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FLUORINE IN MILK, PLANT FOODS, AND FOODS COOKED IN FLUORINE CONTAINING WATER

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SIGNIFICANCE OF THE PROBLEM

Since the discovery was made in this laboratory* that the drinking of water containing as little as one part per million of fluorine by children between the ages of six months and twelve years, is the cause of the defect of their permanent teeth known as mottled enamel which is so widely spread in Arizona and in many other parts of the world, many questions have arisen.

Of prime importance has been the query regarding the possibility of fluorine being present in toxic amounts in foods as well as in water. Do plants grown on Arizona soils, irrigated with fluorine containing waters become impregnated with fluorine in toxic amounts? Does the milk from Arizona dairy cows which are drinking fluorine containing waters contain dangerous amounts of fluorine? Is it safe to cook foods in fluorine containing waters that are not usable for drinking purposes?

It is the purpose of this paper to report findings of the research that has been carried on in the Departments of Agricultural Chemistry and Soils and of Human Nutrition of the Arizona Agricultural Experiment Station which was planned to throw light on these problems.

FLUORINE IN MILK

Experimental Procedure:

Through the courtesy and cooperation of the Dairy Department in the Arizona Experiment Station two cows, one Guernsey, no. 23 of the University of Arizona herd, and the other a Jersey, no. 112 of the University of Arizona herd, were made available for a study of fluorine content of the milk produced during lactation periods in which they were drinking water containing fluorine in different concentrations. These cows were isolated in individual

* More than 25 publications on fluorine vs. mottled enamel research have come from this laboratory.

pens on the University of Arizona farm and fed their usual rations except that they were not allowed to graze in the pasture. They were given waters of known fluorine concentration to drink ad libitum for experimental periods of from two to seven days in length. During this time, one liter samples of milk were collected on the first two and the last two days of each experimental period. Samples of urine were taken at the same time. Composited samples of milk and of urine were analyzed for their fluorine content following the method of Willard and Winter as modified by Armstrong. (J. Am. Chem. Soc. 55: 1741, 1933).

Results:

The fluorine content of the milk samples collected from the Guernsey cow which was given waters containing 0.2, 3.0, 8.0, 12.0, 55.0, 228.0, or 495.0 parts per million of fluorine may be seen in Table I.

TABLE I.

THE FLUORINE CONTENT OF MILK FROM A GUERNSEY DAIRY COW DRINKING WATER CONTAINING FLUORINE

Experimental period no. and length	Average water consumption liters per day	Average milk production lbs. per day	Fluorine content (parts per million) In drinking water	In milk	In Urine
I 3 days	28	11.1	0.2	0.0	0.1
II 7 days	28	10.0	3.0	0.3	3.3
III 8 days	28	8.5	8.0	0.4	3.6
IV 7 days	26	8.7	12.0	0.4	14.1
V 7 days	28	9.4	55.0	0.4	30.9
VI 2 days	6	7.3	495.0	0.3	78.9
VII 2 days	16	6.4	238.0	0.4	108.9
VIII 6 days	34	8.1	0.2	0.3	41.6

Table II presents similar information concerning the milk from a Jersey cow.

TABLE II.

THE FLUORINE CONTENT OF MILK FROM A JERSEY DAIRY COW DRINKING WATER CONTAINING FLUORINE

Experimental period no. and length	Average water consumption liters per day	Average milk production lbs. per day	Fluorine content (parts per million) In drinking water	In Milk	In Urine
I 4 days	24	12.7	0.2	0.2	1.5
II 6 days	22	11.1	56.0	0.3	21.1
III 7 days	13	7.2	500.0	0.5	210.6
IV 13 days	--	7.3	0.2	0.3	--

From Tables I and II, it is evident that there was a small but definite increase in the fluorine content of milk when sodium fluoride was added to the drinking water of the cows. However fluorine is not transmitted in large quantities from the drinking water to the milk. It may be noted that the fluorine content of the milk from the two cows used in this study did not exceed 0.5 parts per million, even when the concentration of fluorine in their water supply had been increased to as much as 500 parts per million, which is far in excess of the amount found in any water supply. The fluorine content of natural waters in Arizona rarely exceeds ten parts per million, although in a few instances, samples of non-potable water which have been tested were found to contain 30 parts per million. Water containing more than 55 parts per million of fluorine in the form of sodium fluoride which was used in this study was found to have a salty taste and the cows consumed much less of it. They lost their appetite and refused all but a minimum amount of water. For example, during the second day in which the Guernsey cow was given water which contained 238 parts per million of fluorine the water consumption dropped from 28 to 3½ liters.

It may also be noted that there was an inverse relationship between the quantity of milk produced and the amount of fluorine in the water. During the period of intake of the highest fluorine-water, milk production reached its lowest level. After returning to normal water, the milk production increased.

Obviously, also, most of the fluorine ingested by the two cows was excreted by the kidneys. Although no attempt was made to collect all of the urine, samples were taken by massaging one side of the vulva at the same time as the milk samples were collected and were analyzed for their fluorine content. It was found that the concentration of fluorine in the urine increased greatly, in direct proportion to the increase in concentration of fluorine in the cow's water supply.

FLUORINE IN PLANT FOODS

Experimental Procedure:

Two widely separated areas of soil were selected and prepared for planting for the purpose of studying the ability of plants to absorb fluorine when grown on soils which contain fluorine. In one area, calcium fluoride was added to one-half of the soil, while the other half served as a control plot without added fluorine. Powdered fluorospar was turned into the soil by machine to a depth of 6 2/3 inches (75 pounds per 318 square feet), the soil containing approximately 500 parts per million of fluorine. Wheat was planted on these plots in March and harvested four months later. The stalks were cut six inches above the ground in order to avoid contamination with the fluorine in the soil and irrigation water. At the time of the cutting they were shocked, thrashed, and the grains and leaves and stalks prepared separately for analysis of their fluorine contents. Hegari, soybeans, and corn were planted in July and harvested toward the end of October.

TABLE III.

THE COMPARATIVE FLUORINE CONTENT OF PLANTS GROWN ON SOILS WITH AND WITHOUT ADDED CALCIUM FLUORIDE

Plant	Fluorine Content (parts per million on dry wt. basis)	
	Grown in control plots with no added fluorine	Grown on soils with calcium fluoride added
Corn	Trace	Trace
Hegari	0.00	2.1
Soybeans	0.83	1.2
Wheat	0.69	1.0
Wheat, Stalks and leaves	3.70	7.2
Alfalfa, 1st cutting	7.0	15.0
2nd cutting	4.5	10.8
3rd cutting	5.0	--
4th cutting	6.0	11.3

The second large area was subdivided into four plots one of which served as a control. To the other three plots, sodium fluoride was added so that the concentration of fluorine in the soils was 800, 1600, and 3200 parts per million respectively. Tomatoes, carrots, beets, string beans, and yams were planted on these plots and harvested when they reached marketable maturity.

TABLE IV.

THE COMPARATIVE FLUORINE CONTENT OF PLANT FOODS GROWN IN SOILS CONTAINING DIFFERENT AMOUNTS OF SODIUM FLUORIDE

	Fluorine Content (parts per million on fresh wt. basis)			
Plant food	Plot 1 No NaF added to soil	Plot 2 800 p.p.m. NaF added to soil	Plot 3 1600 p.p.m. NaF added to soil	Plot 4 3200 p.p.m. NaF added to soil
Beets	1.7	6.5	---	17.7
Carrots	1.0	3.0	0.4	1.3
String beans	0.0	---	0.0	---
Yams	0.0	7.6	---	8.2
Tomatoes	0.7	0.5	2.3	1.2

Results:

Results of the analysis of the plant foods sampled after they were air-dried and ground to a fine powder are presented in Tables III and IV. In general somewhat larger concentrations of fluorine were found in plants grown on fluorine enriched soils. However the amount deposited in the plants did not appear to be proportional to the amount of fluorine in the soil. Also the amount absorbed by the plant appeared to vary with the type of plant food and with the part of the plant itself. For example, beets and yams absorbed more fluorine than string beans and tomatoes, and a higher concentration of fluorine was found in wheat stalks and leaves than in wheat grain (all calculations on the dry weight basis).

FLUORINE IN COOKED FOODS

Experimental Procedure:

For the purpose of determining the extent of absorption of fluorine by foods cooked in fluorine containing waters, vegetables were purchased on the open market and divided into three lots. One lot was analyzed for fluorine content when in the fresh raw state. The second lot was cooked in distilled water, and the third lot was covered with and cooked in the same amount of fluorine containing water for the same length of time. After cooking, the liquid was drained off and the vegetables washed with distilled water. Both the cooking liquid and the cooked foods were then analyzed separately for their fluorine content.

Pink beans, potatoes, carrots, and oatmeal were cooked in water containing 224 parts per million of fluorine, and carrots, cabbage, spinach, cauliflower, Italian squash, Brussel sprouts, and beets cooked in 5 parts per million fluorine water. Results expressed on the fresh weight basis of the analyses of the foods cooked in both distilled and fluorine waters are presented in Table V.

TABLE V.

THE COMPARATIVE FLUORINE CONTENT OF FOODS COOKED IN WATERS WITH AND WITHOUT FLUORINE

Food	Fluorine content (parts per million) Cooked in distilled water without fluorine	Cooked in water containing fluorine 5 p.p.m.	24 p.p.m.
Pinto beans	2.0	---	37.1
Beets	0.0	1.0	----
Potatoes	0.5	---	9.7
Cabbage	0.0	3.6	----
Carrots	0.0	3.5	20.1
"	2.3	3.2	----
Cauliflower	0.0	4.2	----
Oatmeal	0.9	---	22.8
Spinach	2.0	4.0	----
Italian Squash	0.2	3.8	----
Brussel Sprouts	0.2	2.9	----

The results show that foods cooked in fluorine containing waters increased in fluorine content in proportion to the concentration of fluorine in the water. Dry foods like oatmeal increase in volume many times over their dry form by absorbing the water in which they are cooked, and as to be expected retain the fluorine. In the consumable form after cooking, the concentration of fluorine in the oatmeal is only somewhat lower than that of the water in which it was cooked. The increase in the fluorine content of cauliflower and cabbage after cooking in water containing 5 parts per million of fluorine is somewhat greater than the increase found for the less penetrable beets and carrots.

CONCLUSIONS

It may generally be concluded from this study as follows:

1. Fluorine is not transmitted in toxic quantities to milk even when the lactating cows are drinking water which has a fluorine concentration far above that which mottles the teeth of children who drink this water during the period of calcification of their teeth.

2. Since natural soils contain only traces of soluble fluorine and the increase in fluorine content of plant foods grown on soils excessively enriched with fluorine have not been found to be proportionally great, it is reasonable to presume that plant foods grown on Arizona soils are not dangerously high in fluorine content.

3. It appears that vegetables do absorb and retain some of the fluorine from the water in which they are cooked. It is not a wise policy, therefore, to cook in fluorine waters, cereals, beans, and other dry foods which absorb large amounts of water during the cooking process. Again, although vegetables show relatively small increases in fluorine content as compared to cereals when they are cooked in fluorine waters, it is important that as little fluorine water as possible be used.