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ARSENIC IN CATTLE HAIR AFTER FORESTS ARE PRECOMMERCIALLY THINNED WITH ORGANIC ARSENICAL HERBICIDES

by

Logan A. Norris, Supervisory Research Chemist

ABSTRACT

Arsenic levels were measured in hair taken from cattle grazed in northeast Washington forests. The forests were precommercially thinned with MSMA or cacodylic acid. Cattle from two of three study areas showed an increase in arsenic levels during the grazing season. In an untreated area, arsenic levels in cattle also increased during the grazing season. In all cases the levels of arsenic were within the normal range which indicates that herbicides were handled and applied with sufficient care to prevent appreciable exposure of cattle.

KEYWORDS: Herbicide side effects, cattle, thinning (precommercial), arsenic, cacodylic acid, MSMA.

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INTRODUCTION

MSMA (monosodium methanearsonate) and cacodylic acid (dimethylarsinic acid) are effective chemical agents in precommercial thinning of overstocked stands of conifers in northeastern Washington. In these treatments, undiluted herbicide concentrate is injected into axe cuts which are spaced approximately 7 cm around the stem of the tree about 1.5 m above the ground.

Cattle may be grazed for at least part of the year in chemically thinned stands. It is possible, therefore, that cattle could be exposed to arsenic (As) either through careless handling and application of the herbicide or through the movement of chemical residues from treated trees to nearby vegetation which is then eaten by these animals.

Arsenic is commonly accumulated in the hair of mammals. Human hair contains As at concentrations between 0.03 and 1 ppm (Smith 1967, Vallee et al. 1960, Rothman 1954). Exon et al. (1974) reported a threefold increase in As in the hair of rabbits exposed to MSMA in their diet for 12 weeks. The concentration of As in cattle hair nearly doubled during a 10-day exposure to MSMA, and increased 35 times during a 48-day exposure to cacodylic acid (Dickinson; 1972, 1975). On this basis I believe arsenic in hair should be a reasonable index of the degree of exposure cattle receive when they are grazing in forested areas which have been precommercially thinned with the organic arsenical herbicides.

In 1969, the Okanogan National Forest reported the death of eight range cattle in forest areas treated with these herbicides. Arsenic levels were slightly elevated in several body tissue specimens taken from these animals, but were not of diagnostic significance. As a result of this incident and because of concern about applicator exposure, a cooperative study of the behavior and impact of organic arsenical herbicides in the forest was initiated by the Pacific Northwest Forest and Range Experiment Station and Region 6 of the Forest Service.

The study included several State and Federal agencies, universities, companies, and private individuals. As part of the cooperative arsenic study, Dr. Hugh Maycumber, DVM, a private veterinarian in Tonasket, Washington, measured arsenic in hair from cattle grazed in thinned areas. Maycumber's findings are given in the final report of the cooperative arsenic study, but his report has not been widely distributed.1/ I have included a brief description of his study and his results for easy comparison with the findings of my study.

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 $[\]frac{1}{N}$ Norris, Logan A. 1974. The Behavior and Impact of Organic Arsenical Herbicides in the Forest: Final Report on Cooperative Studies. 98 pages on file. Forestry Sciences Laboratory, Pacific Northwest Forest and Range Experiment Station, Corvallis, Oregon.

Maycumber found a statistically significant increase, between spring and fall, in the concentration of arsenic in the hair of cattle grazed in treated areas. A shortage of holding facilities, however, prevented him from sampling hair from cattle grazing in untreated areas. As a result, it was not possible to tell if the increase he detected was caused by exposure to arsenic from thinning operations or from the accrual of arsenic naturally present in the environment. I attempted to resolve this point by repeating a portion of Maycumber's study but included hair samples from cattle which had grazed in untreated areas.

METHODS

THE MAYCUMBER STUDY

The Mt. Hull study area on the Okanogan National Forest was thinned in 1969 with Glowon® (MSMA) Tree Killer and Silvisar 510° (cacodylic acid) and was the site of cattle deaths the same summer. Hair samples were collected from 37 head of cattle on June 26, 1970, before the animals were turned out to graze for the summer and approximately 1 year after the grazing area had been thinned. A second collection of hair from the same animals was made on October 14, 1970.

The Mill Creek study area is on the Colville National Forest. It was thinned with Silvisar 550[®] (MSMA) during the summer of 1970. Hair samples were taken from 28 cattle on May 29, 1970, before they were turned out to graze, and a second hair collection was made on September 9, 1970. The cattle were in the Mill Creek study area during the chemical thinning operation. At the end of the grazing period, tissues from two of these cattle were analyzed for arsenic residues. A female deer, recognized as a regular inhabitant of the Mill Creek area during the 1970 thinning season, was also killed. Various tissues from the deer were analyzed for arsenic residues.

THE NORRIS STUDY

This study was done in a mixed western larch (Larix occidentallis Nutt.), Douglas-fir (Pseudotsuga menziesii (Mirb.) Franco) forest in the Pseudotsuga menziesii Zone near Colville, Washington (Franklin and Dyrness 1973). Approximately 290 ha of a 365-ha grazing allotment were thinned with MSMA between 1969 and 1974. On June 13, 1973, 12 cattle, selected at random, were marked and hair samples were collected for arsenic analysis. These animals grazed throughout the area from June 18 to October 12. On October 16, 1973, hair samples were collected from the same animals. Approximately 75 ha of the allotment were thinned with MSMA during the time the cattle were in the area. In a similar, nearby area which had not been previously thinned chemically, hair from 12 marked animals, selected at random, was also collected in June and again in October.

Arsenic in hair from the Maycumber study was analyzed by neutron activation at Washington State University, Pullman (Thomas and Kristensen 1968). Hair samples from the Norris study and tissue samples from the Maycumber study were oxidized by digestion with nitric-sulfuric acid to remove organic matter. The arsenates were reduced to arsine which was trapped in silver diethyldithiocarbamate in pyridine forming a colored complex which was spectrophotometrically measured. Analyses were done by the Washington Department of Social and Health Services at Wenatchee.

RESULTS

MAYCUMBER STUDY

Levels of arsenic from the hair of cattle grazed on the Mt. Hull and the Mill Creek study areas are shown in table 1.

I tested Maycumber's data with a paired t-test which showed that the increase in arsenic concentration between spring and fall samples was statistically significant (P<0.05) at both sites. A t-test showed no significant difference (P>0.05) between the two sites for samples collected at the same time. The lack of samples from animals grazed in untreated areas prevents a determination of whether or not the increase in arsenic concentration in hair during the summer grazing season was a result of the chemical thinning program.

The results of analysis for arsenic in tissue of two cattle and one deer from the Mill Creek study area are in table 2.

Detectable residues of arsenic were only found in the hair of the two cattle and the deer from the Mill Creek study area. Lack of detectable residues in soft body tissues is expected because arsenic is rapidly dissipated from these tissues. The increase in arsenic concentration in hair from animal 28 was more than three times as great as the average increase for animals from the Mill Creek study area; but despite this, there were no detectable residues of arsenic in body tissues. The concentration of arsenic in hair from the deer is lower than the average for hair from cattle from the

Table	1Arsenic in hair from cattle grazed in forest
	areas thinned with MSMA or cacodylic acid, Maycumber's study

	<u> </u>				
Mt. Hull Study Area <u>1</u> /			Mill Creek Study Area ^{2/}		
Animal number	Spring 1970	Fall 1970	Animal number	Spring 1970	Fall 1970
	ppm ³	/		ppm ³	/
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37	0.56 0.39 0.47 0.96 0.18 1.28 1.04 0.27 0.73 0.41 0.35 0.69 0.54 0.31 0.23 0.53 0.44 0.31 0.23 0.53 0.49 0.35 0.19 0.33 0.35 0.12 0.58 0.62 0.10 0.66 0.71 0.58 0.27 0.73 0.41 0.35 0.53 0.12 0.58 0.62 0.10 0.58 0.27 0.73 0.28 0.62 0.10 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.58 0.27 0.71 0.75 0.75 0.77	0.33 0.65 0.74 1.20 0.56 0.81 1.53 1.14 1.07 1.28 0.72 0.62 1.04 0.72 0.62 1.04 0.72 0.68 1.04 0.72 0.68 1.050 0.97 0.68 1.050 0.97 0.68 1.04 0.57 0.36 0.47 0.23 0.36 0.47 0.23 0.65 0.32 0.47 0.65 0.32 0.47 0.65 0.32 0.47 0.65 0.32 0.47 0.65 0.32 0.47 0.65 0.32 0.47 0.68 0.47 0.36 0.47 0.36 0.47 0.36 0.47 0.36 0.47 0.36 0.47 0.32 0.481 0.481 0.481 0.482 0.657 0.34 0.841 0.34 0.811 0.82 0.657 0.32 0.577 0.74 0.82 0.681 0.657 0.32 0.481 0.482 0.657 0.74 0.82 0.34 0.811 0.812 0.82 0.657 0.74 0.82 0.681 0.82 0.657 0.74 0.82 0.681 0.82 0.657 0.74 0.82 0.681 0.82 0.657 0.74 0.841 0.82 0.681 0.841 0.82 0.681 0.841 0.82 0.681 0.841 0.82 0.681 0.841 0.821 0.681 0.841	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 4 22 23 24 25 26 4 /28	0.18 0.20 0.24 0.23 0.48 0.25 0.64 0.21 0.24 0.58 0.26 0.26 0.23 0.26 0.23 0.58 0.73 0.26 0.23 0.58 0.97 1.28 0.73 0.38 0.66 0.44 0.94 0.26 0.44 0.55 0.57	0.44 0.27 0.61 0.98 0.45 0.59 0.94 0.90 0.35 1.02 0.82 1.18 0.79 0.58 0.50 1.08 1.15 0.53 0.98 0.42 0.57 0.43 0.53 1.16 1.30
1/ _	<u>5/</u> 0.46(a))	0.49(a)	0.73(b)

 $\frac{1}{2}$ Thinned with Glowon $^{\rm I\!R}$ tree killer and Silvisar 510 $^{\rm I\!R}$, 1969.

 $\frac{2}{}$ Thinned with Silvisar 550 $^{\textcircled{R}}$, 1970.

 $\frac{3/}{2}$ Arsenic determined by neutron activation, Washington State University, Pullman.

 $\frac{4}{4}$ Animal slaughtered for tissue arsenic residue

analysis, fall 1970. $\frac{5}{}$ Average values. Values with the same letter are not significantly different (P>0.05).

Tissue	Animal 21	Animal 28	Deer	
	<u>ppm</u>			
Hair Blood Urine Liver Kidney Lung Heart Tongue Spleen Muscle Brain Stomach contents Udder Feces	2/ND ND ND ND ND ND ND ND ND ND ND ND ND	1.3 ND ND ND ND ND ND ND ND ND ND ND	0.3 3/ ND ND ND ND ND ND ND ND ND ND ND	

Table 2--Arsenic residues in tissues from two cows and one female deer which grazed in the Mill Creek study area during thinning operations 1/

 $\frac{1}{1}$ The area was thinned with Silvisar 550[®] during the summer of 1970. Cows were slaughtered in the fall of 1970 and tissues were analyzed at the Washington State Department Social and Health Services Laboratory, Wenatchee, Washington.

 $\frac{2/}{\text{ND}}$ - Nondetectable means less than 0.20 ppm arsenic.

 $\frac{3}{}$ --- Not analyzed.

Mill Creek area. The presence of arsenic indicates that the deer received some exposure. It is not possible, however, to determine whether this represents arsenic from the thinning operation or arsenic which is naturally present in the environment.

NORRIS STUDY

Levels of arsenic in hair from exposed and unexposed animals are shown in table 3. I used paired t-tests on spring and fall residue levels in hair from (1) exposed cattle and (2) unexposed cattle. The results show no significant difference (P>0.05) between spring and fall values in the exposed group. In the unexposed group, on the other hand, the difference between spring and fall values is significant (P<0.05), indicating that the increase in arsenic concentration in hair is real. A t-test showed no difference (P>0.05) between arsenic levels in hair collected in the fall from exposed and unexposed cattle.

	Exposed Group	<u>2/</u>	Control Group <u>3</u> /		
Animal	Spring	Fall	Anima1	Spring	Fall
	<u>p</u>	om		pj	om
1 2 3 4 5 6 7 8 9 10 11 12	0.50 0.71 1.20 0.50 0.71 0.60 0.83 0.60 0.60 0.71 0.83 0.52	0.20 1.10 0.40 0.30 0.60 0.20 4/ 0.60 0.60	1 2 3 4 5 6 7 8 9 10 11 12	$\begin{array}{c} 0.11\\ 0.22\\ 0.15\\ 0.10\\ 0.11\\ 0.11\\ 0.10\\ 0.33\\ 0.16\\ 0.10\\ 0.11\\ 0.33\\ \end{array}$	$\begin{array}{c} 0.30\\ 0.50\\ 0.20\\ 0.30\\ 0.90\\ 0.10\\ 0.40\\ 0.10\\ 0.40\\ 0.40\\ 0.30\\ 0.30\\ 0.30\\ \end{array}$
1/	5/6/0.69	0.48		0.16	0.35

Table 3--Arsenic in hair from cattle grazed in forest areas, Norris study 1

 $\frac{1/}{}$ Arsenic determined at Washington Department of Social and Health Services, Wenatchee, Washington.

 $\frac{2}{}$ Animals grazed from June 18 to October 12, 1973, in a 900-acre grazing allotment. Approximately 700 acres were thinned with MSMA, 190 acres during the 1973 grazing season.

 $\frac{3}{2}$ Animals grazed in untreated forest area.

 $\frac{4}{---}$ Not analyzed; these animals were not resampled in the fall.

 $\frac{5}{}$ Average values.

 $\frac{6}{}$ Average value of 0.72 for the same nine animals sampled in the fall.

DISCUSSION

On the surface these results seem confusing, but they are not. The values reported in both studies are within a normal range of values for arsenic in hair from animals not specifically exposed to arsenic. Dickinson (1972, 1975) determined the toxicity characteristics of MSMA and cacodylic acid in cattle. He measured arsenic in hair before he began administering the herbicide and reported values of 2.1, 1.1, 0.8, 2.4, 0.9, and 1.0 ppm (average 1.4 ppm) arsenic for previously unexposed cattle. Dickinson's values are substantially higher than the mean values found by Maycumber or Norris for either exposed or unexposed cattle. Dickinson (1972, 1975) also reported arsenic residues in hair from the same cattle after exposure to a diet including MSMA or cacodylic acid. The two animals receiving MSMA at 10 mg/kg per day had 1.4 and 3.3 ppm arsenic in hair after 10 days exposure. The three animals fed cacodylic acid at 10 mg/kg per day had 4.3, 2.6, and 2.0 ppm arsenic in hair after 10 days; arsenic accumulation continued with exposure. By 48 days, the arsenic concentrations were 32.8, 15.7, and 12.9 ppm, respectively. Rabbits receiving MSMA at 50 ppm in their diet showed a steady increase in arsenic residue levels in hair. Residues of 1.3 ppm, 2.4 ppm, and 3.6 ppm after 4, 7, and 12 weeks' exposure, respectively, were reported (Exon et al. 1974).

The levels of arsenic in hair detected by Maycumber and me apparently are not affected to an important degree by grazing in chemically thinned areas. The small changes in arsenic concentration detected with time or nature of exposure in these studies are not sufficient to elevate arsenic concentrations in hair to a diagnostically significant level.

Do these results indicate arsenic in hair is not a good indicator of arsenic exposure? No! Arsenic is everywhere in the environment. The natural levels of arsenic vary widely from nondetectable levels in some forest streams to 5 ppm in soil to more than 100 ppm in some shellfish (Norris et al. 1977, Schroeder and Balassa 1966). Man's activities cause some redistribution of arsenic. A copper smelter in Tacoma, Washington, emits approximately 2 x 10^5 kg As per year as As₂0₃ (Crecelius et al. 1975). All living things ingest at least small amounts of arsenic daily. The presence of detectable levels of arsenic in cattle hair before grazing in treated areas shows arsenic in hair is an indicator of exposure. Both the Dickinson (1972, 1975) and Exon et al. (1974) data show that the concentration of arsenic in hair increases with exposure to MSMA and cacodylic acid in the diet. Therefore, the data from both Maycumber's and my study indicate that (1) the organic arsenicals were handled and applied with sufficient care and (2) arsenic accumulation in understory vegetation was small enough so that arsenic in hair from cattle grazed in these chemically thinned areas did not increase beyond the normal range. Т believe MSMA and cacodylic acid can be used operationally for thinning purposes without significant exposure to cattle grazing in thinned areas, even while the application is being made. Careful handling and application procedures are required, however. $\frac{2}{}$

^{2/} Procedures for handling and application of MSMA and cacodylic acid in thinning programs developed by the Siuslaw and Colville National Forest are available from the author.

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