 in high use areas located in Ventura, Monterey and Santa Barbara Counties as part of DPR's Toxic Air Contaminant Program (Vidrio et al., 2015). The Santa Barbara County sampling location was inside of the S10N34W township. The highest chloropicrin air concentrations detected in both 2014 and 2015 were measured in township S10N34W. Additionally, due to an increase in chloropicrin use in the S10N34W township from 2012-2014, this region currently ranks as the highest use 36-square-mile area in the state. Therefore, DPR requests that CARB conduct seasonal ambient air monitoring for chloropicrin in 6-8 locations in the Santa Maria City region near areas of high chloropicrin use (S10N34W, S11N34W, S09N34W, S10N35W, S11N35W, S10N33W, and S09N33W) in addition to one background site located away from areas of use.

Monitoring sites should be located on schools or other public properties, near the edge of the community, and downwind of high use areas (Figure 3). Monitoring should be conducted over a 12-week period corresponding to the high-use chloropicrin season in this region, which occurs during the months of August, September, and October (weeks 33-44). At each sampling site, four 24-hour samples per week should be collected in sequence during each week of the 12-week sampling period. Each week, the four-day sampling period should begin on randomly chosen days over the full seven-day week, including weekends. In addition to the primary samples, replicate (co-located) samples are needed for at least 10% of the total number of samples collected during the 12-week period. Field spike samples should be collected under the same environmental conditions (e.g., temperature, humidity, exposure to sunlight) and monitoring study conditions (e.g., air flow rates, sample transportation and storage) as those occurring at the time of ambient air sampling. DPR recommends target 24-hour quantitation limits of at least 0.003 ppb for chloropicrin.

DPR requests that CARB provide the following in its ambient air monitoring report submitted to DPR:

1. Location of each sampler (if at a school, location of sampler within school grounds);
2. Sampling height and any information relevant to the study (presence of trees, buildings, particular industrial or commercial facilities and activities);
3. Proximity of the sampler to treated or potentially treated fields, including distance and direction;
4. Latitude and longitude coordinates for sampling sites, including the Datum used (e.g., NAD 27 or NAD 83); and
5. A map of the monitoring site locations and photographs of sample locations.

Figure 3. Total chloropicrin mass applied by township in California and in the Santa Maria Region during 2012–2014.



Suggested Communities for Air Monitoring

DPR evaluated 1,267 communities for chloropicrin use. The communities included all cities and census designated places (CDPs) in California, except those within the urban counties of Los Angeles, Orange, San Diego, and San Francisco (Andrews and Verder-Carlos, 2014).

DPR evaluated and rated each of the 1,267 California communities using the following criteria:

a) Regional use density (lbs of active ingredient used) during 2012-2014 (between 1 and 5 miles outside community boundary)
b) Local use density during 2012-2014 (within 1 mile outside community boundary)
c) Community use density during 2012-2014 (within the community boundary)

Ratings were given to the communities within the Santa Maria City region of Santa Barbara County according to the statewide rankings of the 1,267 communities evaluated. Table 7 summarizes the results of the evaluation of pesticide use and the Office of Environmental Health Hazards Assessment's (OEHHA) CalEnviroScreen 2.0 (CES 2.0) Population Characteristics percentile for those communities. For any California census tract, the CES 2.0 Population Characteristics percentile factors the following parameters: percent of children and elderly in the population, percent of low birth-weight births, and

the rates of asthma emergency department visits, educational attainment, linguistic isolation, poverty, and unemployment (OEHHA, 2014).

The highest use of chloropicrin from 2012 to 2014 occurred in the cities of Santa Maria and Guadalupe, followed by the Woodlands, Orcutt, and Nipomo Census Designated Places (CDPs). Santa Maria and Guadalupe Cities also had the highest CES 2.0 Population Characteristics percentile ratings among the communities in Santa Barbara and San Luis Obispo Counties.

DPR recommends CARB select communities where high use of chloropicrin occurs, guided by the community rating from Table 7, and where the CES 2.0 Population Characteristics percentiles are the highest. DPR suggests that CARB consider the communities of Santa Maria, Guadalupe, Orcutt, and Nipomo as these communities are ranked highly for both use and environmental justice factors. Multiple sampling locations can be placed within a single community.

Table 7. Pesticide use and environmental justice factor ratings for communities in Santa Barbara and San Luis Obispo Counties during 2012-2014.

Community Name	County	Chloropicrin Use		CES 2.0 Population Characteristics	
		Statewide Rank[1]	County Rank[2]	Percentile	Rank[3]
Santa Maria city	Santa Barbara	13	1	74	1
Guadalupe city	Santa Barbara	13	2	73	2
Woodlands CDP	San Luis Obispo	18	3	30	4
Orcutt CDP	Santa Barbara	21	4	18	9
Nipomo CDP	San Luis Obispo	22	5	40	3
Callender CDP	San Luis Obispo	23	6	30	4
Garey CDP	Santa Barbara	28	7	23	7
Sisquoc CDP	Santa Barbara	35	8	23	7
Blacklake CDP	San Luis Obispo	49	9	30	4

[1] Community pesticide reported use ranking is out of possible 1,267 California communities evaluated.
[2] Community rankings based on reported use in San Luis Obispo and Santa Barbara Counties, with lower numbers indicate higher nearby pesticide use.
[3] Rankings for the top chloropicrin reported use communities in San Luis Obispo and Santa Barbara Counties, with lower numbers indicating higher environmental justice priority.

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