ATTACHMENT A

Rational for use of the 1,3-D runoff study as a surrogate for chloropicrin runoff
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As an EPA requirement for the re-registration of 1,3-Dichloropropene (1,3-D) in 1999, Dow AgroSciences conducted a field study near Blacksburg, VA to examine the potential for runoff of 1,3-D resulting from a shallow shank (12 inch deep) application of 1,3-D in a soil vulnerable to runoff exposed to worst-case natural and simulated rainfall events.[1] The 1,3-D was broadcast shank applied (no tarp) at a depth of 12 inches below the soil surface at a rate of 32.4 gallons per acre (367 kg/ha) in triplicate plots measuring 12.2m x 33.6m. In addition to natural rainfall that occurred at the site, a 2-hour simulated rainfall event applied 9.4 cm of rain (4.7 cm/hour) at the time that the peak emission (flux) of 1,3-D was expected to occur, about three days after application. The runoff study showed that 0.002% of the applied mass of 1,3-D was measured in water runoff at the edge of the treated plot, ranging from 4 to 17 ppb. The study was subsequently published in the Journal of Soil and Water Conservation.[2]

EPA (2019) states the following in its draft risk assessment for 1,3-D[3]:

> "*Although previous assessments used the best available models and data, the surface water modeling scenario used did not parameterize soil temperature or albedo, important parameters determining the extent of volatilization for incorporated volatile pesticides. The run-off field study is considered more reliable than the current models for estimating 1,3-D aquatic exposure from soil injection uses because it inherently accounts for volatility. Exposure can be estimated for registered uses by linear scaling of the application rate used in the study to maximum labelled application rates. Estimating exposure in this manner results in exposure estimates slightly lower than previously reported modeled EECs.*"

Although a similar field scale runoff study has not been conducted for chloropicrin, the similarity in physical/chemical and environmental fate properties of 1,3-D and chloropicrin suggest that the results of the 1,3-D runoff study would be a reasonable, yet conservative, surrogate for

estimating chloropicrin runoff. A comparison of 1,3-D and chloropicrin physical-chemical and environmental fate parameters is shown in Table 1.

Table 1. Physical/chemical and environmental fate parameters for 1,3-D and chloropicrin.

Parameter	1,3-D	Source	Chloropicrin	Source
Molecular Weight (g/mol)	110.97	EPIWin[4]	164.38	EPIWin[4]
Vapor Pressure (mm Hg @ 25ºC)	28.7[a]	MRID 40483803[5] MRID 40483804[6]	23.8	EPIWin[4] and Merck Index[7] (used by USEPA-EFED)
Solubility (mg/L) (25C)	2180[a]	MRID 40483801[8] MRID 40483802[9]	1620	EPIWin[4]
Log Kow	2.03	MRID 40483805[10] MRID 40483806[11]	2.09	EPIWin[4]
Henry Law Constant (20ºC) atm*m3/mole	3.55×10^{-3}	EPIWin[4]	2.05×10^{-3}	Kawamoto and Urano (1989)[12] - used by USEPA
Koc (L/kg)	44.7[b] (n=7)	MRID 40538901[13]	36.1	EPIWin[4]
Aerobic Soil Metabolism (days)	14.1[b] (n=18)	MRID 42642301[14] MRID 49382009[16]	3.7 days[c] 1.3 days[d]	MRID 43613901[15] USEPA DER (2008)[37]
Aerobic Aquatic Metabolism	4.6 days	MRID 44975502[17]	2.3 days	MRID 49559702 USEPA DER[18]
Anaerobic Soil Metabolism	2.4 days (25ºC) 8.4 days (15ºC)	MRID 40025901[19]	0.03 days[e]	MRID 43759301[20]
Anaerobic Aquatic Metabolism	stable	MRID 00040727[21]	0.03 days[e]	MRID 43759301[20]
Air photolysis (25ºC)	stable	MRID 40390101[22]	≤ 8 hours	MRID 49472401[23]
Aqueous photolysis	stable		1.3 days	MRID 42900201[24]
Hydrolysis (20ºC, pH 5,7,9))	13.5 days @20ºC 11-13 days@15ºC 2 days@29ºC	MRID 00158442[25]	Stable	MRID 43022401[26]

[a] average of *cis*- and *trans*- isomers
[b] average of all study replicates
[c] adjusted from 4.5 days to 3.7 days by EPA via linear regression (USEPA DER, 2008)
[d] adjusted for 175 lb/ac, the maximum chloropicrin application rate for 12-inch deep non-tarped applications, using regression in Ashworth et al. 2018
[e] adjusted to 0.03 days USEPA DER, 2008. EPA recommends using the same value for both anaerobic aquatic and anaerobic soil metabolism half-life.

The vapor pressure and solubility of 1,3-D and chloropicrin, two important properties that affect the fate and transport of chemicals in environmental media are within 20-25% of each other and therefore the Henry's Law Constant for 1,3-D[4] and chloropicrin[12] are within 30% of each other. The octanol-water partition coefficient (log K_{ow}), a predictor of bioaccumulation is nearly identical, 2.03 for 1,3-D[10] and 2.09 for chloropicrin,[4] and indicates that chloropicrin and 1,3-D are not expected to accumulate in aquatic organisms.

Both 1,3-D and chloropicrin degrade via numerous degradation pathways in both soil and water. Chloropicrin[15] degrades up to 10-fold faster than 1,3-D in aerobic soil, especially at lower starting soil concentrations (i.e. Lower application rates). Ashworth et al.[27,28] studied the rate dependent degradation of chloropicrin in aerobic soil and developed a regression equation between soil starting concentration and chloropicrin degradation rate, allowing the degradation rate to be corrected for the application rate of chloropicrin. Sorption to soil (K_{oc}), an important parameter affecting chemical leaching and transport is also very similar; 47 L/kg for 1,3-D[13] and 36.1 L/kg for chloropicrin[4].

The mechanisms for degradation in water differ with each compound degrading in different guideline aquatic studies. For example, 1,3-D undergoes hydrolysis ($T_{1/2}$=9-11 days)[25] while chloropicrin is hydrolytically stable[26]. Conversely, chloropicrin undergoes rapid aqueous photolysis ($T_{1/2}$=1.3 days)[24] while 1,3-D is photolytically stable in water.[22] An anaerobic aquatic metabolism study showed that 1,3-D was stable[21] while chloropicrin degraded rapidly with a half-life of 0.03 days.[20] (MRID 43759301). Although 1,3-D did not degrade in the anaerobic aquatic metabolism study, it demonstrated rapid degradation in the anerobic soil metabolism study with a half-life of 2.4 days at 25ºC. [19]

Chloropicrin also volatilizes rapidly from water due to its relatively high volatility (23.8 mm Hg).[7] Li (2009) measured the loss of chloropicrin from water due to volatility and showed that chloropicrin volatilized very rapidly under all conditions studied (i.e., dark and light, turbulent and static water) with half-lives ranging from 4.1 to 34.5 minutes (0.02 days). [29]

The aquatic dissipation studies summarized above demonstrate that both 1,3-D and chloropicrin would dissipate rapidly via multiple dissipation pathways should they enter an aquatic system.

<u>Water Monitoring</u>

The EPA, as part of its draft risk assessment for chloropicrin (EPA, 2018)[30] reviewed the Water Quality Portal (WQP; https://www.waterqualitydata.us/) and found only 9 (0.3%) of 3063 samples analyzed had detectable chloropicrin, all <0.5 µg/L.[31] EPA concluded that since volatilization is a significant route of dissipation for chloropicrin, the potential contribution of other competing processes, such as leaching and runoff, to aquatic exposure is considered low. and thus, is unlikely to have a negative effect on aquatic organisms.[30] Similarly, EPA reviewed the WQP for the 1,3-D DRA[3] (EPA 2019) and found only 3 (0.3%) out of 1008 monitoring samples were reported above detection limits.

The results of the WQP are consistent with the results of a monitoring study conducted by the Florida Department of Environmental Regulation in 1985 on caladium bulb fields on the ridge in central Florida.[32] That study showed that chloropicrin is unlikely to reach, or persist, in shallow groundwater or surface water. Applications of MeBr containing up to 33% chloropicrin were made to the monitored fields for several years prior to monitoring, at rates of at least 100 pounds of chloropicrin per year.

The data base of pesticides in groundwater (U.S. EPA, 1992)[33] showed that chloropicrin was found at less than 1.00 µg/L in three of the 15,175 wells sampled in Florida. The source of the chloropicrin was unlikely to be from an agricultural application, however. Note that trace chloropicrin concentrations in municipal drinking water may result from the chlorination process itself, where chloropicrin can be generated when chlorine products are added to water sources that also have trace nitrate concentrations. [34, 35]

A significant amount of water monitoring for 1,3-D has also been conducted in the USA and Europe and has been reviewed by van Wesenbeeck and Knowles (2019).[36] Their review found that over 50,000 water samples analyzed by state and federal agencies since 1980 has resulted in

151 (0.3%) detections of 1,3-D. Additionally, 4,000 samples analyzed in groundwater studies specifically targeting high 1,3-D use areas in Europe and the United States resulted in 74 detections of 1,3-D or its primary metabolites. The combined detection rate of 1,3-D and its primary metabolites in high use areas of EU and NA was 0.7%.

Monitoring programs have resulted in very few detections of chloropicrin or 1,3-dichloropropene in groundwater or surface water which is not surprising given the rapid dissipation of both fumigants due to their high volatility, rapid degradation in aerobic soil, rapid aerobic and anaerobic aquatic degradation and in the case of chloropicrin, rapid photolytic degradation in water.

Conclusions

The similarity between chloropicrin and 1,3-D physical/chemical parameters and soil degradation half-lives suggest that the 1,3-D runoff study could serve as a surrogate to estimate chloropicrin runoff under similar climatic and edaphic conditions. Furthermore, given that chloropicrin degrades more rapidly in soil than does 1,3-D, and other relevant dissipation processes are similar in terms of half-life, the results of the 1,3-D runoff study would be a conservative surrogate for chloropicrin runoff under similar worst-case conditions.

Finally, in the unlikely event that chloropicrin does reach surface water it would dissipate rapidly by volatilizing to the atmosphere, degrading via aerobic and anaerobic aquatic processes, and by aqueous photolysis. All these routes of dissipation proceed rapidly, with half-lives ranging from 0.03 days to 2.3 days as demonstrated in laboratory studies.

The rapid dissipation of both chloropicrin and 1,3-D via volatilization and multiple biotic and abiotic degradation pathways in both soil and water are corroborated by the low frequency of detections of these fumigants in water monitoring programs.

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Appendix A- Determination of chloropicrin aerobic soil metabolism half-life corrected for starting application rate

In 2005, EPA used a half-life value of 15.71 days for Aerobic Soil Metabolism, which was calculated as the 90th Percentile upper bound from available half-life data. However, after further consultation with the CMTF and reanalysis of the study by EPA, the EPA revised the chloropicrin Aerobic Soil Metabolism half-life to 3.7 days [37](EPA 2008b).

In addition, the 3.7-d aerobic soil half-life (EPA 2008b DER for MRID 43613901) was based on an equivalent field application rate of 500 lbs chloropicrin/acre.[15, 37] The actual labeled maximum application rates for chloropicrin are 175 lbs/acre for non-tarped applications with injection depths of 12 inches or less and 350 lbs/acre for other shank-applied methods (tarped and/or deeper injection, 18 inches or greater). As Ashworth et al. (2018)[27] demonstrated, the degradation rate of chloropicrin correlates to its initial mass in soil (i.e., the application rate).[27] EPA's value for MRID 43613901[37] is less than the Ashworth (2018) study estimated for this same rate (500 lbs/acre) which had a $T_{1/2} = 7.8$ days. Therefore, the degradation rate for lower application rates of chloropicrin would presumably also be lower than predicted by Ashworth (2018).[27]

Both studies used a similar sandy loam soil; the main difference between the soils was the starting soil moisture. The soil moisture in MRID 43613901 was 75% FC[15] which is within the range of optimum soil moisture for applying chloropicrin. The soil moisture in the Ashworth (2018) study was 4% gravimetric soil moisture content or about 6% volumetric water content,[27] while the label recommended volumetric soil water content for that soil would have been in the range of 12-20% (v/v), significantly higher than the test soil used in the Ashworth (2018) study.

Since the objective of the Ashworth study was to examine the relationship between the starting chloropicrin concentration in soil and chloropicrin degradation rate, the soil moisture was not adjusted to meet current label requirements as if an application of chloropicrin was being made. The primary degradation mechanism of chloropicrin in soil has been shown to be microbially mediated [38,39,40,41](Gan et al. (2000), Zheng et al. (2003), Zhang et al. (2005), and Qin et al. (2016)), however soil microbes need sufficient soil moisture to be biologically active. Gan et al. (2000) estimated that microbial degradation accounted for 68 to 92% of CP degradation,[38] and a similar finding (84%) was reported by Zheng et al. (2003).[39] Castro et al. (1983)[42] reported that certain Pseudomonas species possessed the ability to degrade CP and that the main pathway for CP degradation was via sequential dechlorination to dichloromethane, chloronitromethane, and nitromethane.

Table 1 shows the relationship between aerobic soil metabolism half-life and application rate from Ashworth (2018). Figure 5 below is taken from Ashworth et al., 2018 and shows the relationship graphically.
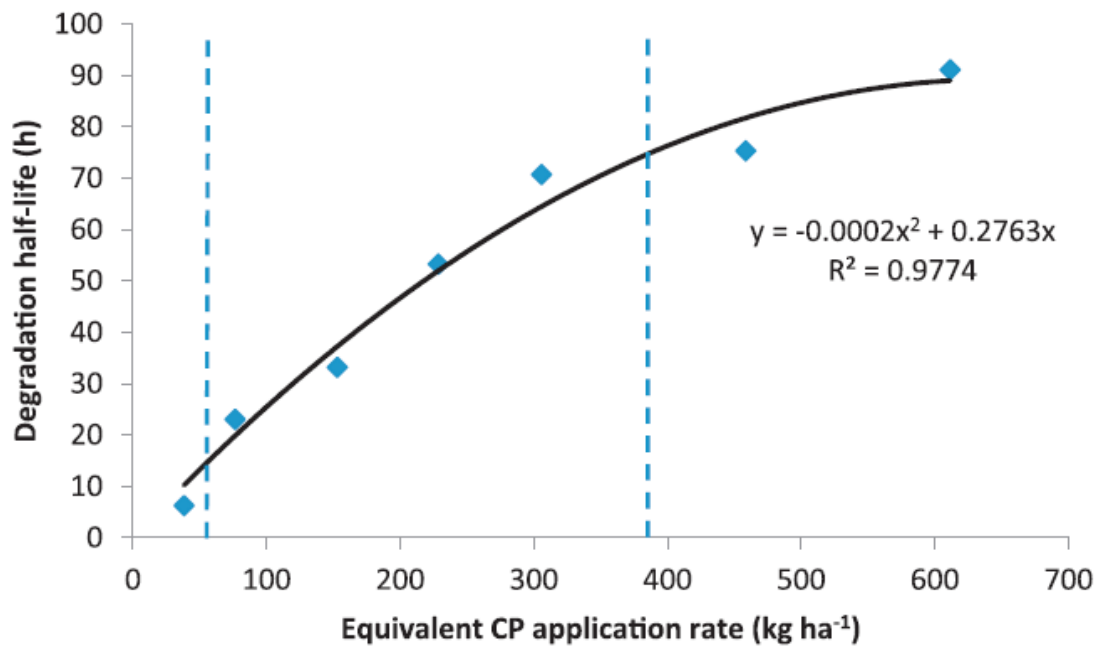
Fig. 5. Second–order polynomial relationship between CP application rate and half-life. The region between the vertical dotted lines represents the application rate range used in the soil columns.

The chart shows Degradation half-life (h) on the y-axis (0 to 100) versus Equivalent CP application rate (kg ha^{-1}) on the x-axis (0 to 700), with the fitted equation:

$$y = -0.0002x^2 + 0.2763x$$
$$R^2 = 0.9774$$

Table 1. Relationship between aerobic soil metabolism half-life and application rate from Ashworth et al., 2018, with study-measured soil moisture below compliance with federal labels.

Application Rate (lbs/acre)	Aerobic Soil Half-Life, in days, from Ashworth et al.[27] (2018) , with study-measured soil moisture (below compliance with federal labels on available soil moisture).	
	Hours	Days
50	14.3	0.6
100	29.6	1.2
150	45.9	1.9
175*	54.5	2.3
200	63.3	2.6
250	81.6	3.4
300	100.9	4.2
350	121	5.0
400**	142.5	5.9
450**	164.8	6.9
500**	188	7.8

* 175 lbs/chloropicrin is the maximum application rate for non-tarped shallow broadcast applications.

** Rates above 350 lbs chloropicrin/acre are not relevant because they exceed the federal maximum application rate of chloropicrin for any application methods.

However, because soil microbes need sufficient soil moisture to be biologically active, and because chloropicrin labels have minimum soil moisture requirement that were not represented in the Ashworth et al., 2018 study, we believe that the soil half-lives presented in Ashworth et al., 2018, at all tested application rates, could have been proportionally shorter had the proper soil moisture been used in the test system. The value of the Ashworth et al., 2018 study however lies in the confirmation of a quantifiable and highly correlated (r2=0.977) relationship between chloropicrin's degradation rate in soil and its initial mass in soil (i.e., its application rate). Lower rates of chloropicrin degrade more quickly than higher rates.

By indexing the EPA's reanalysis of MRID 43613901[15] to Ashworth et al., 2018[27], and adjusting for proper soil moisture, one can estimate soil half-life values based on initial mass as follows: 3.7 days/500 lbs = x days/y pounds, where y = 50 to 450 lbs/acre in 50-lb increments as shown in Table 2.

Table 2. Estimates of chloropicrin aerobic soil half-life (days) for application rates from 50-500 lbs/acre, based on EPA (2008b) value of 3.7-days, but corrected in reference to Ashworth et al. (2018) study on mass-dependent degradation rates for chloropicrin.

Application Rate (lbs/acre)	Estimated Aerobic Soil Half-Lives for different chloropicrin application rates, after EPA (2008b)[37] revised data point from MRID 43613901, and bridged to Ashworth et al. (2018) mass-dependent degradation rate concept.	
	Hours	Days
50	8.9	0.37
100	17.8	0.74
150	26.6	1.11
175*	31.2	1.30
200	35.5	1.48
250	44.4	1.85
300	53.3	2.22
350	61.2	2.59
400**	71.0	2.96
450**	79.9	3.33
500**	88.8	3.70

* 175 lbs/chloropicrin is the maximum application rate for non-tarped shallow broadcast applications.

** Rates above 350 lbs chloropicrin/acre are not relevant because they exceed the federal maximum application rate of chloropicrin for any application methods.

Therefore, the predicted chloropicrin half-life in soil for the maximum rate of chloropicrin that can be applied in a non-tarped shallow shank application of 175 lb/ac would be approximately 1.3 days. This is consistent with the reduction in peak chloropicrin flux and total atmospheric emission of chloropicrin observed in field volatility studies conducted at lower application rates.