Distribution of Ethylene Dibromide within a Fumigation Chamber during Fumigation of Citrus Fruit

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The distribution of EDB during fumigation of citrus fruit was examined using three dosage schedules approved for control of Queensland Fruit Fly (*Dacus tryoni* Frogatt). During a 2-h fumigation, EDB initially sorbed rapidly onto the chamber walls and associated structures; sorbtion onto cartons was less rapid and sorbtion by fruit slower still. EDB distribution during fumigation was similar for each fumigation schedule, with an average of 29% of total EDB sorbed onto the chamber walls and structures, 31% onto the cartons, and 11% and 16% onto Valencia oranges and Eureka lemons, respectively. Similar amounts of EDB were sorbed by fruit for each schedule tested.

Ethylene dibromide (EDB) has been the most widely used fumigation treatment for fruits and vegetables (Balock and Lindgren 1951; Leggo et al., 1965; Monro, 1969; Rigney and Wild, 1975). Unfortunately the major fruit fly in Australia, Queensland Fruit Fly, is relatively tolerant to this fumigant, with a fumigation of 24 g/m^3 EDB at 20 °C required for citrus (Leggo et al., 1965; Rigney and Wild, 1975) compared to 12 g/m^3 at 20 °C required for the Mediterranean, Oriental, Caribbean, and Cherry Fruit Fly (Monro, 1969).

A study of EDB distribution during fumigation, particularly at these higher doses, is required to explain both why only a small amount of EDB is sorbed by the fruit (Sinclair and Lindgren, 1952; Sinclair et al., 1962; Dumas and Bond, 1975) and also why the decline of gaseous EDB is so rapid during fumigation (Sinclair and Lindgren, 1952; Seo et al., 1970; Dumas and Bond, 1975).

Miller et al. (1981) have performed the only distribution study of EDB resulting from a fumigation, however, the distribution was studied after a 2-h aeration period and not during fumigation. Consequently the largest component in their budget was EDB expelled through ventilation (81%); this component is irrelevant when studying EDB distribution during fumigation.

Recently, Morris et al. (1982) developed a sampling and analytical technique which enabled EDB distribution during fumigation to be studied.

MATERIALS AND METHODS

Epoxy coated, galvanized iron, experimental fumigation chambers of 0.283 m^3 were used, with a fan in the bottom to circulate air during fumigation and a water seal around the lid. Four removable lengths of annealed stainless steel tubing (1.5 mm i.d., 3.0 mm o.d.) were fitted through a wall to enable samples to be drawn from various positions in the chamber. Sampling and analytical techniques for gaseous EDB are already described (Morris et al., 1982).

The three currently recommended fumigation schedules for Queensland Fruit Fly (Gellatley et al., 1978), namely 24 g/m^3 at 20 °C, 32 g/m^3 at 15 °C, and 41 g/m^3 at 10 °C, were examined. Load conditions used were, an empty chamber, a chamber with two empty telescopic corrugated fibreboard cartons with waxed inners (Australian Export Citrus Package No. C26), or two cartons packed with either Valencia oranges or Eureka lemons. When loaded with fruit the fumigation chamber was at 27% capacity. three replicate fumigations were done for each load condition and dose/temperature schedule. Samples were taken above the carton, in the middle of the top carton, the middle of the bottom carton, and below the cartons. Additional samples were taken directly from the fumigation chamber through a septum to enable corrections for sorbtion of EDB onto the inside of the sampling tubes (typically 3–10% reduction).

EDB fumigation was commenced by injecting the required dose through a silicone septum into a hot porcelain evaporating dish. After allowing 2 min for fumigant dispersal, sampling commenced and was repeated at 15-min intervals during the 2-h fumigation. All chambers, cartons, and fruit were left in the fumigation room overnight until they had equilibrated with room temperature (fumigation temperature) before fumigation. Following fumigation each chamber was aerated for 24 h, with the fan on during the first 4 h. The stainless steel tubing was cleaned by placing in an oven at 180 °C and gently blowing air through them.

The ability of various regression equations to describe the decline of gaseous EDB over time was compared. The best fits (as determined by correlation coefficients) were logarithmic and parabolic polynomial (transformed in $x^{1/2}$) regressions. The simpler logarithmic fit was preferred because the slope component of the equation is entirely described by the term "b". The data was analyzed by fitting a logarithmic curve for each combination of factors according to the equation $y = a + b \ln x$, where y = concentration of EDB in g/m^3 and x = time in minutes.

A three way factorial analysis of the effects of temperature/dose, chamber loadings, and sampling positions was then performed with the slope term "b" (or rate of EDB decline over time) for each replicate as the variable, rather than EDB concentration. The data were analyzed like this in order to avoid the serious errors that can arise if time is used as a factor in factorial analysis. These errors arise because successive measurements on one subject over time are usually not independent but positively correlated (Snedecon and Cockran, 1980).

Significance comparisons between means were made by using the Waller-Duncan k ratio LSD rule (Chew, 1977). This test is superior to other means comparisons tests, since the k ratio LSD varies with the F value, number of means, error mean square, and error degrees of freedom. The k ratio LSD is thus smaller in a large experiment with a highly significant treatment effect, but considerably larger in a poorly designed or small experiment with a not very significant treatment effect.

RESULTS AND DISCUSSION

The three-way factorial analysis (temperature/doses × loading conditions × sampling positions) found that only the first-order interactions were significant (Table I), with the differences between the three temperature/dose levels

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0021-8561/85/1433-0801$01.50/0

and between the for different chamber loadings being highly significant. The trends were for an increased rate of EDB decline at lower temperatures (higher doses) and increased rates of decline as the chamber loading increased. The differences between the four chamber sampling positions were significant, with a marginally slower rate of EDB decline at the lower sampling positions. This would most likely be due to the density of EDB vapor being six times that of air (Monro, 1969).

The regression curves for each combination of temperature/dose and chamber loadings are shown in Figure 1 parts a–c. Since the effect of sampling positions was only just significant and no second-order interactions (i.e., interactions between different levels of temperature/dose, chamber loadings, and sampling positions) were significant, each point in this figure is the average of twelve individual measurements (4 sampling points, repeated 3 times). The EDB reduction in the gaseous phase during fumigation with an empty chamber was substantial, with final levels of EDB being 65% of the initial dose at 20 °C, 69% at 15 °C, and 61% at 10 °C. The most rapid reduction was in the first few minutes with levels declining by 20% during this time.

The addition of empty cartons to the fumigation chamber resulted in large reductions in gaseous EDB during fumigation. Final EDB levels were 35% at 20 °C, 34% at 15 °C, and 31% at 10 °C, with most of the loss occurring in the first 15 min. The rate of reduction due to cartons, especially at lower temperatures, was slower in the first 4 min than occurred in the empty chamber.

The addition of cartons of Valencia oranges to the fumigation chamber resulted in a further large drop in gaseous EDB, with final levels of 23% at 20 °C, 22% at 15 °C, and 21% at 10 °C. Reduction in EDB levels due to fruit was not as rapid as that due to cartons, with no significant reduction occurring after 4 min. The addition of cartons and Eureka lemons resulted in the largest loss of EDB in the gaseous phase, with final levels of 17% at 20 °C, 14% at 15 °C, and 16% at 10 °C. Although Eureka lemons caused a greater reduction in final EDB level than Valencia oranges the rates of reduction of EDB with time were similar; again no significant reduction of EDB occurred after 4 min.

EDB loss into the water seal around the chamber lids was determined by monitoring levels in the water during fumigation. Between 5.1 and 7.4% of EDB was taken up in the water. Given the amounts of EDB present in the water during fumigation and given the surface area exposed to the fumigation room and EDB vapor pressure (Monro, 1969), the EDB loss through the water seal during fumigation could also be calculated (see Table II).

A summary of the final EDB distribution at the end of the fumigation can be calculated from this data and is presented in table II. The major sink for EDB is the chamber walls and associated structures with 32.2–38.7% of total dose sorbed. After allowing for losses into and through the water seal this gives levels of 581 mg/m^2, 644 mg/m^2, and 1026 mg/m^2 sorbed onto the chamber surfaces at 20, 15, and 10 °C, respectively. Large reductions of gaseous EDB in an empty chamber have been reported previously (Sinclair and Lindgren, 1952; Dumas and Bond, 1975). The EDB levels that sorbed onto the chamber walls and internal structures are similar to the 390 mg/m^2 reported for galvanized iron fumigated with 20 g/m^3 EDB for 2 h (Coggiola and Huelin, 1964).

The cartons were another major sink for EDB with 29.1–33.3% of the total dose sorbed. Actual EDB amounts sorbed by the cartons were 5372 mg/kg, 8134 mg/kg, and

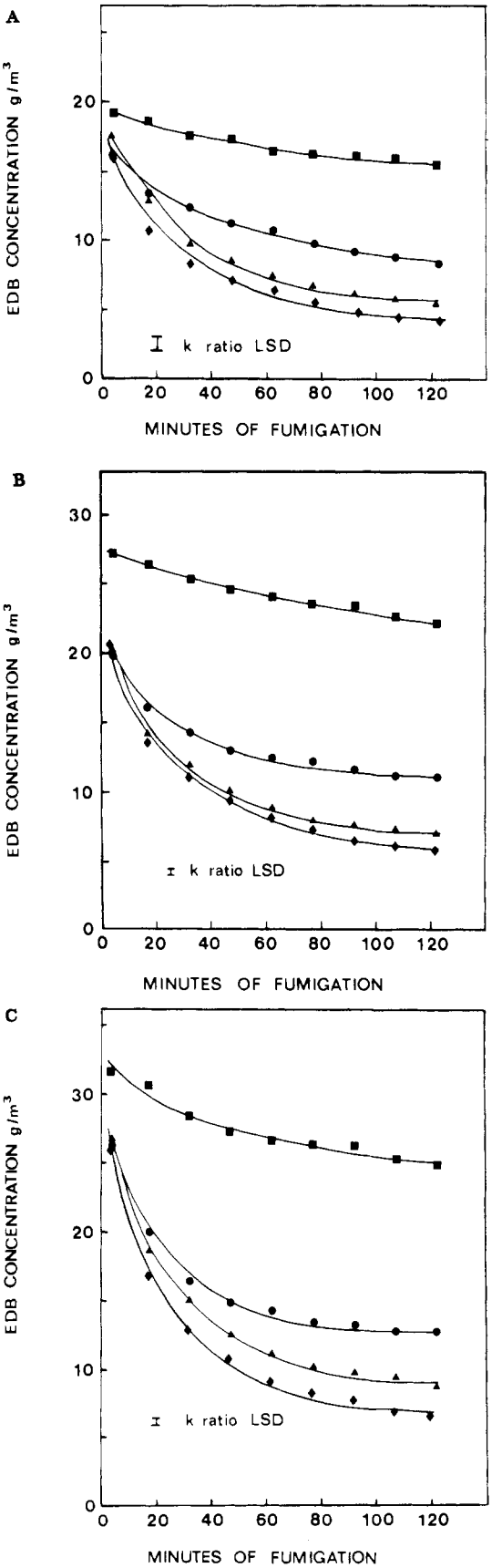

Figure 1. The losses of EDB in the gaseous phase during fumigation with different chamber loadings and under different conditions: (A) fumigation at 20 °C with 24 g/m^3 EDB; (B) fumigation at 15 °C with 32 g/m^3; (C) fumigation at 10 °C with 41 g/m^3 EDB. Fumigation load conditions are (■) empty chamber, (●) chamber and empty cartons, (▲) chamber and cartons of Valencia oranges, (♦) and chamber and cartons of Eureka lemons. Error bars indicate k ratio LSD levels for k = 100 (Chew, 1977).

Table I. Significant Differences between Levels of the Main Fumigation Factors and Their Effect on the Decline of EDB during a 2-h Fumigation[a,b]

factor	level of factor				sig level, %
temp/dose	20 °C/24 g m^3	15 °C/32 g m^3	10 °C/41 g m^3		
	−2.62 c	−3.14 b	−4.29 a		<0.01
chamber loading	empty	cartons	cartons + oranges	cartons + lemons	
	−1.46 d	−2.62 c	−3.74 b	−4.52 a	<0.01
chamber positions	above cartons	top carton	bottom carton	below cartons	
	−3.06 ab	−3.15 a	−2.83 b	−2.83 b	4.2

[a] As measured by the slope term "b" from the equation $Y = a + b \ln x$. [b] Means within a row followed by different letters are significantly different at the $k = 100$ level (Chew, 1977).

Table II. Final Distribution of EDB during a 2-h Fumigation under Three Different Conditions[a]

components of fumigation chamber	fumigation conditions		
	20 °C, 24 g/m^3	15 °C, 32 g/m^3	10 °C, 41 g/m^3
chamber walls and internal structures	30.2	25.1	31.2
water jacket	5.1	6.9	7.4
loss through water jacket	0.13	0.17	0.14
cartons	29.1	33.3	30.3
oranges[b]	11.7	12.5	8.8
lemons	17.6	16.3	13.4
remainder in air-oranges[b]	23.8	22.9	22.2
-lemons	17.9	18.1	17.6

[a] All values as percent of initial fumigant. [b] Only oranges or lemons were present, not both.

9483 mg/kg at 20, 15, and 10 °C, respectively. Since EDB is highly soluble in nonpolar compounds such as wax (Monro, 1969), the high and rapid EDB uptake by these cartons would seem mainly due to the waxed inner cartons.

The high amounts of EDB sorbed by cartons would help explain the results obtained by Swaine et al. (1976), who found fumigation treatments were much more effective in killing fruit fly when the fruit was held in waxed cartons, than when removed from the cartons after fumigation. The cartons in effect provided a secondary fumigation as the EDB desorbs. The work reported in this paper is the only study of EDB sorbtion by cartons during fumigation. King et al. (1979) examined EDB levels in cartons after fumigation and reported a level of 300 mg/kg 2 h after fumigation. However, from their data it was impossible to accurately determine uptake of EDB by cartons at the end of fumigation.

The EDB amounts sorbed by the fruit were less than any other sink (except for the water seal). For oranges they ranged from 8.8 to 12.5% of the total EDB dose resulting in fruit levels of 181 mg/kg, 258 mg/kg, and 233 mg/kg after fumigation at 20, 15, and 10 °C, respectively. Lemons sorbed significantly more EDB than oranges (13.4–17.6%) resulting in levels of 273 mg/kg, 337 mg/kg, and 354 mg/kg at 20, 15, and 10 °C, respectively. A greater EDB uptake by lemons than by Valencia oranges following fumigation has been reported (Sinclair et al., 1962).

After fumigation at each EDB level/temperature combination the final EDB levels in the fruit were similar. This similarity is explained by the empirical derivation of these dose/temperature combinations. The EDB levels used at each temperature were found to be the minimum necessary to assure a probit 9 level of mortality of Queensland Fruit Fly (Gellatley et al., 1978). The similarity of EDB levels in fruit after fumigation at each temperature are then, both an indicator of the potential usefulness of bioassays (in this instance mortality of unsuspecting fruit flies) and of the accuracy of the original fumigation work with this insect.

ACKNOWLEDGMENT

I thank P. J. Nicholls and M. O'Connor from the Biometrical Branch, NSW Department of Agriculture, Haymarket, Sydney, for assistance with the statistical analysis in this paper.

Registry No. EDB, 106-93-4.

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Received for review September 24, 1984. Revised manuscript received February 19, 1985. Accepted June 12, 1985.