A study of the occurrence of fluorine in the drinking water of New Mexico, and the menace of fluorine to health.

John D. Clark

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A STUDY OF THE OCCURRENCE OF FLUORINE IN THE DRINKING WATER OF NEW MEXICO and THE MENACE OF FLUORINE TO HEALTH

by

JOHN D. CLARK AND EDWARD H. MANN
UNIVERSITY OF NEW MEXICO
FLOURINE IN DRINKING WATER

That fluorides had toxic properties was discovered when two chemists, Thenard and Davy became seriously ill from breathing the vapors of hydrofluoric acid while attempting to prepare fluorine in a pure state. Louyet and Nickles later lost their lives from the inhalation of hydrogen fluoride. Soon after the toxic properties of fluorine were well established, concern arose as to the probable toxicity of fluorine contaminated drinking waters and foodstuffs in which the fluorine may occur naturally or in which it might have been used as a preservative.

Black and McKay (1) were the first to describe mottled enamel in the United States when they found it in certain Italian immigrants. Although their findings seemed to point toward a local water supply as being responsible, it was not until 1931 that it was found to be due to fluorine in the water. Proof of this consisted of the facts that water which was suspected of causing mottled enamel in the teeth of humans also caused it in rats, and that the feeding of fluorides to other rats brought about the same condition.

Chemical analyses of waters used by those having defective teeth, and waters not associated with the defect gave added proof to the assumption that fluorine was the causal factor, as the waters associated with mottling contained high concentrations of fluorine and those waters not so associated showed low concentrations. Churchill (2) added further evidence by analysing waters from several parts of the United States, finding high fluorine contents of the waters in all regions in which mottling is endemic.

Mottled enamel is perhaps most prevalent in the arid states of the Southwest, although it has been reported in at least one section in twenty-seven states in the United States, and in every country in the world. The following map, as taken from H. V. and Margaret C. Smith (3), and extended by the use of data kindly furnished by them, shows these
States in which mottled enamel occurs.  
(Also see Text.)

According to H. V. and Margaret C. Smith
regions in the United States; the states in which mottled enamel has been reported (4) being:

Arizona   Louisiana   South Carolina
Arkansas   Minnesota   South Dakota
California Nevada   Tennessee
Colorado   New Mexico Texas
Florida    North Carolina Utah
Idaho      North Dakota Virginia
Illinois   Ohio        Washington
Iowa       Oklahoma    Wisconsin
Kansas     Oregon

In 1931, H. V. Smith and Margaret C. Smith (5) of the University of Arizona made a statewide survey of Arizona, finding more than forty-five scattered communities in which mottled enamel is endemic. The fluorine content of the water supplies in these communities varied from one to six parts per million. The only other extensive statewide survey that had been made at that time was one in Colorado. In April, 1938, Abbott and Voedisch (6) reported the fluorine content of the ground water supplies of North Dakota.

In 1933 the United States Public Health Service sent questionnaires to dentists and health officers of each county in New Mexico, regarding mottled teeth. This survey revealed approximately one-third of New Mexico to be endemic area, particularly the eastern and southwestern sections.

In the first part of 1935, the city of Albuquerque was investigating the possibility of obtaining an additional water supply from the Jemez Mountains. Analyses by the senior author revealed some fluorine, and further study for a period of nearly a year in duration was initiated.

This study was conducted by Clark and Beahm (7) from November, 1935, until August, 1936. During this time samples were obtained from numerous points along streams of the Jemez Country, and, in addition to these, from many other localities whose waters flowed toward Albuquerque.
Mr. Beahm also obtained samples from some fifteen towns throughout the state. Although from most of these, samples were sent in only once, their analyses showed a need for the work now being reported upon.

With some Federal assistance, the State Department of Public Health was able to finance the statewide fluorine survey of New Mexico which was conducted by the authors. Work on this survey was started early in July, 1937, and continued until July, 1938. Endeavor was made to analyze water from every source supplying communities of any size throughout the state. In view of the fact that the fluorine content of a water supply varies over wide limits from time to time, the attempt was made to repeat the analysis of each supply every two weeks. In many cases it was impossible to do this for various reasons, however the averages of the results in these cases are probably not far from correct, when a sufficiently large number of samples were supplied.

The samples were collected by the sanitary inspectors in each of the ten health districts and shipped to the University where they were analyzed by the Sanchis (8) method. At times the waters from certain supplies could not be analyzed by this method, however, these same waters at another date yielded very well to the procedure. In general these waters are not potable for reasons other than their high fluorine content, so that it is not important that these results be included in the survey, although they are given. This survey covered waters from 359 different supplies, of which 157 were in communities of considerable size. Of these 157 communities some thirty-five have water definitely above the danger point of 0.9 parts per million, averaging from 1.1 to well above 12.0 parts per million of fluorine. Twenty-two more communities are on the dividing line between toxic and nontoxic, their averages varying from 0.8 to 1.0 part per million. In this average no community is considered, if from it there were fewer than four samples, and of these there are sixty-five. The map shows the conditions in differ-
ident areas throughout the state. The green* indicates those areas whose water supplies studied, average below 0.8 parts per million of fluorine; the yellow areas are those averaging from 0.8 to 1.0 p.p.m. inclusive; and the red areas are those whose averages are from 1.0 p.p.m. up. The following are data concerning fluorine, which were secured in this work. It is obvious that reliance cannot be placed on the average when few samples were supplied.

*The map has been generalized from data at hand. With increased volume of data, doubtlessly the areas given in color on the map would be changed here and there.
## FLUORINE IN PARTS PER MILLION

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Copies of such detail furnished the fluorine laboratory at the University by the sanitarian or health officer who took the samples, concerning location of samples taken within any community, have been filed with the district health officer of that community, the fluorine content of each water sample being marked on the record. Any citizen interested in securing detailed information concerning his own locality can get it from the health officer of his district.
The results of the survey show that many of our water supplies are high in fluorine, being well above the critical point of 0.9 parts per million at all times, while a great majority of those not above the critical point, as far as averages are concerned, are hovering around it, and perhaps 50 per cent of the time surpass it by a small margin.

When we bear in mind that the map has been prepared from data secured from the analysis of a limited number of samples we cannot be too rigid in our conclusions. However, any community located in the yellow area would do well to insist that more study* be made of its water supplies. The ill effects of intermittent ingestion of fluorides has been pointed out by Dr. Smith (9) in her nutrition studies with rats.

*The Las Vegas High School Science Club, under the direction of Warren E. Portenier, has set a good example in the matter of this further study. This club has covered waters from many sources, particularly waters from privately owned wells in the rural districts. The club has written up a number of case histories of mottled enamel on the teeth of people residing within the field of influence of the school. This is the type of work which can be carried on under direction of a high school chemistry teacher. The University cooperated with Mr. Portenier by supplying materials, etc., and at the initiation of the work, gave some guidance.
THE MENACE OF FLUORINE TO HEALTH

Fluorine poisoning comes under the head of a public health problem because of the widespread occurrence of fluorine in many public and private water supplies.

Cases are on record which show that ingestion of small amounts of fluorides is fatal to both man and other animals. Tappeiner (10) describes the effects of fluorides on dogs, cats, rabbits, and guinea pigs when given by oral ingestion. When 0.5 of a gram of sodium fluoride (NaF) per hundred grams of body weight is given orally, or 0.15 of a gram is given by subcutaneous or intervenous injection, the symptoms are as follows: a condition of drowsiness and weakness; cramps which may attack a single organ or the entire body, and are epileptic in character; paralysis of the vasomotor centers; acceleration and deepening of the breathing with paralysis following; vomiting; secretion from the salivary and tear glands; and, finally death.

Certain investigators have observed that vegetation growing in the vicinity of aluminum factories, from which fluorine is given off in the waste gases, absorbs this fluorine. The herbiverous animals of these vicinities exhibit an endemic toxicosis which has been attributed to the consumption of this vegetation, and the above-stated characteristic symptoms of fluorine poisoning have been reproduced by feeding the suspected forage to guinea pigs.

From the results of many experiments by Smith and Leverton (9) it has been found that the compound of fluorine used, the method and length of time of administration, and individual susceptibilities vary the toxic effects of fluorine to a very great extent. There is evidence, however, that fluosilicates such as sodium fluosilicate (Na₂SiF₆) are more toxic than either sodium fluoride (NaF) or calcium fluoride (CaF₂) and that sodium fluoride is more toxic than the calcium salt.
Typical cases of mottled enamel, showing the destructive effect of fluorine upon human teeth.
There are at least three types of mottled enamel recognized: the mild chalky type, the more severe stained type, and the pitted, corroded type. Plate I, as taken from a paper by Margaret C. Smith (9) of the University of Arizona, shows the three types of mottling, and, for comparison, a set of normal teeth.

In addition to being disfigured, mottled teeth are defective in formation and calcification and are consequently structurally weak. The defect is permanent and, once it has taken place, is irreparable. Certain dental associations have estimated that it would cost $1,000.00 for the dental care of the teeth of the average person with mottled enamel, up to adulthood, at which time the natural teeth must usually be replaced by false.

Mottled enamel has been produced experimentally by H. V. Smith and Margaret C. Smith (9) of the University of Arizona. Their experiments consisted of feeding small amounts of sodium fluoride or water from a supply which contained fluorides, to rats, guinea pigs, and dogs. An idea of the severity of such mottling may be had by comparing the two pictures of Plate II, taken from Dr. Smith’s paper. Picture A shows normal rat incisors and B incisors of a rat which had been fed sodium fluoride.

Experiments show that the fluorine passes into the blood stream and interferes with the calcification of the unerupted teeth. Contrary to common belief, it does not act in the mouth upon the enamel of the erupted portion of the teeth. The teeth of children and adults who have not begun the drinking of water containing fluorine until their second set of teeth have erupted, show no mottling or visible effects of fluorine poisoning.

In humans, as soon as the enamel of the permanent teeth is completely formed and calcified, the enamel organ disappears. The enamel does not regenerate itself but behaves like dead tissue, thus once the teeth are mottled during childhood the mottling is permanent.
a. Normal rat incisors.

b. Mottled incisors of a rat which received sodium fluoride. Note loss of luster and pigment and corrosion of enamel.
The incisors of rats grow continually and regenerate themselves about every thirty days. Experiments at the University of Arizona in which rats were fed fluorides show that it is not the erupted portion of the teeth which is mottled, but the new growth which erupts within two to three weeks after ingestion. Subcutaneous injections of sodium fluoride made on rats also indicated that the fluorine goes to the site of enamel formation. Intermittent injections show that mottling occurs after each injection and that during the intervals between injections normal enamel formation again occurs. Plate III shows the effect of intermittent injections of sodium fluoride. Different effects can be produced by varying the interval between the injections and the amount of fluoride injected. Normal enamel areas alternate with the areas of defective calcification, the extent of which corresponds with the interval between in-

Plate III

Photograph by H. V. and Margaret C. Smith
Effect of intermittent injection of sodium fluoride.
Photograph by H. V. and Margaret C. Smith

a—top; b—center; c—bottom
jections. The mottled appearance of human teeth may be explained by an intermittent use of water which contains fluorine, or a daily variation in the amount of drinking water consumed.

Although it is true that the enamel of adult teeth is unaffected by fluorine, the dentine, which receives nutrients from the blood stream continually and whose composition is subject to change, will suffer. Histological examination shows this to be the case. Plate IV, taken from the work of Dr. Smith (11), shows a microphotograph of the incisor of a rat which received eight injections of sodium fluoride. The incisor shows eight stripes in the dentine. Plate IV B shows a cross-section of the incisor of a rat which had received four injections of sodium fluoride. Stratification in both the enamel and the dentine may be observed.

Conclusions drawn from the above experiments show that the dentine portion of the teeth of adults is effected if they drink water having a high fluorine content. This results in a weakening of the entire tooth structure, even though there be no outward signs of decay.

The bones of a young growing dog, fed very small amounts of sodium fluoride by Brandl and Tappeiner (12) for a period of twenty-one months, were examined microscopically. Deposited in the bones were found vivid glistening crystals which were regarded as being crystalline calcium fluoride. It is believed that soluble fluoride salts are deposited in the bones in considerable amounts, giving rise to an increased brittleness. Feeding fluorine to a rat produces a short, square, and stocky appearance in the skeleton, with the enlarged deformed bones and bowing of the legs typical of rickets. The bones of fluorine fed animals are always chalky and fragile as are the teeth.

Measuring the storage of calcium and phosphorus, the bone forming elements, by determining the balance between the intake and the output shows that fluorine-fed animals retain only half as much as normal animals serving as controls. Fluorine increases the loss or elimination of these elements, and bone development is retarded proportionally.
Addition of calcium to the diet, however, prevents this loss
to a great extent, but has no effect in retarding the mottling
and decay of the teeth. No signs of the bone defects have
so far been observed in children whose teeth are mottled and
it is thought that water has to contain at least six parts per
million before any appreciable effects upon the bone occur.
It seems that the teeth are more sensitive to fluorine than the
bones since a more specific effect is produced on them.

It has been shown by experiments on dogs that the
interference of fluorine with calcium metabolism cannot be
prevented by the intake of vitamin D in the form of cod liver
oil or viosterol, and the effects on the teeth cannot be pre-
vented by an increase in the calcium content of the diet.

Having seen the effects of fluorine on the teeth and
bones, we naturally wonder whether there are not other
effects produced in the body from a continued use of fluorine
contaminated water. It is known that small doses of fluo-
raine, which are still relatively large compared to the amounts
found in natural waters, may go so far as to produce death.

An inhibitory effect on the action of enzymes, char-
acteristic of antiseptics in general, is a property of all inor-
ganic fluorides. Evidence has been established that there is
a specific influence of fluorides on certain enzymatic changes
associated with carbohydrates and fats. The results of a
study conducted by Kastle and Loevenhart (13) on the effect
of antiseptics on the reactions of pancreatic and liver ex-
tracts revealed a harmful effect of most substances studied.

Particularly harmful is the action of sodium fluoride
on the reactions of lipase. Loevenhart and Pierce (14) in-
vestigated the halides of sodium and potassium; the chlor-
ides of calcium, cadmium, barium, and manganese; sodium
nitrate (NaNO₃) and potassium nitrate (KNO₃); disodium
phosphate (Na₂HPO₄); potassium chromate (K₂CrO₄); and
ammonium thiocyanate (NH₄CNS), but none of these sub-
stances showed an inhibitory effect comparable to the action
of fluorides. Solutions of sodium fluoride with a fluorine
content as low as one part in 15,000,000 may inhibit the action of lipase as much as 50 per cent.

Certain phases of the enzymatic breakdown of carbohydrates are particularly sensitive to the fluorine ion. The ability of amylase to produce dextrose is increased by weak solutions of sodium fluoride.

It has been shown that the fluoride ion retards glycolysis in the blood by preventing glycerophosphoric acid from changing into phosphoglyceric acid and hence into pyruvic and lactic acids.

There is a small amount of fluorine in normal blood and it has been shown that continued ingestion of fluorides may increase this amount many fold. Stuber and Lang (15) observed a number of cases of hæmophilia in which the fluorine content of the blood was abnormally high. There seemed to be a correlation between the high fluorine content of the blood and the prolonged time of coagulation, and they suspected that the fluorine may be the cause of this condition. Knowing that goose blood and rabbit blood clots slowly, an investigation was made, and it was found that fluorine was present in large amounts in the blood of these animals. Cat and dog blood, on the other hand, clots rapidly and was found to be free from fluorine.

Continuing their observations they found that as a whole, individuals residing in places where the fluorine content of their drinking water was high, had a coagulation time of six to twenty times that of a normal individual drinking fluorine-free water.

Stuber and Lang are of the opinion that the retarding effect on glycolysis may be responsible for the long clotting time.

In closing the topic of the physiological effects of fluorine, it may be stated that recently Goldemberg (16) reports that by the ingestion of only two milligrams of fluorine per day for a period of six months, the thyroid gland in rats may be enlarged five to six times in volume, thus showing that fluorine poisoning exhibits a very specific effect on the
thyroid gland. Others do not confirm this work. It has been suggested that fluorine may play a very important part in endemic goiter.

REMOVAL OF FLUORINE FROM DRINKING WATER

A good summary of many of the chemical and physical principles which investigators have sought to use in the removal of fluorine from water is to be found in the American Journal of Public Health of April, 1935. Industrial and Engineering Chemistry, in many of its numbers printed during the past ten years, gives accounts of researches, many by large commercial organizations, seeking large-scale methods for fluorine removal.

H. V. and Margaret C. Smith reported a moderate cost method of fluorine removal in the November 10, 1937, Water Works Engineering. Their work is cited in the January, 1938, Industrial and Engineering Chemistry as one of the outstanding accomplishments in chemistry of 1937.

The authors of this bulletin concerning fluorine in New Mexico, have also sought a method of removal of fluorine, restricting their very few and preliminary studies to such methods as might, if successful, be used by the average housewife, in the average kitchen (and the authors have had in mind the isolated ranches in the Southwest). They have taken commercial bone ash, which can be purchased already for use, and have heated water to which small amounts of this material was added. After heating, the bone ash settled to the bottom of the container. About 80 per cent of the water samples so treated (waters not too high in fluorine) have had their fluorine lowered from toxic to non-toxic quantities by this treatment. For waters of over 3.0 parts per million of fluorine, the bone ash was first put into a flask and covered with water. Hydrochloric acid was added little at a time. The bone ash dissolved. Then to the solution of the bone ash in the acid, baking soda was added until effervescence ceased. This neutralized the acid. With the neutralization of the acid solution, by the baking
soda, the bone ash reprecipitated in flocculent form. This precipitate was filtered, washed and used, as was the untreated bone ash as mentioned above. In every case tried, in this preliminary study, this treated bone ash has made an almost complete removal of all fluorine. A five-gallon bottle of water and an electric "bayonet" or immersion heater were found very convenient in the laboratory.

The authors are now starting a detailed investigation of the possibility of removal of fluorine by the use of treated and untreated bone ash. Work already done has to be repeated. Varying quantities of bone ash have to be used, varying rates and time of heating must be tried, and varied treatments of bone ash must be attempted before any one best method, for water from any locality, can be determined, if the results obtained in the few preliminary studies, are confirmed and amplified by further investigation.

BIBLIOGRAPHY OF LITERATURE CITED