# Management of Fungicide Residues on Processing Tomatoes

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**ABSTRACT**

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In 1988 and 1989, residues of ethylenebisdithiocarbamate, chlorothalonil, and anilazine on raw, unwashed, unpeeled processing tomatoes from field experiments were 16–25% of the Environmental Protection Agency tolerance levels. Decreasing fungicide rates early in the season resulted in an additional 33–53% reduction in residues without adverse effects on fruit yield or quality. The total amount of fungicide applied during the growing season, and not just the last applications before harvest, seems to determine residue levels. Ethylene thiourea, chlorothalonil, and anilazine residues were at or below detection limits in tomato juice processed from field-grown tomatoes in both years of the study.

Additional keyword: anthracnose

Anthracnose, caused by *Colletotrichum coccodes* (Wallr.) S. J. Hughes, is an important disease of processing tomatoes in the Midwest (9). Weather conditions such as high humidity, high rainfall, and warm temperatures in the midwestern states favor development of this disease. Wilson and Runnels (10) found 43, 22, 70, and 44% of tomato fruit infected with anthracnose in field tests during 1944–1947. In field tests in 1986, anthracnose reduced yields of processing tomatoes by 69% (8).

Anthracnose lesions on fruit are a primary factor in determining the use and price of tomatoes at the processing plants. One or two lesions per fruit make the individual fruit unusable. As little as 1% infected fruit can decrease the price paid by processors to the grower. Four percent diseased fruit leads to a significant loss in price per ton. Spray programs using mancozeb, chlorothalonil, and anilazine or combinations of these fungicides are the basis for control of anthracnose. At current prices, production of profitable crops of processing tomatoes in the Midwest without these fungicides is impossible in most years.

In 1987, a National Academy of Sciences (NAS) report (3) attributed nearly 60% of the daily dietary oncogenic risk to fungicides. Tomatoes and tomato products contributed almost 15% of the total dietary oncogenic risk estimate (3). Risk estimates in the NAS report were calculated using a model that assumes fungicide residues were present on food or the crop at the published tolerance level and that 100% of the crop hectarage was treated with fungicide (3). Although this method almost certainly overestimated the actual dietary exposure, the Environmental Protection Agency (EPA) assumes this procedure introduces a prudent safety factor into its overall assessment of risk (3). One further assumption made in the NAS report was that residues on raw tomatoes are concentrated when food is processed by drying or removing water (3).

How calculated residue levels in food relate to those actually occurring in raw and processed food products from crops treated with fungicides incorporated into practical grower disease management programs under field conditions is largely unknown. Chaisson and Peterson (4) suggested that the assumed oncogenic risk from fungicides on California tomatoes could not be determined because only one of five fungicides assumed by NAS to be used on tomatoes was used in California and then on only 19% of the total hectarage.

The control of many fungal diseases on tomatoes in the Midwest is much different from that in California. High humidity, high rainfall, and warm summer temperatures necessitate increased dependence on fungicides to control the development of anthracnose and foliar diseases on processing tomatoes. In most seasons with present cultivars, these diseases would destroy the crop. Because growers must have 99% control of anthracnose to obtain top price for their crop, sprays containing mancozeb, chlorothalonil, or combinations of these fungicides are applied to nearly 100% of the hectarage in the Midwest. Twelve or more sprays may be required in a wet season. Rates of weathering and breakdown of pesticides on tomato foliage may differ in California and the Midwest.

The objective of this study was to determine how the selection of a disease control program and the rate of fungicide application affect fungicide residue accumulation on raw (tomato fruit) and processed products (tomato juice).

## MATERIALS AND METHODS

The effect of fungicide spray programs on residue accumulation was tested in 1988 and 1989 at the Vegetable Crops Branch of the Ohio State University at Fremont on Hoytville silty clay loam (pH 6.3, 4.0% organic matter). Georgia-grown transplants (cv. Heinz 1810) were planted in double rows to simulate grower conditions. Plots were double 9.1-m rows bordered by untreated rows. Row centers were 1.5 m apart with plants 0.3 m apart in the row. The experiment used a randomized complete block design, and each fungicide treatment was replicated six times.

Plots received 0-26-26 (896 kg/ha) plowed down on 3 November 1987, 6-24-24 (1,120 kg/ha) disked in on 14 April 1988, and 34-0-0 (224 kg/ha) on 25 April 1988. In year 2, plots received 0-26-26 (896 kg/ha) plowed down on 2 November 1988, 34-0-0 (224 kg/ha) and 0-0-60 (224 kg/ha) on 14 April 1989. Each plant received at transplant 236 ml of starter solution containing 1 L of 11-34-0 in 194 L of water. Georgia-grown transplants were set 15 May 1988. Devrinol (napropamide, 4.5 kg/ha) and Sencor (metribuzin, 0.6 kg/ha) were used for weed control. Fungicide treatments, listed in Table 1, were applied on a 7- to 10-day schedule beginning 1 wk after transplanting at 561 L/ha and 481 kPa. The fungicides Bravo 720 (chlorothalonil), Dithane 4F (mancozeb), and Dyrene 4F (anilazine) were applied with a R & D (Opelousas, LA) $CO_2$-powered, tractor-mounted sprayer through a one-row boom with five Delavan (Delavan Inc., West Des Moines, IA) hollow cone #8 nozzles.

Rates of fungicides applied are in terms of formulated product. Dyrene 50W is labeled for tomatoes; Dyrene 4F is not labeled. Dyrene 4F was included in the program to maintain uniformity of formulation. Ethephon (1.12 kg/ha)

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was applied on 10 August 1988 and 4 September 1989. Rainfall in May, June, July, and August was 2.3, 1.6, 7.2, and 14.4 cm in 1988 and 14.5, 11.2, 3.5, and 5.8 cm in 1989, respectively. Plots were irrigated (2.5 cm) on 14–15 June 1988. Fruit was machine-harvested on 31 August 1988 and 25 September 1989. Marketable red fruit was used for yield and residue analysis.

**Residue analysis.** In both years, samples for fungicide residues were taken from the five treatments and an unsprayed control listed in Table 1. They were tested for mancozeb on raw fruit in 1988 and for mancozeb, chlorothalonil, and anilazine in 1989 (Table 1). Two to 3 hr after sampling, the fruit samples were divided into two lots. One was frozen at −26 C. Within 5 days, frozen fruit was shipped in dry ice via overnight express service to the National Food Laboratory (NFL) for residue analysis. One day after harvest, the other lot was processed into juice in the The Ohio State University Horticulture Department pilot plant. Raw fruit was placed in a dump tank for 45–50 sec. Dump tank water was changed between fungicide treatments. The fruit then was exposed to a high-pressure water spray for 20–30 sec and then a low-pressure spray for 10 sec before it was put into the chopper. Chopped fruit received a hot break treatment and then was juiced (5). Canned juice went through a steam table before sealing. To achieve commercial sterility, the cans were heat processed at 104.4 C for 20 min (5). Canned samples were shipped to the NFL for residue analysis.

Mancozeb was determined by the method for dithiocarbamates listed in the Pesticide Analytical Manual, volume 2 (1). Ethylene thiourea (ETU), a breakdown product of mancozeb, was determined by the gas chromatographic method (2). Chlorothalonil was determined by the multiresidue procedure in the Pesticide Analytical Manual, volume 1 (7), and anilazine was determined by the high performance liquid chromatography method found in Analytical Methods for Pesticides and Plant Growth Regulators (6). The study included spiked recoveries of each fungicide at the rate of one per every 10 samples analyzed.

## RESULTS

**Disease development.** Anthracnose development on fruit was moderate in 1988 and 1989. Dry weather limited development of foliar diseases each year of the test. Fungicide treatments significantly reduced anthracnose infected fruit in comparison to the control only in 1988 (Table 1).

**Raw fruit residues.** In 1988, 12 sprays of mancozeb (5.6 L/ha) left a residue of 0.98 ppm of ethylenebisdithiocarbamate (EBDC) on unwashed fruit, 25% of the tolerance levels permitted by the EPA. A spray program with rates of mancozeb increasing as the crop matured (2.8 L/ha at transplant, 3.7 L/ha at fruit set, and 5.6 L/ha at pink fruit) left EBDC residues of 0.57 ppm, 42% lower than the season-long Dithane program (Table 2). The control sample had EBDC levels less than 0.20 ppm. In 1989, residues from 10 sprays of mancozeb (5.6 L/ha) were 0.18 ppm or only 5% of the EPA tolerance. Mancozeb applied at increasing rates in 1989 (2.8 L/ha at transplant, 3.7 L/ha at fruit set, and 5.6 L/ha at pink fruit) left a residue of 0.12 ppm on raw, unwashed fruit, a 33% decrease in residues compared with to the mancozeb high rate (5.6 L/ha). This response is similar to 1988 results. Overall, EBDC residue levels in 1989 were much lower than 1988 levels probably because of the fact that there were only 10 sprays vs. 12 in 1988. Also, there was 10 cm more rain in the 1989 growing season.

In 1988, 12 applications of chlorothalonil at 3.5 L/ha resulted in chlorothalonil residues of 0.82 ppm on raw, unwashed fruit, 16% of the EPA tolerance level on raw fruit (Table 2). The application of chlorothalonil at increasing rates as the crop matured (1.8 L/ha at transplant, 2.3 L/ha at fruit set, and 3.5 L/ha at pink fruit) resulted in residues of 0.72 ppm, a 12% reduction compared with the season-long high rate, 3.5 L/ha (Table 2). In 1989, 10 applications of

**Table 1.** Fungicide rates, number of applications, yield of good red fruit, and anthracnose-affected fruit with tomato (cv. H1810) in trials at Fremont, OH

| | | 1988 | | | 1989 | | |
|---|---|---|---|---|---|---|---|
| | | | Fruit yield[b] (kg/plot) | | | Fruit yield (kg/plot) | |
| Treatment | Rate[a] (L/ha) | No. of sprays | Good red | Anthracnose infected | No. of sprays | Good red | Anthracnose infected |
| Control | 0 | 0 | 40.5 | 0.57 | 0 | 24.8 | 0.18 |
| Mancozeb | | | | | | | |
| Dithane 4F | 5.6 | 12 | 46.8 | 0.19 | 10 | 30.7 | 0.22 |
| Dithane 4F | | | | | | | |
| Transplant | 2.8 | 3 | | | 1 | | |
| At fruit set | 3.7 | 5 | | | 5 | | |
| At pink fruit | 5.6 | 4 | 48.1 | 0.19 | 4 | 30.3 | 0.25 |
| Chlorothalonil | | | | | | | |
| Bravo 720 | 3.5 | 12 | 48.5 | 0.13 | 10 | 31.6 | 0.24 |
| Bravo 720 | | | | | | | |
| Transplant | 1.8 | 3 | | | 1 | | |
| At fruit set | 2.3 | 5 | | | 5 | | |
| At pink fruit | 3.5 | 4 | 49.1 | 0.09 | 4 | 31.5 | 0.23 |
| Anilazine | | | | | | | |
| Dyrene 4F | 11.7 | 12 | 47.4 | 0.03 | 10 | 37.6 | 0.05 |
| LSD ($P = 0.05$) | | | 10.8 | 0.27 | | 7.3 | 0.31 |

[a] Rates of fungicides applied are in terms of formulated product.
[b] 9.1 m of double rows. Row centers 1.5 m apart, plants 0.3 m in row.

**Table 2.** The effect of rates of application of mancozeb, anilazine, and chlorothalonil on residue accumulation in raw, unwashed, unpeeled, tomato fruit in 1988 and 1989

| | Fungicide residues (ppm)[a] | |
|---|---|---|
| Treatment | 1988 | 1989 |
| Ethylenebisdithiocarbamate (EDBC)[b] | | |
| Check | <0.20 | 0.120 |
| Dithane 4F (5.6 L/ha)[c] | 0.98 | 0.182 |
| Dithane 4F[d] | 0.57 | 0.120 |
| HSD 0.05[e] | 0.36 | 0.879 |
| Anilazine[f] | | |
| Check | ... | <0.100 |
| Dyrene 4F (11.7 L/ha) | 2.16 | 0.847 |
| HSD 0.05 | 0.54 | 0.20 |
| Chlorothalonil[g] | | |
| Check | ... | 0.045 |
| Bravo 720 (3.5 L/ha) | 0.82 | 0.952 |
| Bravo 720[h] | 0.72 | 0.452 |
| HSD 0.05 | 0.54 | 0.20 |

[a] EPA tolerance levels: chlorothalonil = 5 ppm; anilazine = 10 ppm; EBDC = 4 ppm.
[b] Detection limit = 0.20 ppm.
[c] Rates of fungicides applied are in terms of formulated product.
[d] 2.8 L/ha at transplant, 3.7 L/ha at fruit set, and 5.6 L/ha at pink fruit.
[e] Tukey's *w*-procedure; HSD = honestly significant difference.
[f] Detection limit = 0.10 ppm.
[g] Detection limit = 0.01 ppm.
[h] 1.8 L/ha at transplant, 2.3 L/ha at fruit set, and 3.5 L/ha at pink fruit.

**Table 3.** The effect of rates of application of mancozeb, anilazine, and chlorothalonil on residue accumulation in tomato juice in 1988 and 1989

| Treatment | Fungicide residues (ppm) 1988 | 1989 |
|---|---|---|
| Ethylene thiourea[a] | | |
| Check | ... | <0.010 |
| Dithane 4F (5.6 L/ha)[b] | 0.01 | <0.010 |
| Dithane 4F[c] | 0.02 | <0.010 |
| Anilazine[d] | | |
| Check | ... | <0.100 |
| Dyrene 4F (11.7 L/ha) | ND[e] | <0.100 |
| Chlorothalonil[f] | | |
| Check | ... | <0.030 |
| Bravo 720 (3.5 L/ha) | ND | <0.030 |
| Bravo 720[g] | ND | <0.030 |

[a] Detection limit = 0.01 ppm; no variance in results from any replication.
[b] Rates of fungicides applied are in terms of formulated product.
[c] 2.8 L/ha at transplant, 3.7 L/ha at fruit set, and 5.6 L/ha at pink fruit.
[d] Detection limit = 0.10 ppm; no variance in results from any replication.
[e] Not detectable.
[f] Detection limit = 0.01 ppm; no variance in results from any replication.
[g] 1.8 L/ha at transplant, 2.3 L/ha at fruit set, and 3.7 L/ha at pink fruit.

chlorothalonil at 3.5 L/ha gave residues of 0.95 ppm, only 20% of the EPA tolerance, similar to the levels seen in 1988. Chlorothalonil applied at increasing rates (1.8 L/ha at transplant, 2.3 L/ha at fruit set, and 3.5 L/ha at pink fruit) resulted in residue levels of 0.45 ppm, 53% less than the season-long high Bravo rate. As in 1988, chlorothalonil applied at increasing rates reduced residue levels on fruit. A control sample was tested in 1989 and was less than 0.05 ppm.

Twelve sprays of anilazine (11.7 L/ha) in 1988 and 10 sprays in 1989 left residues 20% (2.16 ppm) and 9% (0.85 ppm) of the EPA tolerance levels (Table 2). A check sample was run only in 1989, and residues were less than 0.1 ppm, the detection limit.

**Tomato juice residues.** Tomato juice from fruit receiving 12 sprays of mancozeb (5.6 L/ha) contained 0.01 ppm of ETU. Juice from fruit receiving mancozeb at increasing rates (2.8 L/ha at transplant, 3.7 L/ha at fruit set, and 5.6 L/ha at pink fruit) in 1988 contained 0.02 ppm of ETU (Table 3). The detection limit for ETU was less than 0.01 ppm. The juice from check tomatoes was not tested for ETU.

Residues of chlorothalonil or anilazine were not detectable in tomato juice in 1988 (Table 3). The detection limit for these fungicides was 0.01 and 0.1 ppm, respectively. Residues in tomato juice in 1989, as in 1988, were at or less than the detection level (Table 3). ETU levels from fruit treated with mancozeb were less than 0.01 ppm. Fruit treated with anilazine and chlorothalonil had residue levels less than 0.1 and 0.03 ppm in juice, respectively.

## DISCUSSION

In 1988 and 1989, residues of EBDC, chlorothalonil, and anilazine on raw, unwashed, unpeeled processing tomatoes from field experiments were 16–25% of the EPA tolerance levels. Our results indicate that total fungicide applied per growing season, rather than just the last fungicide sprays before harvest, determines the residue on raw tomatoes. For both mancozeb and chlorothalonil, the season-long high rate resulted in higher residue levels than levels found with increasing fungicide rates as the season progressed. The only difference between these two programs is that lower rates are used in the beginning of the season based on the stage of plant growth. It is this early fungicide rate reduction that results in significantly lower residue levels. The level of EBDC residue reduction was 41 and 33% for 1988 and 1989, respectively. Chlorothalonil residue reduction was 53% in 1989 and 12% in 1988. This spray program would be the most desirable because it lowers residue levels and does not affect tomato fruit quality or yield.

NAS estimates that residues would increase in tomato puree or paste. The NAS report states, "If one assumes that all acres are treated, that residues on raw tomatoes are at the current tolerance level, and that these residues in processed tomato products undergo a 10-fold concentration, then total estimated oncogenic risk from all fungicide residues in tomatoes would increase more than 300% above the committee's risk estimates, which assume no concentration of residues in processed foods. If residues on raw tomatoes are assumed to be one-tenth of the published tolerance and undergo a 10-fold concentration, then estimated oncogenic risk from fungicide residues would decline about 51%" (3). Because of the large quantity of fruit needed, it was not possible to process fruit harvested from this study into puree or paste. However, because juice contained no detectable levels of chlorothalonil or anilazine, further concentration probably would still result in insignificant levels of these pesticides. Admittedly, while our processing of fruit may not be typical of most commercial operations and this could have affected residue levels detected in these tests, our washing times should be similar; washing more than most other factors should be related to removal of residues of nonsystemic fungicides. A California study shows that processed tomato products (i.e., tomato juice, canned whole tomatoes, and paste) contain less than 4% of the original residue after processing (4).

In the past, disease control programs have been selected largely on the basis of efficacy and pesticide costs. In the future, the control of residues may be considered a higher priority. Disease control programs will need to be designed to reduce pesticide residues but allow growers to produce profitable tomato crops. The potential for even greater residue reduction exists by combining host disease resistance, alteration of fungicide rates and schedules, and disease forecasting systems into a useful commercial disease prevention system. To reach this goal, more data are needed on the actual accumulation of residues of raw product and in foods processed from them resulting from use of pesticides.

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