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# THE REMOVAL OF FLUORINE FROM DRINKING WATERS IN THE STATE OF IOWA

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The recent researches of Smith, Lantz, and Smith (1), Churchill (2), McKay (3), Kehr (4), Ostrem, Nelson, Greenwood and Wilhelm (5), Boissevain (6), Dean (7), Sebrell, Dean, Elvove and Breaux (8) and Boruff and Abbott (9) have contributed greatly to our knowledge of the distribution of fluorine in drinking waters and the effect of such waters in the production of mottled enamel of the teeth. Smith, Lantz, and Smith (1), of the University of Arizona, were the first to show that this tooth defect is due to fluorides in the drinking water; they have analyzed the fluoride concentration of waters in endemic areas and have reported it to be high. Dr. H. T. Dean (7), of the United States Public Health Service, has made a recent survey of the occurrence of mottled enamel in the United States and reports that this condition is known to occur in twenty-two states. It is found in other countries also, notably China, Japan, Africa, Italy, Holland, Argentina, Mexico, and England.

The first report of mottled enamel in Iowa and its correlation with fluorides in the drinking water was made by Ostrem, Nelson, Greenwood, and Wilhelm (5) in 1932. These investigators found this condition prevalent at Ankeny, Iowa, and vicinity. Spectrographic examination of the water from this region gave unmistakable evidence of the presence of fluorine. Quantitative analysis of two samples of water from this area by the Churchill modification of the Fairchild Method (2) showed the presence of 10 and 15 parts of fluorine per million of water. The fluorine is confined to deep wells; shallow wells give no qualitative test for fluorine. To the people living in endemic areas the presence of fluorine in the water supply is a grave problem, especially when it occurs in concentrations sufficient to cause mottled enamel. Although mottled enamel has been observed in deciduous teeth, it is far more prevalent in the permanent teeth; and, if the concentration of the fluorine exceeds 2 parts per million in the water, all of the children reared in the community will develop abnormal enamel of the teeth.

Since this condition occurs in the State of Iowa, it became necessary to study the problem from the standpoint of removal of the fluorine, so as to render the water safe to drink without the development of mottled enamel. Work was therefore undertaken in this laboratory along this line and attempts made to remove the fluorine by the addition of aluminum salts. This work had just about started when there appeared an article by Boruff (10) from the University of Illinois in which he states that fluorine may be removed from waters by means of aluminum salts — especially alum. This work seemed so important that we decided to verify results and determine the applicability of the method to some Iowa waters.

## EXPERIMENTAL

Fluorine in the water was determined by the Boruff and Abbott (9) modification of the Willard and Winter method. The water was made definitely alkaline with NaOH to phenolphthalein; 200 cc. of water so treated were placed in a 250 cc. distilling flask and concentrated to approximately 50 cc. Glass beads were employed to prevent bumping. 50 cc. of 1:1 $H_2SO_4$ were then added and the distillate collected; the boiling point of the mixture was kept between 130°C. and 140°C. by adding distilled water through a dropping funnel attached to a capillary tube. The temperature for most efficient results should be held between 130°C. and 140°C. 200 cc. of distillate were collected in an alkaline medium, sufficient to be distinctly alkaline to phenolphthalein. The distillate containing $Na_2SiF_6$ was then concentrated to 20 cc. by evaporation over a free flame and six drops of a 2 per cent alizarin sodium sulphonate solution were then added. 0.2 N HCl was run in drop by drop until the color just disappeared, and then an equal volume of neutral alcohol was added. It is important that the solution at this point be only faintly acid. The mixture was finally titrated with 0.02 N Th $(NO_3)_4$, until there appeared a faint reappearance of a pink color.

Having convinced ourselves that the method of estimating fluorine is satisfactory, we next desired to determine the rate and extent of removal of fluorine from Ankeny city water. The sample of water had a pH of 8.4 and a fluorine content of 7.5 parts per million of water. The pH of the water was adjusted to 7.15 and 20 grains of anhydrous alum, $Al_2(SO_4)_3$, were added to 4 liters of the water in a balloon flask; immediately after the addition of the alum it was stirred for one-half hour with a mechanical stirrer. At the end of this time the contents of the flask were trans-

ferred to a medium-sized glass percolator, corked at the bottom, and allowed to stand. At intervals samples were removed, filtered through qualitative filter paper, and analyzed for fluorine.

Two hours after treatment was begun the fluorine concentration was 1.25 p.p.m.; six hours after treatment was instituted the fluorine concentration dropped to 0.85 p.p.m.; and 24 hours after treatment was begun the concentration of fluorine in the water was 0.40 p.p.m. At this time the pH of the water was 6.78.

Hydrogen ion concentration is also a factor in the removal of the fluorine; a sample of Ankeny city water containing 8.5 parts of F per million was treated with varying amounts of alum, $Al_2(SO_4)_3$, at two pH values. One pH value was that of the original water, namely 7.88, and the other pH was 6.95. The water with a pH of 7.88, when treated with 110 parts per million of alum, contained at the end of two and one-half hours 6.5 parts per million of F; whereas, similar treatment of the water at pH 6.95 removed the fluorine to a level of 5.85 parts per million. 220 parts of alum per million water gave values after $2\frac{1}{2}$ hours of 3.75 p.p.m. F for the water at pH 7.88 and 3.25 p.p.m. for the other water. Four hundred forty parts per million of alum added to the waters with pH of 7.88 and 6.95 gave values after two and one-half hours of 1.13 p.p.m. of F and 0.45 p.p.m. F respectively.

## Summary

The fluorine concentration of Iowa waters may be considerably decreased by alum treatment as suggested by Boruff. Hydrogen ion concentration is a factor in precipitation of the fluorine by alum.

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