

# Exposure to Fluoride in Smelter Workers in a Primary Aluminum Industry in India

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## Abstract

**Background:** Fluoride is used increasingly in a variety of industries in India. Emission of fluoride dust and fumes from the smelters of primary aluminum producing industries is dissipated in the work environment and poses occupational health hazards.

**Objective:** To study the prevalence of health complaints and its association with fluoride level in body fluids of smelter workers in a primary aluminum producing industry.

**Methods:** In an aluminum industry, health status of 462 smelter workers, 60 supervisors working in the smelter unit, 62 non-smelter workers (control group 1) and 30 administration staff (control group 2) were assessed between 2007 and 2009. Their health complaints were recorded and categorized into 4 groups: 1) gastro-intestinal complaints; 2) non-skeletal manifestations; 3) skeletal symptoms; and (4) respiratory problems. Fluoride level in body fluids, nails, and drinking water was tested by an ion selective electrode; hemoglobin level was tested using HemoCue.

**Results:** The total complaints reported by study groups were significantly higher than the control groups. Smelter workers had a significantly ( $p < 0.001$ ) higher urinary and serum fluoride level than non-smelter workers; the nail fluoride content was also higher in smelter workers than non-smelter workers ( $p < 0.001$ ). The smelter workers with higher hemoglobin level had a significantly ( $p < 0.001$ ) lower urinary fluoride concentration and complained less frequently of health problems. Only 1.4% of the smelter workers were consuming water with high fluoride concentrations. A high percentage of participants was using substances with high fluoride contents.

**Conclusions:** Industrial emission of fluoride is not the only important sources of fluoride exposure—consumption of substance with high levels of fluoride is another important route of entry of fluoride into the body. Measurement of hemoglobin provides a reliable indicator for monitoring the health status of employees at risk of fluorosis.

**Keywords:** Fluorides; Fluorosis, dental; Industry; Occupational diseases; Early diagnosis; Prevention and control

## Introduction

Fluorine, a powerful oxidizing agent, is used increasingly in a variety of industries in India. Due to rapid industrialization, health problems among

industrial workers due to fluoride poisoning are on the rise. Emission of fluoride dust and fumes from the smelters of primary aluminum producing industries is dissipated in the work environment and poses occupational health hazards.<sup>1-4</sup>

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Use of cryolite (tri-sodium hexafluoroaluminate,  $\text{Na}_3\text{AlF}_6$ ) as a flux in the conversion of alumina to aluminum, is the major source for fluoride emission responsible for the spread of industrial fluorosis among smelter workers. Waldbott showed emission levels of fluoride from various industries,<sup>5</sup> of which the second highest emission was from aluminum industries. Grandjean reported on skeletal fluorosis as an occupational disease among Danish cryolite workers.<sup>6</sup> Radiologically-confirmed changes in skeletal system of aluminum smelter workers have been established.<sup>7-9</sup> Higher than normal prevalence of asthma among pot-room workers in aluminum industries has been documented in literature.<sup>10-13</sup> Guo showed a relationship between damage to the central nervous system and occupational exposure to fluoride.<sup>14</sup> Grandjean, *et al*, provided evidence of neurotoxicity from industrial compounds,<sup>15,16</sup> through a meta-analysis, they also confirmed neurotoxicity from fluoride in children. These reports suggest that smelter workers are prone to a variety of health problems due to fluoride poisoning. Cancer in fluoride-

exposed workers has also been reported by Grandjean.<sup>17</sup> People may be exposed to fluoride by consuming drinking water, food, dental products and drugs. Fluorosis has no treatment—prevention is the only solution.

Monitoring health complaints and fluoride level in body fluids has introduced to prevent and control the occurrence of industrial fluorosis. India is a highly endemic country for water-borne and food-borne fluorosis,<sup>18-22</sup> therefore, adequate precautions have been established to develop an effective protocol for dealing with industrial fluorosis. Diets with high concentrations of fluoride for the use of condiments, especially black rock salt ( $\text{CaF}_2$ ), is a major concern. Black rock salt is used to enhance the aroma and tangy taste. In India, street foods, fruit juices, and snacks are heavily loaded with black rock salt. The salt resembles “magadi” used widely in African countries. Both salts have a volcanic origin.<sup>23,24</sup> The Indian protocol developed for combating the adverse health effects of industrial fluorosis is different from other procedures available. The procedures in use differ in the developing vs developed countries. Seixas reported that measurement of urinary fluoride can be used as an exposure index in aluminum smelter workers.<sup>25</sup> As early as 1976, Dinman demonstrated a relationship between urinary excretion of fluoride and bony fluorosis among aluminum smelter workers.<sup>26</sup> The practice in the developed countries is to test urinary fluoride level before and after shift. However, in our experience, it is inadequate and testing serum and nail fluoride, as well as urinary fluoride level, is necessary to evaluate the extent of damage caused by the accumulation of fluoride in the body over a period of time. Our protocol for assessing the health status of smelter workers has laid emphasis on a few additional yardsticks for the detection of the

For more information on fluoride concentration in water of a city in Iran and Nigeria see [www.theijoem.com/ijoem/index.php/ijoem/article/view/167](http://www.theijoem.com/ijoem/index.php/ijoem/article/view/167) and [www.theijoem.com/ijoem/index.php/ijoem/article/view/123](http://www.theijoem.com/ijoem/index.php/ijoem/article/view/123)

#### TAKE-HOME MESSAGE

- Due to rapid industrialization in India, health problems among industrial workers due to fluoride poisoning are on the rise. Emission of fluoride dust and fumes from the smelters of primary aluminum producing industries is an important cause of occupational exposure to fluoride.
- Fluorosis has no treatment—prevention is the only solution.
- In India, besides industrial exposure, consumption of diets with high concentrations of fluoride, especially black rock salt ( $\text{CaF}_2$ ), is an important cause of fluorosis.
- Measurement of hemoglobin provides a reliable indicator for monitoring the health status of employees at risk of fluorosis.

**Table 1:** Four categories of health complaints assessed

Category	Health complaints
1	Gastrointestinal complaints including i) nausea/loss of appetite, ii) gas formation, iii) pain in the stomach, iv) constipation, v) diarrhea (intermittent), and vi) headache
2	Non-skeletal health complaints including i) frequent tendency to urinate/itching in the region, ii) excessive thirst, iii) extreme weakness/muscle weakness viz. unable to walk short distances, iv) anemia due to low Hb/paleness, v) allergic reactions (intermittent perivascular inflammation)
3	Complaints in skeletal system and pain in major joints viz. i) neck, ii) back, iii) hip, iv) knee, and v) shoulder
4	Complaints in respiratory system including i) asthma, ii) bronchitis with violent cough, iii) nasal irritation, and iv) chest congestion with difficulty in breathing

disease at an early stage. The protocol is management-friendly with focus on the well-being of the workers and their families.<sup>27</sup>

We conducted this study to determine the prevalence of health complaints and its association with fluoride level in body fluids of smelter workers of a primary aluminum producing industry.

### Materials and Methods

HINDALCO Industries Ltd, one of the largest primary aluminum producing industries in India located in the north-eastern part of the state of Uttar Pradesh, was chosen for the study. Smelter workers (n=462) and supervisors/officers (n=60) working in the smelter units were randomly selected as study groups 1 and 2 for health assessment. Control groups 1 and 2 were comprised of non-smelter workers (n=62) and administrative staff (n=30) working in the same industry but away from the smelter unit, respectively. Smelter workers and supervisors were chosen for the study, as the socioeconomic and educational status of the two groups were different, though they worked in the same work environment; the two groups were also different in terms of nutritional

status.

Prior to the commencement of the study, we explained the objectives for all senior officers and smelter workers. Those who agreed to participate in the study agreed to provide samples of blood, urine, nail and drinking water for testing for fluoride.

All participants were asked about baseline information on health complaints and tested for hemoglobin concentration and fluoride level in serum, urine, nails and drinking water samples.

### Epidemiological survey

Background information of the smelter workers, supervisors and controls was collected and recorded in a data collection sheet. Health complaints for ascertaining fluoride toxicity manifestations were sought through interview. Health complaints were recorded and categorized under four categories (Table 1). For each set of health complaints, those who experienced the symptoms more frequently were further assessed. Category 3 included five major complaints focusing on pain/rigidity in major joints. Three physical tests were carried out to assess pain in the major joints (Fig 1).<sup>28</sup> The answer was considered 'yes' if the participant could

**Normal Individuals**

**Those individuals with joint pain**

1. Normal, healthy individual can bend his body and touch the floor/toes



Unable to bend, without folding his knees



2. Normal, healthy individual can touch the chest with chin



Unable to bend the neck, touching the chest with chin not possible



3. Normal, healthy individual can stretch the hands, fold the arms and touch the back of the head



Unable to stretch the hands, fold the arms and touch the back of the head



**Figure 1:** Three physical tests used for assessing pain in major joints

not perform at least one of the tests. In the study of skeletal fluorosis studies, x-ray study is usually done; however, we did not use x-ray as there were a large number of participants and persistent joint pain was informative enough for our purpose.

Fluoride content in samples was estimated by a potentiometric method using ion selective electrode technology—the most sensitive method for the measurement of fluoride. Body fluids were collected on the day of interview and during the working hours.

Untreated ground water samples, if used for cooking and drinking, were obtained from the participants' native village and their residential area (if residing off campus) and collected in plastic bottles. Fluoride content was estimated

by the standard method using an ion meter (Model No. 4 Star, Thermo-Fisher, USA).<sup>29</sup>

Spot urine samples were collected between 10:00 and 13:30. Fluoride content was estimated using an ion meter (Model No. EA 940, Thermo-Orion, USA).<sup>30</sup> Serum samples were prepared from the blood drawn. Fluoride content was estimated using an ion meter (Model No. EA 940, Thermo-Orion, USA).<sup>31</sup> Nail clippings from both toe and finger were collected, washed using diethyl ether-acetone mixture (1:1 v/v), and dried. Dried nail samples were burned to ashes using a muffle furnace set at 550 °C for 2 hrs; the ash was dissolved in concentrated HCl and neutralized by NaOH. Fluoride content was estimated using an ion me-

**Table 2:** The background information about the participants. Data are presented as mean±SD.

Parameter	Smelter workers (n=462)	Non-smelter workers (n=62)	Supervisors working in the smelter unit (n=60)	Administrative staff (n=30)
Age (yrs)	34.5±7.9	44.8±8.1	38.8±6.9	43.7±7.2
Body weight (kg)	64.4±8.2	63.1±9.6	71.5±12.5	68.8±7.7
Length of service (yrs)	11.1±7.5	20.1±9.0	13.8±7.8	19.4±8.5

ter (Model No. 4 Star, Thermo-Fisher, USA).<sup>32,33</sup> Hemoglobin (Hb) level was measured by a portable battery-operated hemoglobinometer (Hemocue Hb 201+, Angelholm, Sweden) by a method described earlier.<sup>34,35</sup>

### Dietary status

The participants were interviewed for a verbal answer ('yes' or 'no') to record if they could possibly receive fluoride through sources other than emission from smelter in the industry. This was achieved by asking whether they chew tobacco, arecanut, ayurvedic digestive tablets; or eating black rock salt laced snacks, street foods, or pickles. Consumption of ready-made spices containing black rock salt, consumption of black tea (without milk), or use of dental products (e.g., fluoridated toothpaste, mouth rinse) were also studied.

### Statistical analysis

Data were analyzed by Stata ver 11.1. Normally distributed data are presented as mean±SD; non-normally distributed data are presented as median (interquartile range). The mean of two groups was compared by Wilcoxon rank sum or *Student's t* test for independent samples depending on the distribution of the data. A p value <0.05 was considered statistically significant.

### Results

The background information of participants is shown in Table 2. More than 70% of participants—347 (75%) of 462 studied smelter workers, 44 (71%) of 62 non-smelter workers, 46 (77%) of 60 studied supervisors, and 21 (70%) of 30 administrative staff studied—were from the state of Uttar Pradesh; the remaining participants were from other states of India.

The frequency of health complaints reported by the participants is shown in Table 3. The prevalence of gastrointestinal, non-skeletal, and skeletal (category 1, 2 and 3) complaints in smelter workers (group 1) was significantly ( $p < 0.001$ ) higher than non-smelter workers (control group 1); the prevalence of respiratory (category 4) complaints was not significantly different. Supervisors working in smelter unit (group 2) complained more frequently from non-skeletal (category 2) complaints compared to administrative staff (control group 2) ( $p < 0.05$ ); the two groups were not different in terms other complaints. The total complaints reported by study groups were significantly higher than the control groups (Table 3).

Table 4 shows fluoride concentration in body fluids and nail samples taken from participants. Smelter workers had a significantly ( $p < 0.001$ ) higher urinary and serum fluoride level than non-smelt-

**Table 3:** Health complaints in participants. For categories see Table 1.

Group	Median (IQR*) number of complaints in each category				
	1 (6 complaints)	2 (5 complaints)	3 (5 complaints)	4 (4 complaints)	Total (20 complaints)
Smelter workers (n=462)	2 (1)	2 (1)	1 (1)	0 (0)	4 (3)
Non-smelter workers (n=62)	1 (1)	0 (1)	0 (1)	0 (0)	1 (3)
p value	<0.001	<0.001	<0.001	NS <sup>†</sup>	<0.001
Supervisors working in the smelter unit (n=60)	1 (1.5)	1 (2)	1 (1)	0 (0)	3 (3)
Administrative staff (n=30)	1 (2)	0 (1)	0 (1)	0 (0)	2 (3)
p value	NS	0.0135	NS	NS	<0.005

\*IQR: Interquartile range; <sup>†</sup>NS: Not significant

er workers; the nail fluoride content was also higher in smelter workers than non-smelter workers ( $p<0.001$ ). The urinary and serum fluoride levels were also significantly higher in supervisors working in smelter unit than administrative staffs (Table 4); the nail fluoride content of the supervisors was also significantly higher than that of the administrative staff ( $p<0.001$ ).

The prevalence of health complaints and urinary fluoride level of smelter workers and supervisors working in the smelter unit, stratified by Hb level, is presented in Table 5. The smelter workers with higher Hb level had a significantly ( $p<0.001$ ) lower urinary fluoride concentration and complained less frequently of health problems (Table 5). Supervisors working in the smelter unit who had high-

**Table 4:** Fluoride levels in samples taken from the studied participants. Data are presented as median (IQR\*).

Group	Urine (mg/L)	Serum (mg/L)	Nail ashes (mg/kg)
Smelter workers (n=462)	5.09 (5.97)	0.14 (0.10)	1.49 (2.16)
Non-smelter workers (n=62)	1.14 (1.49)	0.05 (0.05)	0.13 (0.20)
p value	<0.001	<0.001	<0.001
Supervisors working in the smelter unit (n=60)	2.18 (3.10)	0.08 (0.04)	0.63 (0.93)
Administrative staff (n=30)	0.78 (0.74)	0.06 (0.04)	0.14 (0.17)
p value	<0.001	<0.01	<0.001

\*IQR: Interquartile range

**Table 5:** Frequency of health complaints and urinary fluoride level in smelter workers and supervisors working in the smelter unit stratified by Hb level. Values are median (IQR\*).

Group	Hb level (g/dL)	Urinary fluoride concentration (mg/L)	Number of health complaints
Smelter workers	>13.0 (n=360)	4.95 (5.67)	4 (3)
	≤13.0 (n=102)	6.64 (6.75)	6 (3)
p value		0.015	<0.001
Supervisors working in the smelter unit	>13.0 (n=47)	2.08 (3.06)	2 (4)
	≤13.0 (n=13)	2.77 (5.98)	3 (3)
p value		NS <sup>†</sup>	NS

\*IQR: Interquartile range; <sup>†</sup>NS: Not significant

er Hb level had no significant differences with those who had lower Hb in terms of urinary fluoride and prevalence of the complaints.

Only 1.4% of the smelter workers were consuming water with fluoride concentrations >1.0 mg/L—the remaining 98.6% smelter workers were consuming safe water with fluoride level <1.0 mg/L, supplied by the industry. All the studied supervisors, non-smelter workers, and administrative staff were consuming safe drinking water (Table 6). Other sources of fluoride might also be important. A high percentage of participants was using substances with high fluoride contents (Table 6).

## Discussion

The frequency of studied health complaints was significantly ( $p < 0.001$ ) higher in the study groups (smelter workers and supervisors working in the smelter unit) compared with the control groups (non-smelter workers and administrative staff). Non-skeletal manifestations are the earliest signs of fluoride poisoning; skeletal manifestations surface years after exposure to fluoride. Inhalation or ingestion of fluoride would cause—within

a few weeks—gastrointestinal discomfort, polyuria, polydipsia, muscle weakness, and anemia.<sup>36,37</sup> Fluoride would also damage the gastrointestinal mucosa resulting in loss of microvilli in intestinal lining with resultant non-ulcer dyspepsia and irritable bowel syndrome. Fluoride can inhibit production of antidiuretic hormone and may cause diabetes insipidus.<sup>38</sup> The above-mentioned health complaints would disappear within a short period of 7–10 days, after cessation of fluoride exposure. The non-ulcer dyspepsia and irritable bowel syndrome need no drug therapy; withdrawal of fluoride ingestion would alleviate the signs and symptoms.

Excess intake of fluoride would cause derangement of muscle structure resulting in loss of muscle power.<sup>39,40</sup> Fluoride also reduces erythropoiesis. Alternatively, due to loss of calcium from the erythrocyte membrane, fluoride may cause echinocytosis.<sup>41,42</sup> Allergic reactions presenting as painful skin rashes due to perivascular inflammation, was also reported among the smelter workers and supervisors working in a smelter unit.<sup>43</sup>

Therefore, it seems that manifestations caused by derangement in soft tissue and organs come first during the course of

**Table 6:** The frequency (%) of participants using other common sources with high fluoride contents

Source	Smelter workers (n=462)	Non-smelter workers (n=62)	Supervisors working in the smelter unit (n=60)	Administrative staff (n=30)
Water with fluoride level >1.0 mg/L*	6 (1.4)	0 (0)	0 (0)	0 (0)
Chewing tobacco	324 (70.1)	43 (69)	22 (37)	6 (20)
Chewing arecanut (Supari)	231 (50.0)	23 (37)	19 (32)	5 (17)
Chewing ayurvedic digestive tablets (Hajmola/Churan) containing black rock salt	94 (20.3)	10 (16)	10 (17)	5 (17)
Consuming snacks/street foods/pickles laced with black rock salt	228 (49.4)	17 (27)	30 (50)	4 (13)
Using black rock salt in cooking (Kalanamak)	74 (16.0)	5 (8)	15 (25)	6 (20)
Using ready-made spices added with black rock salt in cooking	73 (15.8)	3 (5)	13 (22)	9 (30)
Drinking black tea (without milk)	18 (3.9)	5 (8)	3 (5)	3 (10)
Using fluoridated toothpaste (contains >1000 ppm of fluoride)	259 (56.1)	36 (58)	41 (68)	19 (63)

\*According to the Bureau of Indian Standards for fluoride level in drinking water, a concentration of 1.0 mg/L is the upper acceptable limit.

fluoride poisoning; later on, bones get affected. This would explain our observation that the first three studied categories of health complaints were significantly ( $p < 0.001$ ) more frequent among smelter workers than non-smelter workers; the supervisors and the administrative staff were only different in terms of prevalence of category 2 health complaints ( $p < 0.05$ ), *i.e.*, non-skeletal complaints which would attributed to soft tissue manifestations of excess exposure to fluoride.

Skeletal manifestations (category 3 complaints) are due to long-term exposure to excess fluoride causing pain and stiffness in major joints.<sup>7,8</sup> When excess

amount of fluoride is consumed, it tends to accumulate more in cancellous bone compared to cortical bone.<sup>44</sup> As the cancellous bone predominates in the region of joints, the higher fluoride accumulation leads to damage to the bone matrix with resultant complaints from joints. Joint pain would arise in an advanced stage of fluorosis leading to rigidity and stiffness of joints—when the reversal of the damages is not likely. This emphasizes the importance of the early diagnosis of fluoride poisoning.

Considering respiratory problems (category 4 complaints), although other investigators<sup>11-13</sup> have reported occupational

asthma as an important health problem among aluminum industry workers, we found no significant difference between the prevalence of respiratory problems in exposed and non-exposed participants. However, we did not measure the fluoride dust and fumes in the work environment and the smelting area. The studied employees were provided with masks to wear while at work; compliance with this provision could also help a great deal to reduce pot-room asthma.

The results of testing fluoride in body fluids and nail reported in this study have revealed a striking correlation with the prevalence of gastrointestinal and non-skeletal manifestations among the smelter workers and supervisors. Therefore, for the assessment of early manifestations of fluoride poisoning among exposed employees, besides measuring urinary fluoride level, measurement of serum fluoride level<sup>45,46</sup> and nail fluoride content<sup>47,48</sup> should also be considered. The studied control groups (non-smelter workers and administrative staff) also had a considerable level of fluoride in their body fluids and nail reflecting that industrial emission is not the only source for fluoride entry into the body and that it may enter the body through food, water and dental products. Ionic fluoride in serum and urine of non-exposed people has been reported previously.<sup>49</sup>

We found that assessing Hb would be an important test to be done in conjunction with fluoride tests to assess the overall health status of employees. High fluoride level in body leads to a drop in Hb, as fluoride interferes with Hb biosynthesis. Fluoride is also reported to interfere with thyroid hormones leading to diminished stimuli for erythropoiesis causing anemia.<sup>50</sup> Fluoride is also known to destroy probiotics responsible for production of vitamin B<sub>12</sub>, an essential constituent for Hb biosynthesis.<sup>51</sup> Fluoride is also re-

sponsible for loss of microvilli in the gastrointestinal mucosa leading to anemia due to malabsorption. Among the studied participants, when the urine had high level of fluoride, the Hb concentration was low and people were complaining more frequently of health problems (Table 5). According to WHO guidelines,<sup>52,53</sup> the lower limit of normal Hb concentration in adult men is 13.0 g/dL. To the best of our knowledge, this is the first observation reporting a negative correlation between urinary fluoride concentration and Hb level.

In this study, we showed the importance of collecting data on dietary habits, as fluoride can enter the body not only through occupational exposure but also by intake of food and water. The employees in India should be aware that their current food habits would result in health problems in them and their family members.<sup>19-22,54-56</sup> Condiments used for cooking are adding to fluoride burden of the body leading to ill-health. A number of substances with high fluoride content consumed<sup>57,58</sup> and the high prevalence of ill-health among the smelter workers and the supervisors clearly illustrates that besides fluoride emission from smelter units, consumption of certain substances would contribute to fluorosis.<sup>59</sup> It is also suggested to advise the employees and their family members to switch from fluoridated brands of toothpaste to less fluoride containing dental products.

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**Conflicts of Interest:** None declared.

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