

CALCIUM REQUIREMENT FOR MINIMIZING FLUORIDE TOXICITY

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ABSTRACT. Total intake values per day of fluoride and calcium through water and food by selected human subjects living in fluorotic and non fluorotic areas in Tamilnadu of South India were determined. The maximum possible levels of Ca^{2+} ions in solution without precipitating fluoride at different fluoride concentrations were calculated using solubility product principle. The necessary intake of calcium per day required to reduce fluoride toxicity in the subjects studied was estimated. The results reveal that the high intake of calcium through water and food leads to a marked reduction in the ionic fluoride levels in two of the three fluorotic areas, thereby reducing fluoride toxicity.

INTRODUCTION

Fluoride toxicity is characterized on the basis of fluoride intake through water and food. Generally fluorosis is interpreted in terms of the contribution of fluoride through water alone. It is recognized that fluoride intake through food cannot be ignored in assessing the magnitude of fluorosis and it is the total fluoride intake which should be taken into consideration [1,2]. Several studies indicate that higher intake of calcium reduces fluoride toxicity [3-8]. However, quantitative attempts have not been made so far to determine the amount of total calcium intake per day necessary at a given fluoride concentration to reduce fluoride toxicity. The role of total calcium in defining the degree of fluorosis in different fluorotic environments is discussed in this paper along with the necessary calcium intake which precipitates fluoride as calcium fluoride.

EXPERIMENTAL

Criteria for selection of control and fluorotic areas. The criterion for the selection of controls is the state of absence of fluorosis in any of its forms in that particular area. One control (C1) was selected from a non endemic area and the second control (C2), from an endemic area. The criteria for selecting fluorotic areas involved (i) the difference in degree of fluorosis and (ii) the difference in the fluoride concentrations in drinking water. Based on these criteria three fluorotic areas were identified as F1, F2 and F3. Surveys for fluorosis were conducted among the primary and middle school students in the villages of two control areas and three fluorotic areas of Tamilnadu in South India using random sampling technique [9]. A door to door survey for fluorosis was also conducted among adults of the areas of study with the help of experienced clinical examiners. Selection of families was done using purposive sampling technique [9]. From each family two adults and two children were selected, totalling eighty subjects (40 children and 40 adults). This was done in order to determine the total fluoride

and total calcium intake. Control area C1 consisted of 12 villages of Thiruchuli block of Kamarajar District. Control area C2 contained 9 villages of Athoor block of Dindigul district. Fluorotic area F1, where dental fluorosis is prevalent and fluoride in drinking water is less than 1 mg/L (0.34-0.91 mg/L) consisted of 10 villages of Athoor block of Dindigul district. Fluorotic area F2, where dental fluorosis is prevalent and drinking water fluoride is between 1 and 3 mg/L consisted of 10 villages of Nilakkottai block of Dindigul district. Fluorotic area F3, where dental and skeletal fluorosis were prevalent and fluoride in drinking water varied from 3.8-8.0 mg/L consisted of 4 villages of Ayothiyapatnam block of Salem district.

Analysis of drinking water and food samples. The exact quantity of diet and the volume of water, each subject consumed in a day were collected separately in high density polyethylene containers. The normal diet consumed by the people of the region was analysed to determine the fluoride intake through food. This consisted of vegetarian food comprising cereals, pulses, locally grown and available vegetables and fruits, besides beverages like coffee, tea and milk. The liquid diet was filtered and whatever solid present in it was added to the solid diet. The solid diet was weighed and ground to a homogeneous mixture. The liquid diet and water were measured separately. In a similar way collection of diet samples from each subject was done for four alternate days in order to assess the total fluoride intake.

The food samples for fluoride analysis were distilled based on the classical Willard-Winter distillation method [10] and the distillate was analysed by the fluoride ion selective electrode. 25 g of homogenised food sample was taken in a platinum crucible to which 10 mL of calcium hydroxide suspension was added and mixed thoroughly. The sample was evaporated to dryness in a hot air oven and then ashed in a muffle furnace at 600 °C for 5 h. The ash was transferred to a distillation flask. Sufficient AnalaR silver sulphate was added to precipitate any chloride present in the sample. Pieces of glass and porcelain were also put in the flask to avoid bumping. 40 mL of 60% AnalaR perchloric acid and 20 mL of distilled water were added and then distillation was carried out. The distillate, collected between 135-139 °C was neutralised using 0.2 N sodium hydroxide and was analysed for fluoride using fluoride ion selective electrode 9409 and an expandable ion analyser EA 920 Orion USA [11]. Similarly fluoride content of drinking water samples was measured using fluoride ion selective electrode.

The procedure adopted for determining calcium in diet samples was based on the method prescribed in the manual of Atomic Absorption Spectrophotometer, Perkin-Elmer 380 [12]. The flame system used was acetylene-nitrous oxide and measurements were made at the wavelength 422.7 nm. 5 g of homogenised food sample was taken in a Kjeldahl flask, to which 10 mL of concentrated nitric acid was added. 5 mL of concentrated sulphuric acid was added to this mixture and the contents were digested for an hour. After the completion of digestion, the digest was diluted to 100 mL and was used for analysis. The amount of calcium present in drinking water was also analysed using the same instrument used for diet samples.

RESULTS AND DISCUSSION

Magnitude of the problem of fluorosis. Studies conducted in this laboratory for the above mentioned five areas established a direct correlation between the mean fluoride level in drinking water and the % of incidence of dental fluorosis [13].

Total fluoride intake through water and food. Fluoride intake through water and food and the total fluoride values per day were determined and the mean values were computed separately for adult male, female and children and these values are compared in Figure 1. A comparison of the results of different areas lead to the following conclusions.

- The values of total fluoride, water fluoride and food fluoride for the two controls are nearly the same. The increase in water fluoride from controls to fluorotic areas is significant, as expected. The increase is higher from F1 to F2 and still higher from F2 to F3. In fluorotic areas F2 and F3, it is the water fluoride which mainly contributes to the total fluoride intake and the percentage values for the same are 54 and 90, respectively. For the two control areas C1, C2 and fluorotic area F1, in which concentration of fluoride in water is 1 mg/L, fluoride contribution through food to total intake is quite significant.
- Total intake of fluoride is in the order $C1 < C2 < F1 < F2 < F3$. This is as expected and in accordance with the levels of water fluoride of the respective areas.

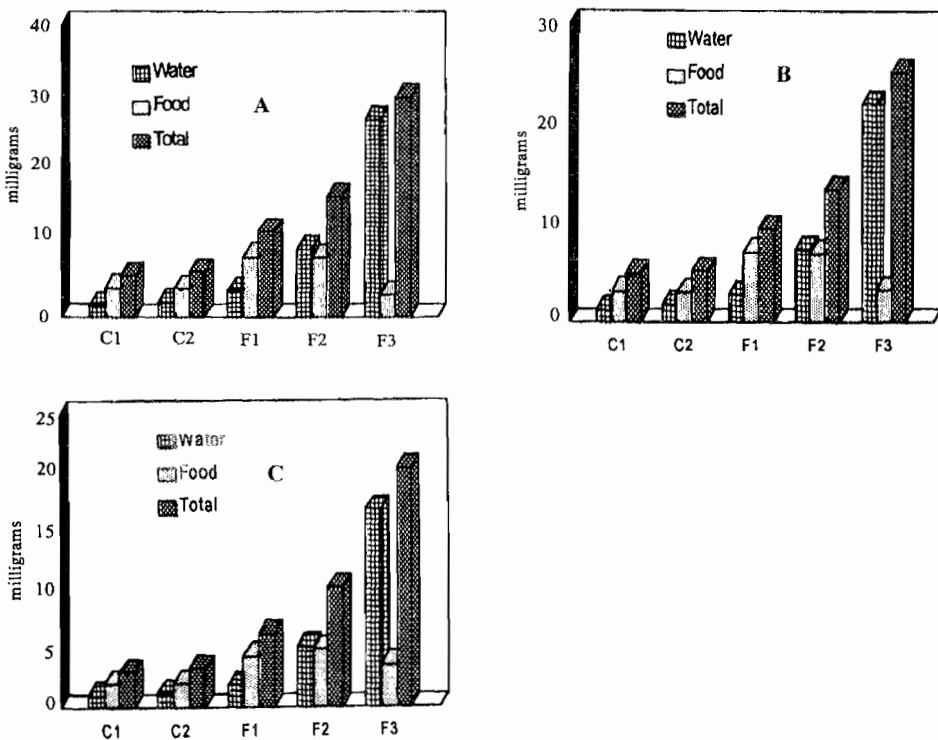


Figure 1. Fluoride through water, food, and total intake per day: mean values (A) adult male, (B) adult female, and (C) children.

Spencer *et al.* conducted similar fluoride intake studies in Chicago and reported that the total fluoride intake is 3-4 mg/day in adults where the drinking water fluoride is 0.9 mg/L and in non fluorotic areas where water fluoride is 0.3 mg/L, the total fluoride intake is of the order 1.1-1.3 mg/day [14-16]. According to a report from WHO [17] the total fluoride intake in

persons residing in low water fluoride areas (less than 0.4 mg/L) did not exceed 1 mg/day unless it is modified by higher amounts of fluoride through diet. McClure [18] in his fluoride balance studies found that fluoride balance did not become significantly positive until the intake was above 4-5 mg/day. In comparison to 4-5 mg/day given as the minimum threshold value beyond which there is fluoride toxicity, the total fluoride intake values in controls C1 and C2 are slightly higher than the above value and even then there is no prevalence of fluorosis in these areas. In fluorotic area F1, the total fluoride intake exceeded this limit to a very great extent and hence fluorosis is observed. In fluorotic area F2, total fluoride is about three times the threshold value, while in fluorotic area F3, the total fluoride intake is 6 times higher. Such higher fluoride intake values clearly explain the magnitude and severity of the problem of fluorosis in fluorotic areas F2 and F3.

Total calcium intake and fluoride toxicity. The mean values of intake of calcium through water and food and also the total intake by the adult male, female and children living in controls and fluorotic areas were computed and the trend is given in Figure 2. Calcium intake per day through water is in the order: C1 > C2 > F2 > F3 > F1. Calcium intake through food is in the order F2 > C1 > C2 > F3 \cong F1. Total calcium intake per day follows the order C1 > C2 > F2 > F3 > F1.

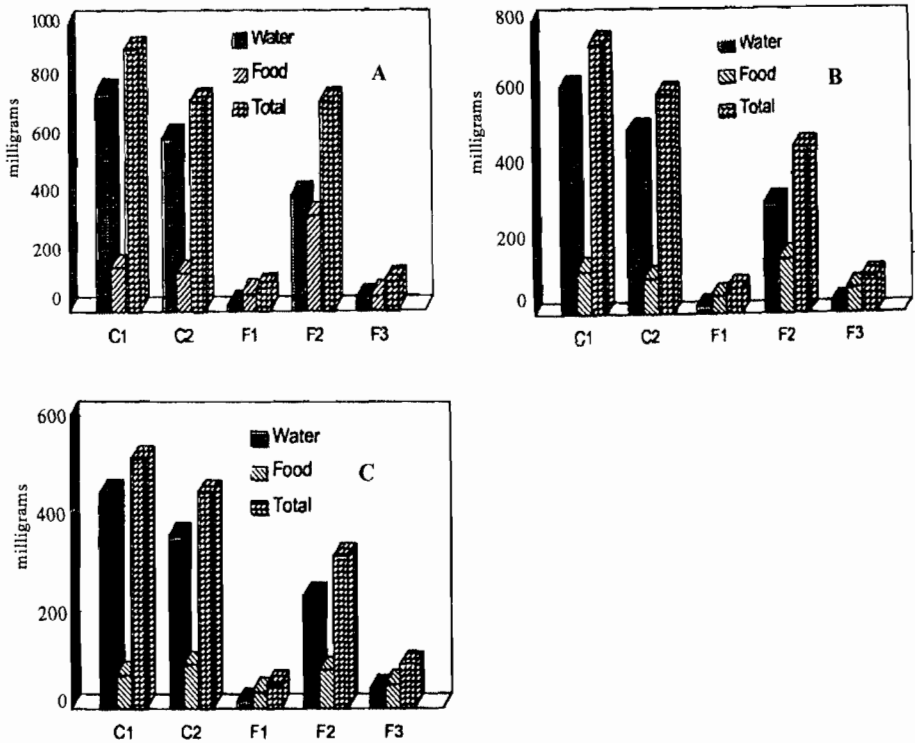


Figure 2. Calcium through water, food, and total intake per day: mean values (A) adult male, (B) adult female, and (C) children.

The maximum possible amount of calcium available as ions, at various fluoride concentrations of the control and fluorotic areas of the present study, are calculated based on the solubility product principle as applied to aqueous solutions [19]. The results obtained explain the significance of total calcium intake on fluoride toxicity. It must be clearly borne in mind that these calculations and results are of academic interest only. No conclusions can be drawn on the exact quantities of calcium ions needed to precipitate fluoride in the human system based in these results which are given in Table 1.

Table 1. Maximum possible Ca^{2+} ions at various F^- levels.

Area	Total intake of F^- by adult male	Corresponding F^- concentration		Maximum possible concen- tration of Ca^{2+} in solution		Corresponding total intake value for Ca^{2+} in solution
	mg	mg/L	mol/L	mg/L	mol/L	mg
C1	5.58	1.223	6.436×10^{-5}	376.61	9.415×10^{-3}	1718
C2	5.98	1.305	6.868×10^{-5}	330.72	8.268×10^{-3}	1515
F1	11.47	2.510	1.322×10^{-4}	89.28	2.232×10^{-3}	407
F2	16.53	3.610	1.901×10^{-4}	43.17	1.079×10^{-3}	197
F3	30.17	6.950	3.658×10^{-4}	11.66	2.910×10^{-4}	51

It is obvious from the table that a daily intake of calcium by an adult male, to precipitate any fluoride at a concentration of 1.2228 mg/L (total intake of 5.58 mg/day) and to reduce the ionic fluoride level has to be greater than 1718 mg.

Similarly the amount of calcium ions in solution at a given concentration of fluoride (based on the total intake value) without precipitating as calcium fluoride in control area C2 and three fluorotic areas F1, F2 and F3 were found to be 330.72, 89.28, 43.17 and 11.66 mg/L respectively. The corresponding values for total calcium in the two control areas and three fluorotic areas are 1718, 1515, 407, 197 and 51 mg, respectively. That is, the amount of calcium intake needed to precipitate any fluoride must exceed 1718, 1515, 407, 197 and 51 mg in the two control areas and three fluorotic areas with reference to adult male subjects. But the actual total intake values per day for adult male in the control areas C1 and C2 and fluorotic areas F1, F2 and F3, respectively, are 909.7, 734.2, 80.3, 727.8 and 100.2 mg, as shown in Figure 2A. The total calcium intake values in the control areas C1 and C2 and fluorotic area F1 are far less than the required calcium to precipitate any fluoride and this indicates that the total fluoride in these areas is completely in the ionic form. In the case of fluorotic area F2, the actual total calcium intake per day viz., 727.8 mg is much higher than the calcium needed to precipitate fluoride. In this area the excess calcium may cause precipitation of fluoride and thus reduce ionic fluoride content significantly. Similarly, in fluorotic area F3, the actual calcium intake is 100.2 mg which is also sufficiently above the required amount to effect precipitation of fluoride.

The amount of fluoride remaining in solution in fluorotic area F2 is again calculated on the basis of solubility product principle. Out of the total intake of 16.53 mg per day, about 7.9 mg of fluoride precipitates out and the balance 8.63 mg remain in solution in the form of ionic fluoride and this is responsible for fluoride toxicity of that area, F2 [19]. Therefore it is concluded that the entire 16.53 mg per day of fluoride corresponding to a daily intake of 727.8 mg of calcium do not contribute fully for fluoride toxicity.

The amount of ionic fluoride in solution corresponding to the recommended dosage of 600 mg of dietary calcium [20-21] among children is calculated. 4.3 mg of fluoride per day

(equivalent to 1.592 mg/L) is found to be in the ionic form irrespective of any greater amount of fluoride intake per day. Hence at this level of recommended dietary allowance of 600 mg of calcium per day by children (11-15 years) the problem of fluoride toxicity will be greatly reduced, irrespective of the high fluoride levels in drinking water. At the worst, the fluoride toxicity will be limited to the stage of dental fluorosis only. It could never reach the stage of crippling skeletal fluorosis.

However, the dietary intake of calcium per day in adults has to be at least twice the recommended allowance of 400-500 mg/day in order to achieve the desired reduction of fluoride toxicity. As such, for adult male and female subjects, the amount of necessary intake of calcium per day is calculated on the basis of solubility product principle and the values are given in Table 2. The amount of calcium intake per day for adult male and female is 1000 mg and 800 mg, respectively. Our findings lead us to conclude that at these levels of calcium, fluoride toxicity will be greatly reduced irrespective of the amount of fluoride intake.

Table 2. Maximum possible levels of fluoride in ionic form at various levels of total intake of calcium.

Subjects	*Necessary Ca ²⁺ to reduce F ⁻ toxicity	Corresponding Ca ²⁺ concentration		Maximum F ⁻ in solution		*Total F ⁻ in solution
	mg	mg/L	mol/L	mol/L	mg/L	mg
Children (10-11 years)	600	222.2	5.555x10 ⁻³	8.379x10 ⁻⁵	1.592	4.30
Adult female	800	222.2	5.555x10 ⁻³	8.379x10 ⁻⁵	1.592	5.73
Adult male	1000	222.2	5.555x10 ⁻³	8.379x10 ⁻⁵	1.592	7.16

*Note. When the concentration of Ca²⁺ is 222 mg/L, the corresponding fluoride ion in solution (maximum) can be only 1.59 mg/L. The differences in the total calcium intake and fluoride present in ionic form between adult male, adult female and children are only due to the differences in the quantity of water and diet consumed by them under Indian conditions. In this calculation, the quantities of drinking water and diet together by children (10-11 Years), adult female and adult male are taken as 2.7 kg, 3.6 kg, and 4.5 kg respectively.

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