

## Fluoride-induced impact of aluminium industrial power plant on plants and human inhabiting areas

Prem Chandra Pandey<sup>1</sup>, Pavan Kumar<sup>2</sup>, Meenu Rani<sup>3</sup>, Swati Katiyar<sup>2</sup>  
and Vandana Tomar<sup>4</sup>

<sup>1</sup> University of Leicester, Centre for Landscape and Climate Research,  
Department of Geography, Leicester, United Kingdom

<sup>2</sup> Banasthali University, Department of Remote Sensing, Newai, Rajasthan, India

<sup>3</sup> Project Directorate for Farming Systems Research, Modipuram, Meerut, UP, India

<sup>4</sup> Haryana Institute of Public Administration, Gurgaon, India

*Received 14 July 2014, in final form 14 November 2014*

Fluorine is a highly reactive common element that does not occur in nature in the elemental state. It exists in the form of fluorides and accounts for about 0.3 g/kg of the Earth's crust. Generally, it is found in the form of a number of minerals like fluorspar, cryolite and fluor-apatite. Fluoride has both positive and negative effects on individual health. Fluoride, in the form of fluorspar and cryolite is distributed extensively in the lithosphere, and is renowned as the thirteenth most common among elements in the earth's crust. Hydrogen fluorides in gaseous form accumulate in the leaves of generally sensitive plants against a concentration gradient and therefore, considered as a most phytotoxic air pollutant and affects plants at extremely low concentration. As per our study, it is found that the fluoride impacts on fauna are in normal condition, but in coming times it may have adverse impact on fauna and flora of surroundings of Hindalco Industries Limited.

*Keywords:* fluoride, human health, detrimental effects, social aspects

### 1. Introduction

Major part of fluoride in water, either naturally occurring or added, is found in the form of the free fluoride ion (IPCS, 2002). Water hardness in the range 0 to 500 mg CaCO<sub>3</sub> per litre has very little effect on ionic dissociation, and therefore, imparts negligible effect on the fluoride bioavailability (Jackson et al., 2002). In a standard dose of fluoride absorption will vary from 100% to 60% on a fasting stomach and when taken with a calcium-rich breakfast. Prior to the Grand Rapids intervention Moulton in 1942 examined the ill effects of ingested fluoride on health and thereafter, regularly analyzed by several organizations and individuals. More recently comprehensive assessment of fluoride and its potential

impacts on health was carried out by IPCS. The major thrust of most of the studies and reviews has been concentrated around bone fractures, cancers, skeletal fluorosis and birth defects, but at the same time, many other disorders argued to be caused by fluoridation were also studied well thought-out (McDonagh et al., 2000; Demos et al., 2001; Fottrell, 2001; Knox, 1985; Choubisa et al., 2010). There is a lack of proper evidences regarding any adverse medical effects coupled with the consumption of water with fluoride added at a concentration of 0.5–1.0 mg/l except enlarge in dental fluorosis described above. The recommended concentration of fluoride lies in the range of 0.5 mg–1.0 mg/l in different climatic conditions (WHO, 1994). Around 355 million people worldwide are receiving fluoridated water artificially. More to the point, approximately 50 million people obtain naturally fluoridated water with 1 mg/l concentration. In some developing countries, mainly some parts of India, Africa and China, drinking water can hold more than the WHO's recommended Guideline Value of 1.5 mg/l concentration. In nature, Fluorides found in many naturally occurring materials like as coal, clay, and minerals. These substances, when heated to high temperatures in aluminium smelters; glass, brick, tile works; and plastic factories, the possibility of fluoride release to the atmosphere are higher.

Workers involved in certain machinery, air transportation, medical and other health services, textile and metal manufacturing, or petroleum and coal production had a higher risk of being exposed to high levels of cryolite in the air. Labour in these types of task breathes in levels as high as 2.5 milligrams of fluoride per cubic meter ( $\text{mg}/\text{m}^3$ ) of the air. Some hazardous waste sites contain containers of hydrofluoric acid (Choubisa et al., 2010). Cleanup workers are at the risk of being exposed to hydrofluoric acid in case of material leakage from the containers. In the nearby region of hazardous waste sites, peoples are at less risk to be exposed to hydrofluoric acid as the acid form fluoride salts when react with soil and before it reaches the people. Fluoride is also contained in pressurized containers used at some hazardous waste sites.

The effects of fluoride on vegetation can be summarized as when gaseous fluoride is absorbed by the leaves, it dissolved in the aqueous phase, is transported acropetally and accumulates where the vascular system terminates. This accumulation of fluoride may lead to marginal necrosis that being at leaf tips and progress to leaf bases. Long term, low concentration exposure to HF result in chronic injury characterized by general chlorosis or chlorosis along leaf veins. Short term, high concentration exposures result in acute injury, characterized by tip and marginal necrosis that progress to leaf bases, or if rapid absorbed causes local concentration of fluoride in the leaf to exceed the toxic threshold, irregular patches of necrosis may occur in the intercostal areas.

## 2. Materials and method

The current survey was conducted on workers and residents of Hindalco Industries Limited (HINDALCO), a flagship company of the Aditya Birla Group.

It is the largest integrated Al plant in Asia, which took up the production in May 1962 with an annual capacity 20,000 tons per annum (TPA) of Al metal. It is presently an integrated Al manufacturing complex with a production capacity of 3, 56,000 TPA of primary metal. It is located at Renukoot, UP, India (refer to Fig. 1).

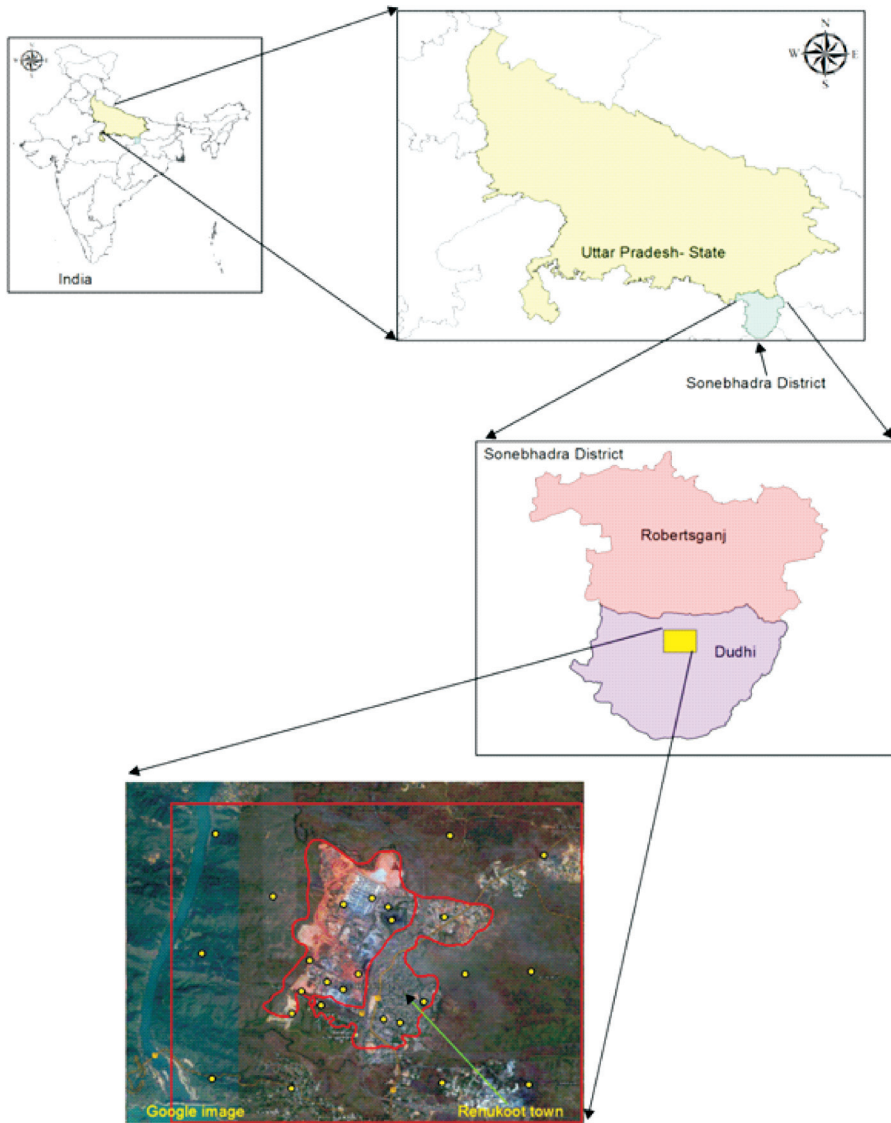


Figure 1. Location map of study area.



product is washed in crucible with 25 ml of distilled water and placed in a beaker. It is again washed in crucible with 35 ml of Perchloric acid ( $\text{HClO}_4$ ) and poured in the same beaker placed earlier. This washed solution was performed with distillation using steam distillation unit. Thereafter, approximately 250 ml of this distilled solution was collected in a beaker, and 5 ml of citrate buffer was mixed with this 250 ml solution to make it 250 ml by volume. Now, 10 ml of solution from the 250 ml volume was taken in a small beaker and mixed with TISAB (total ionic strength anionic buffer) for fluoride analysis. Thereafter, fluoride values were read from this solution (TISAB and 10 ml mixed solution) by using the expandable ion Analyzer.

$$f = \frac{R \times V}{W} \quad (1)$$

where:  $f$  = fluoride concentration,  $R$  = final reading of expandable ion analyzer (in mg/l),  $V$  = final volume (in ml), i.e. 250 ml and  $W$  = weight of the leaf sampled (2–4 mg).

### *2.2. Sampling and sample preparation for industrial workers*

Fluorosis can be diagnosed using estimation of fluoride level in serum, urine and bone. Among the widely used methods available for the determination of fluoride content, most common methods are calorimetric and fluoride specific ion electrode. The fluoride specific ion electrode is more popular because of its speed than other methods. Selective electrode method introduced by Frant and Ross (1966) offer an electrochemical response which is proportional to the fluoride ion activity in sample.

It must keep in mind that determination of fluoride level in the first step may hamper the overall accuracy or validity of the final data in a series of investigation. Errors incorporated during sampling as well as handling may be much higher in comparison to those arise because of lack in accuracy as well as reliability in analytical technique. The media of observation are many and varied; so it is wiser that each condition should be evaluated and a scheme devised accordingly for collection, preparation and analysis of samples for reliability. Usually fluoride selective electrode method is used for collection and subsequent analysis of Serum and Urine sample. Before the intervention of fluoride selective electrode method, generally body fluids (e.g., serum, saliva, blood and urine) were evaporated gently to dryness and dry ash was extracted before fluoride separation. Since, ash results in refractory fluorides frequently, the residue solubility is ensured by fusing with alkali carbonate or hydroxide. Subsequent to fusion, melt is dissolved and afterward fluoride is separated for evaluation.

### *2.3. Geospatial approach*

The study also includes the remote sensing approaches to assess the fluoride concentration. The points from where samples were collected are recorded in the

form of their spatial locations like latitude and longitude. This spatial information is recorded using the GPS (global positioning system) and then transferred into the system as a shape file. The shape file helps in the generation of the output maps representing the location points and study area (refer to the Fig. 2). The information from the point is used as the point file to interpolate the measured values using the spatial analyst of the Arc GIS to assess the fluoride concentration over the study area.

In the present study, fluoride selective electrode method was chosen due to its excellent speed, satisfactory performance and general convenience. The method has turned out to be a suitable method for fluoride estimation in a diverse environment and a variety of industrial samples. The electrode selectivity method is based upon membrane properties of sparingly soluble single crystals of lanthanum, praseodymium or neodymium fluoride. It generates an electro-chemical signal that is proportional to the fluoride ion activity in the sample. The major contribution of fluoride selective electrode is in determination of fluorides level in drinking-water, sea water, soils and minerals, plants, industrial effluents, air, aerosols, flue gases, urine, serum, plasma, and other biological materials. For automated monitoring of fluoride level, instruments are available in fluoride selective electrode method. In case of electrode method, the precision and accuracy even equal or exceed when compared with colorimetric techniques in most of the samples.

### 3. Results

The results found with the sample collection from several levels were studied and analyzed for the fluoride concentration. This study also interpolated the fluoride concentration using geospatial approaches for the possible association of its concentration over a region not accessible or not sampled.

#### 3.1. *Effect of fluoride on flora*

Plants are generally gets exposed to air and soil and therefore gets fluoride through from the soil by means of passive diffusion which is further reaching the shoot through transpiration. Some species of plant accumulate high concentrations of fluoride, even grown up on soil having low fluoride level possibly due to complex formation with aluminium. Fluoride in gaseous form enters into the leaves through pores in stomata. Fluoride may affect metabolic process when enters the internal tissue of leaves or when accumulate on active surfaces of the stigmata which may hamper the chlorophyll contents, growth or reproduction.

The effect of toxic fluoride on plants is visible in the form of chlorosis, peripheral necrosis, leaf distortion and malformation, and abnormal fruit development. Fluoride uptake in the plants, mainly occurs through the root and leaves from

Table 1. Fluoride concentration in different plant.

Location	Botanical name	Fluoride (ppm)
Below Vishwakarma Statue	Cynadon dactylon	35.6
Potline # III	Ficus religiosa	25.0
Potline # III	Cynadon dactylon	43.6
Potline # II	Mangifera indica	33.4
G-6, Hindalco Colony	Azadiracha indica	18.6
Near Guest House (Rihand dam)	Mangifera indica	18.6
Potline # IV	Ficus religiosa	44.1
Plant # 2 Colony	Cynadon dactylon	28.4
Near STP	Solanum tuberosum	19.7
Near WTP	Mangifera indica	21.0
Near STP	Zizyphus mauritiana	17.4
Near Potline # IV	Ficus religiosa	23.6
Near Banvashi ashram	Azadirachta indica	34.1
Near village Katauli	Azadirachta indica	31.7
Near village Murdhawa	Lantana camra	10.7
Near village Pipri	Bauhinia variegata	28.7
Near Hanuman Temple	Mangifera indica	33.4
Near Guest House	Mangifera indica	9.7
Near Guest House	Cynodon dactylon	16.4
River Side	Cynodon dactylon	8.4
Near village Turra	Ficus religiosa	19.7
Near village Turra	Madhuca longifolia	31.0
River Side	Zizyphus mauritiana	28.7
Near village Muirpur	Tamarindus indica	18.2
Near village Muirpur	Zea mays	23.4
Near village Katauli	Triticum aestivum	17.4
Sagaria Nallah	Ficus religiosa	17.2
Plant II Colony	Anthocephalus cadamba	34.0
Railway Colony	Mangifera indica	15.3
Tharpathar Village	Bauhinia variegata	16.0

the soil and air, respectively. The harmful effect of Fluoride occurs in slow in metabolism, reduce plant growth and yield, leaf necrosis, and worst plant death in excess amount. Significant differences exist in plant sensitivity to atmospheric Fluoride, but small or no injury will come about when the most sensitive plant



species are exposed to around  $0.2 \mu\text{g}/\text{m}^3$  air, and many species can tolerate concentrations much greater than this.

Application of 100-ppm fluorine as calcium, to field plots increased the yield in some cases. The fluorine is a necessary element for corn, but it is found that it had a toxic effect upon the observed plants. It was also found that the tobacco plant responds to sodium fluoride by transforming free water into bound water in such quantities as to indicate that this compound includes a state of physiological drought. The concentration of fluoride as high as 50 ppm did not significantly decrease the germination of Sudan grass, cowpeas, soybean, or red clover. The addition of soluble fluorides to the amount of 10 ppm did not decrease the amount of dry matter produced by cowpeas growing in a nutrient solution. The fluoride was found mostly in the roots of these plants.

The tea family, *these* in the best known of these contaminant accumulators, but there are several others that warrant further investigations (Kumar and Rani, 2011). Gaseous fluorides present in the air enter leaves through stomata pores and deposited on the surface of exposed plant surfaces. Surface deposited fluoride account for over 60% of the total fluoride content present in the leaf. Although such type of superficial deposited has a negligible toxic effect of the plant, but it may be hazardous for grazing animals. The basic metabolic process affected adversely by fluoride that penetrates and enters in the internal tissue of leaves and visible on the surface of plant life. In fact, many other visible plant stresses appeared are closely similar, none of these symptoms are specific to fluoride. In most of the conditions, fluoride uptake by root form soil is very and therefore, the concentration in the shoots of the plant is typically less than 10 mg F per kg dry weight in non-polluted atmospheres. However, in exceptional cases, a plant having unusual physiology may accumulate high fluoride when grown on high-fluoride soils.

In industrial areas, atmospheric hydrogen fluoride pollution can be a serious case of fluoride toxicity to plants. Several workers in different plants of India have reported chronic endemic Fluorosis, due to fluoride toxicity. A list of the some plants that are sensitive to fluoride has been compiled and is presented as Tab. 1.

### 3.2. Serum fluoride estimation

Wide disagreement was noticed in the findings when chemical methods of estimating fluoride levels in serum were used, and the results came from different research workers. However, when we analyzed serum samples using fluoride ion electrode for investigation, considerably lower levels of fluoride in serum were found i.e., 0.4 to 0.9 ppm in fluorotic patients and 0.19 to 0.4 ppm in normal subjects. In non-endemic areas, the average value for serum lies between 0.002 to 0.008 mg/100 ml. In endemic areas, serum fluoride levels were found in the range of 0.02 to 0.15 mg/100 ml; whereas, it was 0.02 to 0.19 mg/100 ml in patients with skeletal fluorosis. The level of fluoride in serum of 500 normal, healthy adults was



lying between 0.03 to 0.13 ppm with a mean value of 0.08 ppm and they were 0.04 to 0.28 ppm with a mean of 0.16 ppm in 17 fluorotic patients in our laboratory. The fluoride levels of urine of these 17 fluorotic patients varied between 0.68 to 7.80 ppm with an average value of 3.28 ppm shown in Tabs. 2 to 4.

*Table 2. Fluoride level in serum of pot-room workers.*

Employee code number	Fluoride in serum (mg/l)
027702	0.13
027522	0.08
027616	0.06
027666	0.07
027511	0.15
027339	0.09
027571	0.09
027697	0.12
027767	0.17
027679	0.09
028291	0.11
027483	0.09
027647	0.12
027695	0.08
027484	0.08

*Table 3. Fluoride level in serum of non pot-room workers.*

Employee code number	Fluoride in serum (mg/l)
025944	0.03
006419	0.03
031277	0.02
006476	0.04
006388	0.02

*Table 4. Summary of serum fluoride content.*

	No. of workers investigated	Fluoride content in serum (mg/l)	Average (mg/l)
Pot-room workers	30	0.06 – 0.15	0.10
Non pot-room workers	5	0.02 – 0.04	0.03

### 3.3. Urine fluoride estimation

Best indicator of fluoride consumption is assumed to be level of urinary fluoride. As excretion of fluoride is flexible all through the day, it is wiser to collect 24-hour urine samples instead of collecting random or morning samples for more reliability in fluoride estimation. Generally, urinary fluorides level varies individually and ranges between 0.1 to 2.0 ppm (average 0.4 ppm approximately) when fluoride content of drinking water is 0.3 ppm. Usually, urinary fluoride level rise and fall proportionately with fluoride intake and varies extensively from day to day within the range of 0.5 to 4.48 ppm minimum and 1.5 to 13.0 ppm maximum in case of skeletal fluorosis and 26 ppm or more in case of high endemic regions. Analysis and assessment of excretion pattern of urinary fluoride found in the range of 1.5 to 7.5 ppm with a mean value of 4.2 ppm (Tabs. 5 to 7)

Table 5. Fluoride level in urine of pot-room workers.

Employee code number	Urine pH	Fluoride in urine (mg/l)
027702	6.0	6.10
027522	6.0	0.84
027616	5.5	0.73
027666	5.0	6.50
027511	6.0	1.22
027339	6.0	0.53
027571	5.0	3.52
027697	5.0	0.86
027767	5.6	3.10
027679	5.0	2.86
028291	5.5	0.57
027483	5.5	3.67
027647	5.5	1.67
027695	5.5	2.35
027484	5.5	5.10

Table 6. Fluoride level in urine of non pot-room workers.

Employee code number	Urine pH	Fluoride in urine (mg/l)
025944	5.2	0.78
006419	5.8	0.29
031277	5.2	0.86
006476	4.6	0.82
006388	4.6	0.49

Table 7. Summary of urine fluoride content.

	No. of workers investigated	Urine pH	Workers having the urine pH
Pot-room workers	50	<5.5	20 (40%)
		5.5–6.5	25 (50%)
		>6.5	5 (10%)
Non pot-room workers	10	<5.5	6 (60%)
		5.5– 6.5	4 (40%)

in case of fertilizer workers. In general, urinary fluoride levels are found low in renal disease case. It is observed that fluoride excretion is higher than intake in patients of low fluoride regime.

### 3.4. Geospatial approach

This study has also included the use of remote sensing and GIS to incorporate the present the interpolated estimation of the fluoride concentration over a distance. Figure 3 represents the spatial interpolation of the points into the distance and evaluating fluoride concentration over the study area. It predicts the fluoride values in other places against the measured points. Thus, it provides a better assessment of fluoride concentration over the entire region in very less time and accurately. In Fig. 3, it can be seen the nearest area corresponds to higher fluoride concentration as compared to the far places which is obvious from the study. This estimate the concentration of fluoride over a region those are not sampled or estimated in short duration of time. It can be inferred from the Fig. 4 that fluoride is accumulated more in the proximity to the industrial emission source points as compared to the other places.

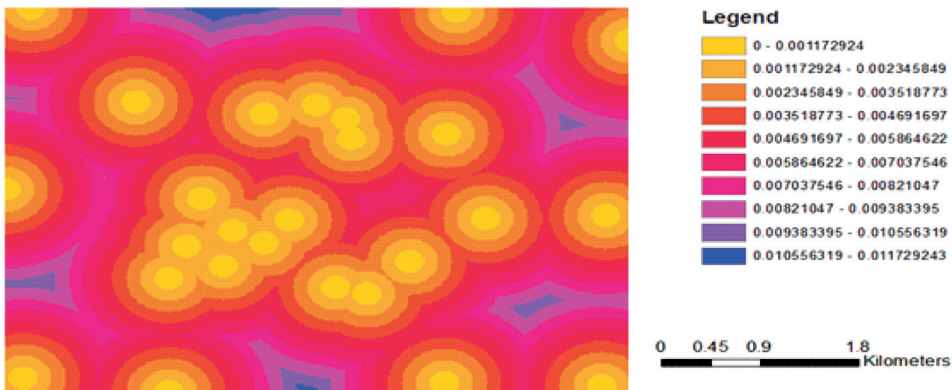


Figure 3. Weighted distance interpolation results generated from the fluoride samples.

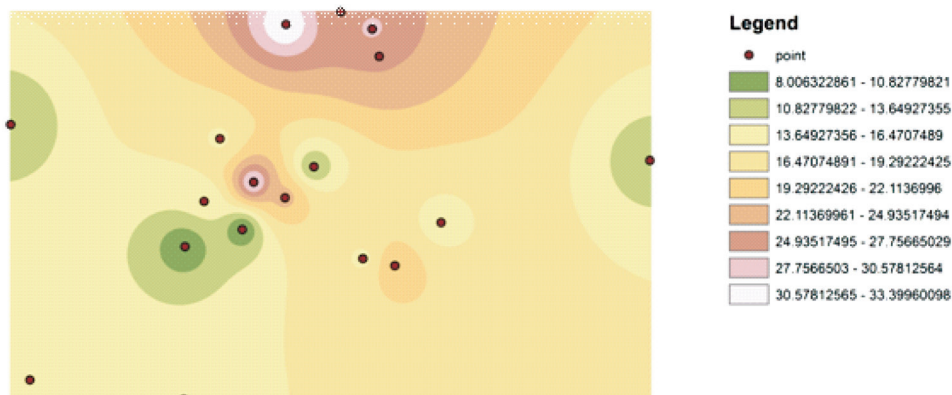


Figure 4. Distance interpolation results generated from the fluoride samples.

#### 4. Discussion

The present report is based on the extensive field and laboratory analysis carried out within 15 km radius of the Hindalco Industries Ltd. According to our experimental estimation, the fluoride content was found about 43.6 ppm as the maximum near Potline 3 in flora sample and 8.4 ppm as minimum near riverside. This result indicates that the fluoride content is much below the permissible limit in the flora sample, i.e.; 80 ppm as stated in CREP. It is also observed that sensitive plant with respect to fluoride accumulation like *Occidentalis*, *Prunus persica*, *Pinus contorta* etc. are growing properly within the premises of Hindalco industry. In this way fluoride content was analyzed in the fauna also. Samples of urine, serum and nails were taken for the analysis. Fluoride content variation in serum from 0.06–0.17 mg/l in case of pot-room workers and 0.01–0.04 mg/l in case of non pot-room workers. In nails it varies from 0.09–3.77 mg/l and 0.39–1.15 mg/l in case of pot room workers and non-pot-room workers respectively. In urine it varies from 0.53–9.50 mg/l of pot room workers and 0.29 to 1.80 mg/l in non-pot room workers shown in Fig. 5. Figure 6 indicated that fluoride concentration is higher in urine as compare to nails and serum.

The most hazardous phytotoxic pollutants released while the aluminium reduction process is fluoride that is found in the form of Hydrogen fluoride, particulate Fluoride i.e. cryolite ( $\text{Na}_3\text{AlF}_6$ ) and calcium fluoride ( $\text{CaF}_2$ ). These are formed mainly by volatilization of fluoride-containing electrolyte. Tables 2 to 10 give a brief description of fluoride in samples of urine, serum and nails. In serum, fluoride content varies from 0.06–0.17 mg/l and 0.01–0.04 mg/l in the case of pot room workers and non-pot room workers, respectively. On the other hand, it ranged from 0.09–3.77 mg/l and 0.39–1.15 mg/l in nails of pot room workers and non-pot room workers respectively. In the urine, it varies from 0.53–9.50 mg/l of

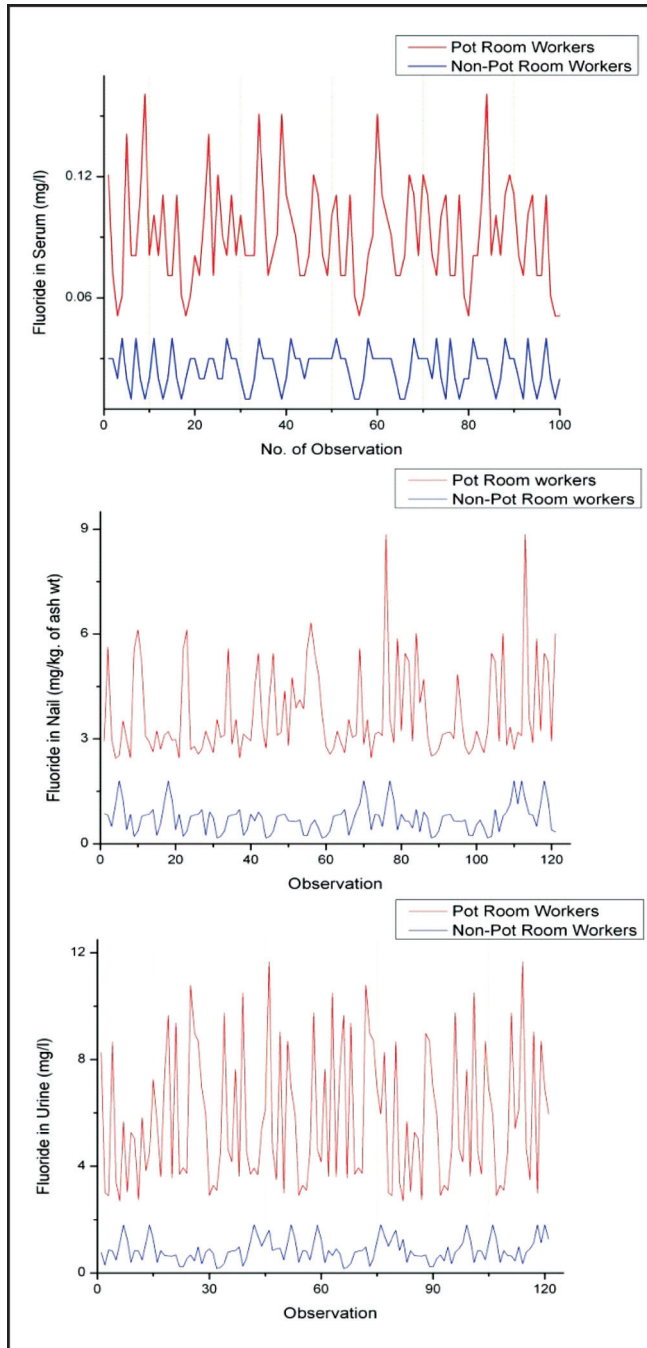
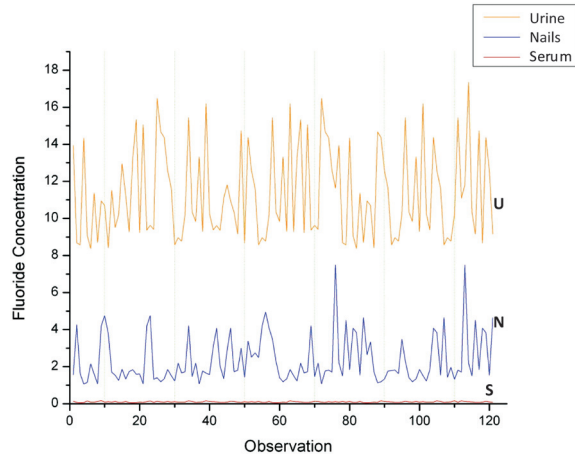


Figure 5. Fluoride concentration in serum, nails and urine as conducted on workers.



**Figure 6.** Fluoride concentration in serum and urine as conducted on workers, where N represents Nails, U represents Urine, and S represents Serum.



**Figure 7.** Dental fluorosis level in different workers of industry.

*Table 8. Fluoride level in nails of pot-room workers.*

Employee code number	Fluoride in nail (mg/kg of ash weight)
027702	0.58
027522	3.28
027616	0.65
027666	0.09
027511	0.18
027339	1.17
027571	0.63
027697	0.10
027767	3.21
027679	3.77
028291	2.81
027483	0.72
027647	0.56
027695	0.28
027484	0.88

*Table 9. Fluoride level in nails of non pot-room workers.*

Employee code number	Fluoride in nail (mg/kg of ash weight)
025944	0.52
006419	0.83
031277	0.39
006476	–
005856	1.15

*Table 10. Summary of nails fluoride content.*

	No. of workers investigated	Fluoride in nail (mg/kg of ash weight)	Average (mg/kg of ash weight)
Pot-room workers	50	0.09–3.77	1.10
Non pot-room workers	10	0.39–1.15	0.65



pot room workers and 0.29 to 1.80 mg/l in non-pot room workers. The average fluoride content was recorded as 1.10 mg/kg in the pot room and 0.65 mg/kg in and non-pot room workers. The fluoride concentration in some of the above reported events were noticed higher with regard to approved limits. However, no visible sign of fluorosis was found in any of the pot room worker or non-pot room worker. Dental fluorosis (Fig. 7) is irretrievable situation caused by extreme fluoride intake during tooth formation stage. It is the earliest *visible* symptom of over consumption and accumulation of fluoride in body of a child. Excess fluoride resulted in dental fluorosis by damaging enamel-forming cells, known as ameloblasts. The destruction of these cells consequences in mineralization disturbance of teeth, and further sub-surface enamel porosity is increased.

## 5. Conclusion

The present study illustrated the fluoride concentration impact on plant, and human beings living near the industries. This impact cannot be overruled by the organization as it has severe effects on the health of human beings, i.e. workers as well as residential persons living in the surrounding area. The concentration of fluoride should be within the standard limit as described by WHO. The analysis of samples indicated that the ground water of nearby villages was found to be highly fluorinated; some cases of fluorosis were also observed due to consumption of fluoridated water. This all has an ill effect on the human health particularly, dental problems as indicated in the Fig. 7. Sometimes, fluoride accumulates in the roots, grains, shoots or leaves and the level of accumulation depends upon the fluoride concentration in air, and water (Gautam and Bhardwaj, 2010). The accumulation of fluoride seriously effects the plant and associated ecosystems (Maňkovská and Steinnes, 1995). It has been found that an accumulation of fluoride is dependant upon the seasons like summer and winter, as well as foliar spread and quantity of trees, shrubs and herbs (Bowen, 1998). Thus, plant accumulates more during winter, closer to industries (Fig. 4) and big foliage and canopy as compared to summer, distant trees and shrubs or herbs (Bowen, 1998).

Hindalco industries took first initiation of providing clean, and safe water to people of affected villages. As per results from the study, it is found that the fluoride effect on flora and fauna is in normal condition around Hindalco. Hindalco has set-up the latest technology i.e. advanced Dry Scrubbing System (DSS) and Electrostatic Precipitators (ESPs) to minimize the emission of fluoride to the environment. All above discussed the results and the introduction of new technology suggests that the Hindalco is an eco-friendly Aluminium plant to the environment. Still, it needs a look over the emissions of fluoride that can become more dangerous to the human, animals and plants if the emission continued at the same pace into the environment.

*Acknowledgement* – Authors are thankful to Dr. A. K Susheela for providing the valuable suggestion and guidance during lab analysis. Authors are also thankful to Hindalco industries, Renukoot, UP, India for providing all necessary support throughout carrying the research.

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#### SAŽETAK

### **Utjecaj fluorida proizvedenih aluminijskim industrijskim postrojenjem na biljke i područja nastanjena ljudima**

*Prem Chandra Pandey, Pavan Kumar, Meenu Rani, Swati Katiyar  
i Vandana Tomar*

Fluor je vrlo reaktivan element koji se u elementarnom obliku ne pojavljuje u prirodi. Postoji u obliku fluorida u Zemljinoj kori, gdje se nalazi u omjeru od oko 0,3 g/kg. Općenito, nalazi se u obliku velikog broja minerala poput fluorita, kriolita i apatita. Fluorid ima i pozitivne i negativne učinke na ljudsko zdravlje. Fluorid, u obliku fluorita i kriolita, široko je rasprostranjen u litosferi, a poznat je i kao trinaesta najčešća tvar u Zemljinoj kori. Fluorovodici u plinovitom obliku nagomilavaju se u listovima uglavnom osjetljivih biljaka prema gradijentu koncentracije. Stoga se smatraju najfitotoksičnijim onečišćujućim tvarima u zraku, a djeluju na biljke i pri ekstremno niskim koncentracijama. Prema našoj studiji utvrđeno je da su učinci fluorida na faunu u uobičajenim okruženjima, ali u budućnosti može imati štetan utjecaj na faunu i floru u okruženju tvornice Hindalco Industries Limited.

*Ključne riječi:* fluorid, ljudsko zdravlje, štetni učinci, društveni aspekti

*Corresponding author's address:* Pavan Kumar, Banasthali University, Department of Remote Sensing, Newai, Rajasthan, India; e-mail: [pawan2607@gmail.com](mailto:pawan2607@gmail.com)