

Retention of Mercury in Tissues of Cattle and Sheep Given Oral Doses of a Mercurial Fungicide, Ceresan M

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A mercurial fungicide, formerly used, Ceresan M (ethyl mercury *p*-toluene sulfonamide), was given in daily oral doses at 15 mg/kg to cattle for 7 or 12 days and to sheep for 4 or 7 days. Residues of mercury in hair, blood, and tissues were determined by atomic absorption spectrophotometry for 20 weeks after dosing had ceased. Resi-

dues of mercury increased in the hair of cattle to a maximum point and then began to decrease. Residues in brain and muscle of both cattle and sheep were lower initially than in liver and kidney and then decreased steadily during the 20-week postexposure. Mercury persisted in tissues of cattle throughout the study.

Organomercurial fungicides have been widely used in the past for the treatment of seed prior to planting to control plant diseases. These fungicides are no longer registered for seed treatment. One of the mercurial fungicides formerly used in this manner was Ceresan M, which contains 7.7% active ingredient of ethyl mercury *p*-toluene sulfonamide (mercury equivalent of 3.2%).

In previous work by Palmer *et al.* (1973), adult cattle and sheep were poisoned acutely after 27 and 13 daily doses, respectively, with Ceresan M at 15 mg/kg based on total formulation. The dose, 15 mg of total formulation/kg of body weight, was comparable to that which animals might consume if in accidental contact with grain treated with the fungicide in the recommended manner. Residues of mercury were generally higher in kidney of cattle and sheep than in liver; lesser amounts were in muscle and brain. During that study, no effort was made to determine the persistence of the mercury in animals after dosages with the fungicide had ceased. Therefore, the objective of this study was to determine the retention of mercury in tissues, blood, and hair of cattle and sheep for 20 weeks after exposure to daily oral doses of Ceresan M for various lengths of time.

MATERIALS AND METHODS

Reagent. Ceresan M (E. I. duPont de Nemours and Co., Wilmington, Del.) has 7.7% active ingredient of ethyl mercury *p*-toluene sulfonamide (3.2% mercury equivalent).

Apparatus. Model 403 atomic absorption spectrophotometer (Perkin-Elmer Corp., Norwalk, Conn.) set at 2537 Å resonance line and equipped with a mercury hollow cathode lamp was used in this study.

Retention Study. Yearling cattle and sheep of mixed breed and sex, obtained at local auction, were dosed with 15 mg/kg/day of total Ceresan M. The lengths of dosing times were chosen so that no noticeable signs of poisoning would be apparent (Palmer *et al.*, 1973). One group of six cattle was dosed for 7 days, another group of six was dosed for 12 days, and two cattle were untreated. One group of five sheep was dosed for 4 days, one group of six sheep was dosed for 7 days, and one group of two sheep served as a control.

Samples of blood and hair were obtained from all animals before the start of the study and weekly for 20 weeks at the end of dosing period. One animal from each dosed group of cattle and sheep was killed 1 day after dosing had ceased and monthly during the study. Tissues analyzed for mercury were kidney, liver, muscle, and brain.

The procedure used for the determination of mercury in

the samples consisted of a wet-acid digestion followed by partitioning into methyl isobutyl ketone and final analysis by atomic absorption spectrophotometry (Wright *et al.*, 1973). Recovery of mercury from fortified tissues averaged 85% (range, 75–94%) of the total mercury present, and the sensitivity of the method was 0.1 ppm. Residues of mercury reported in the Results Section have not been corrected for percentage recovery.

RESULTS AND DISCUSSION

None of the dosed animals had signs of poisoning during the test period. Table I shows residues of mercury in tissues of the cattle dosed for 7 days. Highest residues were found in kidney, then liver, muscle, and brain. Residues in all tissues decreased steadily during the postexposure period. Kidney and liver still had excessive residues at 20 weeks.

Table I also shows residues of mercury in tissues of the cattle dosed for 12 days. The pattern of these residues is very similar to that in the group given seven doses except the residues are considerably higher. Residues again decreased steadily through the 20-week postexposure period.

Table III shows the residues of mercury in sheep. After 4 days of dosing, residues of mercury in liver and kidney were similar at 1 day postexposure. Residues in kidney increased after that time to 83.3 ppm at 8 weeks; however, residues in liver decreased from 1 day postexposure. Residues in muscle and brain were low at 1 day postexposure and were not detectable by 16 weeks postexposure.

The residues in all tissues of sheep after 7 days of dosing were much higher than those in sheep after 4 days of dosing. The residues in kidney increased rapidly and were over 200 ppm after 12 weeks postexposure.

Tissues from control cattle and sheep contained no detectable mercury.

Table II shows the residues of mercury in the blood of cattle and sheep sampled weekly after cessation of dosage. The greatest residues of mercury were in blood collected at 1 day postexposure from cattle given 12 daily oral doses. Residues in blood of both cattle and sheep decreased slowly but were still detectable at 16 weeks postexposure. No residues of mercury were detected in the blood of control cattle and sheep.

Table IV shows the mercury residues in the hair of dosed cattle and sheep. The values obtained in this portion of the study may be misleading because new areas on the animals were sheared each week. Therefore, the reported values are the sum of the old residues already present in the hair plus that deposited during the last week. Residues of mercury increased to 56 ppm in the hair of cattle at 11 weeks postexposure in the 12-dose group. Mercury in wool of sheep did not climb this high. Again, this could be due to the fact that some wool was on the

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Table I. Mercury (ppm) in Tissues of Cattle Dosed Daily with Ceresan M at 15 mg/kg

| Time post-exposure, weeks | Residues (ppm) in indicated tissue ^a | | | |
|---------------------------|---|--------|--------|-------|
| | Liver | Kidney | Muscle | Brain |
| Dosed for 7 days | | | | |
| 0 ^b | 25.5 | 71.5 | 4.3 | 0.9 |
| 4 | 24.5 | 50.7 | 2.4 | 1.1 |
| 8 | 16.0 | 58.8 | 2.0 | 1.0 |
| 12 | 5.9 | 39.9 | 0.6 | 0.7 |
| 16 | 3.2 | 14.8 | 0.5 | 0.3 |
| 20 | 2.9 | 18.4 | 0.1 | 0.4 |
| Dosed for 12 days | | | | |
| 0 ^b | 40.9 | 146.2 | 7.3 | 3.0 |
| 4 | 37.0 | 71.5 | 3.5 | 2.8 |
| 8 | 27.2 | 34.2 | 3.3 | 1.6 |
| 12 | 24.4 | 64.4 | 2.1 | 1.2 |
| 16 | 4.7 | 47.8 | 1.3 | 1.0 |
| 20 | 3.1 | 18.4 | 1.0 | 0.3 |

^a Each value is an average of residues in duplicate samples.

^b Animal killed and samples taken 24 hr after last dose.

Table II. Mercury (ppm) in Blood of Cattle and Sheep Dosed Daily with Ceresan M at 15 mg/kg

| Time post-exposure, weeks | Average residues (ppm) in animals given indicated number of doses ^a | | | |
|---------------------------|--|-----|-----------------|-----|
| | Cattle | | Sheep | |
| | 7 | 12 | 4 | 7 |
| 0 ^b | 2.4 | 2.7 | 1.5 | 2.0 |
| 1 | 1.5 | 2.0 | 0.7 | 1.5 |
| 2 | 0.6 | 2.0 | 0.6 | 1.0 |
| 3 | 0.6 | 1.3 | 0.1 | 0.6 |
| 4 | 0.2 | 0.8 | 0.1 | 0.6 |
| 5 | 0.3 | 0.9 | 0.2 | 0.6 |
| 6 | 0.2 | 0.7 | ND ^c | 0.3 |
| 7 | 0.5 | 0.8 | ND | 0.7 |
| 8 | 0.8 | 0.8 | ND | 0.1 |
| 9 | 0.4 | 0.5 | 0.1 | 0.1 |
| 11 | 0.2 | 0.7 | 0.2 | 0.3 |
| 12 | 0.3 | 0.2 | 0.2 | 0.1 |
| 13 | 0.1 | 0.2 | 0.3 | ND |
| 14 | 0.1 | 0.5 | 0.4 | ND |
| 15 | 0.4 | 0.9 | 0.3 | ND |
| 16 | 0.2 | 0.2 | 0.4 | ND |
| 17 | ND | ND | 0.5 | 0.1 |
| 18 | 0.5 | ND | 0.1 | ND |
| 19 | 0.1 | ND | 0.1 | ND |
| 20 | ND | 0.1 | — ^d | 0.5 |

^a The values for weeks 17-20 were obtained from one animal.

^b Sample taken 24 hr after last dose. ^c ND indicates none detected. ^d — indicates no sample available.

sheep before the start of the study and, therefore, did not contain mercury. Mercury was deposited in wool grown after dosing.

Even though residues of mercury in tissues of cattle and sheep decreased with time after exposure to Ceresan M, residues were present after 20 weeks postexposure; if detected, the animals would be condemned.

Mercury in the hair of cattle and sheep is evidence of prior exposure of an animal to a mercurial compound.

Table III. Mercury (ppm) in Tissues of Sheep Dosed Daily with Ceresan M at 15 mg/kg

| Time post-exposure, weeks | Residues (ppm) in indicated tissue ^a | | | |
|---------------------------|---|----------------|-----------------|-------|
| | Liver | Kidney | Muscle | Brain |
| Dosed for 4 days | | | | |
| 0 ^b | 22.4 | 18.2 | 1.8 | 1.0 |
| 4 | 15.4 | — ^c | 1.6 | 1.4 |
| 8 | 3.1 | 83.3 | ND ^d | 0.3 |
| 12 | 7.8 | 72.7 | 0.3 | 0.5 |
| 16 | — | 57.4 | ND | ND |
| 20 | — | — | ND | — |
| Dosed for 7 days | | | | |
| 0 ^b | 31.7 | 22.2 | 6.4 | 1.8 |
| 4 | 19.3 | — | 0.9 | 2.1 |
| 8 | 16.3 | 228.8 | 0.1 | 1.0 |
| 12 | 9.6 | 208.2 | ND | 0.4 |
| 20 | — | — | 0.5 | — |

^a Each value is an average of residues in duplicate samples.

^b Animal killed and samples taken 24 hr after last dose. ^c — indicates no sample available. ^d ND indicates none detected.

Table IV. Mercury (ppm) in Hair of Cattle and Sheep Dosed Daily with Ceresan M at 15 mg/kg

| Time post-exposure, weeks | Average residues (ppm) in animals given indicated number of doses ^a | | | |
|---------------------------|--|------|-----------------|------|
| | Cattle | | Sheep | |
| | 7 | 12 | 4 | 7 |
| 0 ^b | 0.1 | 0.8 | ND ^c | 0.4 |
| 1 | 1.2 | 1.6 | 0.1 | 0.1 |
| 2 | 18.3 | 8.0 | 0.3 | 0.3 |
| 3 | 15.6 | 13.8 | 0.3 | 0.7 |
| 4 | 16.8 | 14.0 | 0.2 | 1.5 |
| 5 | 27.5 | 15.5 | 0.1 | 4.2 |
| 6 | 38.5 | 29.7 | 1.0 | 1.8 |
| 7 | 35.5 | 35.3 | 4.0 | 5.4 |
| 8 | 30.0 | 41.9 | 3.8 | 8.4 |
| 9 | 26.0 | 31.3 | 3.5 | 10.4 |
| 10 | 29.9 | 51.2 | 3.6 | 5.5 |
| 11 | 27.7 | 55.9 | 4.5 | 4.9 |
| 12 | 26.3 | 43.3 | 3.2 | 4.3 |
| 13 | 19.9 | 50.5 | 8.1 | 9.3 |
| 14 | 21.5 | 43.8 | 3.1 | 5.7 |
| 15 | 15.7 | 38.2 | 3.6 | 8.3 |
| 16 | 19.4 | 41.0 | 5.0 | 14.5 |
| 17 | 8.4 | 53.6 | 3.7 | 5.7 |
| 18 | 6.0 | 39.0 | 5.1 | 5.3 |
| 19 | 7.2 | 31.0 | 3.3 | 8.7 |
| 20 | 6.4 | 19.4 | 4.7 | 8.7 |

^a The values for weeks 17-20 were obtained from one animal.

^b Sample taken 24 hr after last dose. ^c ND indicates none detected.

LITERATURE CITED

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