Cariostatic Effect and Metabolism Of Ammonium Fluosilicate

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THE EQUAL EFFECTIVENESS of sodium fluosilicate and sodium fluoride in reducing dental caries in the white rat, given at a level of 50 p.p.m. fluorine in drinking water, has been demonstrated by the National Institute of Dental Research, Public Health Service (*1*).

Although sodium fluosilicate, Na_2SiF_6, is cheaper than sodium fluoride, NaF, its use for the fluoridation of municipal water supplies may be limited to some extent by its solubility. Ammonium fluosilicate, $(NH_4)_2SiF_6$, is considerably more soluble than either of these two fluorine compounds, and it is also cheaper than sodium fluoride.

The price (*2*) of the three fluorine compounds and their solubility (*3*) are as follows:

	Price in cents		*Solubility*
	Per lb.	*Per lb. of F*	*Gm. per 100 gm. H_2O at 17.5° C.*
Na_2SiF_6	7.0	10.6	0.65
$(NH_4)_2SiF_6$	11.0	16.0	18.5
NaF	11.5	25.5	4.3

The high solubility and high fluorine content of ammonium fluosilicate make it especially useful in small water plants. It may be used in either liquid or dry chemical feeders. By a special process, it has been made free flowing to prevent clogging of the dry feeder (*4*). The treatment of some water requires the addition of ammonia to reduce the odor and taste, and in this case ammonium fluosilicate may be particularly applicable because it may eliminate the ammoniation operation. It appears, therefore, that ammonium fluosilicate would meet the engineering requirements common to most fluoridation installations as described by Maier (*5*, *6*).

In addition, a fluorine compound suitable for fluoridation must meet certain physiological requirements. This study, therefore, presents laboratory data on physiological properties of ammonium fluosilicate pertinent to its use as a practical water fluoridating agent. Sodium fluoride was used as a basis of comparison.

The properties studied in the growing white rat were: (*a*) the inhibition of dental caries; (*b*) development of incisor striations; (*c*) the deposition of fluorine in bones and teeth; (*d*) calcification of bones as determined by ash content; (*e*) average daily weight gain; and (*f*) any gross evidence of untoward physiological effects.

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Materials and Methods

The sodium fluoride used in this study was an analytical grade reagent and its purity was accepted according to assay. The purity of the

Table 1. Daily gain in weight, caries inhibition, deposition of fluorine, and percent ash in bones and teeth of rats receiving distilled water and different fluoride solutions

Item	H_2O	NaF	$(NH_4)_2SiF_6$	NaF+ Na_2SiO_3+ $(NH_4)_2CO_3$
Number of rats	22	24	21	23
F intake (mg.)	0	60. 7	59. 8	59. 9
Days on experiment	99	99	99	99
Initial weight (gm.)	50	50	51	49
Final weight (gm.)	190	190	180	185
Average daily gain (gm.)	1. 4	1. 4	1. 3	1. 4
	Caries diagnosis			
Number rats with caries	19	15	13	15
Percent rats with caries	86. 4	62. 5	61. 9	65. 2
Number carious teeth per carious rat	3. 0	2. 3	2. 4	2. 3
Number carious areas per carious rat	6. 2	3. 1	3. 2	2. 8
Caries score per carious rat	14. 1	6. 6	6. 7	5. 6
Fluorosis index	0	1. 9	2. 0	2. 0
	Percent fluorine in bones and teeth (ash basis)			
Molars	. 006	. 216	. 226	. 236
Incisors	. 002	. 168	. 180	. 163
Mandibles	. 005	. 419	. 439	. 430
Femurs	. 005	. 452	. 462	. 477
	Percent ash in bones and teeth			
Molars	75. 8	76. 1	75. 6	75. 3
Incisors	76. 1	76. 0	76. 5	75. 7
Mandibles	70. 0	71. 3	70. 7	71. 1
Femurs	63. 9	63. 1	64. 3	64. 8

ammonium fluosilicate was assessed by analysis for F, Si, and N (Kjeldahl), and the percent of theoretical purity found was 101.7, 96.0, and 100.1, respectively.

Thirty litters of four female weanling Osborne-Mendel rats were divided into 4 groups of 30 rats each. Group I received distilled water (control); group II received 50 p.p.m. F as NaF; group III received a solution of $(NH_4)_2SiF_6$ containing 50 p.p.m. F; and group IV received a composite fluoride solution of the following composition: 0.1105 gm. NaF, 0.3538 gm. $Na_2SiO_3 \cdot 9H_2O$, and 0.0421 gm. $(NH_4)_2CO_3$, made up to 1 liter. The composite solution contained 50 p.p.m. F and furnished a concentration of silicon and ammonium ion equivalent to that provided by the ammonium fluosilicate solution. Use of this solution made it possible to observe the relation of these ions to the effect of fluoride. The pH of all drinking solutions was adjusted to 5.5–6.0. The cariogenic diet (*1*) and all drinking fluids were ingested ad libitum, and fluoride intake was equalized throughout the experiment by substituting distilled water for the fluoride drinking fluid for short periods of time.

At the end of 99 days the surviving animals were killed and the molar teeth scored for dental caries according to Cox and associates (*7*). The degree of incisor fluorosis was arbitrarily scored 0, 1, 2, or 3, that is, none, mild, moderate, and severe, and a mean fluorosis index was calculated for each group. The femurs, mandibles, and molar and incisor teeth of 10 rats of each group, nearest the mean weight for that group, were pooled by tissue, dried, extracted with alcohol and ether, and ground to pass through a 60-mesh sieve. All tissues were then ashed at

550° C. for 3 hours and analyzed for fluorine (*8, 9*).

Results

The caries inhibiting effect of the fluoride solutions and the deposition of fluorine in the bones and teeth are shown in table 1.

There was no gross evidence of toxicity in the rats receiving either ammonium fluosilicate or sodium fluoride, and the average daily weight gains were comparable for each group. In addition, no difference was found in the severity of fluorosis produced in the three groups of experimental rats receiving the different fluoride solutions. The fluorosis indexes for the four groups of rats were 0.0, 1.9, 2.0, and 2.0, respectively (table 1).

Ammonium fluosilicate was as effective as sodium fluoride in inhibiting caries. There was a 28.4 percent and 27.7 percent reduction in the caries incidence and a 52.5 percent and 53.2 percent reduction in the caries score per carious rat in the animals receiving the ammonium fluosilicate and sodium fluoride solutions, respectively.

The chi square test was used to test the difference in the caries incidence between the control and treated rats according to the following formula:

$$\chi^2=\frac{T(AD-BC-T/2)^2}{(A+B)\ (C+D)\ (A+C)\ (B+D)}$$

where T=Total number of rats (90)
A=Number of carious control rats (19)
B=Number of noncarious control rats (3)
C=Number of carious experimental rats (43)
D=Number of noncarious experimental rats (25)

The difference was found to be significant at the 5 percent level using a one-sided significance test ($\chi^2=3.14$).

The caries scores per carious rat, shown in table 1, were compared by analysis of variance (*10*), and the probability (p) values are found in table 2.

For each of the caries categories tested as shown in table 2, the caries results for the experimental groups are significantly lower (table 1) than those for the control group, and no significant differences exist between the groups receiving different fluoride solutions.

No significant differences are apparent in the deposition of fluorine in the bones and teeth of rats on the various fluoride regimens (table 1). Similarly, no significant differences are found in the ash content of bones and teeth of rats on the various fluoride regimens. These current data on caries inhibition, fluorine deposition, and ash agree closely with similar data obtained in a previous study (*1*).

Table 2. Statistical comparison of caries experience: "p" values of differences between groups

Group	Carious teeth per carious rat	Carious areas per carious rat	Caries score per carious rat
Significance of differences between control and experimental fluorine groups	<0. 05	<0. 01	<0. 01
Significance of differences between experimental fluorine groups	>. 05	>. 05	>. 05

Hence, it appears that ammonium fluosilicate and sodium fluoride were similar with respect to all the criteria investigated. There were no significant differences in the average daily weight gains, the degree of fluorosis, the reduction of dental caries, and in the amounts of fluorine and ash deposited in the bones and teeth.

Discussion

It is not surprising that ammonium fluosilicate proved to be similar to sodium fluoride at a level of 50 p.p.m. F in drinking water in its cariostatic effect and in its ability to deposit fluorine in bones and teeth of the white rat. Previous comparisons between sodium fluoride and sodium fluosilicate at 50 p.p.m. F in drinking water have shown no differences in the amount of fluorine deposited in bones and teeth (*8*), in the extent of incisor fluorosis, and in the reduction of caries (*1*).

Theoretically, ammonium fluosilicate would yield fluorine ion quantitatively upon hydrolysis according to the following scheme (*11*):

$$(NH_4)_2SiF_6 \rightleftharpoons 2NH_4^+ + SiF_6^=$$
$$SiF_6^= + 2HOH \rightarrow SiO_2 + 6F^- + 4H^+$$
$$(NH_4)_2SiF_6 + 2HOH \rightarrow 2NH_4^+ + 6F^- + 4H^+ + SiO_2$$

These reactions also indicate that solutions of ammonium fluosilicate would have an acid reaction and in fact did have a pH of 3.4–3.6.

In an acid medium, such as the gastric juice, the silicon of Na_2SiO_3 would be present as SiO_2 and hence should react like the silicon of $SiF_6^=$ according to the following reactions:

$$Na_2SiO_3 \rightleftharpoons 2Na^+ + SiO_3^=$$
$$SiO_3^= + 2H^+ \rightarrow SiO_2 + H_2O$$
$$Na_2SiO_3 + 2H^+ \rightarrow 2Na^+ + SiO_2 + H_2O$$

The composite solution was similar to the ammonium fluosilicate and sodium fluoride solutions according to the criteria investigated. It appears, also, that neither the ammonium ion nor silicon interfered with the physiological reactions characteristic of the fluoride ion.

The results of this study indicate, therefore, that $(NH_4)_2SiF_6$ is hydrolyzed in dilute solution and behaves physiologically like the solution of NaF.

Conclusions

1. A comparison was made of the ability of sodium fluoride, NaF, and ammonium fluosilicate, $(NH_4)_2SiF_6$, solutions containing 5 p.p.m. F to reduce caries and deposit fluorine in the bones and teeth of the white rat.

2. No differences were observed in the amount of fluorine and ash deposited in the molars, incisors, mandibles, and femurs.

3. There was no difference in the rate of growth among all groups of rats.

4. There was no difference in the production of incisor striations.

5. Ammonium fluosilicate was as effective as sodium fluoride in inhibiting caries in the white rat.

6. The data suggest that ammonium fluosilicate may be equally as effective as sodium fluoride as a fluoride carrier for the fluoridation of municipal water supplies.

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