Volatile Aldehydes Are Promising Broad-Spectrum Postharvest Insecticides

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A variety of naturally occurring aldehydes common in plants have been evaluated for their insecticidal activity and for phytotoxicity to postharvest fruits, vegetables, and grains. Twenty-nine compounds were initially screened for their activity against aphids on fava bean leaf disks. Application under reduced pressure (partial vacuum) for the first quarter of fumigation increased insecticidal activity severalfold. The 11 best aldehydes were assayed against aphids placed under the third leaf of whole heads of iceberg lettuce using the same two-tier reduced-pressure regime, which caused no additional detriment to the commodity over fumigation at atmospheric pressure. Phytotoxicity to naked and wrapped iceburg lettuce, green and red table grapes, lemon, grapefruit, orange, broccoli, avocado, cabbage, pinto bean, and rice at doses that killed 100% of aphids was recorded for three promising fumigants: propanal, (*E*)-2-pentenal, and 2-methyl-(*E*)-2-butenal. These three compounds have excellent potential as affordable postharvest insect control agents, killing 100% of the aphids with little or no detectable harm to a majority of the commodities tested. Preliminary assays indicate that similar doses are also effective against mealybugs, thrips, and whitefly.

Keywords: *Aldehyde; insecticide; fumigant; disinfestation; quarantine*

INTRODUCTION

Occurrence of pests, especially insects, is a major rationale for the rejection of agricultural imports at the port of entry. Agricultural loads found to bear live insects, even in minuscule quantities, are subject to thorough fumigation, usually with methyl bromide. Fumigation with methyl bromide can dramatically lower the shelf life of some commodities, so the threat of rejection or destructive fumigation has led some producers and transporters to abandon their attempts to export raw produce to certain countries, most notably Japan. Consequently, there is a need for effective postharvest pest control methods that are well tolerated by agricultural crops.

In 1993, the EPA took regulatory action to freeze U.S. production of methyl bromide in 1994 at 1991 levels and to prohibit the production and import of methyl bromide after January 1, 2001 (U.S. Environmental Protection Agency, 1990). Since then, the date for phase-out has been pushed back to 2005, partially due to the perception that no realistic alternative presently exists. Use of methyl bromide for quarantine or preshipment purposes is for now exempt from the legislation. Nevertheless, in light of growing controversy around the use of methyl bromide and the approach of its deadline for phase-out, the pressure to find viable alternatives has intensified.

Short-chain aldehydes are small, volatile compounds that occur throughout the plant kingdom and are fundamental flavor and fragrance constituents in both natural and processed foods (Fenaroli and Burdock, 1995). In this paper we describe the use of volatile aldehydes to achieve inexpensive, broad-spectrum disinfestation of postharvest crops. Although several of the aldehydes we studied here were previously tested and discounted as insecticides on the basis of low potency, we have found that by applying them at reduced pressure for the first segment of treatment, their potency can be increased significantly.

The two-carbon acetaldehyde is the only aldehyde that previously seems to have been the subject of thorough investigation as an insecticide. Acetaldehyde (or ethanal) gas was reported to show activity against the green peach aphid (*Myzus persicae* Sulzer) at a concentration of 0.25% for 2 h (Aharoni et al., 1979). Aharoni and Stewart also tested acetaldehyde against western flower thrips (*Frankliniella occidentalis* Pergande) at various reduced pressures and exposure periods and found activity, but no statistical difference between treatments at different pressures (Aharoni and Stewart, 1980). Use of acetaldehyde as a commercial fumigant was not pursued further, apparently due to concerns about getting sufficient kill, damage to the commodity itself, and the safety of workers handling acetaldehyde.

Insecticidal studies involving aldehydes with three or more carbons have been few and cursory. Ferguson and Pirie tested 4 aldehydes among the 94 diverse compounds they assayed against the grain weevil (*Calandra granaria*) in exposures of 5 h at 25 °C and atmospheric pressure (Ferguson and Pirie, 1948). A categorical screening of 189 different chemicals against the oriental fruit fly (*Dacus dorsalis* Hendel) included propanal (Hinman, 1954), but the author apparently did not consider it a promising candidate for further study, because in a second screening of compounds for control

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10.1021/jf000233+ CCC: $19.00
Published on Web 08/03/2000

of oriental fruit fly and Mediterranean fruit fly larvae conducted by Hinman and colleagues, acetaldehyde, butyraldehyde, and isobutyraldehyde were among the 108 compounds tested in 2 h exposures at 75 °F (≈25 °C) and atmospheric pressure, but propanal was not (Burditt et al., 1963). The authors concluded that the low to moderate toxicity of aldehydes made them too weak for commercial insecticide applications. However, by applying some of these same chemicals as gaseous fumigants under reduced pressure for the first quarter of treatment, we have found that potency and efficacy can be increased over applications at atmospheric pressure. Several of the aldehydes tested here act as rapid and potent pesticides while being well tolerated by key commodities.

MATERIALS AND METHODS

Chemicals. All aldehydes were obtained from Aldrich, Inc. (Milwaukee, WI), or from Wako Pure Chemical Industries Ltd. (Osaka), were of the highest grade available, and were used without further purification. All aldehydes were ≥95% purity, with the exceptions of (*E*)-2-octenal (94%); (*E*)-2-nonenal (93%); octanal and heptanal (92%); 2-methylbutanal, (*E*,*E*)-2,4-decadienal, (*E*)-2-undecenal, (*E*)-2-dodecenal, and (*E*)-2-tridecenal (90%); and (*E*,*E*)-2,4-heptadienal (88%).

Target Pests. Green peach aphids (*Myzus persicae* Sulzer) and black bean aphids (*Aphis fabae*) were graciously provided by the University of California Berkeley Insectary and Quarantine Facility and were reared on broccoli starts and fava bean plants, respectively, in a growth chamber (CMP-3023, Controlled Environments Ltd., Winnipeg, Canada) set at 23 °C, a 16:8 h light/dark cycle, and ∼50% humidity. Preliminary assays were conducted against western flower thrips (*Frankliniella occidentalis*), adult white fly (*Bemisia* spp.), and two-spotted spider mites (*Tetranychus urticae*), all collected in the wild in Berkeley and subsequently reared on fava bean plants under the same conditions. Obscure mealybugs (*Pseudococcus viburni* Signoret or *Pseudococcus affinis* Maskell) were provided by Kent Daane and reared on sprouted potatoes in a 2-gal plastic container kept in a room at ambient temperature in continuous darkness.

Leaf Disk Bioassays. Initial screening assays were conducted using 1 cm fava bean leaf disks in 103 mL plastic cups equipped with lids (Plastics Inc., St. Paul, MN). A fine-tipped paintbrush was used to place 10 black bean aphids on each leaf disk, which had been floated on 7 mL of water to prevent the aphids from walking off or coming into direct contact with the insecticide. The pure aldehyde was applied with a microsyringe to a small piece of No. 1 filter paper (Whatman International Ltd., Maidstone, U.K.) stapled to the underside of the lid.

Atmospheric Pressure Fumigations. All fumigation assays were conducted according to the method of Burditt et al. (1963) with minor modifications in a temperature-controlled room set to 23 (± 1) °C. Leaf sections containing ∼50 aphids of mixed stages were placed in 20 mm sections cut from cardboard cylinders of 60 mm diameter, and the ends were screened with nylon mesh to prevent escape. Each cylinder was then placed in a 9.5 L sealed metal fumigation chamber—modified industrial paint sprayers (Speedaire) obtained from W. W. Grainger, Inc. (Emeryville, CA). Pure aldehydes were introduced through a rubber septum in the lid using a 25 μL or 2000 μL syringe (Hamilton Co., Reno, NV) onto a filter paper mounted 2 in. below the chamber lid. Controls were subjected to the same conditions but without injecting the aldehydes. After treatment, the cardboard cylinders were removed from the fumigation chambers and stored at 23 °C until 24 h later, when mortality of the aphids was determined with the help of a microscope. Each aphid was gently prodded with a dissecting needle and categorized as alive or dead; aphids that were moribund or capable of the slightest movement whatsoever were nonetheless counted as alive.

Table 1. LD_{50} Values (± Standard Error) of Short-Chain Aldehydes to Aphids at Atmospheric Pressure versus a Two-Tier Reduced-Pressure Fumigation

aldehyde	LD_{50}[a] (mg/L) atmospheric fumigation[b]	LD_{50}[a] (mg/L) two-tier reduced-pressure fumigation[c]	potentiation factor[d]
propanal	7.3 (± 2.6)	2.1 (± 1.0)	3.5
butanal	26.1 (± 9.0)	10.8 (± 8.8)	2.4
isobutyraldehyde	24.4 (± 3.2)	4.6 (± 2.3)	5.3
hexanal[e]	19.4	<5	3.9
heptanal[e]	15	5	3

[a] Values were determined from a minimum of three replicates at a minimum of three doses at 23 (± 1) °C. [b] 760 mmHg, 2 h [method of Burditt et al. (1963)]. [c] Present study, 30 mmHg for 0.5 h and then 760 mmHg for 1.5 h. [d] Column 2 divided by column 3. [e] One replicate only.

Reduced Pressure Fumigations. Experiments intended to determine the effects of vacuum-assisted aldehyde fumigation were first conducted on insects in 9.5 L tanks with no commodity load, as described above for atmospheric pressure fumigations, with a few modifications. A pump was used to reduce the pressure inside the chamber to 30 mmHg, when an appropiate dose of pure aldehyde was introduced. The reduced pressure conditions were maintained for one-fourth of the 2 h fumigation period, when air was slowly allowed to restore the pressure to atmospheric (760 mmHg) for the remaining 1.5 h of fumigation. After treatment, a vacuum was briefly pulled one more time (often referred to as an "air wash") in an effort to volatize and remove any residual aldehyde smell, and the cardboard cylinders were then removed and stored at 23 °C until 24 h later, when mortality of the aphids was determined. Controls were subjected to the same conditions but without injecting the aldehydes.

Fumigations in the Presence of Commodities. Experiments against aphids in the presence of fruits, vegetables, and grains were conducted as described above except that the treatments lasted for 1 h rather than 2 h. Broccoli, naked iceburg lettuce, wrapped iceburg lettuce, green cabbage, red grapefruit, Valencia oranges, Meyer lemons, Hass avocados, Fuji apples, green seedless grapes, and red seedless grapes were obtained from a local wholesale distributor (Ja-Mar Produce Co., Oakland, CA). Long-grain white rice and pinto beans were purchased at a local retail market. Commodities were stored at 5 °C until 30 min before fumigation, except for rice and beans, which were stored at ambient temperature. Immediately before fumigation, a cardboard cylinder containing a leaf section with ∼50 aphids was inserted under the third outermost leaf of the lettuce and cabbage heads or in the center of the sample for all other commodities. Commodity samples ranging from 800 to 1000 g and representing a 30−40% load factor (defined as the approximate percent of total vessel volume occupied by the sample) were inserted into perforated polyethelene bags (60−70 slits of 4 mm in each bag) prior to treatment except for naked lettuce, which was treated unwrapped. The bagged samples were placed in the 9.5 L chambers and fumigated for 1 h (15 min at 30 mmHg followed by 45 min at 760 mmHg). At the end of the fumigation period the commodity was "air washed" as described above, aerated for 1 h in a ventilated area at 23 °C, and stored at 5 °C for 24 h, when aphid mortality was scored. Controls were subjected to the same conditions but without injecting the aldehydes. Visual and olfactory qualities were also rated 24 h after fumigation using a scale previously established for lettuce (Kader et al., 1973) and adapted to the other commodities.

Fumigation at Distinct Temperatures. Experiments investigating the effect of fumigation temperature on aldehyde potency were conducted on broccoli leaf sections bearing ∼50 aphids. The fumigation lasted for 2 h in a temperature-controlled room set to 15 or 23 °C following the same methods and reduced-pressure regime described above.

Data Anlysis. The LD_{50} and LD_{90} values and their respective standard errors for Tables 1−3 were determined using a

Table 2. Effect of Temperature on Toxicity of Aldehydes to Aphids, Fumigated under the Two-Tier Reduced-Pressure Scheme

aldehyde	LD_{50}[a] at 15 °C	LD_{50}[a] at 23 °C (± 1)
propanal	7.47[b]	2.1 (± 1.0)
butanal	31.0 (± 7.1)	10.8 (± 8.8)
isobutryaldehyde	29.3 (± 9.9)	4.6 (± 2.3)

[a] Approximate LD_{50} (mg/L) in exposures of 0.5 h at 30 mmHg followed by 1.5 h at 760 mmHg. [b] One replicate only.

Table 3. LD_{50} and LD_{90} Values (± SE) for Various Aldehydes against Green Peach Aphids Placed under the Third Leaf of Naked Iceburg Lettuce Heads and Fumigated under the Two-Tier Reduced-Pressure Scheme[a]

aldehyde	LD_{50}[b] (mg/L)	LD_{90}[b] (mg/L)
propanal	87.0 (± 19.0)	124.5 (± 13.9)
butanal	130.1 (± 12.4)	177.5 (± 14.6)
isobutyraldehyde	193.3 (± 11.4)	224.1 (± 12.6)
2-methylbutanal	138.4 (± 20.3)	184.4 (± 7.6)
2-methyl-(*E*)-2-butenal	76.6 (± 22.4)	123.6 (± 14.1)
3-methyl-(*E*)-2-butenal	73.9 (± 29.8)	139.1 (± 6.8)
pentanal	71.4 (± 13.2)	115.7 (± 8.9)
(*E*)-2-pentenal	13.1 (± 2.4)	25.8 (± 7.3)
2-methylpentanal	39.9 (± 6.9)	54.2 (± 4.0)
2-methyl-(*E*)-2-pentenal	33.5 (± 13.9)	85.8 (± 5.7)
(*E*)-2-hexenal	24.3 (± 0.5)	54.4 (± 13.0)

[a] 30 min at 30 mmHg and 1.5 h at 760 mmHg at 23 (± 1) °C; mortality was recorded 24 h later. [b] Values were determined from a minimum of three replicates at a minimum of three doses.

four-parameter logistic regression curve (Sigma Plot 4.0). All other LD_{50} and LD_{95} values and their respective 95% confidence limits were determined using a probit analysis (Finney, 1971). Unless otherwise stated, dose−mortality curve fitting for each compound was based on a minimum of five different concentrations tested in a minimum of three separate replicates.

RESULTS

Twenty-nine aldehyde compounds were initially screened for their insecticidal activity at atmospheric pressure and ambient temperature against aphids on leaf disks in 103 mL plastic cups. All of the compounds were insecticidal, so the procedure was scaled up to 9.5 L tanks.

Aphid mortality from aldehydes replicating the previously established method of 2 h at atmospheric pressure (Burditt et al., 1963) was compared with mortality from similar doses applied at 30 mmHg (Stewart et al., 1980) for the first 30 min followed by gentle entrance of air to the chamber so that the final 1.5 h of treatment was effected at atmospheric pressure (760 mmHg). The results of this assay for several aldehydes are presented in Table 1 and show that use of a two-tier vacuum regime increases the effectiveness of aldehyde fumigants by 2−5-fold.

Preliminary data indicate that ambient temperature of the test environment (and of the lettuce itself) plays an important role in toxicity of the aldehydes. The amount of compound needed to obtain 50% control at 15 °C is several times greater than that needed to elicit the same results at 23 °C (Table 2).

Vacuum fumigation was eventually found to be impractical for the following 18 compounds because they did not volatize completely, appeared to polymerize and precipitate onto the inside surfaces of the test chamber, or caused excessive damage to the lettuce in terms of both cosmetic quality and unpleasant odor: hexanal; heptanal; octanal; nonanal; decanal; (*E*)-2-heptenal; (*E*)-2-octenal; (*E*)-2-nonenal; (*E*)-2-decenal; (*E*)-2-undecenal; (*E*)-2-dodecenal; (*E*)-2-tridecenal; (*E*,*E*)-2,4-hexadienal; (*E*,*E*)-2,4-heptadienal; (*E*,*E*)-2,6-nonadienal; (*E*,*E*)-2,4-decadienal; 2,6-dimethyl-5-heptenal; and 2-isopropyl-5-methyl-2-hexenal.

Potency against Aphids on Lettuce. The results from the other 11 compounds yielded a dose−mortality relationship against aphids, and their LD_{50} and LD_{90} values for applications in the presence of naked lettuce are presented in Table 3.

Tolerance of Naked Lettuce to Treatment. Phytotoxicity impacts on naked lettuce as a consequence of fumigation treatment with 11 aldehydes were scored in terms of visual quality and residual aldehyde odor; the effects from doses that caused 100% aphid mortality are given in Figure 1.

The worst damage to naked lettuce was restricted to the outermost (“wrapper”) leaves, which sometimes became severely wilted, slimey, and necrotic, even in heads of which the inner leaves remained undamaged. The same tests were therefore performed on heads of iceburg lettuce that had been commercially prewrapped in polyethylene bags because the prewrapped lettuce has had the outer leaves removed before packaging. Interestingly, the results showed that insecticidal effects could be achieved on plastic-wrapped lettuce at lower aldehyde doses and with much less detriment to the lettuce appearance and odor (see next section).

By running assays of various time scales, it was learned that substantially equivalent results (in both aphid mortality and impact on lettuce quality) are obtained by applying the compounds for 1 h rather than 2 h (data not shown), so all subsequent assays were for 1 h.

Although all 11 aldehydes caused significant damage to naked lettuce, the following 4 were selected for further study because the product retained, or nearly retained, quality considered to be acceptable for commercial sale: propanal, (*E*)-2-pentenal, 2-methyl-(*E*)-2-butenal, and 3-methyl-(*E*)-2-butenal.

Potency and Phytotoxicity of Four Aldehydes Applied to Various Commodities. The four best aldehydes were further assayed against aphids in the presence of naked lettuce, wrapped lettuce, and a variety of other agricultural commodities. Although aphids do not infest all of the commodities tested, the assays give an idea how much of the aldehydes are being absorbed by a given commodity because a load factor of 30−40% was used throughout.

Figure 2 presents the LD_{50}, LD_{95}, and respective 95% confidence limits for propanal (a), (*E*)-2-pentenal (b), and 2-methyl-(*E*)-2-butenal (c); 3-methyl-(*E*)-2-butenal was similar in potency to 2-methyl-(*E*)-2-butenal but caused greater damage to the commodities, so it was eventually abandoned in favor of the first three candidates. As is evident from the bar graphs, much higher doses were needed to achieve insecticidal effects on broccoli, lettuce, and cabbage than on the other commodities, presumably because they absorbed more of the aldehydes. This hypothesis is supported by the phytotoxicity results (Figure 3a−c), which show that broccoli, naked lettuce, and cabbage suffered by far the worst damage.

The effects of aldehyde fumigations causing 100% aphid mortality in the presence of various commodities are summarized in Figure 3a−c. Propanal caused no detectable visual damage to any commodity besides naked lettuce and caused olfactory damage within the

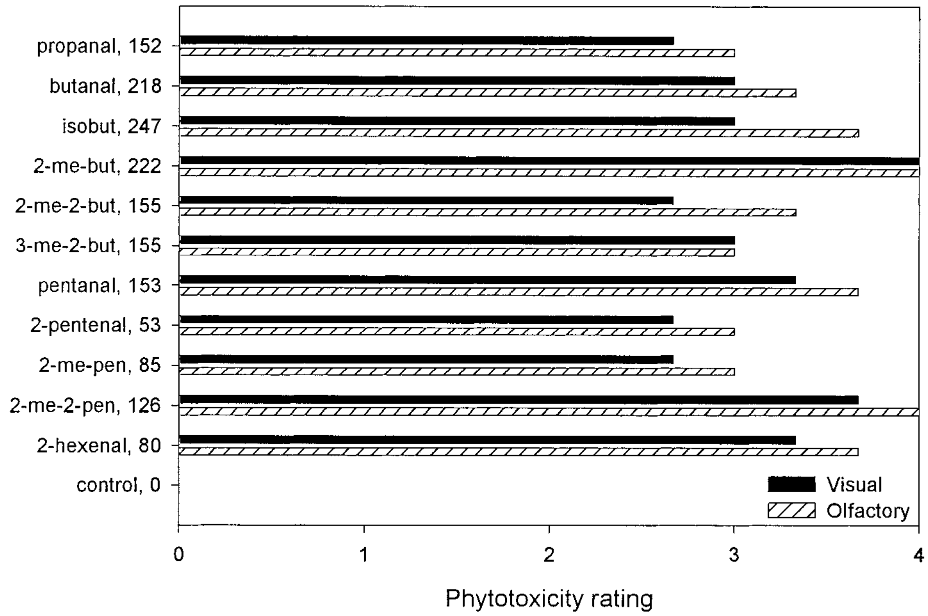

Figure 1. Visual and olfactory quality of naked lettuce 24 h after 2 h, two-tier, reduced-pressure treatments with 11 aldehydes. The amount of each aldehyde applied (given in mg/L following the compound name) was the lowest dose that caused 100% aphid mortality in all three replicates. Visual quality: 0, excellent (free from defects); 1, good (minor defects; not objectionable); 2, fair (moderate defects but still acceptable); 3, poor (substantial defects; limit of marketability); 4, extremely poor (not usable). Olfactory quality: 0, excellent (normal, no change); 1, good (questionably detectable); 2, fair (slightly detectable); 3, poor (detectable; limit of tolerance); 4, extremely poor (pungent and unpleasant).

limits of what may be considered commercially acceptable for all commodities except naked lettuce, broccoli, and cabbage. 2-Methyl-(*E*)-2-butenal was well tolerated by citrus, avocado, apple, and table grape, but left unacceptable olfactory residues in naked lettuce, wrapped lettuce, broccoli, cabbage, and rice. 2-Pentenal could not be sensorily detected at all after treatment of orange, lemon, apple, and grape, caused minor defects in grapefruit, avocado, rice, and bean, and caused unacceptable levels of damage to naked and wrapped lettuce, broccoli, and cabbage. Avocados retained significant aldehyde odors in the skin but the flesh itself did not.

Preliminary results against other target pests indicate that mealybugs, thrips, and whitefly adults are more susceptible to aldehydes than green peach aphids are but that spider mites and cabbage aphids are somewhat more resistant (data not shown).

DISCUSSION

The technique of applying gaseous fumigants in conjunction with a vacuum has been known for many years (Sasscer and Hawkins, 1915), but the effect of vacuum on fumigant efficacy can vary considerably, with the action of some compounds being increased (Bhambhani, 1964), that of others being decreased (Monro, 1969; Monro et al., 1966), and that of still others showing little difference (Aharoni and Stewart, 1980). For instance, a recent study of hydrogen peroxide treatment of teliospores contaminating wheat and barley seeds found no difference between the effectiveness of the treatment under a "deep" vacuum of 38 mmHg and that under a "shallow" vacuum of 680 mmHg (Smilanick et al., 1994). The advantage attributed to vacuum fumigation in the past—and especially of the two-tier system whereby the latter portion of treatment is effected at atmospheric pressure—has been its capacity for increasing penetration of a dense load (Stewart and Aharoni, 1983), for example, of densely packed dates (Brown and Heuser, 1953), or for reducing the time needed for exposure (Monro, 1969). To our knowledge, nothing has previously been written about whether two-tier, vacuum-assisted fumigation with aldehydes increases insecticidal potency in a load-free vessel. The present study shows that the use of a two-tier vacuum regime increases the insecticidal effectiveness of aldehydes even in the absence of a load, for example, against aphids in an otherwise empty vessel, perhaps by improving penetration into the insect itself.

Control aphids exposed to reduced pressure alone were temporarily slowed but suffered no mortality. The lethality of vacuum alone on insects is insignificant except at extraordinarily low pressures impractical for commercial treatments, for example, 0.05−0.03 mmHg (Thornton and Sullivan, 1964), or at very long exposure periods, on the order of 2 days (Aharoni et al., 1986). The effects of vacuum fumigation on target pests is a complex phenomenon, potentially influenced by water loss, relative levels of CO_2 and oxygen in the test chamber, and physiological changes rendering the insect more vulnerable. The two-tier fumigation technique creates initial conditions favorable to volatizing the fumigant and may then employ air to carry the fumigant thoughout the chamber and into the insect itself via its spiracles.

The enormous difference between the dose required to kill aphids on a commodity as opposed to aphids alone suggests that the vast majority of the compound is absorbed by the commodity itself, perhaps due to high water content. The LD_{50} values for aphids on lettuce in a vessel with ~30−40% load factor are manyfold the concentrations needed to kill aphids in an otherwise empty container (compare Tables 1 and 3). A similar phenomenon appears to occur with broccoli, cabbage, and, to a lesser extent, the other commodities as well.

Comparison of panels a, b, and c of Figure 2 shows that (*E*)-2-pentenal is the most potent, followed by 2-methyl-(*E*)-2-butenal, and then propanal. Although propanal is the least potent of the three, it is also much

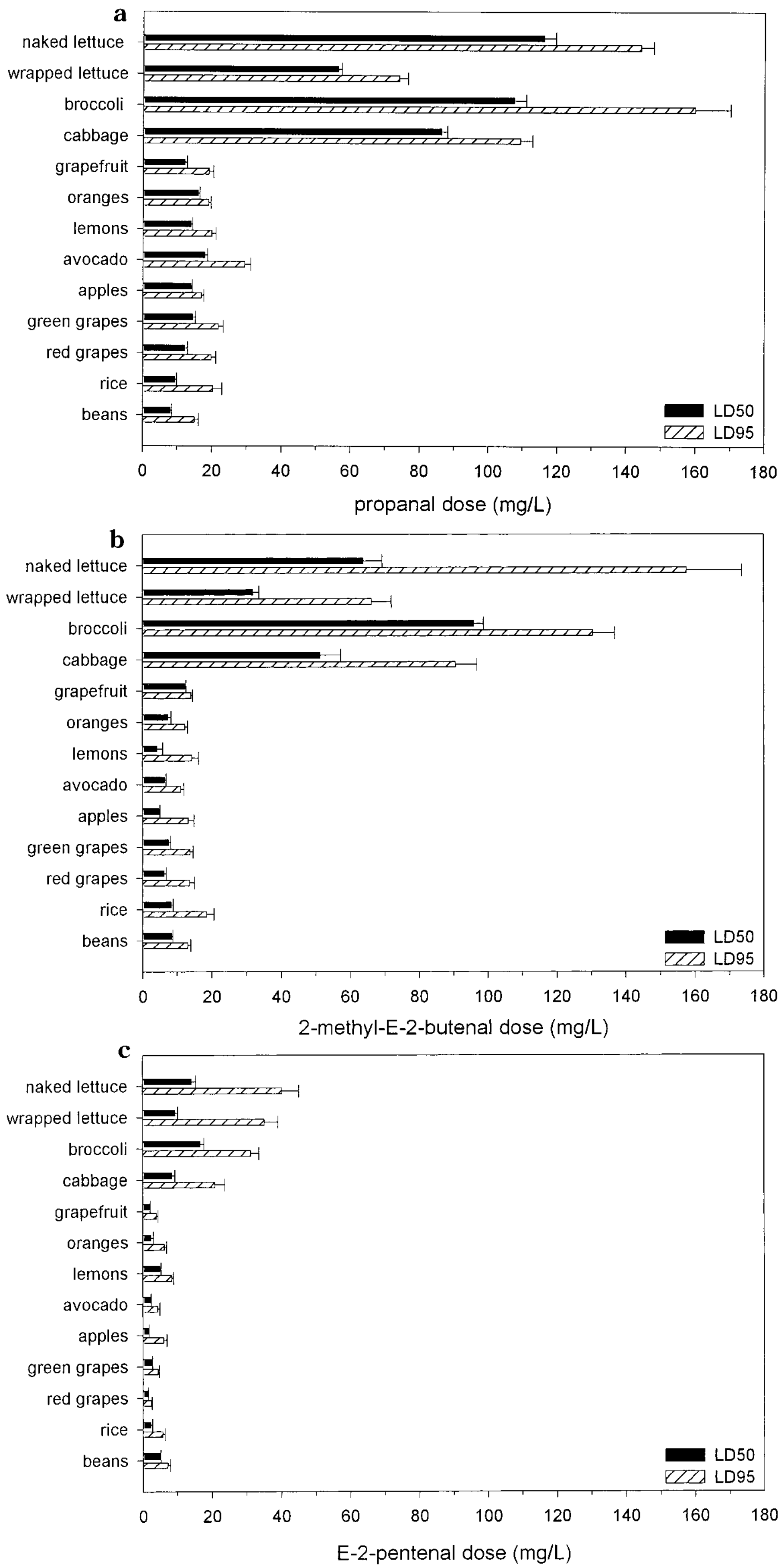

Figure 2. LD_{50} and LD_{95} values for propanal (a), 2-methyl-(*E*)-2-butenal (b), and (*E*)-2-pentenal (c) against green peach aphids (bars indicate 95% confidence limits) in 1 h exposures at 23 °C using a two-tier vacuum regime of 30 mmHg for 15 min and 760 mmHg for the remaining 45 min. Commodities bearing higher LD values are those that absorb the aldehydes more greatly.

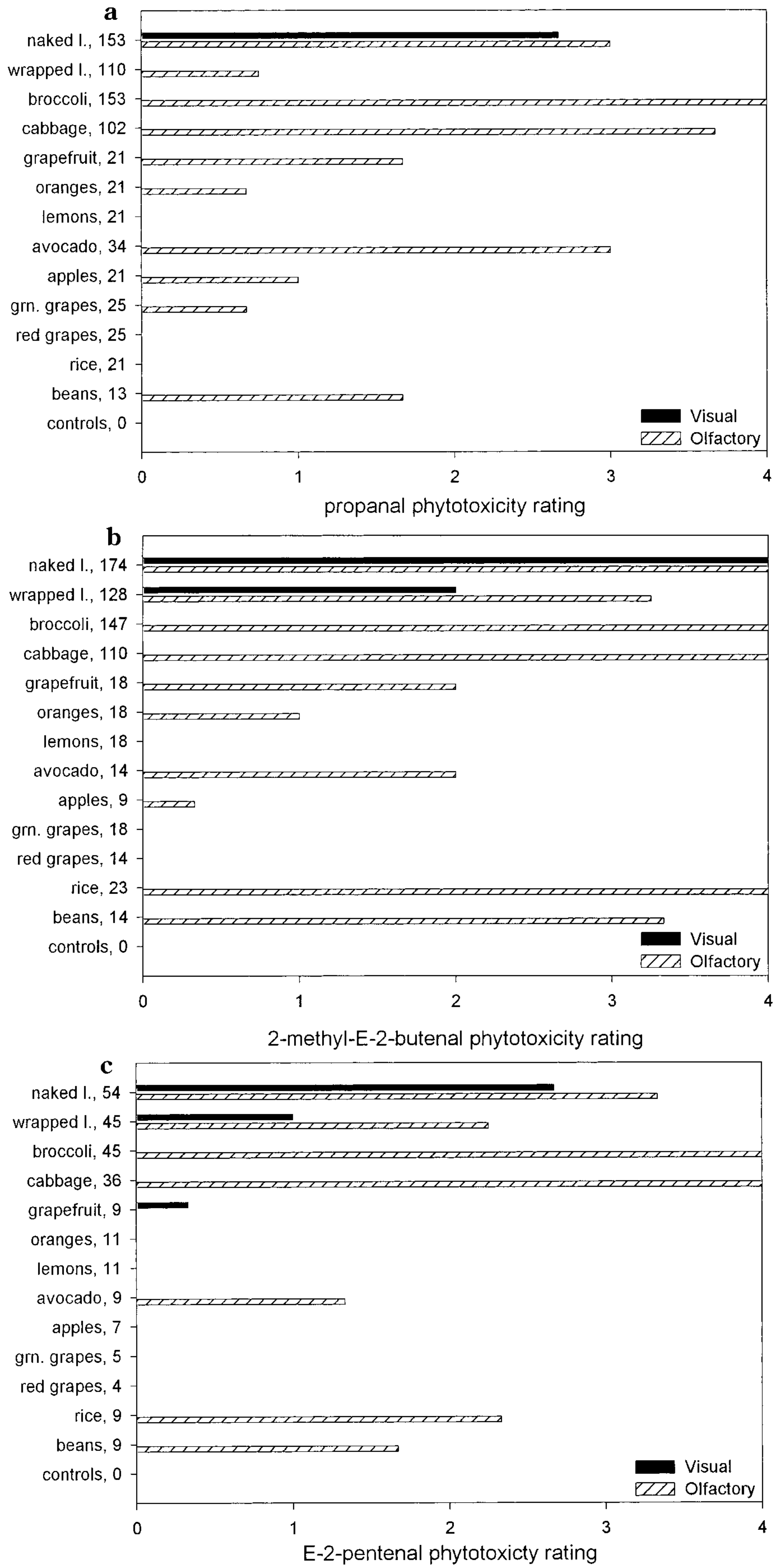

Figure 3. Visual and olfactory quality of various commodities 24 h after 1 h, two-tier reduced-pressure treatments with propanal (a), 2-methyl-(*E*)-2-butenal (b), and (*E*)-2-pentenal (c). The rating is the average damage from three replicates at the lowest dose (given in mg/L following the compound name) that effected 100% mortality in all three. The phytotoxicity scale is the same as that used for Figure 1.

less expensive and therefore may be more promising for commercial situations.

Comparison of panels a, b, and c of Figure 3 shows that propanal is the best-tolerated fumigant for all commodities assayed here, with (*E*)-2-pentenal performing nearly as well. All three compounds caused intolerable levels of visual and/or olfactory damage to naked lettuce, broccoli, and cabbage. Nevertheless, all three compounds also show potential given that at doses that killed 100% of aphids, the visual and olfactory effects to most of the commodities were below a level of 3 on a scale where 3 roughly corresponds to the cutoff for marketable quality.

In our selection of candidate compounds for further study, minimizing cosmetic damage to the commodities was assigned greater importance than residual odor because whereas appearance of a damaged commodity can only be expected to worsen during storage and transport, undesirable aldehyde odors may dissipate with time or with additional air washes immediately following fumigation. It is unclear why odor is so much more greatly retained by broccoli and leafy vegetables than by the other commodities. Grapes, apples, lemons, and oranges performed the best, with treated samples usually indistinguishable from controls. At high concentrations, aldehydes may cause product defects in flavor, and this topic merits further attention.

It is imprecise, and perhaps inadvisable, to attempt a direct comparison among compounds evaluated in separate studies that are conducted under even slightly different conditions. Nevertheless, the present study suggests that several of the aldehydes tested are roughly comparable in insecticidal potency to the widely used quarantine fumigant methyl bromide and may therefore constitute a feasible alternative. Methyl bromide required concentration time products (CTP) of ~100−150 mg L^{-1} h^{-1} against aphids, thrips, mealybugs, and mites on flower bulbs (European and Mediterranean Plant Protection Organization, 1993), and ~25−50 mg L^{-1} h^{-1} against larval and adult western flower thrips in otherwise empty containers (MacDonald, 1993). Our data indicate that the aldehyde CTP values required for 50% mortality of aphids in a fumigation vessel with no comodity load are ~4.2 mg L^{-1} h^{-1} for propanal, ~21.6 mg L^{-1} h^{-1} for butanal, and ~9.2 mg L^{-1} h^{-1} for isobutyraldehyde (calculated from Table 1). In the presence of lettuce (which due to its high water content might be expected to sorb considerably more fumigant than a flower bulb), aldehydes killed 100% of aphids in three replicates at CTPs of ~304 mg L^{-1} h^{-1} for propanal, 310 mg L^{-1} h^{-1} for 2-methyl-(*E*)-2-butenal, and 106 mg L^{-1} h^{-1} for (*E*)-2-pentenal (calculated from Figure 1). With regard to relative toxicity to nontarget organisms, methyl bromide is 14 times more toxic than propanal when inhaled by mice (Lewis, 2000) and 6 times more toxic (Danse et al., 1984) than propanal (Smyth et al., 1951) when administered orally to rats, suggesting that a transition from methyl bromide to aldehydes may lower the health hazards to workers and handlers.

Results seem to justify further study in which the best compound candidates would be applied to larger loads and more varied products and against other target pests. The effects of aldehydes on products after a more extended shelf life are also important to determine, but this was beyond the scope of this study. It seems likely that less delicate commodities, such as unshelled nuts or imported timber, are promising candidates for vacuum fumigation with aldehyde pesticides. The potential for use of aldehydes in soil disinfestation is still untested.

ACKNOWLEDGMENT

We are grateful to UC Berkeley Insectary for providing insects.

LITERATURE CITED

Aharoni, Y.; Stewart, J. K. Thrips mortality and strawberry quality after vacuum fumigation with acetaldehyde or ethyl formate. *J. Am. Soc. Hortic. Sci.* **1980**, *105*, 926−929.

Aharoni, Y.; Stewart, J. K.; Hartsell, P. L.; Young, D. K. Acetaldehyde—a potential fumigant for control of the green peach aphid on harvested head lettuce. *J. Econ. Entomol.* **1979**, *72*, 493−495.

Aharoni, Y.; Apelbaum, A.; Copel, A. Use of reduced atmospheric pressure for control of the green peach aphid (*Myzus persicae*) on harvested head lettuce (*Lactuca sativa*). *HortScience* **1986**, *21*, 469−470.

Bhambhani, H. J. Recent advances in vacuum fumigation. *World Rev. Pest Control* **1964**, *3*, 53−56.

Brown, W. B.; Heuser, S. G. Behaviour of fumigants during vacuum fumigation: I. Penetration of methyl bromide into boxes of dates. *J. Sci. Food Agric.* **1953**, *4*, 48−57.

Burditt, A. K.; Hinman, F. G.; Balock, J. W. Screening of fumigants for toxicity to eggs and larvae of the oriental fruit fly and Mediterranean fruit fly. *J. Econ. Entomol.* **1963**, *56*, 261−265.

Danse, L. H.; van Velsen, F. L.; van der Heijden, C. A. Methylbromide: carcinogenic effects in the rat forestomach. *Toxicol. Appl. Pharmacol.* **1984**, *72*, 262−271.

European and Mediterranean Plant Protection Organization. Quarantine procedure: Methyl bromide fumigation of flower bulbs to control aphids, *Taeniothrips simplex*, *Rhizoglyphus* spp. and *Phenacoccus* spp. *Bull. OEPP* **1993**, *23*, 206.

Fenaroli, G.; Burdock, G. A. *Fenaroli's Handbook of Flavor Ingredients*, 3rd ed.; CRC Press: Boca Raton, FL, 1995.

Ferguson, J.; Pirie, H. The toxicity of vapours to the grain weevil. *Ann. Appl. Biol.* **1948**, *35*, 532−550.

Finney, D. J. *Probit Analysis*, 3rd ed.; Cambridge University: New York, 1971.

Hinman, F. G. Screening tests of compounds as fumigants for eggs and larvae fo the oriental fruit fly. *J. Econ. Entomol.* **1954**, *47*, 549−556.

Kader, A. A.; Lipton, W. J.; Morris, L. L. Systems for scoring quality of harvested lettuce. *HortScience* **1973**, *8*, 408−409.

Lewis, R. J., Sr. *Sax's Dangerous Properties of Industrial Materials*, 10th ed.; Wiley: New York, 2000; Vol. 3.

MacDonald, O. C. Susceptibility of western flower thrips, *Frankliniella occidentalis* (Pergande) to fumigation with methyl bromide. *Ann. Appl. Biol.* **1993**, *123*, 531−537.

Monro, H. A. U. Vacuum fumigation. In *Manual of Fumigation for Insect Control*; FAO: Rome, Italy, 1969.

Monro, H. A. U.; Dumas, T.; Buckland, C. T. The influence of vapor pressure of different fumigants on the mortality of two stored product insects in vacuum fumigation. *J. Stored Prod. Res.* **1966**, *1*, 207−222.

Sasscer, E. R.; Hawkins, L. A. *A Method of Fumigating Seeds*; USDA Bulletin 186; U.S. Department of Agriculture: Washington, DC, 1915.

Smilanick, J. L.; Goates, B. J.; Denis-Arrue, R.; Simmons, G. F.; Peterson, G. L.; Henson, D. J.; Rij, R. E. Germinability of *Tilletia* spp. teliospores after hydrogen peroxide treatment. *Plant Dis.* **1994**, *78*, 861−865.

Smyth, H. F., Jr.; Carpenter, C. P.; Weil, C. S. Range-finding toxicity data. *List IV. Am. Med. Assoc.: Arch. Ind. Hyg. Occup. Med.* ***1951***, *4*, 119−122.

Stewart, J. K.; Aharoni, Y. Vacuum fumigation with ethyl formate to control the green peach aphid in packaged head lettuce. *J. Am. Soc. Hortic. Sci.* **1983**, *108*, 295−298.

Stewart, J. K.; Aharoni, Y.; Hartsell, P. L.; Young, D. K. Acetaldehyde fumigation at reduced pressures to control the green peach aphid on wrapped and packed head lettuce. *J. Econ. Entomol.* **1980**, *73*, 149−152.

Thornton, B. C.; Sullivan, W. N. Effects of a high vacuum on insect mortality. *J. Econ. Entomol.* **1964**, *57*, 852−854.

U.S. Environmental Protection Agency. Clean Air Act, Title VI, Amendments of 1990 (for more information, see http://www.epa.gov/docs/ozone/mbr/mbrqa.html).

Received for review February 22, 2000. Revised manuscript received June 28, 2000. Accepted June 29, 2000. We are grateful to TransFRESH Corp. for providing financial support for this research.

JF000233+