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Site specific toxicological risk from fluoride exposure through ingestion of vegetables and cereal crops in Unnao district, Uttar Pradesh, India

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ABSTRACT

A study was carried out to assess toxicological risk from the fluoride (F) exposure due to ingestion of vegetables and cereal crops such as rice and wheat grown in potentially fluoridated area (brick kiln and sodic areas), of different age groups in Unnao district, Uttar Pradesh, India. Fluoride contents in vegetables and cereal were found to be in the order brick kiln sites > sodic sites > normal sites. Among vegetables maximum F concentration was found in spinach and mint, whereas in cereal crops, wheat accumulated more F than rice. The exposure dose of F was determined using estimated daily intake (EDI) and bio-concentration factor (BCF) of F. The children of age group 3–14 years in the potentially fluoridated area were found to be at the risk of fluorosis. The mean BCF value of F was the highest in mint (36.6 mg/kg_{dwt} plant/mg/kg_{dwt} soil), followed by spinach (33.99 mg/kg_{dwt} plant.mg/kg_{dwt} soil).
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1. Introduction

Fluoride has both beneficial and detrimental effects on human health. When it is consumed in inadequate quantities (less than 0.5 ppm), it causes health problems such as dental caries and lack of formation of dental enamels but when consumed in excess (more than 1 ppm) it causes fluorosis (WHO, 1996). Worldwide around 355 million people are receiving artificially fluoridated water. In addition, around 50 million people receive water naturally fluoridated at a concentration of around 1 ppm (WHO, 2004). The level of natural fluoride that occurs in ground water ranges from 0.5 to 48 ppm or more (Susheela, 2003) and has become a worldwide problem. Human beings are exposed to fluoride (F) not only through drinking water but also through foods and dentifrices. The excessive ingestion of this ion leads to dental and skeletal deformities in the body, popularly known as fluorosis. The disease 'fluorosis' has now become a global problem and the health impairment due to fluorosis has occurred in the citizens of about 25 nations across the globe, and more than 200 million people worldwide are at the risk of fluorosis (UNICEF, 1999). It was thought previously that the principal source of F that causes fluorosis in human beings is the sources of drinking water. However, some food materials also contribute considerable amount to the total intake of F (Singer and Ophauge, 1979; Singh et al., 1993). Several factors influence the level of fluorides in food. These include the sites in which the food is grown and whether there were sources of fluoride emissions in the area, the amount of fertilizer and pesticides applied and whether fluoridated water is used in food preparation (McClure, 1949; Myers, 1978). Fruits and vegetables grown in industrial areas where fluoride emissions are high contain elevated fluoride levels. Jha et al. (2008) reported the accumulation of air borne F in the vegetations grown in the vicinity of brick fields. Vegetables grown 1.5 and 5 km from a steel plant contained average fluoride levels of 0.54–8.82 and 0.39–4.95 mg/kg, respectively, compared with 0.02–0.41 mg/kg for controls (Krelowska-Kulas, 1994). The Unnao district of Uttar Pradesh has been reported widely for the occurrence of F in the ground water (Agarwal and Choudhary, 2001; Rai, 1997; Kumar et al., 2004), which is used for drinking as well as irrigating the vegetables and cereal crops. The exposure doses of F in different sources of water in shallow aquifers of Makur in Unnao district, Uttar Pradesh, were calculated for infants, children and adults and were found to be high against the standard value of 0.05 mg/kg d, the minimum risk level (MRL) calculated by the Agency for Toxic Substances and Disease Registry (ATSDR; Jha et al., 2009). It has been reported that the solubility of F varies with the soil type as well as pH and has the tendency to be higher in the alkaline pH range (Larsen and Widdowson, 1971; Gilpin and Johnson, 1980), leading to higher bio-availability. The importance of pH of soil in solubility of fluoride is shown by the fact that the percentage of

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soluble fluoride in normal soil, saline soil and alkaline soil are 0.05%, 0.78% and 1.24% of the total fluoride, respectively (Lavado and Reinaudi, 1979).

Researchers in the past have measured daily intake rates of fluoride from various sources such as diet, toothpaste and infant formula (Fomon et al., 2000; Levy et al., 2001; Pendrys and Stamm, 1990). Since food is also a major source of F intake in human beings, there is a need for site specific risk assessment (SSRA) of F as a part of exposure evaluation from vegetable intake particularly in the potential fluoride contaminated area.

In this study, an attempt was made to evaluate (i) the site specific risk assessment (SSRA) due to F ingestion through vegetables and cereal crops grown in the vicinity of brick kilns, sodic area (the area where surface soil has pH >8.5 and sodium adsorption ratio (SAR) >15) and in non-fluoridated area (referred to as normal area) by examining bio-concentration factor (BCF) magnitudes for F from soil to vegetation and (ii) the risk to fluorosis in different age groups, using quantitative health risk assessment method.

2. Material and methods

2.1. Study area

The study area selected lies between 26°18′–27°02′N latitude and 80°27′–81°03′E, and falls in Hasanganj and Purwa tehsils of Unnao district in Uttar Pradesh, India. Two potentially fluoride contaminated sites such as brick kiln area and fluoride rich sodic area and one normal area (F uncontaminated) were selected for the present study indicated in Fig. 1. The potentially fluoridated areas were classified on the basis of higher F in ground water used for irrigation and drinking purpose and fluoride contamination due to atmospheric dust in brick kiln sites and possible higher soluble F in the soils with high pH. The soil samples were taken from those fields from where the vegetable and cereal crop samples were collected. These sampling sites were within the periphery of 5 km from the brick kilns where the vegetables were grown by the farmers either in their kitchen garden or in the fields.

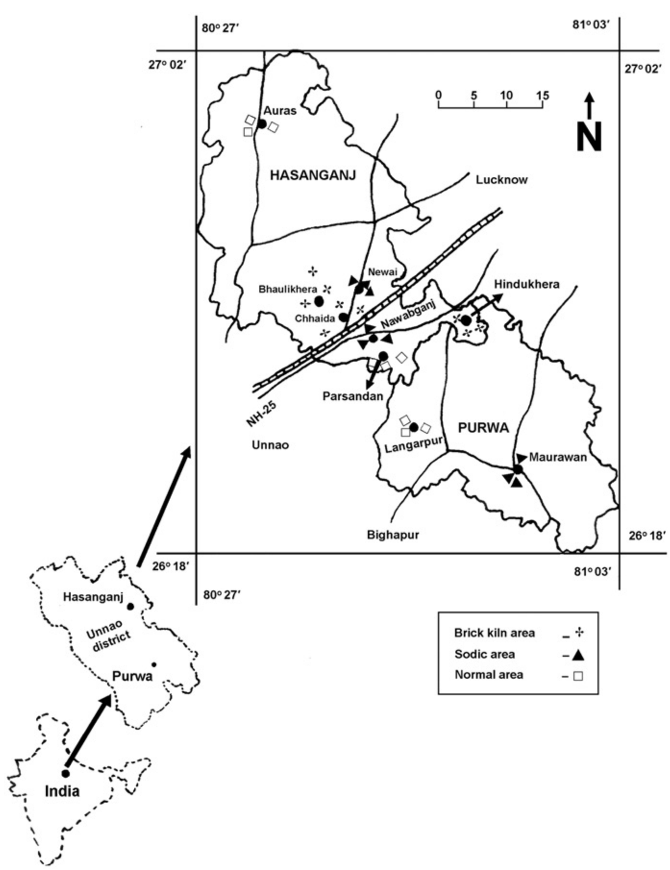

Fig. 1. Location map of the study area showing sampling points.

2.2. Collection and processing of the samples

Common vegetables such as spinach (*Spinacea oleracea*), ridge gourd (*Luffa acutangula*), lady's finger (*Abelmoschus esculentus*), mint (*Mentha arvensis*), tomato (*Lycopersicon esculentum*), pumpkin (*Cucurbita moschata*), bottle gourd (*Lagenaria siceraria*), cauliflower (*Brassica oleracea* var. *Botrytis*), cabbage (*B. oleracea* var. *Capitata*), carrot (*Daucus carota*) and the major cereal crops such as rice (*Oryza sativa*) and wheat (*Triticum aestivum* L.) are commonly grown in the study area (brick kiln area, sodic area and non-fluoridated normal area). The edible parts, i.e. leaves of spinach and mint, cabbage, roots of carrot, fruit of bottle guard, curd of cauliflower, pumpkin, ridge gourd, tomato, lady's fingers and samples of rice and wheat were collected in triplicate from the farmers' fields during their respective growing seasons in the year 2007–2008 from potentially fluoridated areas such as brick kiln sites, sodic soils and from non-fluoridated areas. The sampling locations within these sites were selected where maximum numbers of vegetables are grown. The number of samples of each vegetable at each site was not the same but varied generally between 2 and 4 numbers. of samples.

The samples were put inside polythene bags and transported to the laboratory for processing and analysis immediately. A part of the sample from each vegetable was kept for the determination of moisture percent, whereas the rest of the samples were firstly chopped into pieces, air dried for 2 d and then oven dried at 70° C. The dried samples were then milled to pass through 0.2 mm sieve and kept for F determination.

Three soil samples each from the respective vegetables and cereal crop fields were collected, mixed thoroughly and composite samples were prepared and collected in a polythene bag and then transported to laboratory for further processing and analysis .

2.3. Nutritional survey

A general nutritional survey was carried out. Five households were selected from each village in the designated sites (brick kiln sites, sodic soil sites and normal area) and the food habits, frequency of eating common vegetables and major cereal crops such as rice and wheat in different age groups were assessed. The population was grouped into three categories according to their ages. The three age groups were children (3–14 years), children (15–18 years) and adults (19–70 years) with their average body weights taken as 25, 50 and 70 kg, respectively. A detailed dietary habit, frequency of eating common vegetables and cereal crops such as rice and wheat were recorded. The intake frequency indicated was just an assumption near the mean values. However, these figures may change with the change in socio-economic conditions of the villagers. The survey was carried out by following the standard guidelines as prepared by National Institute of Nutrition (NIH), Hyderabad, India (Thimmayamma and Rau, 1987). The survey was just based on the questionnaire and is not a bio-medical research in which human subjects are administered some medicine, injection or chemicals to study toxicity symptoms. This study satisfied all the criteria of ethical treatment of human subjects and kept the identity of human subject intact.

2.4. Moisture percent determination in vegetables

Fresh vegetables of about 40 g were chopped into pieces, air dried for 2 d and then kept in a hot air oven at 70° C for 3 d till a constant weight was attained. The moisture percent was calculated using the following formula:

$$\text{Moisture } \% = \frac{(W1-W2)}{W1} \times 100$$

where $W1$ is the fresh weight of the vegetable and $W2$ is the weight of the vegetable after drying at 70° C. The values of the moisture content of vegetables were required for the determination of average consumption of vegetables on dry weight basis for each age group.

2.5. Total fluoride determination in vegetables and cereal crops

The total F in the vegetables, rice and wheat was determined by extracting the dried ground and sieved samples with 0.1 N perchloric acid and measuring F concentration with the help of ORION 5-star series ion analyzer, using ion selective electrode (ISE; Villa, 1979). The average recoveries based on the spiked samples at two different levels of F were 94 ± 6–99 ± 6%.

2.6. Exposure dose determination

The quantitative health risk assessment was measured by assessing the exposure doses of F due to the consumption of various vegetables and cereal

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