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Determination of exposure and probable ingestion of fluoride through tea, toothpaste, tobacco and pan masala

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Abstract

Levels of water soluble and acid soluble fluoride in tea, toothpaste, tobacco and pan masala (mouth freshener) were estimated. These items are, generally, ignored while calculating the total dietary intake of fluoride. Tea, toothpaste, tobacco, pan masala (with tobacco and without tobacco) frequently expose human body to 3.88-137.09, 53.5-338.5, 28.0-113.0, 16.5-306.5 and $23.5-185.0 \mu g$ of fluoride per gram of these items, respectively. An effort was also made to quantify, on the basis of available studies, the probable human ingestion of fluoride through these substances. Increased leaching of fluoride from some of these substances has been observed in acidic conditions in the present study. The results can be extrapolated to acidic conditions of human stomach.

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Keywords: Fluoride; Tea; Toothpaste; Tobacco; Pan masala (mouth freshener)

1. Introduction

Fluorine is the 13th most abundant element in earth's crust. Human beings are frequently exposed to fluoride through food, water and various other products of daily use. Excessive ingestion of fluoride causes dental and skeletal fluorosis. Fluorosis is prevalent in India for the last six decades and the effect of fluoride on human health has been clearly understood by now. Fluoride enters the body through gastrointestinal tract and remains there as hydrofluoric acid [1]. From gut, it enters the plasma and 85% of it binds to serum albumin. It degenerates muscle fiber [2], adversely affects spermatogenesis [3] and influences the calcium current in neurological systems [4]. In India, the number of people consuming tea, tobacco, and pan masala is on the increase. Besides drinking water, these items, too, serve as important sources of fluoride. In addition to these, toothpaste also contains high level of fluoride. Due to the limited information on level of fluoride such items are ignored while calculating the total dietary intake of fluoride. This study, therefore, becomes important in determining the exposure of human body to fluoride through these items. This also stresses the need of inclusion of fluoride level of these items in determining the total dietary intake of the fluoride for consumer of such items. In addition, it will help to understand the leaching of fluoride from these items in simulated stomach conditions. An effort, therefore, has been made to quantify the probable ingestion of fluoride through these items and to discuss the bioavailability, metabolism and health effects of fluoride on human body in the light of recent researches for better understanding of the subject.

2. Materials and methods

In the present study, 8 samples each of tea and tobacco, 15 samples of pan masala (7 without tobacco and 8 with tobacco) and 15 samples of toothpaste were obtained from the local market. The samples were dried at 80 $^{\circ}$ C in hot air oven for sufficient time and were powdered. One gram of each sample was mixed with 50 ml of distilled water and stirred continuously on a magnetic stirrer for 30 min [5]. Tea samples were boiled for 5 min in water, instead of stirring. The samples were analyzed for water soluble and acid soluble (0.2 M HCl solution) fluoride in

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them. Fluoride analysis was done as per APHA-AWWA-WPCF method no. 4500-F⁻ D [6]. For the estimation of acid soluble fluoride, 0.2 M HCl was used in place of distilled water. Blank corrections were also made to minimize error.

3. Results and discussion

Human body is exposed to different levels of fluoride through consumption of different edible products. Ingestion pattern of fluoride varies for each product. Level of exposure and ingestion in case of tea is the same because it is fully ingested while it is different in case of toothpaste, tobacco and pan masala. Only a fraction of these is ingested in the body. Levels of fluoride in tea, toothpaste, tobacco and pan masala are presented in Table 1. Each gram of tea exposes human body to 3.88-137.09 µg of fluoride with a mean value of 63.51 µg while same quantity of toothpaste exposes the human body to $53.5-338.5 \,\mu g$ with a mean value of 183.78 µg. The range of exposure was found to vary between 28.0 and $113.0 \,\mu$ g/g with a mean value of 75.5 μ g/g for tobacco, while it was 16.5–306.5 μ g/g with a mean value of 113.93 µg/g for pan masala with tobacco. In case of pan masala without tobacco it varied between 23.5 and 185.0 μ g/g with a mean value of 63.88 µg/g. Generally, tobacco, toothpaste and pan masala are not swallowed, only some fraction of them is ingested during their consumption, which ultimately becomes available to body for absorption along with sublingually absorbed fluoride. Absorption of fluoride in the body depends on chemical and physical nature of the ingested fluoride. Soluble fluoride present in the food is efficiently absorbed. It has been observed that absorption of fluoride from water ranges from 86 to 97% while 20% of fluoride is soluble in water from less soluble edible items like tobacco, pan masala, etc. [5,7]. Studies on toothpaste indicated that up to 20% of toothpaste was ingested per brushing [8] but ingestion pattern of fluoride for tobacco and pan masala is not yet established. The ingestion is almost similar to that of toothpaste. Therefore, in this study, we have assumed

Level of water and	acid soluble fluoride	in different product

it to be similar to the fluoride ingestion from toothpaste, i.e. 20% for ingestion study. Consumption pattern of tea is totally different from toothpaste, tobacco and pan masala. In tea preparation, tea leaves are removed after boiling in water and this water is consumed directly. So, fluoride present in tea becomes available to body for absorption. Level of fluoride ingested through these items is calculated using mean value of fluoride presented in Table 2. On an average, a person who brushes once in a day (2 g per brushing) and consumes two cups of tea (2 g per cup) is exposed to 621.6 µg/day of fluoride. Out of this 327.55 µg/day is ingested, as there is 100% ingestion of fluoride for tea and 20% for toothpaste. Since consumption habits of pan masala and tobacco differ from person to person so they are exposed to different levels of fluoride, e.g. if a person consumes three sachets of pan masala (6 g/sachet) containing tobacco, then, he/she will further be exposed to 2050.74 µg of fluoride, out of which 410.15 µg/day is ingested. Exposure and ingestion of fluoride from these products would be different at different doses depending on the intake habits of different persons. Thus, highest level of exposure and ingestion of fluoride will be associated with a person consuming maximum quantities of these products.

It is evident from the results that leaching of fluoride from various products increases by few folds in acidic conditions. In the present study leaching of fluoride varying from 1.34 to 2.69 fold in acidic conditions as compared to extract by boiling in water has been observed in tea, toothpaste and tobacco. However, this trend was not observed for pan masala (mouth freshener). Hence, human beings can be exposed to higher fluoride level with same product because similar acidic conditions prevail in our stomach. Most of the children, who brush their teeth every day with high fluoride containing toothpaste, in both urban and rural areas, had gingivitis and periodontal diseases [9]. Net fluoride available to body depends on the bioavailability and its metabolism. Thus, this paper discusses the bioavailability and metabolism of fluoride as well as brief account of dental and skeletal fluorosis.

Sample	Water soluble F ⁻ (µg/g)			Acid soluble F ⁻ (µg/g)		
	Range	Mean	S.D. (±)	Range	Mean	S.D. (±)
Tea	3.88-137.09	63.51	80.31	2.99-329.01	170.94	227.99
Toothpaste	53.5-338.5	183.78	83.73	43.5-560.5	314.6	127.89
Tobacco	28.0-113.0	75.5	30.09	38.5-370.0	101.06	111.79
Pan masala with tobacco	16.5-306.5	113.93	94.78	0.5-148.5	98.145	0.94
Pan masala without tobacco	23.5-185.0	63.88	52.24	4.5-82.6	4.98	23.53

Table 2

Table 1

Daily exposure and ingestion of fluoride through different products

Sample	Quantity taken (g/day)	Exposure of fluoride (µg/day)	Ingestion of fluoride (µg/day)	% of ingestion
Теа	4	254.04	254.04	100
Toothpaste	2	367.56	73.51	20
Tobacco	10	755.0	151.0	20
Pan masala with tobacco	18	2050.74	410.15	20
Pan masala without tobacco	18	1149.84	229.97	20

3.1. Bioavailability of fluoride

Bioavailability is the portion of a nutrient in food, which is absorbed and utilized in living beings [10]. In humans, the bioavailability of fluoride from various food items is reported to vary from 2 to 79%. Parameters like pH and the mineral content of the food are of importance for bioavailability [11–14]. Bone meal, fish bone meal, canned sardines and chicken bone meal are considered as low fluoride availability food products, most probably, because of the high content of calcium [15]. Experiments on rats show that minerals like calcium and magnesium influence absorption and retention of fluoride in different tissues [16,17]. While calcium most likely interferes with the absorption of fluoride, it has been proposed that magnesium reduces the toxicity of fluoride by intracellular mechanisms [18,19].

Majority of studies on bioavailability of fluoride in humans have been focused on comparing fluoride concentration in plasma after intake of the test substance or test food. The results obtained have been compared with plasma values analyzed after oral or intravenous administration of NaF [13–15]. Some studies have reported bioavailability based on both, plasma values and urinary fluoride concentration [12,20]. The methodological approach is based on the assumption that NaF is fully absorbed (100%) in empty stomach, no endogenous loss of fluoride, and understanding that urinary excretion is the major route of fluoride elimination.

3.2. Metabolism of fluoride

Passive diffusion takes place during the absorption of fluoride in stomach and small intestine. If a person has not consumed any food, sodium fluoride (NaF) is absorbed completely and very rapidly [21]. Increase in fluoride level in plasma can be observed a few minutes after ingestion. The reason behind this is that fluoride in stomach, primarily, present as weak hydrogen fluoride (HF), which diffuses from the stomach and easily reaches the blood [22]. The plasma fluoride level is not homeostatically regulated. Fluoride is distributed from plasma to all tissues and organs. However, since the soft tissues generally do not accumulate fluoride most of the fluoride in the human body (99%) is found in mineralized tissue. Fluoride is incorporated into the crystal lattice structure in the form of fluorapatite or fluorhydroxyapatite [23].

In case of young individual, a larger fraction of single fluoride dose deposits in the skeleton as compared to adults [21]. In plasma, fluoride is transported as ionic fluoride and nonionic (bond) fluoride. Ionic fluoride does not bind to plasma proteins, and is easily excreted with the urine. However, in the form of HF, a variable amount of (35–45%) will be reabsorbed and returned to the systemic circulation. pH of tubular fluid and urinary flow are the main factors influencing reabsorption rate [21,22]. The amount of urinary fluoride excreted reflects the amount of fluoride ingested, and elimination of absorbed fluoride occurs mainly via kidney [23]. As reported in literature, the urinary excretion of fluoride (age group 3–6 years) is 32–80% of total fluoride intake [24–26]. Fluoride, in faeces, is that part which was never absorbed [27]. On an average, 6-25% of the fluoride ingested is lost in faeces [23,28,29].

3.3. Health effects

Fluorosis covers a wide range of clinical manifestation related to chronic fluoride intoxication. Fluoride accumulates in mineralized tissues, notably the lattice of bone and tooth crystals [30]. Fluorosis can be easily detected in teeth, in the form of mottling of tooth enamel. The clinical symptoms of mild dental fluorosis vary from thin white striae across the enamel surface to irregular cloudy area. In severe condition, the white areas merge and loss of enamel surface may occur. The loss of enamel occurs only on surface, and not the full thickness of the enamel [31,32]. Enamel becomes brownish in colour due to the uptake of colour from food to the porous enamel. After pre-eruptive maturation of enamel it becomes non-susceptible to dental fluorosis. The pre-eruptive maturation of the anterior teeth is finished by the age of 8 years [33].

Skeletal fluorosis can be defined as excessive deposition of fluoride in bone [34]. If high fluoride drinking water (>8 mg/l) is consumed over a long period of time during the adolescence, severe forms of skeletal deformities may occur [35]. There is strong correlation between level and duration of fluoride exposure and the development of skeletal fluorosis and its severity. The changes in skeletal tissue may result in back stiffness and limb pain due to ossification and fusion of the vertebral column and spinal ligaments [36]. In endemic region, approximately 10% of those with skeletal fluorosis exhibit neurological disease due to the mechanical compression of the spinal cord and nerve by bony growth (osterophytes) in the spinal canal [37]. A good correlation has been observed between the parodontopathy index and fluoride level in systemic fluids [38]. Fluoridated drinking water was found positively associated with cancer of oral cavity and pharynx, colon and rectum, hepato-biliary and urinary organs [39]. Human beings are protected from the adverse effect of high fluoride ingestion by the dietary habits as well as by various functions carried out by their organs, as 40-60% of fluoride ingested daily is excreted by kidneys through urine [26,40]. Calcium and magnesium form insoluble complexes with fluoride anion and multivalent cations in the small intestine and decrease the uptake of fluoride into bones and teeth significantly [40,41]. Besides calcium and magnesium, nutritional supplements involving high intake of Vitamins A, C, and D, high protein diet, etc. save human beings from detrimental effects of high level of ingested fluoride. Minerals also interact with fluoride and decrease intestinal absorption of fluoride and protect human beings from its harmful effects to some extent [42–43].

4. Conclusions

The study confirms that tea, toothpaste, tobacco and pan masala are rich sources of fluoride besides drinking water and food. Thus, these items should not be overlooked during the estimation of total dietary intake of fluoride. It also shows that extraction of fluoride from these items can increase several folds in acidic conditions as in human stomach. The people who are residing in the fluoride-affected areas are at a greater risk due to excessive fluoride intake especially when they are unaware of the amount of fluoride being ingested due to lack of awareness.

References

- A.A. Zahvoronkov, L.S. Strochkova, Fluorosis geographical pathology and some experimental findings, Fluoride 14 (1981) 182–191.
- [2] R.D. Kaul, A.K. Susheela, Evidence of muscle fiber degeneration in rabbits treated with sodium fluoride, Fluoride 7 (1974) 177–233.
- [3] N.J. Chinoy, E. Sequeira, Reversible fluoride induced fertility impairment in male mice, Fluoride 5 (1992) 71–76.
- [4] F.A. Smith, Handbook of Hazardous Materials, Academic Press Incorporation, New York, 1993, p. 277.
- [5] V. Singh, M.K. Gupta, P. Rajwanshi, S. Srivastava, S. Dass, Indian J. Environ. Health 35 (1993) 215–220.
- [6] APHA-AWWA-WPCF, Standard Methods for the Examination of Water and Wastewater, 15th ed., American Public Health Association, Washington, DC, 1994.
- [7] M.A.R. Buzalaf, B.S.D. Almeida, V.E.D.S. Cardoso, K.P.K. Olympio, T.D.A. Furlani, Total and acid-soluble fluoride content of infant cereals, beverages and biscuits from Brazil, Food Addit. Toxicol. 21 (2004) 210–215.
- [8] W. Slooff, H.C. Eevens, J.A. James, J.R.M. Rose, Integrated Criteria Document Fluoride, National Institute of Public Health and Environment Pollution, Bilthover, The Netherlands, 1989.
- [9] N. Sukusu-art, N. Arkasuwan, Survey on oral health status of primary school children in urban and rural areas, Hat Yai, Songkhla, Research/Government Report, Thailand, 2000.
- [10] B.L. O'Dell, Bioavailability of trace elements, Nutr. Rev. 42 (1984) 301–308.
- [11] J. Ekstrand, M. Ehrnebo, L. Boreus, Fluoride bioavailability after intravenous and oral administration: importance of renal clearance and urine flow, Clin. Pharmacol. Ther. 23 (1978) 329–337.
- [12] C.J. Spak, J. Ekstrand, D. Zylberstein, Bioavailability of fluoride added to baby formula and milk, Caries Res. 16 (1982) 249–256.
- [13] E.R. Shulman, M. Vallejo, Effect of gastric contents on the bioavailability of fluoride in humans, Pediatric Density 12 (1990) 237–240.
- [14] A. Goyal, K. Gaupa, A. Tewari, Bioavailability of fluoride in human from commonly consumed diets in India, J. Indian Soc. Pedodontic Prev. Dent. 16 (1998) 1–6.
- [15] K. Trautner, G. Siebert, An experimental study of bio-availability of fluoride from dietary sources in man, Adv. Oral Biol. 31 (1986) 223– 228.
- [16] F.L. Cerklewski, Fluoride bioavailability—research nutritional and clinical aspects, Nutr. Res. 17 (1987) 907–929.
- [17] F.L. Cerklewski, J.W. Ridlington, Influence of type and level of dietary calcium on fluoride bioavailability in the rat, Nutr. Res. 7 (1987) 1073–1083.
- [18] M. Koskinen-Kainulainen, H. Luoma, Excretion, serum, bone and kidney level of F in rats after a high single dose of fluoride and Mg + F, Magensium 6 (1987) 212–219.
- [19] M. Koskinen-Kainulainen, H. Luoma, J. Tuomisto, The LD-50 excretion serum and bone level of F after a high single F and F + Mg dose in rats with findings on cardiac Ca and Mg, Magnes. Trace Elem. 9 (1990) 15–27.
- [20] J. Ekstrand, M. Ehrnebo, Influence of milk products on fluoride bioavailability in man, Eur. J. Clin. Pharmacol. 16 (1979) 211–215.

- [21] J. Ekstrand, Relationship between fluoride in the drinking water and the plasma fluoride concentration in man, Caries Res. 12 (1978) 123–127.
- [22] G.M. Whitford, D.H. Pashley, G.I. Stringer, Fluoride renal clearance: a pH-dependent event, Am. J. Physiol. 230 (1976) 527–532.
- [23] World Health Organization, Fluorides and oral health, Report of WHO Expert Committee on oral health status and fluoride use, WHO, Geneva, 1994.
- [24] M. Haftenberger, G. Viergutz, V. Neumeister, G. Hetzer, Total fluoride intake and urinary excretion in German children aged 3–6 years, Caries Res. 35 (2001) 451–457.
- [25] A. Villa, M. Anabalon, L. Cabezas, The fractional urinary fluoride excretion in young children under stable fluoride intake conditions, Community Dentistry Oral Epidemiol. 28 (2000) 344–355.
- [26] F.V. Zohouri, A.J. Rugg-Gunn, Total fluoride intake and urinary excretion in 4-year old Iranian children residing in low-fluoride areas, Br. J. Nutr. 83 (2000) 15–25.
- [27] J. Ekstrand, Fluoride metabolism, in: O. Fejerskov, J. Ekstrand, B.A. Burt (Eds.), Fluoride in Dentistry, Munksgaard, Copenhagen, 1996, pp. 55– 68.
- [28] H. Spencer, I. Lewin, E. Wistrowski, J. Samachson, Fluoride metabolism in man, Am. J. Med. 49 (1970) 807–813.
- [29] R. Maheshwari, J. King, A.J. Brunetti, H.C. Hodge, E. Newburn, S. Margen, Fluoride balances in pregnant women, J. Occup. Med. (1981) 465–468.
- [30] FAO/IAEA/WHO, Trace Elements in Human Nutrition and Health A Joint FAO/IAEA/WHO Expert Consultation, World Health Organization, Geneva, 1996.
- [31] V.P. Jalili, K. Bobra, The fractional urinary fluoride excretion in young children under stable fluoride intake conditions, Community Dentistry Oral Epidemiol. 28 (2000) 344–355.
- [32] O. Fejerskov, A. Richards, P.K. Denbesten, The effect of fluoride on tooth mineralization, in: O. Fejerskov, J. Ekstrand, B.A. Burt (Eds.), Fluoride in Dentistry, Mungsgaard, Copenhagen, 1996, p. 112.
- [33] Standing Committee of the Scientific Evaluation of Dietary Reference Intakes; Food and Nutrition Board & Institute of Medicine, Dietary Reference Intakes for Calcium Phosphorus, Magnesium, Vitamin D and Fluoride, National Academic Press, Wasington, DC, 1997.
- [34] A. Saraux, D. Boullin, P. Jeandel, L. Abdoulaye, P. Le Goff, Endemc skeletal fluorosis, Revue du Rhumatisme 61 (1994) 753–757.
- [35] R.T. Haimanot, A. Fekadu, B. Bushra, Endemic fluorosis in Ethiopian Rift Valley, Trop. Geogr. Med. 39 (1987) 209–217.
- [36] P.E. McGill, Endemic fluorosis, Baillieres Clin. Rheumatol. 9 (1995) 75–81.
- [37] A.H. Sddiqui, Skeletal fluorosis in Nalgonda district, AP, India, Fluoride 1 (1968) 76–85.
- [38] W. Domazalska, Incidence of periodontal diseases in subjects with various degree of exposure to fluorides, Czas Somatol 25 (1972) 1005.
- [39] K. Takahashi, K. Akiniwa, K. Narita, Regression analysis of cancer incidence rates and water fluoride in the U.S.A. based on IACR/IARC (WHO) data (1978–1992), J. Epidemiol. 11 (2001) 170–179.
- [40] J.L. Shupe, H.B. Petrson, N.C. Leone, G.M. Whitford (Eds.), Fluoride: Effects on Vegetation, Animals and Humans, Paragon Press, 1983, p. 167.
- [41] F.L. Cerklewski, Influence of dietary magnesium on fluoride bioavailability in the rat, J. Nutr. 117 (1987) 496–500.
- [42] L. Levander, Cheng (Eds.), Micronutrient Interaction: Vitamins, Minerals and Hazardous Elements, Ann. N.Y. Acad. Sci., New York, 1980, p. 181.
- [43] M.K. Malde, Dietary fluoride sources in areas with endemic fluorosis, PhD Thesis, University of Bergen, Norway, 2002.