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# FLUORIDE

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## LUNG CANCER IN A STEEL CITY

### A Personal Historical Perspective

Almost twenty years have passed since my first study on "Lung Cancer in a Steel City - Its Possible Relation to Fluoride Emissions" (1). This was followed two years later by "Further Observations on Cancer in a Steel City" (2). Both studies presented evidence of a connection between industrial air pollution in Hamilton and the high mortality rates for cancer of the lung, as well as cancer of the gastro-intestinal tract and genito-urinary system. The zonal distribution of deaths revealed a definite geographic pattern - a "cancer belt" - quite similar to the isopleth lines of equal concentrations for many atmospheric pollutants (including fluorides) previously reported by the Ontario Ministry of the Environment and by Environment Canada. The highest mortality rate for lung cancer was in the northeast area of the city, close to the large steel mills. The victims were mostly men who had worked therein or had resided nearby for many years, although some had moved away after retiring.

The major source of the excessive amounts of fluorides in the Hamilton atmosphere is the huge amounts of fluorspar (Spar) used in steelmaking. Thousands of tons of this calcium fluoride are used daily by the two huge and one medium-sized steel mills in Hamilton; all three mills are located in the northeast section of the city. An analysis of dust from one Hamilton steel mill gave the following result (a "control" dust sample is shown in brackets):

|          |          |         |
|----------|----------|---------|
| Arsenic  | 4.3 ppm  | [1.8]   |
| Cadmium  | 1.6 ppm  | [0.2]   |
| Fluoride | 3.46%    | [0.063] |
| Lead     | 160 ppm  | [13]    |
| Mercury  | 0.26 ppm | [0.08]  |
| Zinc     | 1100 ppm | [70]    |

The most striking aspect of the dust composition is its high fluoride content of 3.46% or 34,600 ppm, 55 times higher than in "control" dust. Fluoride can be emitted from open hearth steel furnaces both as gaseous and particulate matter, i.e. HF and SiF<sub>4</sub> (3); also the fluoride content of SiF<sub>4</sub> is 73%. Therefore, it doesn't take much SiF<sub>4</sub> contamination to yield a dustborne fluoride content that is 3.46%. Also relevant is the observation that fluoride fallout values near a fluoride-emitting factory can be 90 times higher than in a more distant zone (4).

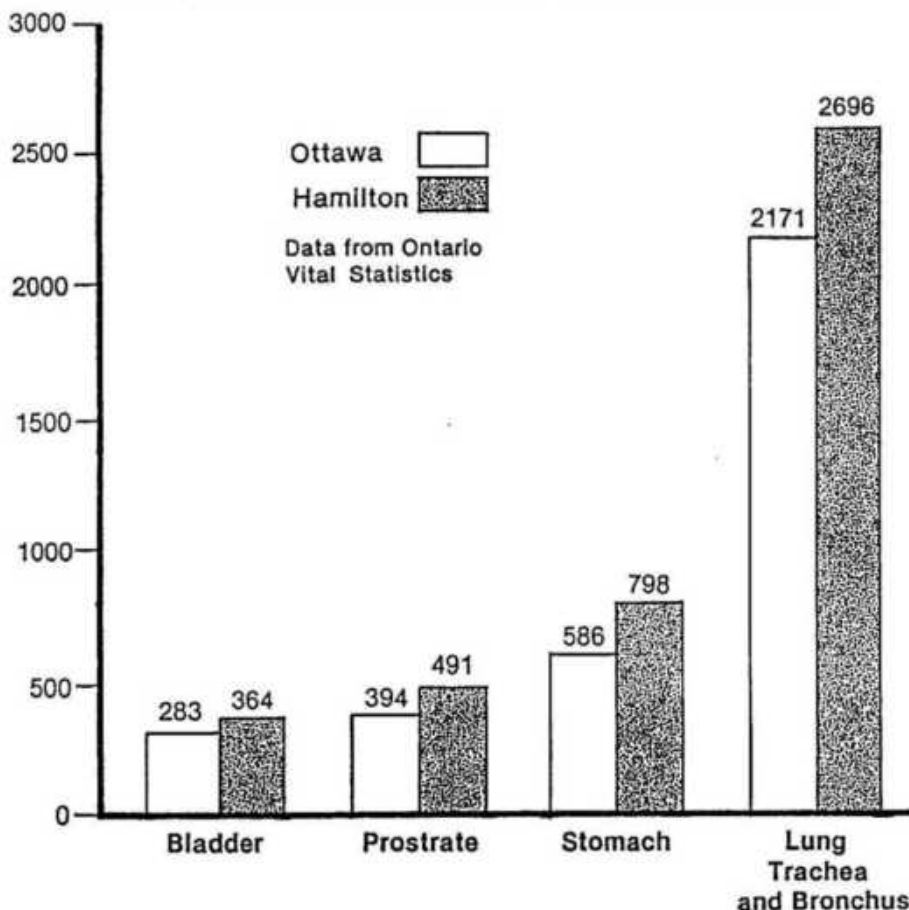
In my 1972-74 studies, the male death rate from lung cancer in the most heavily polluted residential zone was 65/100,000, which was 2.83 times higher than the national average of 23/100,000 (2), compared with a 2.42 times higher rate reported in a 1988 study of the same zone involving a correction for age which reduced the ratio to 1.99, along with an additional correction for smoking which further decreased the ratio to 1.40 (5). In terms of my own experience, I suspect that the correction(s) for smoking might be excessive, because the lung cancer victims which I studied had not smoked as many cigarettes as did their "white collar" colleagues. Nevertheless, the main point to remember is that even a suspected over-correction of the raw data revealed a significant difference in lung cancer deaths between the heavily polluted zone and other areas, even when based on "guesstimates" calculated 14 years later.

Figure

## Smoking vs Heavy Industrial Pollution

Comparison of Cancer Deaths (4 sites) in Hamilton and Ottawa during the 20-year period 1966-1985. Ottawa and Hamilton are the same population size, but they differ greatly in pollution levels. Hamilton is a heavy industry, manufacturing center, while Ottawa is a city of light industry with the Canadian federal government as a major employer.

Both Smoking and Heavy Industrial Pollution are recognized as being able to cause many types of cancer. About twice as many cigarettes were sold in Ottawa during this 20-year period, compared with sales in Hamilton.



Another point to consider is that, of the 300-or-so histological examinations of lung cancer tissue I had reported, 48% were of the small undifferentiated "oat cell" type of cancer. This kind of cancer usually constitutes 20%-or-less of lung cancers in male smokers (1,2). The same type of "oat cell" cancer

also predominated in Newfoundland fluorspar miners (6). In this connection, the conclusions of Little *et al.* are extremely relevant, i.e.,

The high incidence of bronchial cancer in Newfoundland fluorspar miners is of interest . . . . If we assume that the age distribution and smoking histories are comparable, the incidence of bronchial cancer appears to be at least five times higher among the Newfoundland fluorspar miners than among the Colorado uranium miners for similar radiation exposures . . . . [and] suggests that an additional factor or co-carcinogen is present, and the possibility that fluorspar itself is the co-carcinogen (7).

The authors of the 1988 Hamilton study mention co-carcinogens, but did not discuss fluoride(s) (5).

Finally, the fact that fluoride has recently been implicated as a likely carcinogen responsible for osteosarcomas (8) should warn epidemiologists not to ignore the carcinogenic potential of atmospheric fluoride(s).

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\*\*\*\*\*

V.A. Cecilioni  
740 Barton Street East  
Hamilton, Ontario L8L 3B1, Canada

## DISFIGURING, OR "WHITE AND STRONG"?

by

John Colquhoun\*  
Auckland, New Zealand

**SUMMARY:** Two cases of dental fluorosis in 8-year-old children who grew up in fluoridated Auckland, New Zealand, are described. Recent studies are examined with attention to the prevalence, and variability in assessment of acceptable appearance, of dental fluorosis. Recent statistics on caries prevalence suggest that fluoride intoxication has impeded caries reductions.

**KEY WORDS:** Dental caries; Dental fluorosis; Fluoridation; New Zealand.

### Introduction

In another paper (1), the highly subjective aspect of caries diagnosis, and the way such diagnosis can influence and be influenced by fluoridation research, was described. Similar subjective considerations may influence assessments of disfigurement (or "cosmetic defect" - various terms are used) resulting from dental fluorosis.

In 1983, when Principal Dental Officer in Auckland, I stated in a letter to the Director-General of Health: "The evidence now suggests that the provision of fluoride to children is out of control, with many children suffering the effects of toxic levels of intake." I suggested that water fluoridation be discontinued. The evidence was later published (2). Dental fluorosis, the diffuse, symmetrically arranged tooth mottling which is a manifestation of fluoride intoxication, affected 25% of children in fluoridated central Auckland. Visible fluorosis of front teeth affected 10%, and 3.6% had discolored or pitted fluorosed enamel. It was suggested that either of the last two categories of the condition could be described as "disfiguring." In the non-fluoridated areas only 5% of the children had the condition - very mildly except for the few who had been given fluoride tablets.

In this paper two cases, children examined in the survey who grew up in fluoridated Auckland, are briefly described. Other research which reported cases of unsightly dental fluorosis in fluoridated areas is reviewed.

---

\* Education Department, University of Auckland, Private bag, Auckland, New Zealand. Note: The author was formerly Principal Dental Officer, Department of Health, Auckland, New Zealand, Convenor of the Fluoridation Promotion Committee of the N.Z. Dental Health Foundation and President of the N.Z. Society of Dentistry for Children. He is currently a research fellow in the Education Department, University of Auckland.

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### Two Cases

Figures 1 and 2 are photographs of teeth of 8-year-old children with dental fluorosis who grew up in fluoridated Auckland, New Zealand. The first, classified as "mild" by the traditional Dean classification, shows that the condition can be quite visible. This child's parents had noticed it and were concerned. The child was bottle-fed as an infant and according to the parents was extremely thirsty both in infancy and during pre-school years — that is, up to the age of 5 years when the permanent teeth were developing. The only known source of fluoride was the fluoridated drinking water. The second case has a severer form of the condition, called "moderate" in the Dean classification. Discoloration and pitting of the chalky porous enamel has occurred. This child was also bottle-fed, but in addition swallowed fluoride toothpaste from an early age. The parents were unaware of any hazard from fluoride, having been assured that both fluoridated water and fluoride toothpaste were "perfectly safe". In Auckland's non-fluoridated areas the only children with comparable dental fluorosis had been given fluoride tablets regularly since birth — at the then recommended dose which has since been drastically reduced. According to fluoridation proponents, this sign of fluoride intoxication is only a "cosmetic defect", not a health hazard.

Figure 1



Figure 2



#### **Dental Fluorosis in 8-Year-Old Children in Fluoridated Auckland, New Zealand**

The first case (left), classified as "mild" according to the traditional Dean classification, shows that the condition can be quite visible.

The second case has a severer form of the condition, called "moderate" in the Dean classification. Discoloration and pitting of the chalky porous enamel has occurred.

(Reproduced from color photographs by Jan Caris)

### Other Studies

Since the above-mentioned Auckland study was published, other New Zealand studies have reported similar prevalences, in fluoridated areas, of such fluoride-related diffuse tooth mottling (3-6), and significantly lower prevalences in non-fluoridated areas (3-7). In the U.S.A., though some authors claim that prevalences were little or only slightly greater than expected (8-11), most recent studies report very high prevalences (12-15), as Table I shows.

Table 1

Percentages of children with dental fluorosis, from recent comparable New Zealand and U.S.A. studies, in "optimal" (or near-optimal) fluoride, and low-fluoride, water areas.

| Community                               | Age of Children (years) | F conc. (ppm) | Percent with Dental Fluorosis | Source |
|---|-------------------------|---------------|-------------------------------|--------|
| <b>Optimal:</b>                         |                         |               |                               |        |
| Auckland (NZ)                           | 7-12                    | 1.0           | 25                            | (2)    |
| Auckland Region (NZ)                    | 9                       | 1.0           | 25                            | (3)    |
| Hastings (NZ)                           | 10                      | 1.0           | 23                            | (5)    |
| Kewanee, IL                             | 8-10,<br>13-15          | 1.0           | 28 <sup>1</sup>               | (12)   |
| Kerrville, TX                           | 7-18                    | 1.4           | 16                            | (14)   |
| Angleton, TX                            | 7-18                    | 1.3           | 33                            | (14)   |
| Alvin, TX                               | 7-18                    | 1.3           | 29                            | (14)   |
| Kingsville, TX                          | 7-18                    | 1.0           | 39                            | (14)   |
| Richmond, MI                            | 6-12                    | 1.2           | 51                            | (13)   |
| Redford, MI                             | 6-12                    | 1.0           | 48                            | (13)   |
| Hudson, MI                              | 6-12                    | 0.8           | 32                            | (13)   |
| New York State                          | 12-17                   | 1.0           | 27 <sup>2</sup>               | (15)   |
| <b>Low Fluoride:</b>                    |                         |               |                               |        |
| Ricmond (NZ)                            | 12-14                   | 0.2           | 6 <sup>3</sup>                | (7)    |
| Auckland (NZ)                           | 7-12                    | 0.2           | 4                             | (2)    |
| Auckland Region (NZ)                    | 9                       | 0-0.2         | 15 <sup>4</sup>               | (3)    |
| Napier & adjacent small town/rural (NZ) | 10                      | 0-0.2         | 3 <sup>3</sup>                | (15)   |
| Iowa towns                              | 8-16                    | 0.0           | 3                             | (10)   |
| San Antonio, TX                         | 7-18                    | 0.4           | 2                             | (14)   |
| San Marcos, TX                          | 7-18                    | 0.3           | 8                             | (14)   |
| N. Braunfels, TX                        | 7-18                    | 0.3           | 9                             | (14)   |
| Cadillac, MI                            | 6-12                    | 0.0           | 12                            | (13)   |
| New York State                          | 12-17                   | 0-0.3         | 4 <sup>2</sup>                | (15)   |

<sup>1</sup> Estimated. Survey 5 years earlier reported child prevalence 15% (8) and tooth prevalence 15% (9). This survey reported no child prevalence but tooth prevalence 28% (12).

<sup>2</sup> Front teeth were not scored.

<sup>3</sup> Among children who received no fluoride supplements.

<sup>4</sup> 55% of the children received fluoride supplements.

In the New Zealand studies the percentages are for the proportion of the condition described as "diffuse opacities" which were of "homologous", "bilateral" or "contralateral" distribution. In other studies, and Colquhoun's (2), the same condition was termed "dental fluorosis".

The authors of the later New Zealand studies, like the American authors, support fluoridation and presented their data from a proponent perspective. They omitted my first Auckland study from their reference lists. Nonetheless, their data supported my original Auckland findings.

A study the following year in the Auckland Region (3) reported that 19% of 9-year-olds in fluoridated areas had "diffuse mottling" affecting their upper front teeth, and 29% had the condition when all their teeth were examined. However, it was stated that 85% of the diffuse mottling was bilateral in distribution (that is, arranged symmetrically each side of the mouth, which is the description of dental fluorosis). So the prevalences of dental fluorosis were actually 25%, and 15% with front teeth affected. In the non-fluoridated areas, among children who had not been given fluoride tablets, only 4.25% of children had front teeth so affected. The prevalence among such children in the non-fluoridated areas when all teeth were examined was not reported. The authors suggested a reduction in the water fluoride level to less than the existing 1 part per million, in order to reduce the amount of mottling. They claimed that less than 1% of the children had "cosmetically poor" teeth due to fluoride, and another 1.5% were "cosmetically doubtful". But the examiner had used a purely subjective assessment, and had earlier been reported to say that the mild mottling caused by fluoride "can be attractive; it just makes the teeth look very white and strong" (16).

Other studies (4,5) have reported a rise in very high prevalences of diffuse mottling in fluoridated Hastings. The prevalence of the symmetrical kind (dental fluorosis) was 23% in the more recent study, and only 3% in surrounding non-fluoridated areas. Front-tooth "diffuse mottling" of "unsatisfactory appearance" affected 2.7% of children in Hastings and 1.5% of children in the unfluoridated area. However, these percentages include non-symmetrical mottling. The percentages with dental fluorosis of unsatisfactory appearance were not reported. A longitudinal study by one of the authors (6) reports that, over three years, there was an increase in discoloration and pitting of front teeth and "a deterioration in tooth appearance" of Hastings children who at a younger age had only mild fluoride mottling. These authors also suggested ways to reduce fluoride intake, but favored lowered tablet doses and lowered fluoride content of toothpastes, rather than lowered water fluoride content, because of the lower dental decay prevalence among "non-European" (Maori) children in fluoridated Hastings.

The case for a current fluoridation benefit to permanent teeth in New Zealand relies on the above Hastings and environs study. In it, socio-economic differences between the compared groups were not reported. Three studies have reported decay prevalences inversely related to income levels (17-19). Also, since Hastings has been fluoridated since 1953, the decay reductions reported there in recent years could not be caused by fluoridated water, because all the children studied had consumed such water all their lives.

The New Zealand estimates of unsightly fluorosis are summarized in Table 2. Most of the U.S.A. studies report unsightly dental fluorosis in "optimal" water fluoride areas. However, in the study which claimed the lowest overall prevalence (8) it was reported that 2.4% of children in "optimal" Kewanee had "moderate to severe" dental fluorosis, according to the Dean classification. Those categories include some discoloration and pitting of the fluorosed

Table 2

Percentages of children with dental fluorosis assessed by examiners as aesthetically unacceptable, from recent New Zealand studies in fluoridated areas.

| Community       | Age of Children (years) | Percent with Unsightly Dental Fluorosis | Description and Source                                 |
|-----------------|-------------------------|---|--|
| Auckland        | 7-12                    | 10.0<br>3.6                             | "Visible on front teeth"<br>"Discolored or pitted" (2) |
| Auckland Region | 9                       | 2.5                                     | "Cosmetically poor"<br>or "doubtful" (3)               |
| Hastings        | 10                      | 2.7*                                    | "Unsatisfactory appearance"<br>(5)                     |

\* Included non-symmetrical diffuse mottling.

enamel. The authors' follow-up study using a new classification system (9) reported that in the Kewanee children 1% of all tooth surfaces affected by fluorosis were "characterized by staining, pitting or both".

#### Fluorosis and Caries

The justification for continuing to add fluoride to drinking water is the reduction in dental caries which is claimed to result. In 1986, at the ISFR Conference in Utah, I presented the then most recent (1984) data on the dental status of all 12- and 13-year-old children completing their final year of treatment by the School Dental Service, in which 98% of the child population of New Zealand are enrolled. Officially collected School Dental Service data from the six main, largely urban, population areas showed that in non-fluoridated Christchurch children had dental health comparable to that in the five other main population centers, which are fluoridated. In fact, slightly more children were free of dental decay in Christchurch, and the number of decayed, missing and filled teeth ("mean DMFT") differed by only a small fraction of a tooth. The data were later published in *Community Health Studies*, the peer-reviewed journal of the Australian and New Zealand Public Health Association (19). Just before leaving New Zealand I received from the Minister of Health more recent data. The updated information, added to that presented earlier, is shown in Table 3. In Christchurch in 1986 even more children were free of dental decay, and the number of decayed, missing and filled permanent teeth is the same as, or lower than, in many of the large fluoridated centers.

Older (12- and 13-year-old) children in the combined fluoridated areas of New Zealand still have slightly less dental decay than in the combined non-fluoridated areas (the differences being 0.2 DMFT and 1% caries-free). But, as explained, that comparison is of different kinds of population: one mainly in cities and large towns; the other predominately in rural areas or small towns (19). When similar populations are compared, dental decay is now less in non-fluoridated areas.

Table 3

Caries-free percentages and mean DMFT (number of decayed, missing and filled permanent teeth) in 1984 and 1986 for all form II (12- and 13-year-old) children from all school dental clinics in non-fluoridated Christchurch and in the other, fluoridated, main population areas of New Zealand.

| Center                  | 1984              |               |           | 1986              |               |           |
|-------------------------|-------------------|---------------|-----------|-------------------|---------------|-----------|
|                         | (No. of Children) | Caries-free % | Mean DMFT | (No. of Children) | Caries-free % | Mean DMFT |
| <b>Non-fluoridated:</b> |                   |               |           |                   |               |           |
| Christchurch            | (4658)            | 20.9%         | 3.2       | (4419)            | 26.6%         | 2.9       |
| <b>Fluoridated:</b>     |                   |               |           |                   |               |           |
| Greater Auckland        | (14323)           | 20.1%         | 3.1       | (13045)           | 24.8%         | 2.8       |
| Hamilton                | (2865)            | 15.3%         | 3.5       | (2596)            | 22.0%         | 2.9       |
| Palmerston North        | (1401)            | 19.7%         | 3.2       | (1215)            | 20.3%         | 3.1       |
| Wellington              | (6216)            | 20.8%         | 3.0       | (5870)            | 26.1%         | 2.6       |
| Dunedin                 | (1385)            | 17.7%         | 2.9       | (1338)            | 23.5%         | 2.9       |

**Explanatory Note:** Children from lower income areas usually have poorer dental health (17,18). Christchurch has had a lower average income than the fluoridated main centers (19). Christchurch has very few children of indigenous Maori descent (claimed today to have poorer dental health). This point was discussed in the earlier paper (19). No studies show poorer Maori dental health when ethnic groups of similar socio-economic status are compared. Dunedin, with poorer child dental health than Christchurch, also has very few Maori children.

The new information also shows that, nationally, dental decay reductions are now greater in non-fluoridated areas. From 1980 to 1987, mean DMFT has declined by 41% in fluoridated areas (from 4.6 to 2.7) and by 48% in non-fluoridated areas (from 5.6 to 2.9). The percentage of children who are free of decay has increased 3.3 times in the former (7.6% to 25.4%) and 3.8 times in the latter (6.5% to 24.4%). By 1980, the children who grew up in fluoridated areas had drunk fluoridated water all their lives. Therefore, the above reductions in decay could not be due to fluoridated water.

### Discussion

There is thus increased evidence of fluoride intoxication, as well as increased evidence that there is little if any dental benefit from fluoridation. In the past, "population mobility" and "fluoride in the food chain" have been invoked to explain the lack of differences in child dental health between fluoridated and non-fluoridated communities. Such factors cannot explain a reversal of the claimed effect of fluoridation. A reasonable hypothesis seems to be that fluoride in the drinking water has impeded decay reductions.

When fluoridation was introduced it was predicted that only 10 to 12 percent of children would have dental fluorosis, so mild it would be "detectable

only on close expert examination" (20). Could not other adverse effects have been similarly underestimated? Concern should center on degree of toxicity rather than on amount of "cosmetic impairment". The claim that only tooth-forming cells are damaged by fluoride is extremely implausible, contrary to common sense, and can be disputed on scientific grounds. There is evidence of more general harm (21,22).

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### SOME INVESTIGATIONS INTO THE "DMF" MEASUREMENT OF FLUORIDE DENTAL BENEFIT

by

John Colquhoun\*  
Auckland, New Zealand

**SUMMARY:** A survey of 73 dentists' treatment records for 14- to 16-year-old children in central Auckland, New Zealand, is described. Average number of fillings inserted varied between 0.25 and 8.50 per child per year. The effect of such variable diagnostic standards on measured "DMF" scores in some early and recent fluoridation studies is discussed. A hypothesis is advanced to explain the origin of the "DMF" measure.

**KEY WORDS:** Auckland, N.Z.; Dental caries; Dental diagnosis; "DMF" measurement; Filling rates; Schoolchildren.

#### Introduction

In the history of science many examples of unconscious bias of professionals have resulted in malleability of apparently objective quantitative data. The problem was greater when the professionals were firmly committed to acceptance of a theory, as well as firmly convinced of their own objectivity.

One of the founders of modern psychology, Alfred Binet, received his professional education when the prevailing theory about intelligence was that

\* Education Department, University of Auckland, Private Bag, Auckland, New Zealand.

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of phrenology, and the accepted method of measurement that of craniometry. Binet eventually had doubts. "I feared that in making measurements on heads with the intention of finding a difference in volume between an intelligent and less intelligent head, I would be led to increase, unconsciously and in good faith, the cephalic volume of intelligent heads and to decrease that of unintelligent heads" (1). He then proceeded to test his own suggestibility, and discovered that his measurements of heads varied, in conformity with his preconceptions, by an average of 3 mm, which was more than the differences claimed to exist between heads of bright and dull subjects. Binet discontinued his belief in phrenology (now regarded as pseudo-science, but in its day scientifically accepted as respectable). Stephen Gould has commented: "How much better off we would be if all scientists submitted themselves to self-scrutiny in so forthright a fashion" (2).

If such suggestibility can influence simple linear head measurements, it could influence even more the counting of the number of decayed, missing and filled permanent teeth (DMFT), or tooth surfaces (DMFS), in a child's mouth. The counting of a tooth or tooth surfaces as "decayed" involves an assessment of at what stage, in the process of softening and ultimate disintegration of the tooth surface, the lesion should be classed as "carious" or decayed. The numbers of missing and filled teeth are easier to count. Earlier, however they were influenced by the suggestibility of the various operators who had made the decision when to extract or fill the teeth.

In an earlier study (3) I described the problem of ensuring uniform diagnostic standards, which could affect later recorded mean DMFT, within a School Dental Service district where all the operators were under the supervision of a single controlling officer. During a period of transition, following a 1976 national decision to change from a radical to a more conservative method of diagnosis to decide when a tooth required filling, some lack of uniformity among operators' diagnoses was observed. However, as reported in that study, at the time of its data collection (the same time as for the present study) diagnostic standards in central Auckland schools dental clinics were uniform. Filling rates of dental clinics (the average number of fillings per child per year) varied between 0.25 and 0.79. Indeed it was shown that the filling rates were correlated with the socio-economic levels of the residential areas where the clinics were situated. Children in more affluent areas required fewer fillings.

The present study reports on variability of filling rates of privately practicing dentists in the same area of central Auckland, and discusses the relevance of such diagnostic variability to the reliability of DMF scores used in earlier and recent fluoridation studies.

#### Materials and Methods

In New Zealand, children up to 12 or 13 years of age receive regular free dental treatment at school dental clinics which are situated at most schools. After that age the children are enrolled for treatment with privately practicing dentists of parents' own choosing, who provide further regular free dental treatment at state expense until the age of 16 years (or 18 if still attending school). The dentists' claims for payment are sent to the local Department of Health officer responsible for administering both above services. In 1981, while occupying that post in central Auckland, I became concerned



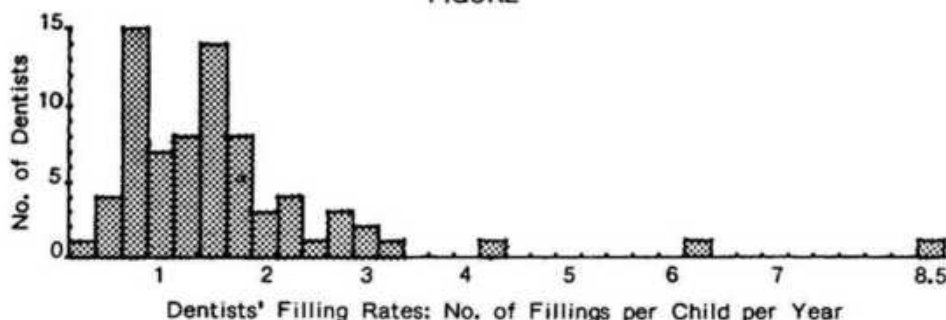
over the wide variations in the amount of treatment being provided by private dentists. The survey described below, initiated that year, was completed in 1982.

Seventy-three such dentists were providing treatment to 14- to 16-year-old children. Forty claims for payment for each dentist were examined, 20 when first received and 20 for succeeding treatments for each child, to calculate the number of fillings per child provided for one year within the period each child was enrolled with the same dentist.

### Results

Filling rates varied from 0.25 to 8.50 fillings per child per year. Most were within the range 0.5 to 3.0. The distribution among the dentists is shown in the Figure.

FIGURE



No socio-economic correlation of the above filling rates is possible, as it was with school dental clinics (3), because most of the private dentists practiced in the central area of the city or in the more affluent suburbs. Most draw their patients from a range of socio-economic backgrounds, with many who leave school dental clinics in the less affluent suburbs "dropping out" (i.e. 15% of those who leave dental clinics do not enroll, while others soon discontinue receiving regular treatment). Only two of the dentists practiced in the city's low-income unfluoridated area. One had a high filling rate (6.10), which was attributed to the lack of fluoridation, whereas the other had a rate (1.83) closer to the city average.

### Discussion

The average filling rate for private dentists was almost three times higher than that in school dental clinics, a situation only partly explained by the difference of patients' ages. Almost half of the private dentists had filling rates comparable to those of the school dentists. A more likely explanation for the high rates of the others was the different kind of remuneration. The private dentists operated under a fee-for-service system (more fillings: more pay). The school dentists earned a fixed salary and were encouraged to keep the number of fillings to a minimum.

The above findings were presented to the local branch of the dentists' professional association, whose officers expressed regret and a desire to achieve more uniform standards. The survey had established that considerable diagnostic variability can and does occur. Early studies have reported similar variability (4,5).

#### Relevance to DMF Studies

The relevance of such diagnostic variability to the reliability of fluoridation studies is obvious. Each of the components of the DMF score used in such studies could be influenced by subjective diagnostic variability among dentists. That observation applies not just to the dentists who carried out the dental examinations and recorded the DMF scores for the studies, but also to the many dentists who had earlier inserted the fillings or extracted the teeth. Diagnostic variability among examiners can be partly adjusted for by standardizing and periodically testing the examination methods. More importantly, to eliminate the possibility of examiner bias, the study can be "blind": examiners are not aware which children come from fluoridated and which from non-fluoridated areas. Actually, most fluoridation studies were not blind. Such precautions, however, take care of only the "D" (decayed) component of the DMF score. To calculate the other components, the examiners merely count up the number of filled and extracted teeth. The dentists who had earlier inserted the fillings and extracted the teeth in the two kinds of areas usually knew which of their patients had drunk fluoridated water and which had not.

The 1976 national diagnostic change within the School Dental Service of New Zealand (6), referred to in the Introduction and reported in various studies (3,7-10), had far-reaching effects. Mean DMFT scores of 12- and 13-year-old children were reduced by 47% in 5 years. The reduction occurred in both fluoridated and non-fluoridated areas; in fact, reduction was greater in the latter (11). It was acknowledged that such a reduction was too steep to be wholly a reduction in dental decay prevalence (10). It has been suggested that, in other countries, a similar change in diagnostic criteria by dentists could account for much of the reported decline in decay in recent years (12). Clearly, a reduction in recorded DMF scores does not necessarily reflect a reduction in dental decay.

#### Further Investigations

The Hastings fluoridation trial in New Zealand (13) is one of the most well-documented examples of bias influencing DMF scores, thanks to official files released under New Zealand's Official Information Act (14-17). In that trial the experimental control community was abandoned when it was discovered, a few years after the trial commenced, that younger children in the non-fluoridated control town had less decay than younger children in fluoridated Hastings, who should have exhibited the greatest benefit. The explanation offered was that a previously unknown trace element (molybdenum), found in the soil of one area of the control town, had gotten into the town's consumed vegetables, thus temporarily lowering decay rates for the younger children.

Large decay reductions were nonetheless eventually reported in Hastings. The released official files reveal that diagnostic criteria there had been

changed after the trial began thus ensuring that Hastings DMF scores were lowered. The change (similar to the latter, 1976, national one) was not reported in the published versions of the trial. It was easy to so ensure that lowered DMF scores among the younger Hastings children were recorded by the trial's examiner, because the younger children were all treated in state school dental clinics, where instructions were given to the operators to insert fillings at a later stage in the tooth softening process than had been done at the commencement of the trial. The published initial examinations for the trial recorded DMF scores which consisted mostly of a count of fillings that had been placed at a very early stage in the decay process. The later examinations, from which the great reductions claimed to be caused by fluoridation were calculated, counted fillings placed at a much later stage in the decay process, while any small unfilled cavities were no longer classed as decayed.

But the older children in the trial were treated by the town's privately practicing dentists, at a younger age (11-12 years in the earlier stages) than occurred in Auckland in 1981-82. The recorded "reductions" in decay were not so spectacular among the older children, but DMF scores were lowered. A part of those reductions could have resulted from the decline in decay now known to have been occurring elsewhere, with or without fluoridation. However, the released files show that the trial's examiner, in addition to instructing state dental clinic staff in the new diagnostic standards, had interviewed private dentists in the town about the importance of insuring the trial's success by not filling teeth too early. It is easy to understand how such persuasion would be effective. Without being in any way consciously dishonest about their changed diagnoses, the dentists could have found fewer cavities requiring filling because of their strong conviction that fluoridation would work. They had earlier campaigned hard to persuade citizens and the local council to fluoridate the water supply, promising that reductions in tooth decay would soon follow.

A similar explanation could account for the findings in the recent Wick (18) and Stranraer (19) non-blind studies in Scotland, which claim to show, by comparing DMF (or "dmf", for primary teeth) of small samples of young children, that dental decay increased suddenly after the local water supplies were defluoridated. In Stranraer the DMFT of 10-year-olds rose, after defluoridation, by only 4% in 6 years, the increase consisting wholly of the "MF" (extractions and fillings) component. In Wick, where the children examined for the study were 5- to 6-year-olds with few if any permanent teeth, the increase in dmf consisted almost entirely of a 61% increase in the number of primary teeth extracted (that is, the "m" component; the "df" component increased by only 0.4%). Such an increase reflects a change in the treatment pattern of Wick dentists, following the decision to defluoridate. No doubt the dentists believed they were responding to real increases in decay. However, statistics obtained from much larger numbers of older children, in both Holland (20) and New Zealand (21), show no increase in dental decay following cessation of fluoridation.

#### Inadequacies of the Measure

The DMF measure gained acceptance a short time before the first fluoridation trials, and has been used for subsequent trials and surveys. Early critics pointed out its serious limitations as a measure of caries prevalence. No other disease is measured by counting the number of lesions. Even under

uniform diagnostic conditions, fillings and extractions are not evidence of present or current decay. They show only that teeth were diagnosed as decayed in the past — like visible scars of earlier disease. A child with fillings or extracted teeth could be free of decay at the time of examination.

Another anomaly of the DMF measure, exemplified in the Hastings and other results, arises because children of different ages have different numbers of permanent teeth. "Decay reductions" are calculated by dividing the difference between an early and a later mean DMF reading by the first reading and expressing the result as a percentage. As Philip Sutton has observed, some remarkably impressive results can be obtained, especially for younger age groups (22). Children aged 6 years with only 4, or perhaps 6, permanent teeth, have low DMF scores. Thus a slight delay in inserting fillings can be interpreted as impressive "reductions." For example, the mean DMFT of 6-year-olds in Hastings was 1.41 in 1954 and 0.82 in 1957, 2½ years later — a "reduction" of 42%, which in reality was less than one filling. Given the subjective variability of caries diagnosis, differences in mean DMF of less than one tooth or tooth surface, though "statistically significant", can be of no clinical significance.

The DMF measure applied to small samples is especially susceptible to subjective variability. Even proponents of fluoridation like J.M. Dunning in the early years urged caution: "Interpretive and other examining errors in DMF studies may be large, easily exceeding 100 percent difference between samples . . ." (23).

Other early critics advocated alternative methods of measurement. Veikko Hurme proposed a biometric technique which allowed for the amount of tooth tissue exposed and the length of time of such exposure (24). In New Zealand, Eric Hewat doubted the value of the DMF measure. In his national caries survey (25) he used a "caries index" which, like Hurme's proposal, took into account the number of tooth surfaces present, and the differing times of tooth eruption. His "annual caries attack rate" went further, in that it measured current disease activity. Hewat used the above measures in 1952 for pre-fluoridation examinations of children for the Hastings fluoridation trial. However his results were later destroyed and never published. The published "initial" examinations were carried out 2 years later by Hewat's successor, after fluoridation had commenced.

### Hypothesis

Donald McKenzie has challenged the assumption that the mathematical sciences are immune from social influences (26). He examined a controversy in the history of the development of statistical theory at the turn of this century (Pearson v. Yule on measurement of association). The controversy reflected different "cognitive interests" of the scientists involved, which, in turn, arose indirectly from social influences. Those scientists strongly committed to eugenics research had a strong interest in and influence upon the methods of measuring association. Stephen Gould described the social influences, and professional commitment to the hereditarian theory of "IQ", which he suggests led to another creation of abstract statistical theory: "Factor analysis, despite its status as pure mathematics, was invented in a social context, and for definite reasons" (27).

The DMF and IQ measures are much less sophisticated devices. But adoption of the DMF method of measuring dental caries prevalence could well have been, like the adoption of the IQ score, a reflection of the cognitive interests of strongly committed professionals. The very limitations of the DMF measure facilitated claims of exaggerated "reductions" in dental decay. There are striking analogies between the vogue for IQ in psychometrics and the vogue for DMF in dental science. Both measures sought to represent, as a simple linear number, human conditions which are of complex origin. Both facilitated the promotion of theories, enthusiastically supported by professions anxious to achieve a new scientific respectability. Both, it has turned out, sometimes facilitated rather shameless manipulation and selection of data. Both, also, by virtue of their long acceptance and use, have achieved rather stubborn entrenchment, in spite of their unsound theoretical bases.

Critics have pointed out that the DMF measure, here criticized, has often been used in my own published studies. An author concerned to explain or reinterpret past events and studies has no option but to use what data are available. It is often the inescapable option of scholars to observe nature or society through inadequate and distorting lenses. The problem of working from "one-sided evidence" is well known to historians. By piecing together records from the past, including those which used deficient methods of measuring, a closer approximation of reality might be perceived. Today it is becoming apparent that fluoridationist claims do not stand up to examination, even with the inadequate measure which, at first, facilitated the claims.

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# FLUORIDES IN HAIR AND URINE OF CHILDREN IN THE VICINITY OF A PHOSPHATE INDUSTRY WASTE DISPOSAL SITE

by

W. Czarnowski and J. Krechniak\*  
Gdańsk, Poland

**SUMMARY:** Fluoride content in urine and hair of children aged 7-16 living in the environs of a phosphate industry wastes disposal site was determined. These wastes which contain 0.2% soluble fluoride may present a considerable hazard for the inhabitants and the environment. Significant elevations of fluorides in urine and hair of exposed children were ascertained.

**KEY WORDS:** Children; Hair fluoride; Phosphate industry waste; Urinary fluoride.

## Introduction

Phosphate industry wastes originate from the production of phosphoric acid and superphosphates of phosphorites. In addition to calcium sulfate these waste products also contain other substances including fluorine compounds. Wastes disposal sites may pollute the surrounding environment as a result of leaching by rainfall and transportation of dust particles by wind.

People living in the vicinity of the wastes are exposed to fluorides by aerosols as well as by drinking water and agricultural products grown in the polluted area. An increased absorption of fluorides results in an elevated level of fluorides in tissues and excreta. Fluorides particularly accumulate in hard tissues (bones, teeth), nails, hair, and are, for the most part, excreted with urine (1-4).

The aim of this paper was to determine the degree of environmental exposure to fluorine compounds of persons living in the vicinity of a phosphate industry waste disposal site.

To exclude the interference of occupational exposure on the obtained results, children living in the same neighborhood and attending the nearby primary school were used in this study.

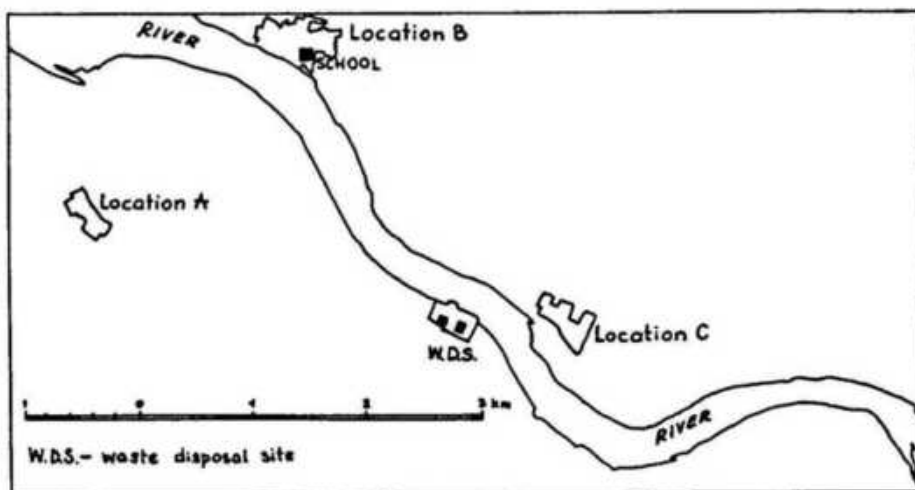
## Materials and Methods

**Experimental Design:** Samples of hair and urine were collected from 56 children aged 7-16 living in the environs of a phosphate industry wastes disposal site. The children under study were divided into three groups according to the distance and location of their residences to the wastes disposal sites: Group A - 3 km W, Group B - 3 km NW, Group C - 1 km E. Controls were 46 children of similar age from a non-exposed area.

\* Department of Toxicology, Medical Academy, Gdańsk, Poland.

Figure 1

Map of Waste Disposal Sites and Affected Localities



Also the content of fluorides in the wastes and drinking water from the investigated area were determined.

Preparation of Samples: Specimens of hair (about 5 cm length) were rinsed on a fritted glass filter with acetone, detergent, 2 N sulfuric acid and redistilled water. After drying 100 mg aliquots were placed into polyethylene test tubes, treated with 70% sodium hydroxide solution and heated in a boiling water bath until completely dissolved (about 1 hr). Cooled and neutralized samples made up with water to 4 mL were diluted with equal volumes of TISAB buffer (5).

Fluorides in samples of water and urine were determined directly after dilution with equal volumes of TISAB buffer. The concentration of creatinine and specific density were also measured in urine.

TISAB buffer: 57 mL of acetic acid, 58 g sodium chloride, 0.3 g sodium citrate were diluted with water and adjusted to pH 5.2 and made up to 1 L.

Determination of Fluoride: Fluoride concentrations were measured by a fluoride-specific electrode (6,7). Calculations were based on a response factor from a standard curve prepared daily. Recovery of F from hair and urine amounted to  $100 \pm 8\%$ . The coefficient of variation in 10 samples of hair obtained from the same person was 9%.

Significance was determined by Student's t-test.

### Results

The investigated phosphate industry wastes contain 0.2% fluoride. The



Table 1  
Fluoride in Urine and Hair of Children

| Groups               | n  |       | Urine  |         |                   | Hair                 |
|----------------------|----|-------|--------|---------|-------------------|----------------------|
|                      |    |       | mg F/L | mg F/L* | mg F/g Creatinine | µg F <sup>-</sup> /g |
| Group A              | 14 | mean  | 2.63   | 2.75    | 2.34              | 3.21                 |
|                      |    | ±S.D. | 0.77   | 0.95    | 0.95              | 1.04                 |
|                      |    | p     | 0.001  | 0.001   | 0.001             | 0.01                 |
| Group B              | 14 | mean  | 2.80   | 2.44    | 2.59              | 3.74                 |
|                      |    | ±S.D. | 1.63   | 1.32    | 1.92              | 0.65                 |
|                      |    | p     | 0.001  | 0.001   | 0.001             | 0.001                |
| Group C              | 27 | mean  | 2.39   | 2.60    | 2.55              | 3.17                 |
|                      |    | ±S.D. | 0.90   | 0.99    | 1.17              | 0.78                 |
|                      |    | p     | 0.01   | 0.001   | 0.001             | 0.001                |
| All Exposed Children | 56 | mean  | 2.56   | 2.43    | 2.51              | 3.33                 |
|                      |    | ±S.D. | 1.10   | 1.07    | 1.33              | 0.85                 |
|                      |    | p     | 0.001  | 0.001   | 0.001             | 0.001                |
| Controls             | 46 | mean  | 0.74   | 0.70    | 0.74              | 2.34                 |
|                      |    | ±S.D. | 0.30   | 0.42    | 0.37              | 0.67                 |

n = number of children; S.D. = Standard Deviation;

p = level of significance

\* normalized to standard specific density 1.024

fluoride concentration in drinking water from wells and water conduits situated in the vicinity of the waste disposal sites was 2.0-2.3 mg F/L. Tap water used by children from the control group contained 0.33-0.51 mg F/L.

See Table I for fluoride content in urine and hair of children from exposed and unexposed areas. Differences in urinary and hair fluoride between exposed children and controls were statistically significant. Urinary fluoride is expressed in milligrams per liter, normalized to the standard specific gravity 1.024 and as rates to creatinine excretion.

#### Discussion

A large phosphate industry wastes disposal site containing 0.2% soluble fluoride may present a considerable hazard for the inhabitants and the environment in the surrounding area. It may be assumed that people living in the vicinity of a wastes disposal site are exposed to fluoride to a greater degree than individuals from remote city districts. Absorption of fluorides may occur orally through water or agricultural products and by inhaling dust particles. Inhalatory exposure is influenced by such factors as atmospheric precipitation and direction of winds.

The estimated fluoride level in drinking water in the investigated area

(2.0-2.3 F/L) surpassed recommended values. On the other hand the concentrations of fluorides in drinking water consumed by children in the control group (0.33-0.51 mg F/L) is lower than that normal and rather typical for Poland (8).

The estimated environmental exposure to children living in the neighborhood of the waste disposal site is rather moderate. Urinary fluoride levels of the exposed group were about three times higher than that of controls, hair fluoride was about 50% higher.

Fluoride levels in occupationally exposed persons estimated by us in another study exceeded the values described in this paper in urine 2-10 times and in hair, 100-300 times (9).

No correlation was observed between fluoride content in urine and hair of the children under study. Similarly no significant differences in fluoride levels in biological media between various groups (A,B,C) of children in the exposed area was established.

#### Conclusions

1. Fluoride levels in urine and hair of children living in the vicinity of a phosphate industry wastes disposal site were significantly elevated.
2. The fluoride level in drinking water in the exposed area is about four times higher than in other districts of the investigated city.

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# FORMATION OF SUSTAINED-RELEASE FLUORIDE MATRIX TABLETS

by

K.S. Aithal, K. Nalini and Y.V. Pathan\*  
Manipal, India

**SUMMARY:** The present work is an attempt to develop a sustained-release fluoride matrix tablet. Formulations were developed that incorporated various rosin derivatives which are widely used in chewing gums and dental products for their film properties. Tablets prepared from these materials as hydrophobic matrices were evaluated for various tablet parameters. Dissolution studies were carried out in buffer media with pH 7.2. All the formulations had adequate hardness, higher disintegration time, and less friability loss. The drug release followed a first-order kinetic pattern controlled by diffusion and was linear to the square root of time function. In pentaerythritol rosin (PR) ester and rosin-maleic acid adduct esterified with glycerol (RMEP) formulations  $T_{80\%}$  was  $> 3$  hr.

**KEY WORDS:** Fluoride, sustained release; Matrix tablets.

## Introduction

Conventional methods of fluoride therapy include various combinations of systemic and topical administration (1-3), most of which rely on a measure of compliance by patients for maximum efficacy. These approaches also are limited in their ability to maintain effective levels of fluoride at the intended site of action (4). The approaches to developing effective delivery systems for prolonged fluoride release have included both the sustained release and controlled release delivery systems. Sustained release delivery systems are those that prolong the release of the drug, but the rate of drug release is not uniform. Conversely, controlled release delivery systems provide a constant rate of drug release for a prolonged period (5-6). The sustained release fluoride preparations that have been investigated include a sustained release tablet or capsule (7-8) and an aerosol system to deliver microcapsules of fluoride directly on the tooth surface (9). The controlled release fluoride delivery systems include a fluoride releasing polymer and a membrane controlled reservoir system that can be placed intraorally (10-12).

Rosin and rosin derivatives have been reported to be useful as coating and microencapsulating materials for sustained release of the drug when administered orally (13-15). These were also reported to be useful as hydrophobic matrix materials in the formulation of sustained release matrix tablets (16). The present communication deals with the development of a sustained-release fluoride formulation that incorporates various rosin and rosin derivatives as matrix materials.

\* College of Pharmacy, Manipal, 576-119, India. Current address: Room 5014, Kresge-11,0576, University of Michigan Medical Center, Ann Arbor, Michigan 48109, USA.

### Materials and Methods

Rosin N grade (ISI), Pentaerythritol rosin ester (PR ester), Glycerol rosin ester (GR ester), Rosin maleic acid adduct esterified with glycerol (RMEG) and with pentaerythritol (RMEP), have been obtained from Prabhat General Agencies, Bombay. Sodium fluoride (U.S.P.) and all other ingredients were of Indian Pharmacopoeial Standard.

Preparation of NaF Matrix Tablets: Formula for each matrix tablet was NaF, 2 mg; lactose, 43 mg; starch, 5 mg; and the rosin or rosin derivative as matrix material, 50 mg.

All ingredients were separately sieved through 60 mesh and then mixed thoroughly in geometric proportions to ensure uniform mixing. Each batch was of 500 tablets. Granules were prepared with starch paste (10%) as binding agent and were dried at 45°C overnight. Granules were evaluated for various properties as discussed below. Granules of mesh size 22/44 were selected for subsequent compression. The tablets, weighing 100 mg with 0.6 mm diameter and 2.5 mm thickness, were prepared on a single punch machine at a pressure of 1500 kg/cm<sup>2</sup>.

Evaluation of the NaF Granules and Matrix Tablets: Granules were evaluated for properties like bulk density, true density, total porosity, angle of repose, friability loss, granule strength, and drug content according to standard pharmacopoeial procedures. The tablets were evaluated for weight variation, hardness, friability loss, disintegration time, dissolution studies, and drug content. Fluoride was estimated by the Orion research microprocessor ion analyser model 901 (Orion, USA).

### Results and Discussion

Physicochemical properties of rosin and rosin derivatives have been discussed earlier (13-15). Because it was observed that these differ from each other in their properties, it was thought worthwhile to know the effect of various rosin derivative matrix materials on the granule and tablet characteristics. Various granule properties studied are given in Table 1. Properties like friability loss, granule breaking strength, and total porosity differing from each other and were significantly different from the values obtained for the control granules. The differences can be attributed to the properties of the matrix materials used since their percentage in the formulation was quite high (50%). Rosin itself was brittle in nature leading to high friability loss and lower breaking strength. The more bulky the alcohol molecule used for the esterification of the rosin, the better were the binding and granule properties. Drug content in 100 mg granules for each formulation showed more or less the same values, thus indicating uniform mixing of the ingredients.

Tablet properties studied are given in Table 2. The weight variation and the drug content of the tablet batches were within the pharmacopoeial limits. Rosin ester formulations have shown better hardness and less friability loss. Disintegration time was significantly higher in all the matrix tablet formulations than the control tablets (without hydrophobic matrix materials, only lactose was used in place of matrix materials). The highly hydrophobic nature

Table 1  
Granule Properties of Various Fluoride Formulations

| Matrix Materials | Parameters* |      |       |       |      |       |     |
|------------------|-------------|------|-------|-------|------|-------|-----|
|                  | A           | B    | C     | D     | E    | F     | G   |
| Rosin            | 0.5         | 1.34 | 62.79 | 24.42 | 12.0 | 150.9 | 7.7 |
| GR-ester         | 0.5         | 0.9  | 47.74 | 24.54 | 6.5  | 166.8 | 7.2 |
| R.M.E.P.         | 0.5         | 1.3  | 59.8  | 28.11 | 4.5  | 180.2 | 7.4 |
| R.M.E.G.         | 0.5         | 1.5  | 67.1  | 27.43 | 3.5  | 192.6 | 7.7 |
| PR-ester         | 0.5         | 1.3  | 57.95 | 59.5  | 2.4  | 304.9 | 7.7 |
| Control          | 0.7         | 1.3  | 47.42 | 22.2  | 9.7  | 138.8 | 7.3 |

\* A. Bulk Density (gm/mL); B. True Density (gm/mL); C. Total Porosity; D. Angle of Repose (°); E. Friability Loss (%); F. Granule Breaking Strength (gm/cm<sup>2</sup>); G. Drug Content in 100 mg of Granules in ppm/100 mL.

Table 2  
Tablet Characteristics of Various Fluoride Formulations.

| Matrix Materials | Parameters* |      |      |       |     |      |      |
|------------------|-------------|------|------|-------|-----|------|------|
|                  | A           | B    | C    | D     | E   | F    | G    |
| Rosin            | 95.39       | 5.84 | 1.2  | >180  | 6.9 | 79.5 | >180 |
| GR-ester         | 94.31       | 6.26 | 0.9  | >180  | 7.0 | 25.0 | 134  |
| R.M.E.P.         | 98.45       | 6.25 | 0.87 | 150   | 7.6 | 52.1 | 150  |
| R.M.E.G.         | 91.63       | 4.80 | 0.94 | 88.25 | 6.5 | 22.0 | 67.5 |
| PR-ester         | 94.27       | 7.3  | 0.73 | >180  | 6.8 | 76.0 | >180 |
| Control          | 105.2       | 6.95 | 0.87 | 13.25 | 7.4 | 3.5  | 7.0  |

\* A. Weight/Tablet; B. Hardness (kg/cm<sup>2</sup>); C. Friability Loss (%); D. Disintegration Time (min); E. Drug Content/Tablet in ppm per 100 mL; F. Dissolution Studies T 50%; G. Dissolution Studies T 80%.

Fluoride estimation was done by Orion Research Microprocessor Ion Analyser Model 901 (Orion, USA)

of rosin and rosin derivatives when used as matrices prevented the penetration of water leading to shorter disintegration time.

Dissolution studies showed that most of the matrix tablets released 80% of the drug in 3 hr in the dissolution media (Figure 1). During the dissolution studies the tablets indicated an inert matrix behavior. The dissolution data were plotted as a function of square root of time (Figure 2 and 3). The data seemed to follow the Higuchi equation (17) for an inert porous matrix. This type of behavior was observed in all the formulations showing diffusion-

Figure 1

Drug Release Profiles *in vitro* in Phosphate Buffer with pH 7.2 from Various Fluoride Matrix Tablet Formulations.

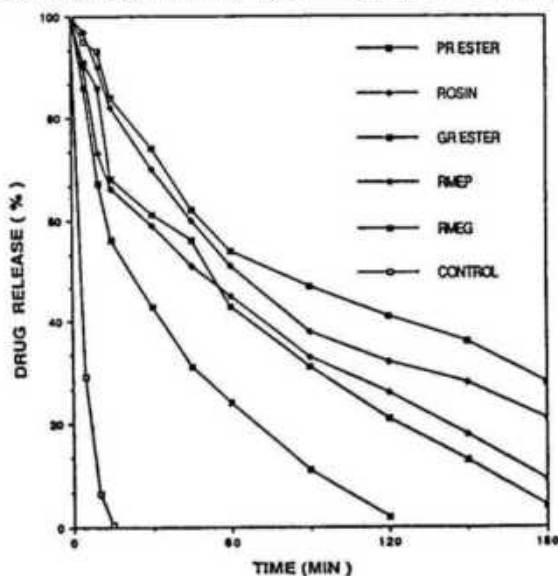


Figure 2

Drug Release Profile from PR Ester Fluoride Matrix Tablet Formulation vs. Square Root of Time.

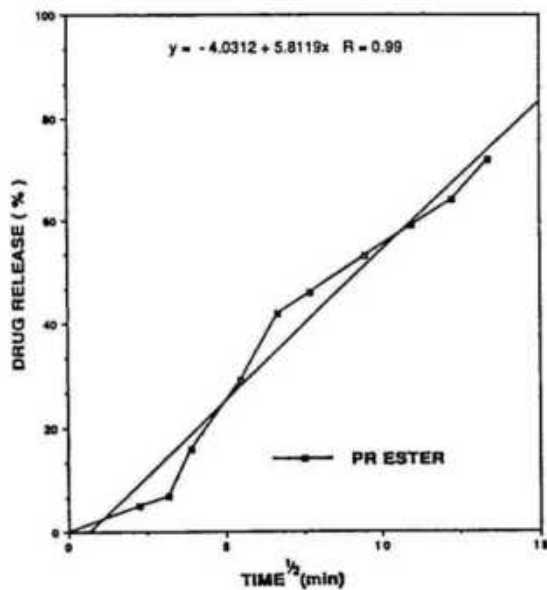
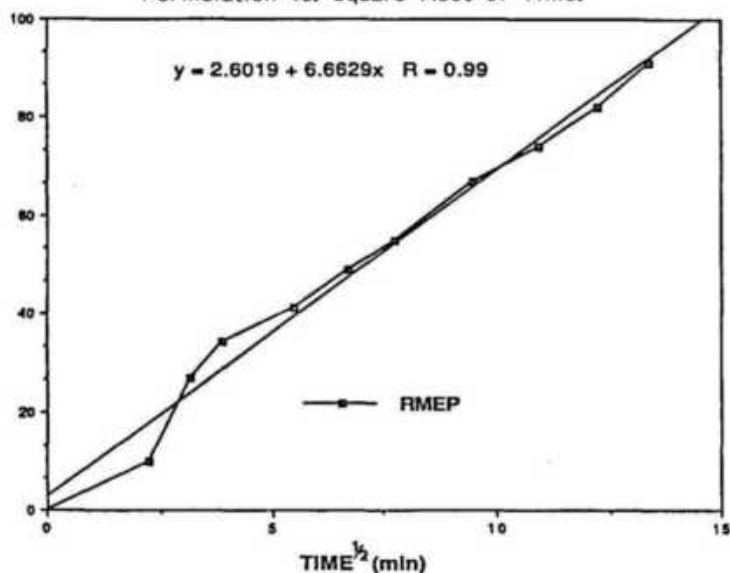


Figure 3

Drug Release Profile from RMEP Fluoride Matrix Tablet Formulation vs. Square Root of Time.



controlled, first-order release kinetics. With a rosin formulation a different pattern was observed. During dissolution the tablet was eroded and when the data were plotted against the square root of time, a straight line was not obtained. Thus drug release from the rosin matrix tablet obeyed a different release pattern than the other matrix tablets. There may be more than one mechanism in the drug release from the rosin matrix tablets. Similar behavior was also observed earlier and discussed in detail (16).

### Conclusions

Overall, the results of the present study showed that rosin and rosin derivatives can be used as hydrophobic matrix materials for controlled release of fluoride in the oral cavity. These matrix tablets can deliver 80% of the drug in 3 hr, i.e. during the school day. Some of the granule and tablet characteristics differed from each other significantly, and a proper control on the matrix concentration may help in developing tailor-made formulations with the desired drug release pattern. Drug release was controlled by a diffusion mechanism and was linear to the square root of time. PR ester was the best matrix material compared to other rosin derivatives. An important advantage of these materials over other matrix materials is their low cost compared to celluloses and other materials as well as their ready availability.

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## EXPOSURE TO FLUORIDES IN THE FRENCH PHOSPHATE FERTILIZER INDUSTRY

by

M. Hery, F. Diebold, G. Hubert, J.M. Gerber and J.C. Limasset  
Nancy, France

**SUMMARY:** Exposure to fluorides was assessed in seven French fertilizer-production plants. Results show that the short-term limit value of hydrogen fluoride is often exceeded and that long-term values are generally respected, except in the case of superphosphate production, where almost fifty percent of sampling results are over limit values.

**KEY WORDS:** Fertilizers; Fluorides; Superphosphate.

### Introduction

Fluorosis in the superphosphate industry has been reported by several authors. For example, Derryberry (1) found bone changes in a group of workers exposed at an air concentration averaging 3.38 mg fluoride/m<sup>3</sup>, although such changes did not appear in workers having an average exposure of 2.65 mg/m<sup>3</sup>. More recently, El Ghawabi (2) reported that 36% of the workers employed in an Egyptian superphosphate plant showed variable degrees of change in the density of bones. These changes were present in all workers with more than twenty years of exposure.

Furthermore, three mortality studies were conducted in the Florida phosphate industry (3-5). The conclusions of these studies are not exactly the same, but they all suggest a dose-reponse of respiratory cancer with long-term duration of employment. No causal agent is precisely evidenced, although the authors indicate the potential exposures include chemical pollutants (fluorides, acid mists), and possibly radionuclide-bearing dusts (uranium is a component of phosphate ore) and radon.

Recently, a study performed by de Labrusse (6) in a French fertilizer factory showed a decrease in pulmonary function of the workers. Age (more than fifty years old) and duration of exposure (more than ten years) were evidenced to be in relation to this decrease.

Ernst *et al.* (7) explored the relationship of respiratory symptoms and lung function to exposure to ambient air pollution consisting of particulate and gaseous fluorides. They showed there was such a relationship for a population composed of young children living near an aluminum smelter.

The aim of our study was to measure the exposure levels to chemical pollutants of workers employed in the French phosphate fertilizer industry, including phosphoric acid production plants, superphosphate and compound fertilizer plants. This report only deals with fluoride pollution. The results will be published elsewhere in their entirety (8).

\* Institut National de Recherche et de Sécurité, BP 27, 54501 Vandoeuvre-les-Nancy, France.

Industrial Processes: The industrial processes are numerous and varied. It is useless to describe them precisely here. The principle is to transform phosphate rock in order to make it assimilable by the soil. Phosphoric acid and superphosphate production begins with an acidulation of the ore, generally with sulphuric acid. Sometimes, phosphoric acid is used for superphosphate. The following general reactions occur:



Hydrogen fluoride, or more probably fluosillicic acid, evolves during these acidulation reactions.

In the production of fertilizers, superphosphate (or phosphoric acid) is used, with the addition of ammonia (or nitrate compounds) and potash (or potassium compounds). The final product (NPK compound fertilizer) is then generally granulated.

#### Materials and Methods

For this study, seven factories were selected. This selection was performed in order to be representative of the manufacturing industry of chemical fertilizers in France. The activities at the various sites are described in Table 1.

Table 1  
Characteristics of the Studied Factories

|                         | Factory |   |   |   |   |   |   |
|-------------------------|---------|---|---|---|---|---|---|
|                         | A       | B | C | D | E | F | G |
| Phosphoric Acid Plants  |         | 1 | 2 | 1 |   |   |   |
| Superphosphate Plants   |         | 1 | 1 | 1 | 1 | 1 | 1 |
| NPK Granulation Plants  |         | 1 |   | 1 | 1 | 1 |   |
| PK Granulation Plants   |         |   |   | 1 | 1 |   |   |
| Ground Phosphate Plants | 2       |   |   |   |   |   |   |

Assessments were restricted to periods of three or four successive days in each plant. Two different types of samples were taken:

1. Personal Samples: Filter cassettes were clipped to the collar of the work clothes, near the breathing zone. These cassettes were sampled for a full-shift time, or when it was not possible, for four or five hours, at a flow rate of 1.0 Lpm. The cassettes were in closed-face configuration.

2. Area Samples: Filter cassettes were placed at fixed location in the room. Sampling duration varied generally from two to four hours. But in some cases, it was limited to fifteen minutes, for comparison with short-term exposure limits. In these cases, samples were taken in a place representative of occa-

sional individual exposures. The cassettes were in open-faced configuration, and the flow rate was 2.0 Lpm.

Sampling and analytical methods for fluorides are listed in Table 2.

Table 2  
**Sampling and Analytical Methods**

|   |  |
|---|--|
| Sampling Procedure                      | Fluorides are collected using 37 mm diameter cassettes, with a teflon* filter (Millepore FALP 0.8 $\mu\text{m}$ ) followed by a cellulose filter (Durieux band grise) impregnated with 2% sodium carbonate.<br><br>* teflon is used in order to avoid sulphuric acid-phosphoric acid neutralization (see [7]). |
| Soluble Fluoride Analytical Procedure   | Fluoride ion specific electrode for the two filters desorbed separately in water.<br>For the teflon filter, the remaining dust (insoluble particulate fluorides) is filtered onto a PVC-acrylonitrile filter (Gelman DM 0.8 $\mu\text{m}$ ).   |
| Insoluble Fluoride Analytical Procedure | Melting of the PVC-acrylonitrile filter in sodium carbonate at 900°C.<br>Fluoride ion specific electrode for the final product of the melting.   |

The results of personal samples were directly compared to long term exposure limits (French VME equivalent to the American TLV-TWA) edicted by the French Ministry of Employment: 2.0 mg/m<sup>3</sup> for soluble fluorides, 2.5 mg/m<sup>3</sup> for total (soluble + insoluble) fluorides. Short-term samples were compared with short-term limit values: 2.5 mg/m<sup>3</sup> for hydrogen fluoride (French VLE equivalent to the American TLV-STEL).

The French system of limit values is presented in the Appendix.

### Results

Phosphoric Acid Plants: None of the 175 personal samples collected in these plants shows a fluoride concentration above the relative long-term limit value. Table 3 gives the ranges of these values.

Areas where short-term limit values are exceeded are not numerous: most acidulation reactions occur in closed vessels, and gaseous emissions are controlled by ventilation. However, sampling by night in the sludge filtration area of Plant I in Factory C gave several values averaging 5 mg/m<sup>3</sup> in areas where workers go from time to time. During this period, all exits were closed.

Otherwise, the exposure of a maintenance operator working above a phosphoric acid storage tank reached 20 mg/m<sup>3</sup>, for a twenty minute sampling duration. Indeed, if most reactions take place in closed vessels, it is not the same for storage which is realized in open air on account of corrosion problems with the tops of the tanks.

**Table 3**  
**Personal Samplings at Four Phosphoric Acid Plants**

| Factory                   | Occupations           | Number of Samples | Soluble Fluoride Concentration (mg/m <sup>3</sup> ) |               |      |
|---------------------------|-----------------------|-------------------|---|---------------|------|
|                           |                       |                   | Minimum Value                                       | Maximum Value | Mean |
| <b>Factory B</b>          |                       |                   |   |               |      |
|                           | Foreman               | 8                 | < 0.02  | 0.16          | 0.07 |
|                           | Switchboard Operators | 8                 | < 0.02  | < 0.02        |      |
|                           | Areas Operators       | 21                | < 0.02  | 0.19          | 0.06 |
| <b>Factory C Plant 1</b>  |                       |                   |   |               |      |
|                           | Foremen               | 13                | 0.13  | 1.26          | 0.44 |
|                           | Switchboard Operators | 5                 | 0.16  | 0.38          | 0.29 |
|                           | Area Operators        | 31                | 0.12  | 1.35          | 0.53 |
| <b>Factory C Plant II</b> |                       |                   |   |               |      |
|                           | Foremen               | 11                | 0.10  | 0.98          | 0.30 |
|                           | Area Operators        |                   |   |               |      |
| <b>Factory D</b>          |                       |                   |   |               |      |
|                           | Foreman               | 14                | 0.05  | 0.16          | 0.10 |
|                           | Area Operators        | 20                | 0.05  | 0.38          | 0.13 |

**Superphosphate Plants:** About fifty percent of the samples taken during full-time working of the plants show results higher than the soluble fluoride or total fluoride long-term limit values (excluding the second assessment in Plant G). If we include this second assessment, which occurred after an improvement in the gas exhaust ventilation system, this number decreases to forty percent.

The majority of fluoride set free during the first steps of the process evolves as hydrogen fluoride (or fluosilicic acid). For most operators (except perhaps some conveyance operators), exposure to hydrogen fluoride must be controlled and compared to the short-term limit value. In all factories except Plants C and D, we found some fifteen-minute samples in which the fluoride concentration was above 2.5 mg/m<sup>3</sup>. These samples were collected in working areas where employees used to go (mixers and densheds). Table 5 gives an example of eleven successive personal samplings on a mixer operator in Plant F. Each sample was taken during a period of a quarter of an hour, except the ninth (half an hour).

In this case, the three-hour average concentration of soluble fluoride was below the long-term value, but three samplings exceeded the 2.5 mg/m<sup>3</sup> hydrogen fluoride limit value.

**Table 4**  
**Personal Samplings at Six Superphosphate Plants.**

| Factories<br>Occupations                             | n  | Soluble Fluoride<br>Concentrations<br>(mg/m <sup>3</sup> ) |      | Total Fluoride<br>Concentrations<br>(mg/m <sup>3</sup> ) |      |
|--|----|--|------|--|------|
|  |    | Range  | Mean | Range  | Mean |
| <b>Factory B</b>                                     |    |  |      |  |      |
| Mixer Operators                                      | 4  | 0.08-24.9  | 7.85 | 0.21-30.3  | 9.40 |
| Storage Operators                                    | 4  | 0.08-2.32  | 1.04 | 0.15-2.55  | 1.19 |
| Switchboard Operator                                 | 4  | 0.06-2.15  | 0.71 | 0.10-2.59  | 0.87 |
| <b>Factory C</b>                                     |    |  |      |  |      |
| Mixer Operators                                      | 7  | 0.11-2.45  | 0.40 | 0.15-1.83  | 0.53 |
| Superphosphate<br>Conveyance Operator                | 8  | 0.12-2.67  | 0.26 | 0.16-0.77  | 0.32 |
| <b>Factory D</b>                                     |    |  |      |  |      |
| Mixer Operator                                       | 5  | 0.35-1.60  | 0.65 | 0.50-2.24  | 1.08 |
| Superphosphate<br>Conveyance Operators               | 5  | 0.07-1.22  | 0.56 | 0.13-1.68  | 0.89 |
| <b>Factory E</b>                                     |    |  |      |  |      |
| All (continuous working)                             | 2  | 1.67-2.00  | 1.84 | 1.84-2.43  | 2.14 |
| All (discontinuous working)                          | 10 | 0.46-1.00  | 0.70 | 0.48-1.42  | 0.84 |
| <b>Factory F</b>                                     |    |  |      |  |      |
| Area Operators                                       | 5  | 0.77-3.47  | 2.31 | 0.80-3.89  | 2.51 |
| Superphosphate<br>Conveyance Operators               | 7  | 0.40-2.24  | 1.27 | 0.43-3.39  | 1.48 |
| <b>Factory G</b>                                     |    |  |      |  |      |
| Area Operators                                       | 11 | 0.86-3.16  | 2.09 | 0.94-4.92  | 2.55 |
| Granulation Operators                                | 4  | 0.77-2.11  | 1.29 | 0.84-2.51  | 1.49 |
| <b>Factory G<br/>(after ventilation improvement)</b> |    |  |      |  |      |
| Area Operators                                       | 12 | 0.52-2.35  | 1.17 |  |      |
| Granulation Operators                                | 4  | 1.05-1.58  | 1.19 |  |      |

Table 5  
 Successive Personal Samplings on a Mixer Operator.

|                | Soluble Fluoride<br>Concentration (mg/m <sup>3</sup> ) | Insoluble Fluoride<br>Concentration (mg/m <sup>3</sup> ) |
|----------------|--|--|
| 8:30 to 8:45   | 1.50   | 0.02   |
| 8:45 to 9:00   | 2.58   | 0.04   |
| 9:00 to 9:15   | 1.60   | 0.07   |
| 9:15 to 9:30   | 4.19   | 0.42   |
| 9:30 to 9:45   | 1.55   | 0.02   |
| 9:45 to 10:00  | 3.18   | 0.05   |
| 10:00 to 10:15 | 0.67   | 0.04   |
| 10:15 to 10:30 | 0.57   | 0.30   |
| 10:30 to 11:00 | 2.00   | 0.02   |
| 11:00 to 11:15 | 1.24   | 0.08   |
| 11:15 to 11:30 | 1.79   | 0.02   |
| Mean           | 1.86   | 0.12   |

Compound Fertilizer Plants: Most granulation plants of compound fertilizers use superphosphate as a source of phosphate compounds. This superphosphate has already been partially defluoridated, when produced. All but a few samples taken in factories D, E and F are below 1 mg fluoride/m<sup>3</sup>.

The plants of Factory A do not use superphosphate: the process consists in blending ground-up phosphate ore with various nitrate and potassium compounds, without any chemical reaction. Therefore, no soluble fluoride is produced.

#### Discussion

About five hundred long-duration personal samples were taken during this assessment campaign. Tables 3, 4 and 6 show that most high values occurred in superphosphate plants. The processes used in these plants are generally less sophisticated than those used in the other activities of the French fertilizer industry: manufacturing troubles are more numerous, human interventions are more frequent, and so are exposures to fluorides. For the same reason, the short-term exposure limit value for hydrogen fluoride was exceeded in superphosphate plants more often than in phosphoric acid plants.

Technological improvements (better controls of the quantities of phosphate ore and acids arriving in the reactors, more efficient ventilation, etc) should decrease the workers' exposure. The factories which have already introduced these improvements in their processes (for example C and D) have shown significant progress in the working conditions of their employees.

Table 6  
**Personal Samplings at Eight Compound Fertilizer Plants**

| Factories        | Occupations                         | n  | Soluble Fluoride Concentrations (mg/m <sup>3</sup> ) |      | Total Fluoride Concentrations (mg/m <sup>3</sup> ) |      |
|------------------|-------------------------------------|----|--|------|--|------|
|                  |                                     |    | Range  | Mean | Range  | Mean |
| <b>Factory A</b> |                                     |    |  |      |  |      |
|                  | PK Plant # 1:<br>All Operators      | 16 |  |      | 0.05-0.83  | 0.31 |
|                  | NPK Plant # 2:<br>All Operators     | 5  |  |      | 0.04-0.14  | 0.09 |
| <b>Factory B</b> |                                     |    |  |      |  |      |
|                  | NPK Plant<br>All Operators          | 52 | <0.02-0.12   | 0.04 | 0.04-0.34  | 0.13 |
| <b>Factory D</b> |                                     |    |  |      |  |      |
|                  | NPK Plant:<br>All Operators         | 52 | <0.02-0.21   | 0.07 | 0.02-0.54  | 0.20 |
|                  | PK Plant:<br>All Operators          | 31 | <0.02-1.22   | 0.28 | 0.10-1.68  | 0.53 |
| <b>Factory E</b> |                                     |    |  |      |  |      |
|                  | NPK Plant:<br>Granulation Operators | 4  | 0.04-0.49  | 0.18 | 0.05-0.49  | 0.20 |
|                  | Survey Operators                    | 10 | 0.11-1.26  | 0.45 | 0.12-1.39  | 0.52 |
|                  | Supply Operators                    | 10 | 0.04-0.44  | 0.21 | 0.05-0.56  | 0.26 |
|                  | PK Plant:<br>All Operators          | 17 | 0.07-0.56  | 0.21 | 0.08-0.79  | 0.27 |
| <b>Factory F</b> |                                     |    |  |      |  |      |
|                  | NPK Plant:<br>Operators             | 8  | 0.69-2.24  | 1.24 | 0.71-3.39  | 1.50 |
|                  | Foremen                             | 3  | 0.26-0.95  | 0.56 | 0.29-1.00  | 0.60 |

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#### Appendix

The French Ministry of Employment recognises two categories of limit values:

Regulatory values for certain compounds.

Indicative values in the general case.

These indicative values are subdivided into two types:

The limit value of mean exposure (VME) is the permitted value for the time-average concentrations to which a worker is effectively exposed during the course of a working day.

The limit value of exposure (VLE) is a ceiling value measured during a maximum period of fifteen minutes, taking into account the nature of the risk, the working conditions and the technical possibilities of the method of sampling and measurement.

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## SYMPOSIUM IN SOUTHERN POLAND

by

J.R. Marler  
Ottawa, Ontario, Canada

A symposium entitled "Health and Environmental Pollution," sponsored by the Polish Academy of Sciences, was held during November 21-22, 1988 in the Upper Silesian town of Zabrze. The conclave was attended by more than 60 scientists and featured 21 speakers (including one each from West Germany, France and Canada), supplemented by 12 poster presentations. Also present were two observers from the U.S. Environmental Protection Agency.

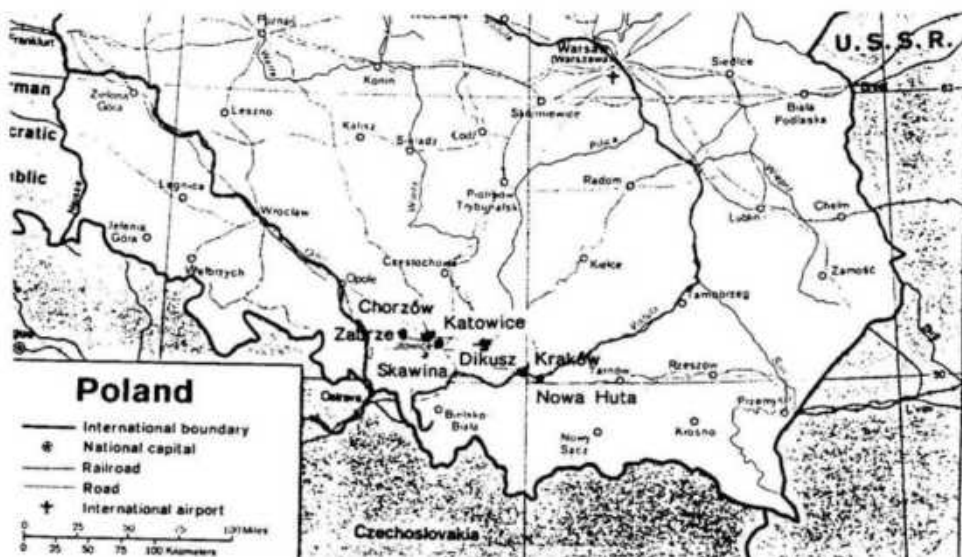
Southern Poland is probably the most polluted region in all of Europe. The environmental blight is caused primarily by mining and metal industries. The symposium emphasized environmental health problems in the region extending from the Upper Silesian town of Zabrze and eastward to the vicinity of Krakow (see map).

The Zabrze-Chorzow-Katowice area abounds in an assortment of iron-and-steel mills along with lead/zinc smelters. There is also a large lead/zinc

Figure 1

## Severely Polluted Region of Southern Poland

Major centers of pollution in this heavily industrialized section in southern Poland include Zabrze, Chorzów, Katowice, Dikusz, Kraków, Skawina and Nowa Huta. The Vistula rises in the southern part of this area and the Oder rises just over the Czech border, both flowing through this area.



smelter at Dikusz. The prevailing winds blow in a west-to-east direction. This means that some of the Silesian pollution impacts on Krakow which, in turn is likewise polluted by the immense iron-and-steel complex at nearby Nowa Huta. Throughout the entire region, brown Silesian coal is used as a fuel with little-or-no attempt at pollution control. A dustfall peak of  $2\frac{1}{2}$  tons/km<sup>2</sup>/yr has been recorded. Other than fluoride, the main pollutants are lead, cadmium, zinc, polycyclic aromatic hydrocarbons (PAH), and sulfur dioxide, along with the chronic problem of "acid rain." One of the consequences is that Krakow's historic buildings are crumbling from the corrosive impact. It is also important to note that this region encloses the headwaters of the Vistula river which has become befouled by metallic contamination and other pollutants.

The symposium topics included alveolar-bronchial respiratory effects, cardiovascular and infarctation incidence, gynecological-perinatal-mutagenic observations, immune system derangements, eye problems, effects on carbohydrate-energy metabolism, and incidence of carcinogenesis and neoplasms. Also discussed was the pollution-induced shortening of lifespan among the local inhabitants, and the successful use of magnesium-containing salts to alleviate fluoride toxicosis.

During the years 1954-1981, Krakow was subjected to severe fluoride pollution emanating from a large electrolytic aluminum smelter at nearby Skawina, and the environmental impact has been documented in Polish publications (1,2). The fluoride contamination in Krakow has abated since the shutdown of the aluminum operation. However, fluoride continues to be a problem in Upper Silesia where bone pains in citizens of Chorzow have been positively correlated with high urinary fluoride concentrations, as have metabolic derangements in the adenosine triphosphate (ATP) cycle.

Recent events in Poland have understandably disrupted the original schedule for appearance of the proceedings of this symposium during 1989. It is hoped that the proceedings will eventually be issued.

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J.R. Marler  
Researcher Emeritus  
National Research Council of Canada  
Ottawa, Ontario K1A 0R6, Canada

INCIDENCE OF HIP FRACTURES IN OSTEOPOROTIC WOMEN  
TREATED WITH SODIUM FLUORIDE

by

B.L. Riggs, D.J. Baylink, M. Kleerekoper, J.M. Lane,  
L.J. Melton, III, P.J. Meunier  
Rochester, Minnesota

(Abstracted from *J. Bone Mineral Res.*, 2:123-126, 1987.)

To determine whether fluoride therapy decreases the occurrence of vertebral fractures and increases the risk of hip fractures, data from five medical centers experienced with this therapeutic measure was reviewed. In 416 osteoporotic patients who were followed for more than 1,000 patient-years of fluoride treatment, 17 nontraumatic hip fractures were recorded. An incidence of 1.6% per year similar to the 1.9% per year incidence for 120 patients in this series who had been followed for 3 years prior to initiation of fluoride therapy. The expected incidence for women of the same age in the general community is 0.5% per year.

Although untreated osteoporotic women are at increased risk for hip fracture, treatment with fluoride seems to induce neither substantial decrease or increase in incidence of hip fracture.

KEY WORDS: Female osteoporosis; Fluoride therapy; Hip fracture.

REPRINTS: Mayo Clinic and Mayo Foundation, Endocrine Research Unit,  
Rochester, MN 55905, USA.

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INCREASED INCIDENCE OF HIP FRACTURES IN OSTEOPOROTIC  
WOMEN TREATED WITH SODIUM FLUORIDE

by

L. Rune Hedlund and J.C. Gallagher  
Omaha, Nebraska, USA

(Abstracted from *J. Bone and Mineral Research*, 4:223-225, 1989)

The incidence of hip fracture was compared in four groups of osteoporotic women: 22 were treated with placebo, 17 with fluoride and calcium, 18 with fluoride and calcitrol, and 21 with calcitrol alone. Four hip fractures had occurred in 3 patients on fluoride and calcitrol, and two hip fractures in 2 patients on fluoride and calcium. No hip fractures occurred in patients receiving either calcitrol alone or placebo. The difference in fracture rates for fluoride versus nonfluoride treatment is significant ( $p = 0.006$ ). Moreover, the six hip fractures occurring in patients receiving fluoride during 72.3 patient-years of treatment is higher than would be expected in normal women

of the same age. The probability of observing 6 fractures in 2 years is extremely small (0.0003). In four of the hip fracture cases, the history suggested a spontaneous fracture.

According to these findings fluoride treatment can increase the risk of hip fracture in osteoporotic women.

KEY WORDS: Calcitrol; Fluoride therapy, Hip fracture; Osteoporosis.

REPRINTS: J.C. Gallagher, M.D., St. Joseph's Hospital, 601 North 30th Street, Omaha, NE 68131, USA.

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### BONE COMPLICATIONS DURING THE TREATMENT OF OSTEOPOROSIS WITH FLUORIDE

by

C. Marcelli, E. Pansard, E. Thomas, C. Hérisson and L. Simon  
Montpellier, France

(Abstracted from *Rev. Med. Interne*, 10:118-126, 1989)

Fourteen osteoporotic women who had been under treatment with fluoride (23 mg/day of fluoride ions) for an average of 12 months developed periarticular pain in the lower limbs or pelvis, corresponding to 21 fractures due to bone insufficiency. Six of these fractures were revealed only by radionuclide uptake in the painful areas. The remaining 15 fractures, including 2 of the sacrum, were visible at radiography. Four patients had a past history of hyperthyroidism or were in a state of active hyperthyroidism detected by hormonal assays and iliac bone biopsy when the fractures were diagnosed.

The decalcification associated with hyperthyroidism facilitates the occurrence of bone insufficiency fractures, as do treatments with high doses of fluoride, inadequate calcium supplement intake (observed in 5 cases) or the presence before treatment of renal impairment or disorders of bone mineralization, sometimes detected only by iliac bone biopsy.

Repeated measurements of blood and urinary fluoride levels during treatment make it possible to adjust fluoride dosage. Lower doses (14 mg/day of fluoride ions) might reduce the incidence of the adverse effects of fluoride on bone.

KEY WORDS: Bone fractures; Calcium deficiency; Fluoride therapy; Hyperthyroidism; Osteoporosis.

REPRINTS: Service de Rhumatologie, hôpital Lapéyronie, CHU Montpellier, France.

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MICRORADIOGRAPHIC STUDY OF BONE AND TOOTH  
ALTERATIONS IN BOVINE FLUOROSIS

by

C. Nyssen-Behets, A. Vandersmissen, M. Ansay and A. Dhem  
Louvain, Belgium

(Abstracted from *Int. J. Tissue React.*, 11:31-37, 1989)

A 6-year-old fluorotic cow showed exostoses in the 5th rib, the cannon bone and mandible. Microradiographic alterations of lamellar bone in these skeletal items included both matrix modification and mineralization troubles. The enamel of the incisors was covered by a layer of cementum; the dentine presented numerous giant tubules and accentuation incremental lines. In the cementum, hypomineralized areas were located along the periodontal fibers.

KEY WORDS: Bovine fluorosis; Dental fluorosis; Microradiographs.

REPRINTS: Human Anatomy Research Unit, Catholic University of Louvain,  
Louvain, Belgium.

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ENVIRONMENTAL CONTAMINATION BY HEAVY METALS AND FLUORIDE  
IN THE SAEFTINGE SALT MARSH (THE NETHERLANDS)  
AND ITS EFFECT ON SHEEP

by

A.J. Baars, H. van Beek, T.J. Spierenburg, W.G. Beefink,  
J. Nieuwenhuize, J.J. Pekelder, J. Boom  
Lelystad, The Netherlands

(Abstracted from *Veterinary Quarterly*, 10:90-98, 1988.)

From May 1983 to May 1985 the local pollution of the Saeftinge salt marsh (The Netherlands) in the Westerschelde estuary (southwestern part) with metals and fluoride was investigated. Samples from soil and vegetation were analyzed monthly for cadmium, lead, copper, zinc, manganese, iron, and fluoride. The amount of these substances in the soil appeared to be related both to the percentage of clay particles and organic matter in the soil and to the frequency of tidal submergences.

In vegetation, the pollutants clearly showed a seasonal variation. Adherent clay, deposited on the plants during submergences, contributed considerably to the total amount of these elements. In sheep grazing in the marsh, investigated for renal and fecal excretion of these elements, these samples and seasonal variation in vegetation were not related. Presumably sheep consumed vegetation selectively, avoiding the more contaminated plants. The sheep which received regular clinical inspection revealed no signs of acute or chronic

intoxication. The organs of sheep that died during the investigation failed to show enough increase in levels of cadmium in the liver and kidney, and iron in the liver to cause alarm. Fluoride found in rib, although slightly increased, did not indicate fluorosis.

In conclusion, contamination with metals and fluoride, as observed in the salt marsh, apparently failed to impair the health of locally grazing sheep.

**KEY WORDS:** Environmental fluoride; Fluoride contamination; Heavy metals; The Netherlands; Salt marsh; Sheep.

**REPRINTS:** Central Veterinary Institute (Centraal Diergeneeskundig Instituut), Dept. of Analytic Chemistry and Toxicology, P.O. Box 65, 8200 Lelystad, Netherlands.

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#### EFFECT OF FLUORIDE INGESTION ON DENTAL FLUOROSIS IN SHEEP

by

G.E. Milhaud, M.A. Borba and S. Krishnaswamy  
Maisons-Alfort, France

(Abstracted from *Am. J. Vet. Res.*, 48:873-879, 1987)

The effects of fluoride ingestion (3.5 mg/kg of body weight) were evaluated in 9 ewes at 6 through 9, 10 through 13, or 14 through 17 months of age (3 ewes/age group). In the 3 groups, the plasma fluoride concentration rapidly increased to 0.45, 0.46, and 0.50 microgram/mL, respectively, and decreased rapidly to 0.1 microgram/mL after fluoride was removed from the ration. In 5 of the 9 ewes, this short-term exposure caused symmetrical, moderate damage to the molars, which is characteristic of fluorosis in sheep. In 7 of the 9 ewes, abnormal gaps were found between incisors, which are not characteristic of fluorosis in sheep. Fluoride accumulation was higher in the roots of the incisors and in the dentine of the molars, which was particularly evident in teeth that were already developing when fluoride was administered; fully developed teeth and teeth that developed several months after administration of fluoride were likewise affected. The increase in fluoride concentration in the enamel was confined to a maximum of 2 molars. The abnormal wear of the molars was not directly linked to fluoride accumulation.

**KEY WORDS:** Dental fluorosis; Ewes, sheep; F in incisors, molars; Plasma fluoride.

**REPRINTS:** Ecole Nationale Vétérinaire d'Alfort, Service Pharmacie et Toxicologie, 7 Avenue Général de Gaulle, 94701 Maisons-Alfort, France.

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FLUORIDE CONTENT OF WATER, DUST, SOILS AND CEREALS  
IN THE ENDEMIC DENTAL FLUOROSIS AREA OF KHOURIBGA (MOROCCO)

by

Y. Haikel, J.C. Voegel and R.M Frank  
Strasbourg, France

(Abstracted from *Archs. Oral Biol.*, 31:279-286, 1986)

A systematic study of the fluoride content of the water was made to determine whether phosphate dust was a source of the fluoride in drinking water or whether uptake of fluoride ions through food, could possibly explain the fluorosis.

The fluoride content of drinking water, dust, soil, straw, raw barley (ears and stems) and barley grains in Khouribga were compared with those from a second Moroccan area, Beni-Mellal, the control area 90 km east of Khouribga, where no fluorosis occurs. Climatic conditions are similar to those of Khouribga but it contains no phosphate factories.

For the 156 rural wells sampled in the Khouribga area the mean value was  $0.7 \pm 0.39$  ppm  $F^-$ ; 82 per cent of the water samples contained less than 1 ppm  $F^-$ . Beni-Mellal, the town used as a control had a water fluoride content of  $0.07 \pm 0.04$  ppm  $F^-$ . Unexpectedly, therefore, larger amounts of fluoride appeared in solution with time and at a saturation when particle size increased. Large phosphate rock particles, probably of francolite, and smaller phosphate particles of clay-contaminated francolite, randomly collected in the factories of Khouribga, never contained more than 1.2 ppm  $F^-$ .

The soil of the Khouribga area contained abnormally high levels of fluoride, averaging  $5525 \pm 4985$  ppm  $F^-$  whereas only 130 ppm was the average in Beni-Mellal. The highest fluoride levels (6,691 ppm  $F^-$ ) were found in the south-west and the lowest in the north (1829 ppm  $F^-$ ). Close to the industrial center, and up to a distance of 5 km from it, the mean content was approximately 3% by weight (30,000 ppm), 40 km distant the fluoride content of the recovered particles was only 21,000 ppm. No dust was found at any of the three sampling sites in Beni-Mellal.

In 1982, the factories located in Khouribga processed 30,000 tons of phosphate rock a day, 0.56% of the total treated phosphate is discharged as small particles into the atmosphere. Thus, through industrial emission, 167 tons of fluoride-containing phosphate particles would be ejected into the atmosphere of the area every day.

The fluoride content of the dried unwashed straw samples was 462 ppm and that of similar washed samples 159 ppm. These levels are high compared to the value of 10 ppm in the control area of Beni-Mellal. Unwashed raw barley (ears and stems) contained an average of 40 ppm washed samples, 23 ppm, much higher than the 6 ppm in similar samples from the control areas of Beni-Mellal. The fluoride concentrations measured in dried barley grain indicated a mean value of 6.5 ppm similar to that found in Beni-Mellal (4.5 ppm).



In the Khouribga area the animals start to limp, their milk production decreases and their teeth are often so irregularly worn that the animals cannot chew and graze. There marked fluorosis in herbivores comes more from ingestion of dust-contaminated food (unwashed straw and unwashed raw barley) with a high fluoride content than from inhalation of fluoride-containing dust. On the other hand endemic human fluorosis in the same region is mostly due to inhalation of fluoride containing phosphate dust – not to fluoride in drinking water – only exceptionally to fluoride deposited during storage on cereals, because grains grown in Khouribga have a low fluoride content.

The grain kernal itself is well protected by its husk, and little uptake of fluoride occurs from the soil to the inside of the grain. People could not ingest sufficiently high levels of fluoride to explain the fluorotic lesions simply by eating bread made from such grains unless the grain becomes contaminated by dust.

Because animals live mainly on the locally-harvested straw which contains large amounts of fluoride originating both from air-borne deposits on plants, and from harvest and storage techniques used in these areas, they suffer more serious fluoride intoxication than humans.

**KEY WORDS:** Air-borne fluorides; Animal fluorosis; Cereals; Dental fluorosis; Fluoride dust; Grains; Morocco; Phosphates; Soil.

**REPRINTS:** Unité de Recherches INSERM U157, Faculté de Chirurgie Dentaire, 4 Rue Kirschleger, 67000 Strasbourg, France.

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CHANGES IN ORAL HEALTH FROM 1973 TO 1982  
OF 13-15-YEAR-OLD SCHOOLCHILDREN RESIDING  
IN THREE DIFFERENT FLUORIDE AREAS IN FINLAND

by

K. Parviainen,\* J. Ainamo and H. Nordling  
Helsinki, Finland

(Abstracted from *J. of Dental Research*, 64:1253-1256, 1985)

To analyze the change in the oral health of 13-, 14-, and 15-year-old schoolchildren residing in Finland, three towns were selected for both the original study in 1973 and the follow-up study in 1982 namely Jyväskylä, with 0.2 ppm, Kuopio, with 1.0 and Hamna, with at least 2.5 ppm F in community drinking water.

Improvement in dental health was mainly due to a dramatic decline of caries in the low-fluoride (0.2 ppm F) area, whereas in the high-fluoride area (2.5 ppm F), the improvement in oral hygiene and gingival health was the greatest.



During the nine-year period, the socio-economic level had changed for the better in all three locations. The clinical examination was performed exactly in the same manner in 1973 and 1982 and by the same examiner (KP). It consisted of assessment of plaque, gingivitis, and caries experience in the right sides of the jaws.

The overall mean VPI (Visible plaque index) score for the total study population had decreased to 39% in 1982 (44% in 1973). This slight overall reduction was mainly due to improved oral hygiene among boys ( $p < 0.05$ ) and among the 13-year-old girls ( $p < 0.01$ ). According to a separate calculation, the total mean VPI score for the boys remained, even in 1982, at a statistically significant higher level ( $p < 0.001$ ) than that for the girls.

The significant difference in 1973 between total mean GBI (Gingival Bleeding Index) scores for boys and girls had disappeared in 1982.

The marked differences in caries experience observed among three fluoride areas in 1973 were no longer found statistically significant in 1982, except that the total mean DFS (decayed or filled surfaces) score for the high-fluoride area was still significantly lower ( $p < 0.01$ ) than those of the other fluoride areas.

The significant negative correlation ( $p < 0.05$ ) between GBI and DFS in the high-fluoride area in 1973 was no longer there in 1982.

The general trend of declining DFS scores was found to originate mainly from the town with the lowest fluoride content in the drinking water. This finding is all the more important since low fluoride levels in drinking water are characteristic of most parts of the country. The striking difference between the low fluoride and the two fluoride areas in this respect would seem to negate to some degree the frequently expressed assumption that the decline in DFS scores might be strongly influenced by a change in diagnostic and treatment criteria. On the other hand, the same observation could mean that local application of fluorides, although more expensive than water fluoridation, can yield a level of caries prevention similar to that provided by consuming fluoridated drinking water.

Most of the differences between age and sex groups in 1973 had disappeared or strongly decreased by 1982. This change may pertain to an increased awareness of the importance of oral hygiene in modern society and is corroborated by a corresponding increase in the sale of both toothbrushes and fluoride toothpaste in Finland (Tala and Ainamo, 1982). That caries reduction was greatest in the area where oral hygiene had improved and gingivitis was lowest supports earlier views on a low correlation between mechanical cleaning of the teeth *per se* and dental caries (Ainamo, 1980).

KEY WORDS: Caries decline; Gingival health; Low and high F areas.

REPRINTS: School of Dentistry, Emory University, Atlanta, Georgia 30322, USA.

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EFFECT OF A MOUTHRINSE CONTAINING CALCIUM LACTATE  
FOR THE FORMATION AND REMINERALIZATION OF DENTAL PLAQUE

by

J.S. van der Hoeven, M.J. Schaecken and T.J. Creugers  
Nijmegen, Netherlands

(Abstracted from *Caries Res.*, 23:146-150, 1989)

A mouthrinse containing calcium lactate was tested for its effect on accumulation of dental plaque and on concentrations of calcium and phosphorus by human volunteers who rinsed four times per day with a calcium lactate (165 mmol/L) solution for 1 week. Plaque samples, collected 16 h after the last rinse were analyzed chemically. Calcium lactate rinses failed to affect the plaque score; calcium and phosphorus in plaque increased approximately twofold. It is noteworthy that incorporation of monofluorophosphate (5 mmol/L) into the rinsing solution failed to influence significantly calcium, phosphorus, and fluoride levels in plaque. Increased mineral deposition in the plaque may provide an explanation for the reduced caries development observed earlier in rats fed a diet containing calcium lactate.

KEY WORDS: Calcium lactate; Dental caries; Plaque mineralization.

REPRINTS: Dept. of Oral Health, University of Nijmegen, The Netherlands.

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COMPARATIVE EFFECTS OF A 4-YEAR FLUORIDE MOUTHRINSE PROGRAM  
ON HIGH AND LOW CARIES FORMING GRADE 1 CHILDREN

by

J.A. Disney, R.C. Graves, J.W. Stamm, H.M. Bohannon and J.R. Abernathy  
Chapel Hill, North Carolina, USA

(Abstracted from *Community Dent. Oral Epidemiol.*, 17:139-143, 1989)

Effectiveness of fluoride mouthrinse (FMR) was compared on high and low caries forming children after a 4-yr exposure to weekly rinse beginning in the first grade. Over 1200 grade 1 children drawn from both low fluoride and fluoridated sites were divided into treatment and concurrent, longitudinal control groups. After 4 yrs these children were stratified according to caries increment; those above the 75th percentile were considered high-caries formers, all others were designated low caries formers. After adjustment of the mean increments for differences in SES, age, race, and sex in rinse and control groups, high-caries formers (approximately 25% of the children) in the rinse and control groups in low-fluoride areas showed increments of 7.00 and 7.79 surfaces, respectively, a saving of 0.79 surfaces. Low caries formers (approximately 75% of the children) demonstrated increments of 1.11 DMFS in the rinse group and 1.40 in the control group (saving 0.29 DMFS).

The pattern was quite similar for children in fluoridated areas except that the increments, as well as the savings realized, were lower.

The results raise questions as to the practical effectiveness of school based FMR programs even for high caries forming children.

**KEY WORDS:** Fluoride mouthrinse; High caries forming children; Low caries forming children.

**REPRINTS:** Dept. of Dental Ecology, School of Dentistry, University of North Carolina, Chapel Hill, NC 27599, USA.

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INVESTIGATIONS ON THE RELEVANCE OF DEFLUORIDATED WATER AND  
NUTRITIONAL SUPPLEMENTS IN FLUOROSIS ENDEMIC AREAS  
IN ANDHRA PRADESH, INDIA

by

Kola Rajyalakshmi, N.V. Ramamohan Rao and Neelam Krishna  
Hyderabad, India

(Abstracted from **Fluoride Research 1985**, Elsevier, Amsterdam, 1986)

In Nalgonda, Andhra Pradesh, one of the districts in India severely affected by dental and skeletal fluorosis, several hundred people are crippled by the disease. Seventy-two patients in each of four villages in the age groups of 1-5, 5-10, 10-18, and above 18 years old, were provided defluoridated water and nutritional supplements following baseline studies on dietary intake and clinical, radiological, and biochemical profiles. The results of investigations, conducted once in three months each year after commencement of the intervention program, were compared with initial data.

Relief in clinical symptoms of body and joint pains was gradual. Radiological profiles of the patients indicated that fluoride remobilization from bones is reversible but the rate of removal is slow. Statistical evaluation of biochemical data revealed significant reduction in concentrations of alkaline phosphatase and serum fluoride. The investigations showed that the affliction can be significantly reduced through simultaneous consumption of defluoridated drinking water and nutritional supplements. Fluoride already absorbed is remobilized and excreted in the urine even after cessation of excessive fluoride ingestion through drinking water.

**KEY WORDS:** Defluoridated water; Dental, endemic, skeletal fluorosis; Fluoride remobilization; India; Joint pains; Nutritional supplements.

**REPRINTS:** Institute of Preventive Medicine, Public Health Laboratories and Food (Health) Authority, A.P., Hyderabad 500 029, India.

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## HUMAN PLASMA FLUORIDE LEVELS FOLLOWING INTAKE OF DENTIFRICES CONTAINING AMINEFLUORIDE OR MONOFLUOROPHOSPHATE

by

K. Trautner and J. Einwag  
Wurzburg, Federal Republic of Germany

(Abstracted from *Arch. Oral Biol.*, 33:543-546, 1988)

After single, oral doses, 8-hour profiles of fluoride (F) concentrations in plasma were determined in healthy human volunteers. Comparison of bioavailability of F from four dentifrices either aminefluoride (AMF) or monofluorophosphate (MFP) with that of NaF revealed no significant differences with respect to F availabilities, plasma F peak levels and plasma F profiles, regardless of the F compound, F content or further ingredients. It is concluded that dentifrices with 0.02 percent F or less markedly decrease the risk of inducing plasma F concentrations that might disturb enamel mineralization, even if large quantities of dentifrice are ingested by small children.

**KEY WORDS:** Aminefluoride; Fluoride dentifrices; Monofluorophosphate; Plasma fluoride.

**REPRINTS:** Dental School, University of Wurzburg, Pleicherwall 2, D-8700 Wurzburg, FRG

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## INFLUENCE OF FOOD ON RELATIVE BIOAVAILABILITY OF FLUORIDE IN MAN FROM Na<sub>2</sub>FPO<sub>3</sub>-CONTAINING TABLETS FOR THE TREATMENT OF OSTEOPOROSIS

by

K. Trautner  
Wurzburg, Federal Republic of Germany

(Abstracted from *Int. J. Clin. Pharmacol. Ther. Toxicol.*, 27:242-249, 1989)

Aqueous solutions of NaF and sodium monofluorophosphate (Na<sub>2</sub>FPO<sub>3</sub>, MFP) and tablets containing MFP (Caflu, Tridin) were given as single oral doses to fasting healthy volunteers in an intra-individual crossover study. Serum fluoride (F) and urinary F concentrations were measured using an F-ion-sensitive electrode; 8 h profiles of F concentrations in serum and 24 h urinary F output were determined. F availability and pharmacokinetic data were identical for all items.

When Caflu tablets given under the following varying experimental regimens a) on a fasting stomach, b) with breakfast, c) with milk, d) with breakfast and milk, serum peak levels were reduced provided tablets were taken with food. Intake with breakfast failed to affect F bioavailability, where-

as milk reduced F availability by 28% compared with the fasting stomach experiment. This effect was partly abolished when milk was taken as part of the breakfast. It is suggested that entrapment of F in coagulation products of milk and formation of Ca-salts are important factors in causing reduction of F availability, and that prolonged stay of the chyme after concomitant ingestion of food allow F to become liberated from bound forms and coagulation products by digestive processes.

KEY WORDS: Fluoride tablet bioavailability; Osteoporosis therapy; Sodium monofluorophosphate.

REPRINTS: Dental School, University of Wurzburg, Pleicherwall 2, D-8700 Wurzburg, FRG.

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#### EFFECT OF SYSTEMIC FLUORIDE ON RAT MOLAR MORPHOLOGY. STEREOPHOTOGRAMMETRIC ANALYSIS

by

C.J. Creath, J.D. Eick and E.P. Hicks  
Tulsa, Oklahoma, USA

(Abstracted from *Caries Res.*, 23:26-31, 1989)

Female Sprague-Dawley rats, divided into three groups, were given the same solid diet (with 1.6 ppm F). Group I was given deionized water; Group II, water to which 5 ppm fluoride was added; Group III, 50 ppm fluoride in water. All females were mated with the same male.

Pups were sacrificed at 28 days, mandibles were removed and cleaned. Three-dimensional representations of occlusal surfaces of the 1st and 2nd mandibular molars were reproduced via computer-digitized images using stereophotogrammetry. Fissure depths and widths were measured, "volumes" of fissures were computed. Fissure widths, depths, and volumes were compared statistically using unpaired t-tests between groups ( $p < 0.05$ ). Differences were significant in fissure depth between Groups I and II between Groups I and III. For fissure volumes, significant differences were found between Groups I and III. Width of fissure and overall tooth size were not significantly different between fluoride-treated groups. Standardization showed a reproducibility to within  $\pm 40$  mm with the stereophotogrammetric method.

KEY WORDS: Molar morphology; Rats; Systemic fluoride.

REPRINTS: Michael Cardone Senior School of Dentistry, Oral Roberts University, Tulsa, OK, USA.

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ANALYZING THE FLUORIDATION CONTROVERSY:  
RESOURCES AND STRUCTURES

by

Brian Martin\*  
Wollongong, NSW, Australia

(Abstracted from *Social Studies of Science*, 18:331-363, 1988)

Fluoridation has been endorsed by most established public health, dental and medical bodies, whereas concerted opposition has been organized by voluntary citizen groups, supported by a few relevant professionals. Many dentists, physicians and lay groups fail to examine the scientific and ethical issue in any detail. Instead they rely upon views of individuals or organizations they trust. Refusal by proponents to debate in local fluoridation controversies — before referenda or administrative decisions — is common. Their stance being that the measure is scientific beyond question but that a concerted political struggle must be waged in order to obtain it. Attacks and criticism without any context or commentary are used to disparage the individual or groups named, rather than to deal with the issues they raise. Professionals who oppose fluoridation are inhibited from speaking out by threats and actions which potentially jeopardize their careers and help to ensure that relatively few reports or articles critical of fluoridation are ever submitted to scholarly journals. Of those that are submitted, evidence is available that it is unusually difficult to obtain publication. The pro-fluoridationists, through their influence over editorial policy of journals and publishers, have used their power to prevent publication of anti-fluoridation findings and views by scientists.

Fluoride is a major industrial pollutant, especially for the aluminum and phosphate fertilizer industries. Release of fluoride, for example, has been opposed by farmers because of its detrimental effects on crops and animals. Fluoridation provides a profitable outlet for some of the fluoride industrial waste. If the public perceives fluoride as a dangerous chemical, pressure for tighter control is likely to be great. On the other hand establishment of a benign image of fluoride lessens this problem for industry. Promotion of water fluoridation as a public health measure has served this purpose admirably. It has been carried out mainly by a tiny fraction of dental and medical professionals, some of whom, especially researchers into fluoride and tooth decay, have built their reputations on work tied to the promotion of fluoridation. "Altruism" at the material level does not apply here. Many others, active in campaigning for fluoridation, develop a personal stake in the issue: their public image and self-image are tied to the benefits of the measure. Control over professional organizations by pro-fluoridationists which makes possible the "suppression" of opponents, for example by threatening to debar a dissident from practice; the control over political resources associated with the dental and medical professions gives rise to the major asymmetry in the fluoridation debate.

KEY WORDS: Fluoridation controversy; Fluoridation opposition; Fluoridation promotion.

REPRINTS: Department of Science and Technology Studies, University of Wollongong, P.O. Box 1144, Wollongong, New South Wales 2500, Australia.

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## INSTRUCTIONS TO AUTHORS

*Fluoride*, the official journal of the International Society for Fluoride Research (ISFR) is published quarterly (January, April, July, October). Its scope is the publication of papers and reports on the biological, chemical, ecological, industrial, toxicological and clinical aspects of inorganic and organic fluoride compounds. Papers presented at the annual ISFR conference are published in *Fluoride*. Submission of a paper implies that it presents original investigations and relevant bio-medical observations. Review papers are also accepted.

### PREPARATION OF PAPERS

1. **General** — No precise limit is given on the length of the paper; it should be written concisely in English, submitted in two copies, doublespaced with generous margins. Measures are given in metric system (SI).
2. **Title** — A concise but informative title should be followed by the name of author(s), the location and state (country) where the research was carried out. The name and address of the institution where the work was done should appear at the bottom of the first page.
3. **Summary** — The paper should begin with a brief, factual summary.
4. **Introduction** — Following the summary, a short introduction should state the reason for the work with a brief review of previous works on the subject. References are given by numbers in parentheses.
5. **Materials and Methods** — should be condensed; however if the methodology is new or developed by the author(s) it can be more detailed.
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8. **Abbreviations or Acronyms** — must be defined either parenthetically or in a footnote when they first appear.
9. **Bibliography** — should be arranged according to the order in which the articles are cited in the text (not alphabetically). An example follows:

Fiske, C.H. and Subba Row, Y.: The Colorimetric Determination of Phosphorus. *J. Biol. Chem.*, 66:375-400, 1925.

For books, the title, editor, publisher, location and year of publication, and pages should be given.

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