

Efficacy of Sulfuryl Fluoride Against Fourth-Instar Pecan Weevil (Coleoptera: Curculionidae) in Pecans for Quarantine Security

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Abstract

The efficacy of sulfuryl fluoride was evaluated for control of fourth-instar pecan weevil, *Curculio caryae* (Horn), at 25°C for a 24-h exposure. Larvae, collected as they naturally emerged from pecans, were used to artificially infest pecan nuts. Infested nuts were fumigated with six concentration by time (CT) treatment dosages of sulfuryl fluoride (0–750 g·h/m³) within air-tight, glass containers. The sulfuryl fluoride concentration in each fumigation container was analyzed 30 min after sulfuryl fluoride introduction and just prior to termination of the experiment. Mean sulfuryl fluoride CT dosages were calculated from sulfuryl fluoride measurements and were used for probit analysis. The lethal accumulated dosage (LAD₉₉) of sulfuryl fluoride for pecan weevil was 1052.0 g·h/m³ with a 95% C.I. of 683.21–2,573.0 g·h/m³. For the confirmatory trial, we used two sulfuryl fluoride CT dosage treatments, 1,100 and 1,300 g·h/m³, and a nonfumigated control. All larvae were dead in both fumigation treatments by 14-d postfumigation. Due to higher mortality in the nonfumigated control in the confirmatory trial compared to that of the dose–response trial, 1300 g·h/m³ was selected as the sulfuryl fluoride CT dosage for a proposed quarantine treatment schedule. Fumigating pecans with sulfuryl fluoride can control larval pecan weevil infestations in commercially traded nuts and maintain compliance with quarantine regulations both within and outside the United States.

Key words: fumigation, *Carya illinoiensis*, export, tree nut

The pecan weevil, *Curculio caryae* (Horn), is native to North America where it is capable of infesting the nuts of all *Carya* spp. (Juglandaceae) (Ring et al. 1991) and, in a single report, was documented to infest Carpathian walnuts, *Juglans regia* L. (Harris et al. 2010). This host range includes pecan (*C. illinoiensis* [Wangenh.] K. Koch) which is the most economically important native nut crop grown in North America. In fact, the pecan weevil is a key pest of commercial pecan where host plant and the pecan weevil occur (Gentry et al. 1973, Payne et al. 1979, Mulder et al. 2012).

Beginning in mid-summer through early autumn, adult weevils emerge from the ground and disperse to nearby pecan trees. The females bore a feeding hole through the shucks and shells of mature nuts to deposit eggs in kernels. Infested pecan nuts remain on the tree as eggs hatch and larvae develop on the kernel endosperm. About 4 wk after oviposition, mature larvae, i.e., fourth instars, chew a hole through the pecan shell, fall to the ground, burrow into the soil, and build a pupation cell where they diapause for 1 or 2 yr before pupating. After pupation, adult weevils remain in the pupation

cell until the following summer when they then emerge to repeat the cycle. This 2- to 3-yr life cycle matches masturing by *Carya* spp. Although most larvae exit the nut before harvest, some remain in the pecan for a longer period and may not emerge until after harvesting, processing, and transporting nuts to market (Harp and Van Cleave 1976, Dutcher and Payne 1981).

Pecan nuts are marketed as shelled or in-shell. Quarantine treatment is required to prevent harvested, in-shell pecans that are infested with nondetected pecan weevil larvae from being shipped to areas where this weevil is not established but pecan production occurs, such as the southwestern United States and to other countries (Harris et al. 2010, Mulder et al. 2012, Sutherland et al. 2017). Currently, the only approved treatment by the USDA Plant Protection and Quarantine is freezing for 7 d at, or below, –17.78°C (USDA 2019). Fumigation currently is not an approved quarantine treatment for pecan weevil. Fumigation research by Leesch and Gillenwater (1976) demonstrated that phosphine failed to control pecan weevil, and only high dosages of methyl bromide (e.g., 1,900

g·h/m³ at 27°C) controlled in-shell weevils. Sorption of phosphine and methyl bromide by pecan kernels (≥95 and >60% in 24 h, respectively) was suggested as a factor for those results.

ProFume gas fumigant (99.8% sulfuryl fluoride, Douglas Products, Liberty, MO) is used to control rodent, insect, and other invertebrate pests infesting postharvest commodities and the structures where these commodities are processed, stored, and transported (Buckley and Thoms 2012). This fumigant was developed in response to a need for a postharvest alternative to methyl bromide resulting from restrictions imposed by the Montreal Protocol (Thoms et al. 2008). The label for this fumigant includes tree nuts in the United States and can be used to fumigate walnuts and almonds for domestic and export markets.

Even though sulfuryl fluoride is labeled for application to tree nuts, including pecans, the efficacy against in-shell pecan weevil larvae is unknown. Therefore, the purpose of this study was to determine the efficacy of sulfuryl fluoride for control of fourth-instar pecan weevil in pecans such that it can be used as a quarantine treatment for postharvest pecans. Based on previous research documenting that high fumigant sorption contributed to an ineffective quarantine treatment, we also conducted an experiment to determine the level of sulfuryl fluoride sorption by pecans during fumigation.

Materials and Methods

Pecan Weevil Larvae

Pecan nuts with a closed, green husk (i.e., a nondehisced involucre) were mechanically harvested from a pecan weevil-infested orchard of 'Stuart' and 'Schley' cultivars on 23 and 24 September 2015 at the USDA, ARS, Southeastern Fruit, and Tree Nut Laboratory (SEFTNRL) in Byron GA. This was done at a time when mature fourth instars begin to naturally emerge from nuts. Pecans remained in harvest wagons which were elevated on one end and emerging larvae were funneled into a collection bin at the other end of the wagon. The bin was checked nearly daily for 1 mo. These larvae were placed on moistened paper towels in lidded plastic bins (32 × 19 × 12 cm) and stored in a walk-in, climate-controlled cooler (7.2 ± 1°C; 0:24 [L:D] h) until used in fumigation assays.

Preparation of Pecans and Weevil Larvae for Fumigation. To obtain a known number of weevil-infested nuts for fumigation, nuts were artificially infested with fourth-instar pecan weevil. The method of artificial infestation was developed after preliminary attempts to infest pecans failed. In preliminary trials, a hole was drilled into the nut, a larva was inserted, and the hole sealed. This process resulted in 100% larval mortality after just 24 h, likely due to desiccation in these harvested nuts (4–6 wk after larvae were collected) with low water content and larvae becoming covered with pecan oil from macerated kernel debris resulting from drilling (T.E.C., personal observation). Even placing and sealing larvae in empty pecan shells resulted in 100% mortality after 48 h, likely due to desiccation (T.E.C., unpublished data).

The artificial infestation method used in this study resulted in low control mortality and still allowed fumigant penetration through the shell to the larva. For this artificial infestation method, 'Desirable' pecan nuts were prepared by cutting off one end of the shell, extracting the kernels and then sanding the cut end smooth (Fig. 1A). Each larva was placed in a plastic 2.0-ml microcentrifuge tube (Online Products for Science, San Diego, CA) that was previously cut to 24 mm in length and had 300–400 µl of 4% agar in the bottom of the tube. The agar provided a moisture source to prevent

larval desiccation during the assay. The tube was then plugged with 8.4 mm of cotton dental wick (Fig. 1B). The tube (containing the larva and agar) was placed within the previously prepared, empty pecan shell and a circular glass cover slip (15- to 18-mm diameter, 0.19- to 0.22-mm thick; Thermo Scientific or BioQuip) was glued (Elmer's Glue-All All Purpose Glue, Elmer's Products, Westerville, OH) over the shell opening (Fig. 1C).

For both fumigation assays (as described later), the infested pecans were placed in an insulated container and shipped via overnight delivery to Kansas State University, Manhattan, KS, where fumigation trials were conducted. In both trials, infested pecans were shipped on the same day larvae were placed into the empty pecan shells.

Fumigation Containers

Fumigation containers were airtight 3.8-liter glass jars equipped with a modified gas-tight, screw-on, metal lid. Each lid had a 1-cm vertical port in the center fitted with a rubber injection septum that allowed for fumigant introduction and headspace gas sampling using gas-tight syringes.

Fumigant Introduction, Measurement, and Aeration

A commercial supply of sulfuryl fluoride (99.8% purity; Douglas Products, Liberty, MO) from a metal, gas cylinder was used for the fumigations which were conducted at normal atmospheric pressure. Sulfuryl fluoride gas was drawn from the cylinder and held in a previously evacuated Tedlar gas bag (CEL Scientific Corp, Santa Fe Springs, CA). The desired sulfuryl fluoride concentration for introduction into fumigation containers was obtained using precisely calibrated gas-tight syringes to draw measured aliquots of a known volume of sulfuryl fluoride and dilute it with a known volume of air in additional Tedlar bags. A dilution volume of sulfuryl fluoride needed to achieve a desired concentration in a given fumigation jar was calculated and that volume of air was removed from the jar using a gas-tight syringe and then replaced with the same volume of sulfuryl fluoride. The 99.8% sulfuryl fluoride was used for high concentrations and diluted sulfuryl fluoride was used for lower gas concentrations. All sulfuryl fluoride concentrations were measured in mg/l in each jar; recommended quarantine treatments were calculated to deliver the desired concentration by time (CT) dosages in g·h/m³ over the 24-h exposure period.

The gas concentration within each jar was measured twice, 30 min after sulfuryl fluoride introduction and just before the termination of the 24-h exposure period, using quantitative gas chromatography/mass spectrometry (GC/MS) and the external standard curve method (Sekhon et al. 2010). The mass spectrometer was set in the selected ion mode to detect a fragment ion characteristic of sulfuryl fluoride. The external standard curve method calculated a precise concentration at a given sample time as described by Phillips et al. (2014). Calculation of an accumulated dosage, expressed in g·h/m³ for a given jar, was the product of the average concentration in a jar and the number of exposure hours. Following the fumigation period, the jar lids were removed and the air space in each jar was ventilated for at least 2 h, after which the burlap bags containing pecans were removed.

Determining Sulfuryl Fluoride Sorption by Pecans

The loss of sulfuryl fluoride in fumigation jars containing in-shell pecans (noninfested), glass marbles, or nothing was evaluated after application of sulfuryl fluoride. Pecans used for this assessment were shipped from the USDA, ARS, SEFTNRL and held at -20°C until

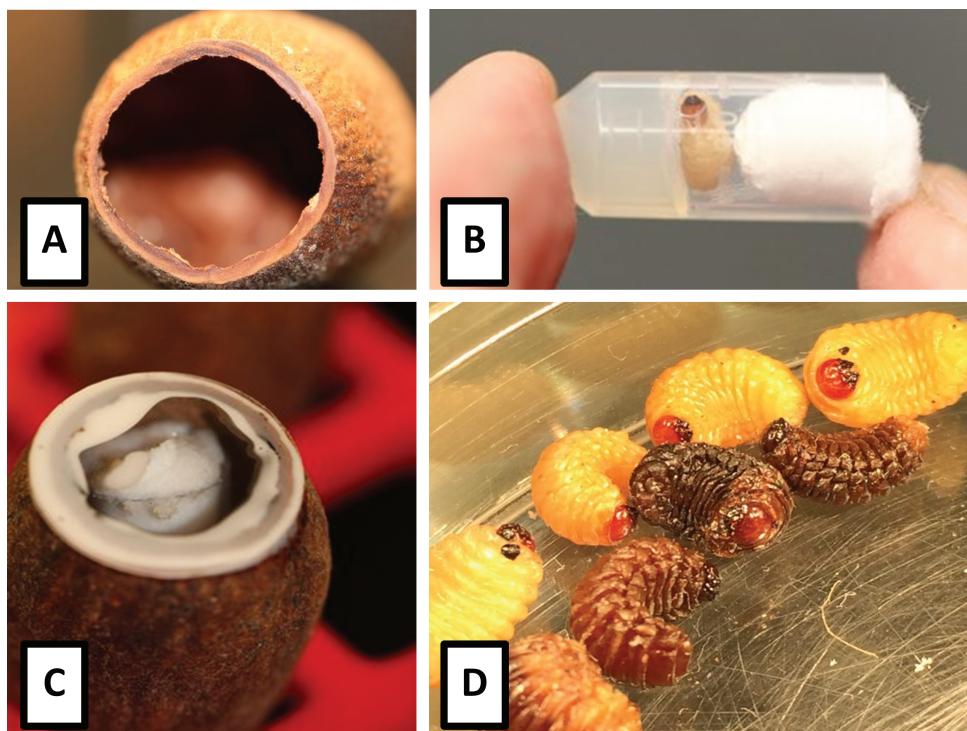


Fig. 1. *A) Pecan nut with end of shell cut off, kernel removed, and open edge smoothed; (B) fourth-instar pecan weevil in plastic microcentrifuge tube containing 4% agar and plugged with a cotton dental wick; (C) plastic microcentrifuge tube, containing agar, fourth-instar pecan weevil larva, and dental wick, placed inside previously prepped, empty pecan shell. Smoothed, open end of pecan shell sealed with circular glass microscope cover slip attached using nontoxic glue; and (D) four dead larvae (light) and four dead but diseased larvae (dark) from the nonfumigated control for the confirmatory trials. Photo was taken at the 7-d postfumigation assessment.

used. The average volumetric displacement of 60 pecans in jars was previously determined. The number of relatively sorption-neutral glass marbles used was equal to the displacement of 60 pecans. Sulfuryl fluoride application was calculated based on available air volume and applied to four jars (two jars at 150 and two jars at 750 g·h/m³) for 24 h at 25°C for pecans, glass marbles, or nothing. Jars were then opened and ventilated within a fume hood for 15 min. To determine if any postventing desorption occurred, the gas-tight lids were re-applied to jars which were then placed in the 25°C incubator for 24 h followed by measurement of sulfuryl fluoride concentrations. This information was used to determine percentage sorption of the contents for each jar of each treatment: pecans, marbles, or nothing. Percentage sorption was calculated as follows: $\frac{(\text{initial sulfuryl fluoride concentration} - \text{terminal sulfuryl fluoride concentration})}{\text{initial sulfuryl fluoride concentration}} \times 100$.

Dose–Response and Confirmatory Fumigation Trials

At least 30 infested pecans (as previously described) were placed in a burlap bag, tied closed, and labeled by target fumigation dosage and replicate. One cloth bag was placed inside each 3.8-liter fumigation jar, as previously described, and equilibrated in environmental chambers set to 25°C ± 1°C for at least 4 h prior to fumigant introduction. Each dosage and the control were replicated four times. The amount of sulfuryl fluoride added to each jar was computed based on the volume of jar space remaining after displacement from the 30 pecans and was adjusted higher than the target based on the presence of pecans as suggested by the sorption data. Negligible displacement was assumed from burlap bags (Phillips et al. 2014). All fumigations were conducted at 25 ± 1°C for an exposure period of 24 h.

For dose–response trials, five target CT dosages (150, 300, 450, 600, and 750 g·h/m³) and one nonfumigated control were evaluated. These CT dosages were based on previous research conducted with methyl bromide for control of pecan weevil larvae (Leesch and Gillenwater 1976). A confirmatory trial, based on results of the dose–response trial, was conducted to validate a proposed quarantine dosage. For this trial, two target CT dosage products (1,100 and 1,300 g·h/m³ representing ~1 and 1.2× the LAD₉₉ calculated in the dose–response trial) and one nonfumigated untreated control were evaluated.

Postexposure Evaluations

Following fumigation and aeration, pecans in labeled burlap bags were wrapped and shipped in an insulated container via overnight delivery to the USDA, ARS, SEFTNRL at Byron, GA. Upon receipt, burlap bags containing pecans were placed by replicate, with treatments randomized within a replicate, in an environmental chamber set at 15.5°C ± 1°C and no light. Three trays (40.5 × 31 × 2 cm) were placed in the chamber and each was filled with water and contained a paper towel wick to increase humidity. Survival, moribundity, and mortality of larvae were assessed 9 d after fumigation for the dose response trial and after 7 d for the confirmatory trial. Each larva was carefully extracted from the pecan shell by removing, in sequence, the cover slip, microcentrifuge tube, and dental wick. The condition of each larva was assessed with the aid of a dissecting microscope and classified based on its movement as alive (normal larval movement with or without prodding), moribund (slight, abnormal movement with prodding), or dead (no movement with prodding). Pecans in which the glass cover slips did not completely cover the opening, or if the glass cover slip

was cracked, were discarded from the test due to possible leakage of sulfuryl fluoride through the opening rather than through the pecan shell. Moribund larvae (which were removed from the pecan shell) were kept in separate plastic trays on moistened paper towels in an environmental chamber (at conditions described above) for seven more days to assess potential recovery.

Statistical Analyses

Proportion data for sorption by jar contents (pecans, glass marbles, or nothing) were analyzed by logistic regression using the Kenward-Roger method to determine degrees of freedom (Warton and Hui 2011, SAS 2014). Proportion data on mortality of pecan weevil larvae in the initial fumigation trial and in the confirmatory trial were analyzed similarly. In each case, treatment differences were elucidated through the lsmeans procedure with a Tukey-Kramer adjustment (SAS 2014). For the initial fumigation trial, the dose-response mortality of pecan weevil larvae 9 d after treatment was subjected to probit analysis (SAS 2014) to determine the lethal accumulated dosage (LAD_{99}) and 95% CI. This information was used to develop proposed quarantine treatment dosage to validate in the confirmatory trial.

Results

Determining Sulfuryl Fluoride Sorption by Pecans

After a 24-h fumigation period, the mean percentage loss (\pm SE) of sulfuryl fluoride was significantly higher for jars containing pecans than empty jars or jars with glass marbles ($F = 153.05$; $df = 2, 9$; $P < 0.0001$; Fig. 2). No significant difference in loss was detected between empty jars and those with glass marbles. The majority of sulfuryl fluoride loss during fumigation was likely from sorption rather than leakage. This was because the empty jars had initial sulfuryl fluoride concentrations closest to target levels when compared with jars containing glass marbles or pecans.

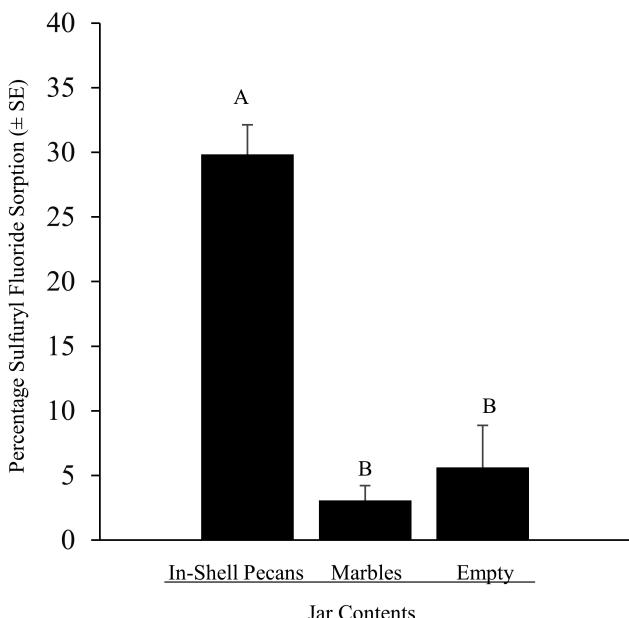


Fig. 2. Percentage sorption of sulfuryl fluoride as calculated from initial and terminal sulfuryl concentrations in individual jars containing in-shell pecans, glass marbles, or nothing following a 24-h fumigation. Treatments followed by the same letter are not significantly different ($P < 0.05$).

After aeration and closure in sealed fumigation jars for 24 h, desorption of about 0.5 g/m^3 (120 ppm) of sulfuryl fluoride was observed for pecans fumigated at 750 g-h/m^3 . All other jars, including the jars with pecans fumigated at 150 g-h/m^3 , had $<0.1 \text{ g/m}^3$ (24 ppm) desorbed.

Dose-Response Trial

Mean (\pm SD) concentrations (g/m^3), mean (\pm SD) CT dosages (g-h/m^3) of sulfuryl fluoride, and larval survival for all treatments are provided in Table 1. During this trial, a significant treatment effect on survival was detected during the larval assessment 9 d after fumigating ($F = 23.19$; $df = 5, 18$; $P < 0.0001$). Larval survival in the nonfumigated control treatment was significantly higher than any dosage of sulfuryl fluoride tested. Similarly, larval survival in the 150 g-h/m^3 target CT dosage was significantly higher than that in all higher CT dosages.

Some larvae were recorded as moribund during the assessment; however, no moribund larvae were detected in the nonfumigated controls. Most moribund larvae, 78.6% (22 of 28), occurred in the lowest target sulfuryl fluoride CT dosage (150 g-h/m^3). When all moribund larvae were reassessed seven days later (i.e., 16 d after fumigation), 57% were dead with the others still moribund. Regardless of dosage, none had recovered. It was assumed that the remaining moribund larvae would die. Therefore, because moribund larvae showed no signs of recovery, moribund larvae were considered dead and the larval survival rates for the fumigated treatments did not change.

The mean (\pm SD) sulfuryl fluoride CT dosages used did not exactly match the target dosages sought. The lowest dosage used ($151.9 \pm 9.8 \text{ g-h/m}^3$) approached the target dosage of 150 g-h/m^3 , whereas the next dosage, $288.8 \pm 8.9 \text{ g-h/m}^3$, was lower than the target dosage of 300 g-h/m^3 . The mean dosages of 533.3 ± 14.3 , 671.0 ± 43.1 , and $862.8 \pm 32.0 \text{ g-h/m}^3$ were all higher than the target dosages of 450 , 600 , and 750 g-h/m^3 , respectively. For probit analysis, mean sulfuryl fluoride CT dosages were used to obtain an estimated LAD_{99} of $1,052.0 \text{ g-h/m}^3$ (slope \pm SE = 3.57 ± 0.67 , $\chi^2 = 27.95$; $df = 1$; $P < 0.0001$; 95% CI of 683.21 – $2,573.0 \text{ g-h/m}^3$).

Confirmatory Trial

Mean (\pm SD) concentrations (g/m^3) and mean (\pm SD) CT dosages (g-h/m^3) of sulfuryl fluoride and larval survival for all treatments are provided in Table 2. The larval assessment was conducted 7 d postfumigation. Dead larvae that were unnaturally dark in color (Fig. 1D, $n = 34$) were observed in all treatments. Only a few of these dark-colored larvae had been observed previously in the dose-response trial and were classified as dead. In the confirmatory trial, these dark-colored larvae were classified as diseased based on prior experience (T.E.C., personal observation). The percentage of diseased larvae in each treatment dosage was comparable ($F = 0.7882$; $df = 2, 11$; $P > 0.497$), indicating that fumigation was not a factor contributing to disease in the confirmatory trial. The higher presence of diseased larvae could be due to larvae being held longer after field collection to conduct the confirmatory trial compared with the shorter time held before the dose-response trial.

Survival in the nonfumigated control was lower than that observed during the previous dose-response trial (Table 2). Larval survival among treatments was significantly different ($F = 115.26$; $df = 2, 9$; $P < 0.0001$) with the nonfumigated control survival higher than either the $1,100$ or $1,300 \text{ g-h/m}^3$ treatments (Table 2).

Unlike the dose-response trial, only four moribund larvae were detected, and they were found in all treatments: one each in the

Table 1. Target and mean (\pm SD) concentration by time (CT) dosages (g-h/m³) of sulfuryl fluoride along with the target and mean (\pm SD) concentrations (g/m³) of sulfuryl fluoride used in dose-response fumigation trials against fourth-instar *Curculio caryae* and the resulting percentage survival (mean \pm SD) observed 9 d after fumigation

Target CT dosage (g-h/m ³) ¹	Mean CT dosage \pm SD (g-h/m ³)	Target concentration (g/m ³)	Mean concentration \pm SD (g/m ³)	Percentage larval survival \pm SD ²
0	0	0	0	97.6 \pm 3.1a
150	151.89 \pm 9.80	6.25	6.33 \pm 0.41	17.4 \pm 5.7b
300	288.81 \pm 8.87	12.5	12.03 \pm 0.37	7.2 \pm 7.3c
450	533.29 \pm 14.29	18.75	22.22 \pm 0.59	1.7 \pm 1.9c
600	670.96 \pm 43.12	25	27.96 \pm 1.8	3.1 \pm 2.6c
750	862.76 \pm 32.01	31.25	35.95 \pm 1.33	2.2 \pm 1.5c

¹Four replicates per target dosage with initially at least 29 larvae per replicate.²Means followed by different letters within this column are significantly different ($P \leq 0.05$).**Table 2.** Target and mean (\pm SD) concentration by time (CT) dosages (g-h/m³) of sulfuryl fluoride along with the target and mean (\pm SD) concentrations (g/m³) of sulfuryl fluoride used in confirmatory fumigation trials against fourth-instar *Curculio caryae* and the resulting percentage survival (mean \pm SD) observed 7 d after fumigation

Target CT dosage (g-h/m ³) ¹	Mean CT dosage \pm SD (g-h/m ³)	Target concentration (g/m ³)	Mean concentration \pm SD (g/m ³)	Percentage larval survival \pm SD ²
0	0	0	0	80.0 \pm 7.2a
1100	1121.0 \pm 25.6	45.83	46.71 \pm 1.07	0b
1300	1303.5 \pm 78.1	54.17	54.32 \pm 3.26	0b

¹Four replicates per target dosage with initially at least 29 larvae per replicate.²Means followed by different letters within this column are significantly different ($P \leq 0.05$).**Table 3.** Proposed quarantine treatment schedule for treatment of in-shell pecans with sulfuryl fluoride to control larval *Curculio caryae*

Temperature	Exposure period	Minimum sulfuryl fluoride (g/m ³) readings at:			
		0.5 h	2 h	12 h	24 h
25°C and above	24 h	75	70	54	39

nonfumigated control and 1,300 g-h/m³ treatments and two in the 1,100 g-h/m³ treatment. When these larvae were reassessed 14 d postfumigation, two of these larvae had died, one was diseased, one remained moribund, and none recovered. As in the dose-response trial, because moribund larvae showed no signs of recovery, moribund larvae were considered dead and the larval survival rates for the fumigated treatments did not change.

Although all larvae were dead at both target fumigation dosage rates by the final assessment at 14-d postfumigation, the higher target CT dosage of 1,300 g-h/m³ is recommended as the quarantine treatment CT dosage. This CT dosage was selected to compensate for the higher mortality observed in the nonfumigated controls in the confirmatory trials. This CT dosage is less than the maximum CT dosage of 1,500 g-h/m³ permitted by the labeling for sulfuryl fluoride for fumigation of food commodities (Douglas Products 2017). The proposed quarantine treatment schedule for sulfuryl fluoride for control of the pecan weevil is listed in Table 3. This schedule is for a 24-h fumigation period and will result in an accumulated CT dosage of 1,300 g-h/m³, as validated in this study.

Discussion

Results from this study demonstrate that sulfuryl fluoride can be used to kill pecan weevil larvae that remain within harvested pecan nuts. Even though a low percentage of fumigated larvae were moribund,

they showed no signs of recovery (i.e., 14 or 16 d after fumigation). In fact, delayed or latent mortality of insects following exposure to sulfuryl fluoride is well documented (Osbrink et al. 1987, Su and Scheffrahn 1990, Thoms and Scheffrahn 1994, Phillips et al. 2014).

We used pecan nuts artificially infested with pecan weevil larvae in this study. This method allowed for fumigation of a manageable number of infested nuts as opposed to a high volume of nuts with a likely low proportion of infested nuts (i.e., as when harvested directly from trees). Artificial infestation was also advantageous due to a short window of opportunity available to fumigate naturally infested nuts because a high percentage of larvae leave the nut over a short period.

The 30% sorption of sulfuryl fluoride by pecans in the current study was similar to the sorption (27.9–30.5%) of sulfuryl fluoride reported by Sriranjini and Rajendran (2008) for in-shell and shelled almonds, shelled walnuts, cashew nuts and shelled pistachio nuts. Nonetheless, the sorption of sulfuryl fluoride by pecans was much less than the sorption of 95% and 60% for phosphine and methyl bromide, respectively, as previously documented by Leesch and Gillenwater (1976). Sulfuryl fluoride has lower sorption characteristics compared with methyl bromide for other food commodities tested, including wheat kernels, gluten and flour, and semolina (Kenaga 1957, Hwaidi et al. 2015).

The level of desorption of sulfuryl fluoride from pecans in this study was dose dependent, which was also observed by Osbrink et al. (1988) on other food commodities. Research evaluating the desorption of sulfuryl fluoride from a variety of food commodities (Meikle and Stewart 1962, Osbrink et al. 1988) has demonstrated that any desorbing sulfuryl fluoride residues are transient and rapidly decrease to extremely low (ppb) or nondetectable levels. Osbrink et al. (1988) documented that longer aeration times resulted in less desorption of sulfuryl fluoride. In this study, pecans were aerated for only 15 min for the sorption trial. In commercial fumigations, commodities are aerated for a longer time to reach the required clearance

concentration of sulfuryl fluoride (e.g., 1 ppm in the United States). The labeling for sulfuryl fluoride also requires that bulk, stored food commodities be aerated for at least 24 h following fumigation (Douglas Products 2017).

Sulfuryl fluoride has become an important treatment in the United States to reliably disinfest of tree nuts of insects (Hosoda 2010, 2013). It is an odorless and colorless inorganic gas that is not known to cause off-flavors in treated foods. An extensive program of food quality studies was conducted on tree nuts (walnuts, pistachios, and almonds) in cooperation with the Dried Fruit and Tree Nut Association of California, and two groups of sensory researchers at the National Food Laboratory in Dublin California and Department of Pomology, University of California, Davis, CA. These research studies confirmed lack of adverse effects on taste, quality, and commercial value of tree nuts fumigated with a sulfuryl fluoride (Buckley and Thoms 2012).

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References Cited

Buckley, S., and E. Thoms. 2012. Current status of ProFume® gas fumigant for disinfestation of commodities, pp. 241–246. In S. Navarro, H. J. Banks, D. S. Jayas, C. H. Bell, R. T. Noyes, A. G. Ferizli, M. Emekci, A. A. Isikber, and K. Alagusundaram (eds.), Proceedings of the 9th International Conference on Controlled Atmosphere and Fumigation in Stored Products, ARBER Professional Congress Services, Turkey.

Douglas Products. 2017. Applicator manual for ProFume® gas fumigant. Douglas Products and Packaging Company, Liberty, MO. pp. 70.

Dutcher, J. D., and J. A. Payne. 1981. Pecan weevil (*Curculio caryae*, Coleoptera: Curculionidae) bionomics: a regional problem. *Misc. Publ. Entomol. Soc. Am.* 12(2): 45–68.

Gentry, C. R., L. B. Bowden, J. A. Payne, and W. L. Tedders. 1973. A bibliography of the pecan weevil, *Curculio caryae* (Coleoptera: Curculionidae). *Bull. Entomol. Soc. Am.* 19: 203–207.

Harp, S. J., and H. W. Van Cleave. 1976. Biology of the pecan weevil. *Southwest Entomol.* 1: 21–30.

Harris, M. K., K. L. Hunt, and A. I. Cognato. 2010. DNA identification confirms pecan weevil (Coleoptera: Curculionidae) infestation of Carpathian walnut. *J. Econ. Entomol.* 103: 1312–1314.

Hosoda, E. 2010. Update on Commercial Acceptance of ProFume® gas fumigant. In G. L. Obenau (ed.), Proceedings of the 2010 Annual International Research Conference on Methyl Bromide Alternatives and Emissions Reduction, Orlando, FL. <https://mbao.org/static/docs/confs/2010-orlando/papers/068HosadaECommercialAcceptanceofProFumeGasFumigant.pdf>. Accessed 3 February 2020.

Hosoda, E. 2013. ProFume® gas fumigant, The First Decade. In G. L. Obenau (ed.), Proceedings of the 2013 Annual International Research Conference on Methyl Bromide Alternatives and Emissions Reduction, San Diego, CA. <https://mbao.org/static/docs/confs/2013-sandiego/papers/35HosodaE.pdf>. Accessed 3 February 2020.

Hwaidi, M., P. J. Collins, M. Sissons, H. Pavic, and M. K. Nayak. 2015. Sorption and desorption of sulfuryl fluoride by wheat, flour and semolina. *J. Stored. Prod. Res.* 62: 65–73.

Kenaga, E. E. 1957. Some biological, chemical, and physical properties of sulfuryl fluoride as an insecticide fumigant. *J. Econ. Entomol.* 50: 1–6.

Leesch, J. G., and H. B. Gillenwater. 1976. Fumigation of pecans with methyl bromide and phosphine to control the pecan weevil. *J. Econ. Entomol.* 69: 241–244.

Meikle, R. W., and D. J. Stewart. 1962. Structural fumigants, the residue potential of sulfuryl fluoride, methyl bromide, and methanesulfonyl fluoride in structural fumigations. *J. Agric. Food Chem.* 10: 393–379.

Mulder, P. G., Jr., M. K. Harris, and R. A. Grantham. 2012. Biology and management of the pecan weevil (Coleoptera: Curculionidae). *J. Integr. Pest Manag.* 3: A1–A9, doi:10.1603/IPM10027

Osbrink, W. L. A., R. H. Scheffrahn, N.-Y. Su, and M. K. Rust. 1987. Laboratory comparisons of sulfuryl fluoride toxicity and mean time of mortality among ten termite species (Isoptera: Hodotermitidae, Kalotermitidae, Rhinotermitidae). *J. Econ. Entomol.* 80: 1044–1047.

Osbrink, W. L. A., R. H. Scheffrahn, R. Ching Hsu, and N.-Y. Su. 1988. Sulfuryl fluoride residues of fumigated foods protected by polyethylene film. *J. Agric. Food Chem.* 36: 853–855.

Payne, J. A., H. C. Ellis, and D. W. Lockwood. 1979. Biology and distribution of the pecan weevil in Georgia and Tennessee. *Pecan South* 6(1): 30–33.

Phillips, T. W., M. J. Aikins, E. Thoms, J. Demark, and C. Wang. 2014. Fumigation of bed bugs (Hemiptera: Cimicidae): effective application rates for sulfuryl fluoride. *J. Econ. Entomol.* 107: 1582–1589.

Ring, D. R., L. J. Grauke, J. A. Payne, and J. W. Snow. 1991. Tree species used as hosts by the pecan weevil (Coleoptera: Curculionidae). *J. Econ. Entomol.* 84: 1782–1789.

SAS Institute Inc. 2014. Base SAS® 9.4 Procedures Guide. SAS Institute Inc., Cary, NC.

Sekhon, R. K., M. W. Schilling, T. W. Phillips, M. J. Aikins, M. M. Hasan, and W. B. Mikel. 2010. Sulfuryl fluoride fumigation effects on the safety, volatile composition, and sensory quality of dry cured ham. *Meat Sci.* 84: 505–511.

Sriranjini, V. R., and S. Rajendran. 2008. Sorption of sulfuryl fluoride by food commodities. *Pest Manag. Sci.* 64: 873–879.

Su, N.-Y., and R. H. Scheffrahn. 1990. Efficacy of sulfuryl fluoride against four beetle pests of museums (Coleoptera: Dermestidae, Anobiidae). *J. Econ. Entomol.* 83: 879–882.

Sutherland, C., J. B. Pierce, B. Lewis, and R. Heerema. 2017. Pecan weevil: wanted dead, not alive. New Mexico State University, Cooperative Extension Service, Las Cruces, NM. circular 683, pp. 8.

Thoms, E. M., and R. H. Scheffrahn. 1994. Control of pests by fumigation with vikane gas fumigant. *Down To Earth*. 49: 23–30.

Thoms, E., J. Busacca, and S. Prabhakaran. 2008. Commercializing a new fumigant: the ProFume® success story, 698–703. In G. Daolin, S. Navarro, Y. Jian, T. Cheng, J. Zuzun, L. Yue, and W. Haipeng (eds.), Proceedings of the 8th International Conference on Controlled Atmosphere and Fumigation in Stored Products, Sichuan Publishing Group, Chengdu, China.

USDA Treatment Manual. 2019. pp. 956. https://www.aphis.usda.gov/import_export/plants/manuals/ports/downloads/treatment.pdf. Accessed 3 February 2020.

Warton, D. I., and F. K. Hui. 2011. The arcsine is asinine: the analysis of proportions in ecology. *Ecology*. 92: 3–10.