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Propylene oxide as a potential quarantine fumigant for insect disinfestation of nuts

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Abstract

Propylene oxide (PPO) at a low pressure of 100 mm Hg was tested for rapid disinfestation of nuts as a potential replacement for methyl bromide, by evaluating its toxicity to the Indianmeal moth, *Plodia interpunctella* (Hübner) in the absence and presence of three species of nuts. Eggs and larvae were the most tolerant stages. Complete mortality of all life stages was achieved at a Ct product of 61.2 mg h/L for empty space fumigation. Dosages of 13.9, 60.3, 72.1 and 93.1 mg/L were required to kill 99 % of the larvae when fumigation of 4-h duration took place in an empty chamber, and in presence of peanuts, almonds and walnuts, respectively. After an initial dose of 68.7 mg/L, and a 5-h exposure time, sorption of PPO by peanuts, almonds and walnuts was relatively high, ranging from 87 % of the initial concentration for peanuts to 91 % for walnuts. PPO residues measured in peanuts, almonds and walnuts were 111, 46 and 80 ppm respectively 0-1 days after termination of fumigation, all of which were below the 300 ppm maximum tolerance set by the FDA of the United States. These data show that the combination of PPO with low pressure has the potential for replacing methyl bromide fumigation for quarantine and pre-shipment purposes.

Key words: Propylene oxide, nuts, quarantine fumigation, toxicity, Plodia interpunctella, sorption.

Introduction

MB is the only fumigant available for quarantine treatment of commodities for which rapid disinfestation techniques and a very high degree of insect mortality are essential. The loss of MB could have a significant negative impact on world agriculture, particularly because no available alternatives to MB currently exist for rapid disinfestation of commodities. Thus, there is a critical need to develop new fumigants for quarantine purposes. Propylene oxide (PPO) is commonly used as a sterilant to reduce bacteria, mould and yeast contamination on processed spices, cocoa and processed nutmeats except peanuts. Several reports on insect toxicity indicated that PPO would be an effective replacement for methyl bromide in some postharvest situations (Creasy and Hartsell, 1999; Griffith, 1999; Isikber et al., 2001; Zettler et al., 2002; Navarro et al., 2004). PPO is a liquid fumigant under normal temperature pressure (NTP) with a boiling point of 35 ºC and a noticeable ether odor (Weast et al., 1986). As a fumigant, PPO has reduced environmental risks

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compared with methyl bromide. It is not an ozone depleter and it degrades into nontoxic propylene glycol in the soil and in the human stomach. Human health and environmental effects of PPO are reviewed in Meylan et al. (1986). A disadvantage of PPO is that it is flammable from 3 % to 37 % in air and therefore, to avoid flammability it should be applied under low pressure or in a CO_2-enriched atmosphere. PPO is considered here for rapid disinfestation of the nuts as a replacement for MB by evaluating its toxicity against a major insect pest of stored nuts, and its sorption and residue on the nuts.

Materials and methods

Toxicity trials were carried out on all life stages of a major insect species of stored nuts: the Indianmeal moth, *Plodia interpunctella* (Hübner). All the tests were carried out in 2.6 L desiccators at a temperature of 30 ºC and 70 % relative humidity at a 4-h exposure time. For PPO treatments under 100 mm Hg vacuum, four to five concentrations of PPO ranging from 1 mg/liter to 20 mg/liter were tested for each stage of the insect. Moreover, the toxicity of PPO under 100 mm Hg to the larvae of *P. interpunctella* in the presence of 0.5 kg of walnuts, peanuts and almonds was also determined by using four to five dosages ranging from 40 mg/liter to 100 mg/liter. Each test was replicated at least twice. Probit analysis was applied to mortality data to determine the LC_{50}s, LC_{99}s and their respective 95 % confidence intervals. Sorption of PPO by walnuts, peanuts and almonds was determined at an initial dosage of 68.7 mg/L using a gas chromatograph. The PPO residues in peanuts, almonds and walnuts were measured after 5 h. fumigation at 30 °C at the sole dose of 112 mg/L PPO. Residue levels in each commodity were determined at the end of fumigation and following a 3 d aeration period. The residue levels in the commodities were determined by a commercial analytical laboratory service (Aminolab Ltd. Israel) following an analytical method that was a modification of the ASTA analytical method of the Official Methods of Analysis of the AOAC (Anonymous, 2000).

Results and discussion

PPO under 100 mm Hg was toxic to all life stages of *P. interpunctella*. Eggs and larvae of *P. interpunctella* by LC_{99} values of 15.3 and 13.9 mg/liter respectively were more tolerant than the adults and pupae by LC_{99} values of 5.9 and 8.8 mg/liter, respectively (Table 1). The complete mortality of all life stages of *P. interpunctella* was achieved at a Ct product of 61.2 mg/liter/h. It required dosages of 13.9, 93.1, 60.3 and 72.1 mg/l to kill 99 % of the larvae of *P. interpunctella* when fumigated in an empty desiccator and in the presence of walnuts, peanuts and almonds, respectively (Table 2). The results indicated that there was a six and half-fold increase in LC_{99} value of PPO at low pressure when the larvae were fumigated in the presence of walnuts as compared to those fumigated in the empty space. Similarly, there was a four to five-fold increase in the LC_{99} value of PPO at low pressure for fumigation in peanuts and almonds as compared to fumigation in the empty desiccators. Thus, the present study indicates that a much higher dose of PPO is required for fumigation in the presence of walnuts, peanuts and almonds to obtain complete mortality of the larvae of *P. interpunctella.*

Sorption of PPO by walnuts, peanuts and almonds after a 4-h exposure time was very high, ranging from 87 % to 91 % of the initial concentration (Figure 1). In all cases, there was an initial rapid decrease in concentrations of PPO during the first hour of exposure followed by a more gradual subsequent drop. The drop in concentrations during the first hour for walnuts was 86 % of the initial dosage applied, while that for peanuts and almonds, was from 77 % to 82 % of the initial dosage applied indicating a rapid sorption of PPO by all three species of nut. The PPO residues in walnuts, peanuts and almonds were a maximum average of 80, 111 and 46 ppm respectively at 0-1 day after termination of aeration (Table 3). These were all below the

maximum tolerance of 300 ppm. Very low PPO residues ranging from < 2 to 26 ppm were detected at 3 days after termination of aeration. These data indicate that the PPO rapidly desorbs from the commodity under conditions of NAP and 30-35 ºC

Table 1. Probit analysis data and Ct products (mg h/L) for propylene oxide at 100 mm Hg for all life stages of *P. interpunctella* resulting from 4-h laboratory fumigations at 30 °C.

Life stage	n[a]	Slope[b]±SE	LC_{50} (Fiducial limit)[c] (mg/L)	LC_{99} (Fiducial limit)[c] (mg/L)	H[d]	Ct product for LC_{99} (mg h/L)
Egg	1000	6.4±0.66	6.7 (6.03 – 7.25)	15.3 (13.42 – 18.48)	0.75	61.2
Larva	236	9.1±1.52	7.7 (7.10 – 8.38)	13.9 (11.89 – 18.59)	0.26	55.6
Pupa	276	12.3±2.65	5.7 (5.01– 6.25)	8.8 (7.69 – 11.79)	0.70	35.2
Adult	202	4.7±0.71	1.9 (1.58 - 2.22)	5.9 (4.41 - 9.72)	0.89	23.6

[a] Number treated, excluding controls.
[b] Slopes among life stages of tested insect are unparallel and unequal where noted.
[c] Numbers in brackets give the 95 % confidence range.
[d] Heterogeneity factor, chi-square/degrees of freedom (chi-square is significant, P < 0.05).

Table 2. Probit analysis data for propylene oxide at low pressure of 100 mm Hg for the larvae of *P. interpunctella* resulting from 4-h laboratory fumigations of 1 kg of peanuts, almonds, walnuts and empty chambers.

Treatment	n[a]	Slope[b]±SE	LC_{50} (Fiducial limit)[c] (mg/L)	LC_{99} (Fiducial limit)[c] (mg/L)	H[d]
PPO at 100 mm Hg with peanuts	240	21.2±3.16	46.8 (44.70 – 48.53)	60.3 (56.95 – 66.25)	0.51
PPO at 100 mm Hg with almonds	238	19.4±3.38	54.7 (52.12 – 57.01)	72.1 (67.09 – 82.86)	0.37
PPO at 100 mm Hg with walnuts	240	19.8±3.77	71.0 (67.84– 73.66)	93.1 (86.67 – 107.60)	0.03
PPO at 100 mm Hg in empty chamber	236	9.1±1.52	7.7 (7.10 – 8.38)	13.9 (11.89 – 18.59)	0.26

[a] Number treated, excluding controls.
[b] Slopes among life stages of tested insect are unparallel and unequal where noted.
[c] Numbers in brackets give the 95 % confidence range.
[d] Heterogeneity factor, chi-square/degrees of freedom (chi-square is significant, P < 0.05).

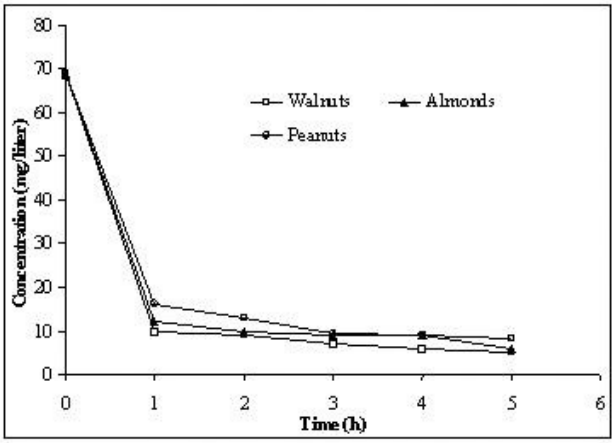

Figure 1. Concentrations of PPO (mg/L) in fumigation chambers of 2.64 L capacity during five hours of exposure after application of PPO at a dose of 68.7 mg/L to 1 kg of walnuts, almonds and peanuts at 30 °C and 60 ± 5 % relative humidity.

Table 3. PPO residues (ppm) on walnuts, almonds and peanuts at 0-1 day and 3 days after 4-h fumigation at 30 ºC and atmospheric pressure with a dose of 112 mg/L PPO.

Commodity	Average PPO residue (ppm) in sample	
Time after fumigation	0-1 day	3 days
Walnuts	80	26
Almonds	46	< 2
Peanuts	111	8

PPO at atmospheric pressure is not realistic due to long exposure times and its flammability from 3 % to 37 % in air (16). However, several studies have shown that vacuum fumigation or the admixture of CO_2 could increase the toxicity of PPO (Isikber et al., 2001; Navarro et al., 2004). Since low pressure treatments are now technically and economically available and feasible, commercial scale fumigation of PPO with low pressure can be implemented for rapid disinfestation in quarantine and pre-shipment (QPS) situations.

Although sorption of PPO by the nuts tested was relatively high, the fumigation still enables a sufficient build up of gas concentrations to achieve insect mortality. Based on its high and rapid toxicity to insects, and its rapid desorption from the commodities, the combination of PPO with low pressure can become a potential fumigant for replacement of MB for quarantine purposes where rapid disinfestation of the nuts is essential.

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