Association of Ethylene Dibromide (EDB) with Mature Cranberry (*Vaccinium macrocarpon*) Fruit

Kang Xia* and Charles W. Rice

Department of Agronomy, 2004 Throckmorton Plant Sciences Center, Kansas State University, Manhattan, Kansas 66506

Ethylene dibromide (EDB), a potential carcinogen, has been used in gasoline mixtures to avoid the accumulation of metallic lead in engines. Ethylene dibromide is present in the environment and in groundwater. Previous analysis has shown that EDB levels have reached up to 16 μg L^{-1} in the groundwater at two fuel spill plumes in the vicinity of the Massachusetts Military Reservation (MMR) Base and up to 1.69 μg L^{-1} in the Coonamessett and Quashnet Rivers in Cape Cod, MA (U.S. Air Force IRP, Fact Sheet #98-10, 1998). Groundwater and river water from this area are used to flood some local cranberry bogs for irrigation and harvesting of cranberry fruits. The potential sorption of EDB by cranberry fruits during harvest has caused concern but information regarding its occurrence is not available. In this study, low levels of EDB (0.04−0.15 μg kg^{-1}) were found to be associated with cranberry fruits that were exposed to EDB at levels ranging from 3 to 12 μg L^{-1} at 10, 20, and 30 °C for up to 7 days. Rinsing EDB-exposed cranberry fruits twice with deionized water or once with 0.01 M NaCl solution reduced the amount of EDB associated with the cranberry fruits by 65−72% to a level of 0.02 μg kg^{-1}. Therefore, the EDB most likely is associated with the water residue on the surface of the cranberry fruit rather than being absorbed into the flesh of the fruit during the EDB exposure.

Keywords: *EDB; cranberry fruit; absorption; cuticle layer*

INTRODUCTION

Ethylene dibromide is a volatile, slightly water-soluble, biodegradable, and weakly sorbing brominated organic compound. Ethylene dibromide has been produced in the United States since the 1920s. It was used widely as a fumigant on more than 40 crops and as a lead scavenger in gasoline until 1983 (*1*). By early 1984, all registered agricultural uses of EDB were phased out as a result of the determination by the Environmental Protection Agency (EPA) that they presented an "imminent hazard" to the health of humans.

Once introduced into the environment through application on crops and contamination by EDB-containing gasoline, EDB can persist in soils and groundwater for up to 20 years (*1*, *2*−*5*). This persistence is largely due to sorption interactions of EDB with soil constituents, hydrodynamic dispersion, and abiotic and biotic transformations. Once dissolved in groundwater, EDB is transported by the bulk motion of flowing water. By late 1983, trace amounts of EDB were detected in groundwater in Florida, Georgia, California, South Carolina, New York, Wisconsin, Washington, Massachusetts, Connecticut, and Hawaii (*6*). Concentrations in an unconfined aquifer sampled at several sites in Whatcom County, Washington ranged up to 6.2 μg L^{-1} (*7*). In 1986, concentrations ranging from 0.02 to about 600 μg L^{-1} EDB were detected in well-water samples taken by the State of Florida's EDB monitoring program (*1*). In 1998, field sampling and analyses detected levels of EDB up to 16 μg L^{-1} in the groundwater at two fuel spill plumes (designated FS-1 and FS-28) in the vicinity of the Massachusetts Military Reservation (MMR) Base and up to 1.69 μg L^{-1} in the surface water in the Coonamessett and Quashnet Rivers in Cape Cod, MA (*8*). Groundwater and river water from this area are used for irrigating and harvesting cranberry fruits.

In Massachusetts, cranberries (*Vaccinium macrocarpon*) are harvested by two methods: wet-pick and dry-pick. The wet-pick method is used for 90% of all cranberries harvested and involves flooding the bogs with surface water (*9*). In the wet-harvest processes, a wide-tired machine called a water reel or an "egg beater" moves through the bog without crushing the vines. The water reel beats the water as it moves, agitating the berries off the vines and allowing them to rise to the surface. The floating berries are corralled with a floating boom and lifted into trucks by a conveyer. The cranberries harvested in this manner usually are used in products such as sauce, juice, jams, and jellies.

Transmission electron micrographs show that the fruit cuticle of one cranberry variety (*Vaccinium macrocarpon* var. Searles) is about 10−13 μm thick and full of embedded waxes which increase as the fruit progresses from the white to the blush stage of maturity (*10*). No pores or channels were found in the cuticle layer. The cuticle layer was devoid of microfibrils, suggesting the absence of hydrophilic paths. Thirteen organic compounds including *n*-paraffins, *n*-aldehydes, *n*-alcohols, *n*-fatty acids, sterol alcohols, pentacyclic tritepene alcohols, and acetates were derived from the cuticle wax of cranberry (*Vaccinium macrocarpon*, var. Howes) by Croteau and Fagerson (*11*). The skin of the cranberry fruit is not completely permeable to O_2, CO_2, ethylene (*12*), and ethephon (*13*). However, enhanced diffusion of ethephon across the cranberry fruit cuticle in the presence of ethanol was observed by Farag et al. (*14*).

* Corresponding author (E-mail: kxia@ksu.edu).

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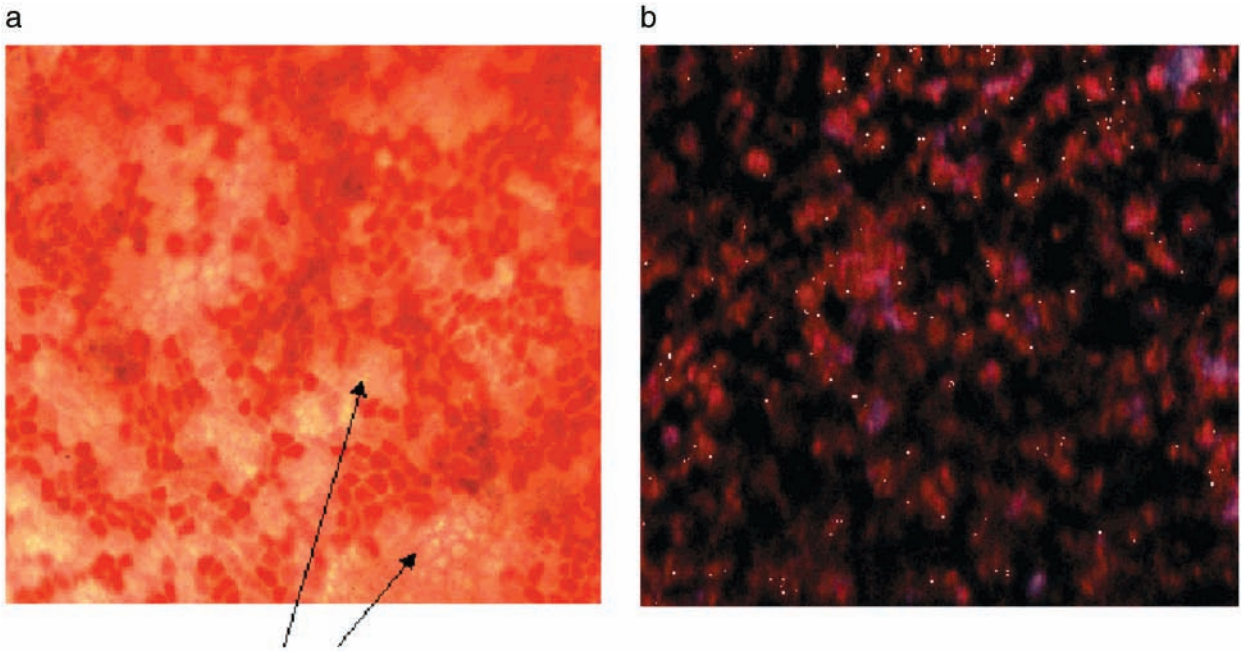

Figure 1. Microscopic image of the surface of (a) surface injured and (b) noninjured cranberry fruits. Arrows indicate injured areas.

The presence of ethanol may increase the surface binding of ethephon to the cuticle outer surface and thus, increase the partitioning of ethephon into the cranberry fruit cuticle (*15*).

The discovery of EDB at levels up to 1.69 μg L^{-1} in the surface water supply on some Massachusetts cranberry farms has affected the local cranberry industry because of the concern of potential EDB contamination during cranberry harvest (*16*). Currently, there is no regulation on the maximum contamination level (MCL) for EDB in cranberry fruits. The federal EPA has set MCLs of 0.05 μg L^{-1} in drinking water, 30 μg kg^{-1} in ready-to-eat cereal products, 150 μg kg^{-1} in foods that need to be cooked before eating, and 900 μg kg^{-1} in raw grains (*17*). No literature on the association of EDB with cranberry fruit has been found. The objective of this project was to investigate the possible association of EDB with mature cranberry fruits that are exposed to EDB in water.

METHODS AND MATERIALS

Sample Collection. Disease-free and EDB-free ripe cranberry fruits (*Vaccinium macrocarpon*, var. Early Black) were collected from seven different locations in a local EDB-uncontaminated bog in Cape Cod, MA during the cranberry harvest season in October 1998. Each cranberry fruit was hand-collected directly from the cranberry vines with an appropriate EDB-free sampling apparatus. The cranberry fruits were stored at 4 °C in 500-mL glass jars for 2 days before they were exposed to water containing different amounts of EDB.

Exposure of Cranberry Fruits to EDB. To investigate the amount of EDB associated with cranberry fruits, cranberry fruits were exposed to water containing different concentrations of EDB (0, 3, 6, and 12 μg L^{-1}) at different temperature conditions (10, 20, and 30 °C) for various exposure periods (1, 2, 3, and 7 days). Because damage of the cuticle layer during harvest might affect the amount of EDB associated with the fruit, surface-injured cranberry fruits were exposed to EDB at the same conditions used for the non-surface-injured fruits.

To injure the cranberry fruit surface, 10 fruits were shaken continuously in a 60-mL glass (Qorpak) bottle without water on an end-to-end shaker at low speed for 2, 4, 12, and 24 h. The appropriate time was determined by observing the changes on the cranberry cuticle under a microscope at the end of each shaking period. A 4-h period was determined to be optimum because of the significant changes on the cranberry fruit surface (Figure 1), although the fruits are not subjected to this duration or level of agitation in the wet harvesting processes. After being shaken for 12 h, the cranberry fruits were damaged severely and had noticeable juice leakage from the fruit.

Ten weighed, surface-injured or noninjured, cranberry fruits were immersed in a 60-mL glass (Qorpak) bottle with an airtight closed lid containing 50 mL of water without EDB or EDB at concentrations of 3, 6, or 12 μg L^{-1} for 1, 2, 3, or 7 days at 10, 20, or 30 °C. Each treatment was replicated four times. The pH and the ionic strength of the solution were adjusted to 6.55 and 0.0012 M, respectively, to match those of the EDB-contaminated surface water samples at Cape Cod, MA . Pure analytical-grade EDB (AccuStandard Inc., New Haven, CT) was used.

As soon as each replication was completed, the cranberry fruits were separated from the EDB-contaminated water. The water drops attached on the fruit surface were absorbed gently with EDB-free, soft tissue paper. The cranberry fruits then were transferred into another bottle and stored in a freezer at −20 °C for about a week until EDB extraction.

To determine whether EDB is associated with cranberry fruits on the surface or in the flesh, separate sets of cranberry fruits that were exposed to 6 μg L^{-1} EDB at 20 °C for 1, 3, and 7 days were rinsed with 50 mL of deionized water, 0.01 M NaCl solution, or hexane.

Extraction of EDB from Cranberry Fruits. The EDB-exposed frozen cranberries (10 berries each replication) were dipped into liquid nitrogen for 2 min right before they were homogenized in a food processor. The processor was decontaminated first with tap water and detergent, then with methanol, and finally with hexane (GC grade, ACROS, Pittsburgh, PA) between samples and at the beginning and end of each day. After each hexane rinse, the food processor was dried in an oven at 100 °C for 10 min in order to remove the hexane residue.

Immediately after the homogenization, bromodichloromethane (BDCM) (GC grade, Aldrich, Milwaukee, WI) was added to each homogenized sample as a surrogate. Ethylene dibromide and the surrogate then were extracted by mixing each homogenized sample with 30 mL of deionized water/acetone

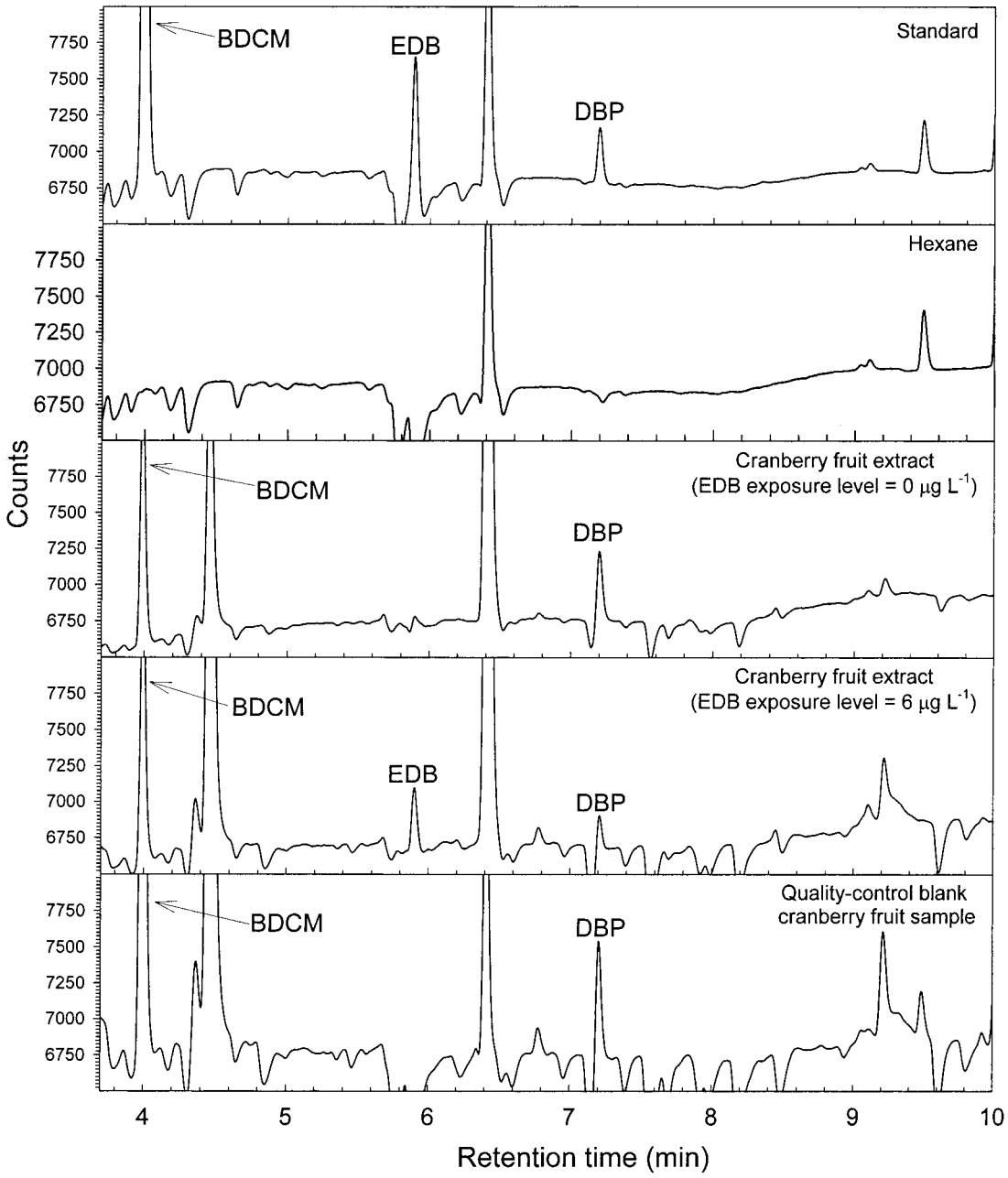

Figure 2. Representative GC−ECD chromatograms for EDB standard; hexane (used as extract for EDB); two extracts of EDB-exposed cranberry fruits (0 μg L^{-1} and 6 μg L^{-1} EDB exposure levels); and a quality control blank cranberry fruit sample. The surrogate, bromodichloromethane (BDCM), has a retention time of 4 min. Ethylene dibromide (EDB) appears at approximately 5.85 min. The internal standard, 1,2-dibromopropane (DBP), appears at approximately 7.20 min. The unidentified peak that has retention time of 4.5 min is from an unknown indigenous compound in cranberry fruit. The unidentified peak that appears at 6.4 min is the hexane solvent peak.

solution (1:1) and 2 mL of hexane (GC grade, ACROS). The mixture was shaken on an end-to-end shaker for 2 min before the hexane layer was transferred to a 2-mL gas chromatography (GC) sample vial (Agilent, Palo Alto, CA). All vials were sealed and were kept upside down at −20 °C in a freezer for about a week until the EDB analysis. Immediately prior to the EDB analysis using GC−ECD, 100 μL of the hexane layer that was stored in the 2-mL GC sample vial was transferred into a 100-μL polypropylene crimp/snap GC sample vial (Agilent, Palo Alto, CA). The sample was spiked with 1,2-dibromopropane (DBP) (GC grade, Aldrich,) as an internal standard. Analysis of EDB was conducted using EPA method 504.1 (*18*).

EDB Measurement using GC−ECD. Quantitation of EDB in cranberry fruit extracts was accomplished using an HP 5890 GC (Agilent, Palo Alto, CA) with a ^{63}Ni electron capture detector (ECD). The column was DB-1 (30 m × 0.32 mm i.d., 0.25-μm film thickness, 100% dimethylpolysiloxane capillary column, J&W Scientific, Folsom, CA). Following is the program for the GC oven: initial temperature, 40 °C for 4 min; ramped at 9 °C/min to 160 °C and then at 35 °C/min to 270 °C; held at a final temperature of 270 °C for 5 min. The injection port was kept at 200 °C. The ECD was operated at 290 °C. A 2-μL splitless injection was used. Helium was used as carrier gas at a flow rate of 25−30 cm sec^{-1}. The makeup gas consisted of 95% Ar and 5% methane and was operated at a flow rate of 58−60 mL min^{-1}. The column head pressure was maintained at 4.83×10^4 pa. All data were collected and processed using HP ChemStation95.

All stock standards were purchased as 100 μg mL^{-1} in methanol. The stock solution was diluted in hexane (GC grade, ACROS) to prepare the calibration solutions: 0.05, 0.15, 0.25, 0.5, and 1.0 μg L^{-1}. The limit of quantitation (LOQ) was 0.03 μg L^{-1} for solution and 0.006 μg kg^{-1} for cranberry fruits. The EDB recovery rate was 100 ± 10%. The surrogate recovery

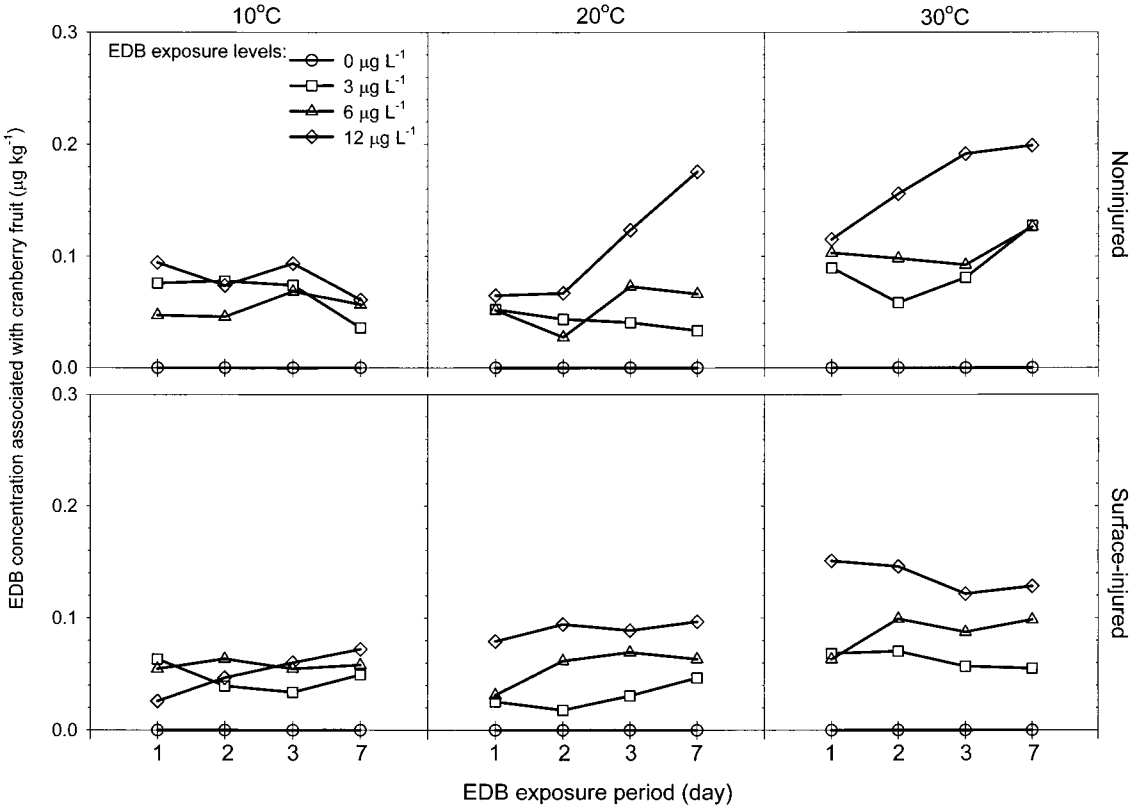

Figure 3. Average concentrations of EDB associated with surface injured and noninjured cranberry fruits exposed to four levels of EDB in water at three temperatures for four exposure periods.

Table 1. Analysis of Variance (ANOVA) Test on the Association of EDB with Surface Injured and Noninjured Cranberry Fruits Exposed to Four Levels of EDB in Water at Three Temperatures for Four Exposure Periods

source	DF	type III SS	mean square	*F* value	P
INJURE[a]	1	0.02484835	0.02484835	22.97	0.0001
TEMP[b]	2	0.13874547	0.06937274	64.12	0.0001
EDB[c]	3	0.55225185	0.18408395	170.15	0.0001
DAY[d]	3	0.00759586	0.00253195	2.34	0.0736
INJURE*TEMP*EDB*DAY[e]	18	0.03439844	0.00191102	1.77	0.0289
TEMP*EDB*DAY[f]	18	0.02204187	0.00122455	1.13	0.3203
INJURE*TEMP*EDB[f]	6	0.01075172	0.00179195	1.66	0.1317
INJURE*EDB*DAY[f]	9	0.0290006	0.00322229	2.98	0.0021
INJURE*TEMP*DAY[f]	6	0.01365038	0.00227506	2.1	0.053
TEMP*EDB[g]	6	0.0776964	0.0129494	11.97	0.0001
TEMP*DAY[g]	6	0.01939007	0.00323168	2.99	0.0076
INJURE*TEMP[g]	2	0.00917925	0.00458962	4.24	0.0153
EDB*DAY[g]	9	0.02590265	0.00287807	2.66	0.0056
INJURE*EDB[g]	3	0.01984356	0.00661452	6.11	0.0005
INJURE*DAY[g]	3	0.00096301	0.000321	0.3	0.8278

[a] Fruit surface condition (noninjured and injured). [b] Temperature (10, 20, and 30 °C). [c] EDB exposure level (0, 3, 6, and 12 μg L^{-1}). [d] Exposure period (1, 2, 3, and 7 days). [e] Four-way interactions. [f] Three-way interactions. [g] Two-way interactions.

rate was 85 ± 10%. Figure 2 shows the GC−ECD chromatograms for hexane, EDB standard, extracts of EDB-exposed cranberry fruits, and quality-control blank cranberry fruit sample.

The experiment follows 2 × 4 × 4 × 3 random block design. Analysis of variance (ANOVA) was conducted using SAS program PROC GLM (*19*, *20*). A *P* value less than 0.01 indicated a statistically significant association of EDB with cranberry fruit.

RESULTS AND DISCUSSION

Association of EDB with Cranberry Fruits. The average concentrations of EDB in cranberry fruits with the various treatments are shown in Figure 3. No EDB was associated with the cranberry fruits exposed to water, but it was associated with cranberry fruits exposed to solutions containing 3, 6, and 12 μg L^{-1} EDB. The higher the EDB level to which the cranberry fruits were exposed, the more EDB was associated with the fruits. Injury of cranberry fruit surface resulted in less EDB associated with the fruits. Results shown in Figure 3 also suggest that increasing temperature and exposure period to EDB increased the amount of EDB associated with cranberry fruits at high EDB exposure levels. Analysis of variance (Table 1) suggested significant ($P < 0.01$) main effects of EDB exposure level, temperature, and injury on the association of EDB with cranberry fruits. The main effect of exposure period was not statistically significant. Certain three-way and two-way interactions were statistically significant as indicated by the *P* values in Table 1.

The amount of EDB associated with the fruits was affected significantly by the two-way interactions be-

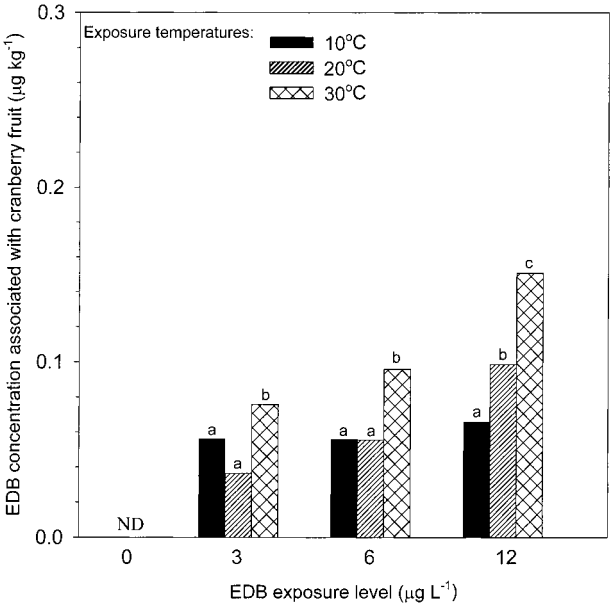

Figure 4. Effects of the two-way interactions between temperature (TEMP) and EDB exposure level (EDB) on the association of EDB with cranberry fruits. Different letters between bars of a given treatment indicate significance at $P < 0.01$. ND means nondetectable amount of EDB.

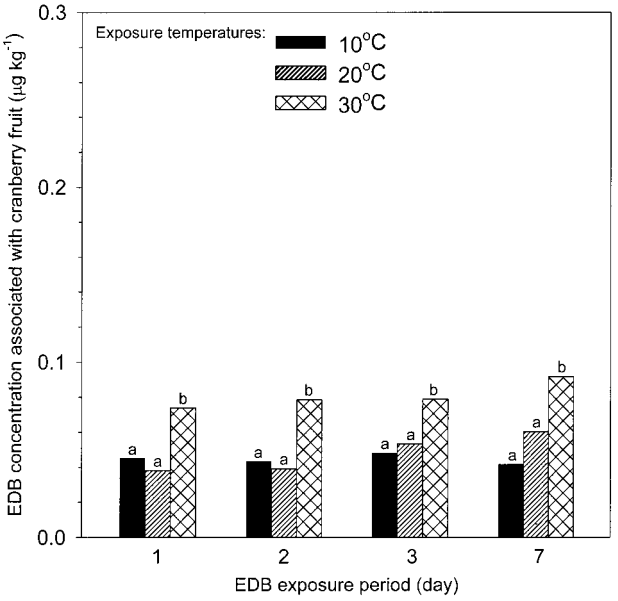

Figure 5. Effects of the two-way interactions between temperature (TEMP) and exposure period (DAY) on the association of EDB with cranberry fruits. Different letters between bars of a given treatment indicate significance at $P < 0.01$.

tween temperature and EDB exposure level and between temperature and exposure period. The amount of EDB associated with the fruits also was affected significantly by the two-way interactions between EDB exposure level and exposure period and between exposure level and fruit surface condition. Two-way interactions between fruit surface condition and temperature and between fruit surface condition and exposure period did not significantly affect the amount of EDB associated with EDB-exposed cranberry fruits. The three-way interactions among fruit surface condition, EDB exposure level, and exposure period had a statistically significant effect on the level of EDB associated with cranberry fruits. The four-way interactions among all experimental factors did not significantly affect the amount of EDB associated with cranberry. Because statistically significant two-way interactions and three-way interactions occurred (Table 1), it is important to discuss their relative importance compared to the main effects.

The effects of the two-way interactions on the association of EDB with cranberry fruits are shown in Figures 4, 5, 6, and 7. At 10 °C, the average EDB concentration associated with cranberry fruits was 0.07 μg kg^{-1}. At 20 °C and 30 °C, the amount of EDB that was associated with cranberry fruit was significantly higher for the fruits exposed to a solution containing 12 μg L^{-1} EDB than those exposed to solutions containing 3 μg L^{-1} and 6 μg L^{-1} EDB. With exposure to 12 μg L^{-1} EDB, the average concentration of EDB associated with cranberry fruits increased from 0.07 μg kg^{-1} to 0.15 μg kg^{-1} when the temperature was increased from 10 °C to 30 °C (Figure 4). The exposure period interactions with temperature affected the amount of EDB associated with the fruit (Figure 5). At 30 °C, significantly more EDB was associated with cranberry fruits than at the two lower temperatures, no matter how long the fruits were exposed to EDB. Significantly more EDB was found to be associated with cranberry fruits exposed to solution containing 12 μg L^{-1} EDB than those that were exposed to 3 μg L^{-1} and 6 μg L^{-1} EDB (Figure 6). At this EDB

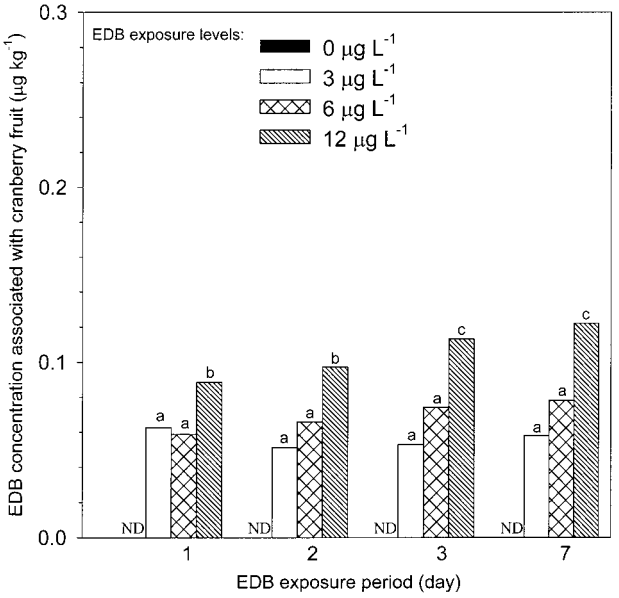

Figure 6. Effects of the two-way interactions between EDB exposure level (EDB) and exposure period (DAY) on the association of EDB with cranberry fruits. Different letters between bars of a given treatment indicate significance at $P < 0.01$. ND means nondetectable amount of EDB.

exposure level, longer exposure periods increased the amount of EDB associated with the cranberry fruits. Longer exposure periods increased the amount of EDB associated with surface-noninjured cranberries exposed to 12 μg L^{-1} at 20 °C and 30 °C (Figure 3). Although the main effect of exposure period was not statistically significant (Table 1), its interactions with temperature and exposure level were important. The temperature interaction effect on the amount of EDB associated with cranberry fruit might be expected, because increasing temperature might increase sorption and also might affect characteristics of the fruit surface. However, a water temperature of 20 or 30 °C would not be expected at the time of cranberry harvest; a temperature of 10 °C would be more likely. At these temperatures, the

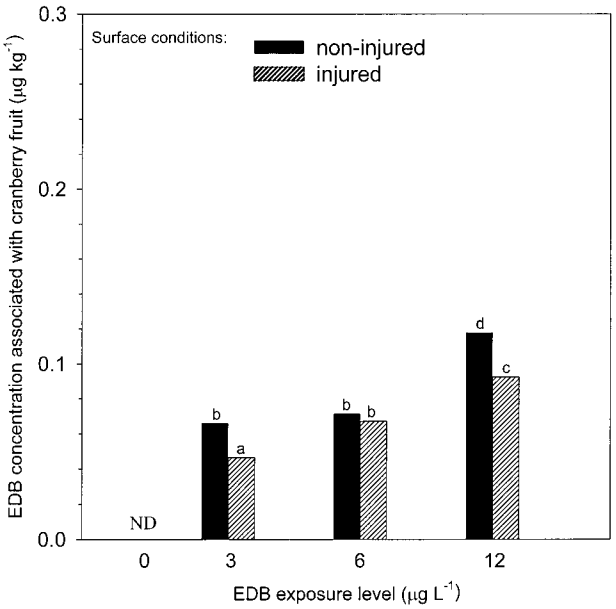

Figure 7. Effects of the two-way interactions between cranberry fruit surface condition (INJURY) and the EDB exposure level (EDB) on the association of EDB with cranberry fruit. Different letters between bars of a given treatment indicate significance at $P < 0.01$. ND means nondetectable amount of EDB.

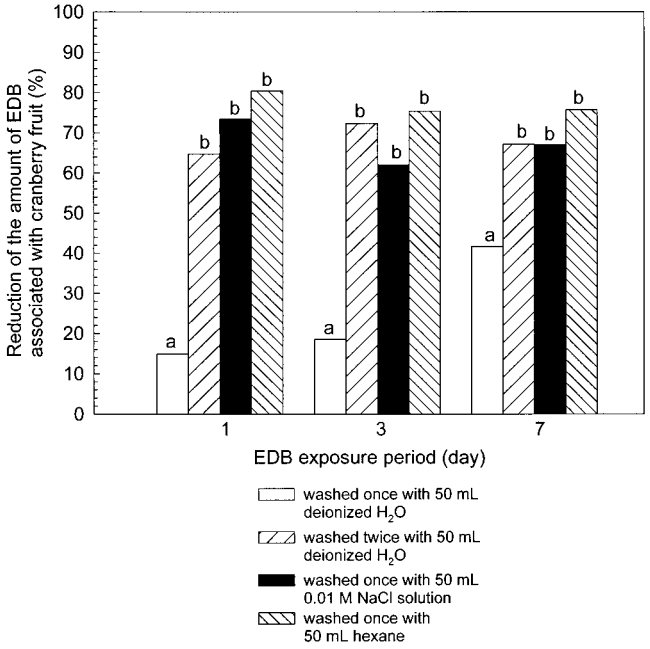

Figure 8. Reductions in the amount of EDB associated with cranberry fruits after the EDB-exposed fruits were washed with deionized water (once and twice), 0.01 M NaCl solution, and hexane. The cranberry fruits were exposed to 6 μg L^{-1} EDB at 20 °C for 1, 3, and 7 days before the washing. Different letters between bars of a given treatment indicate significance at $P < 0.01$.

average EDB concentration associated with the cranberry fruits was approximately 0.05 μg kg^{-1} when the fruits were exposed to 3 μg L^{-1} and 6 μg L^{-1} EDB for all four exposure periods (Figures 4 and 5).

Injury to the cranberry fruit surface resulted in less sorption of EDB, especially when the fruits were exposed to 12 μg L^{-1} EDB (Figure 7). The interaction of injury and EDB exposure level was significant ($P < 0.0005$) (Table 1 and Figure 7). Apparently, damage to the cuticle waxy layer on the fruit surface (Figure 1) minimized the association of EDB with the fruit. Damage to the cuticle layer may reduce the amount of cuticle wax containing compounds that may bind with EDB or a thin water layer that contains EDB. This result strongly suggested that the association of EDB with cranberry fruits most likely occurred on the cuticle waxy layer of cranberry fruits.

The Effect of Rinsing on the Association of EDB with Cranberry Fruits. A significant reduction occurred in the concentration of EDB associated with the cranberry fruits when they were washed with different solutions after exposure to 6 μg L^{-1} EDB at 20 °C (Figure 8). About 15−42% of EDB associated with the cranberry fruits was rinsed off the fruits after one washing with 50 mL of deionized water. When the fruits were rinsed twice with 50 mL of deionized water each time, the amount of EDB that was associated with cranberry fruits was reduced by 65−72%. A similar range of reduction rates (62−73%) also was observed when the fruits were rinsed once with 50 mL of 0.01 M NaCl solution. Rinsing the EDB-exposed cranberry fruits once with 50 mL of hexane reduced the amount of EDB associated with the fruits by as much as 75−80% (to about 0.014 μg kg^{-1}).

The significant reductions in the amount of EDB associated with cranberry fruits by rinsing them with deionized water, saltwater, and organic solvent suggest that EDB was sorbed on the surface instead of being absorbed by the flesh of the fruit. Rinsing the EDB-exposed cranberry fruits twice with deionized water reduced EDB associated with the fruits by the same amount as rinsing once with saltwater. The similar reductions from rinsing the EDB-exposed cranberry fruits with hydrophilic solvent (saltwater) and hydrophobic solvent (hexane) suggest that the EDB is more likely to be associated with the water residues on the cranberry fruit surface than to be bound with the organic compounds in the cuticle wax layer. Ethylene dibromide is a nonpolar organic compound. It is much more soluble in hexane than in water because of its hydrophobic characteristic. If the EDB molecules were bound with the organic compounds in the cuticle wax layer, rinsing the EDB-exposed fruits with hexane should have desorbed significantly more EDB molecules than rinsing the fruits with water or saltwater. Croteau and Fagerson (*11*) found that cuticle wax of cranberry contained organic compounds such as *n*-paraffins, *n*-aldehydes, *n*-alcohols, *n*-fatty acids, sterol alcohols, pentacyclic tritepene alcohols, and acetates. Water molecules can be sorbed on the cuticle wax of cranberry fruits through hydrogen bonding with C═O, COOH, and OH functional groups in those compounds and, therefore, form a water layer on the fruit surface.

Although EDB is a nonpolar organic compound, it is slightly soluble in water. The water solubility of EDB is 3370 mg L^{-1} at 20 °C (*21*). The maximum EDB concentration used in our experiment was 12 μg L^{-1}. In our study, low levels of EDB ranging from 0.04 to 0.15 μg kg^{-1} were found to be associated with cranberry fruits. One cranberry fruit weighs approximately 1 g. At maximum, about 1.5×10^{-4} μg of EDB was associated with one cranberry fruit. It was found in our experiment that one cranberry fruit could sorb about 0.01 g of water on its surface. This gives an EDB concentration of 1.5×10^{-2} mg L^{-1}. It is highly possible for EDB to be present and to accumulate in the water

phase on cranberry fruits for up to 7 days when the fruits are immersed in EDB-contaminated water.

In summary, the amount of EDB associated with cranberry fruits was affected by the interactions of EDB exposure level, exposure period, and temperature. At water temperatures of 10−20 °C, which would be expected during cranberry harvest season, the amount of EDB associated with the fruit at EDB exposure levels up to 6 μg L^{-1} or exposure periods up to 7 days did not change significantly. The lowest average concentration associated with unwashed fruit was 0.04 μg kg^{-1} when exposed to 3 μg L^{-1} EDB in water; the highest was 0.15 μg kg^{-1} when exposed to 12 μg L^{-1} EDB in water. Injury to the surface of the cranberry fruit, as simulated by shaking, resulted in less EDB associated with the fruit. Apparently, damage to the cuticle layer may reduce the surface binding of water residue containing EDB and, therefore, reduce the amount of EDB associated with cranberry fruits. Results of the rinsing experiment suggest that the EDB was most likely to be associated with the water residue on the surface of the cranberry fruit that had been exposed to EDB-contaminated water. Water rinsing of EDB-exposed cranberry fruits can reduce the amount of EDB associated with the fruits by 65−72% (to 0.02 μg kg^{-1}).

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