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FLUORIDE TOXICITY IN RICE FIELDS IN THE PERIPHERY OF AN ALUMINIUM SMELTER IN ODISHA, INDIA

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ABSTRACT: The prevalence of hydrogen fluoride (HF) toxicity in rice plants with visible foliar injury was observed in rice fields in the vicinity of an aluminium smelter in Odisha, India. The crop was damaged up to 2.5 km from the aluminium smelter by the atmospheric transportation of fluoride (F) under the influence of the prevailing wind and rain. Plants growing nearer to the smelter (1 km) were exposed more to the fluoride toxicity, resulting in reduced vegetative growth with a reduction in the total dry matter production and a light green colour due to a lower chlorophyll content. The grain yield of the plants with chronic fluoride toxicity was reduced by 53.7%. The grain yield increased as the distance from the smelter increased indicating that the industrial F pollution persisted in the study area. The F content in the leaves of the rice plants at a distance up to 2.5 km from the smelter was higher (95.8±28.6 ppm) than the threshold limit of 30 ppm. The F content in the upper layer of the soil within 3 km from the smelter varied from 83.3±0.7 to 203.1±5.9 ppm. The F content in water at 1 to 2 km from the smelter was higher (1.7±0.6 to 3.7±0.9 ppm) than the threshold limit of 1.5 ppm, whereas from 2.5 to 3 km from the smelter, it was within the 1.5 ppm standard (0.6±0.3 to 0.8±0.4 ppm). The range of the F concentration in the rice grains was 0.3±0.1 to 1.4±0.9 ppm with those collected from 2 or more km from the smelter having a F content that was lower than the threshold limit of 1.0 ppm. The F-containing emissions of the aluminium smelter were considered to be the likely cause of the F accumulation and the pollution of the soil, plants, and water in the vicinity of the aluminium smelter.

Keywords: Aluminium smelter; Chlorosis; Fluoride content; Necrosis; Rice plant; Yield.

INTRODUCTION

The fluoride ion (F) is widely regarded as the third most important air pollutant after SO_4^{2-} and NO_3^{-1}. In India, it is one of the major pollutants in coal-burning and industrial activities, such as power generating stations and the production of steel, iron, aluminum, zinc, phosphorus, chemical fertilizers, bricks, cement, and hydrofluoric acid, etc., and pollutes not only the air, water and soil of nearby areas but also enters the food chains through vegetation, including the agricultural crops on which man and animals are generally dependant for food.[2] Prolonged exposure to industrial F pollution causes serious health problems in the form of neighborhood and industrial fluorosis in man[3] and animals[4] inhabiting the vicinity of F emitting industries.

Plants can either absorb F from the contaminated soil or from the atmosphere.[5] F released into the atmosphere from an aluminium smelter can hydrolyze to corrosive hydrogen fluoride (HF), after coming in contact with precipitation.[6] It has been found that a HF content as low as 10 g/kg (total air) can initiate injuries in plants.[7] In plants, chronic F toxicity causes physiological complications like necrotic lesions, burning, chlorosis, leaf damage, and inhibition in the developmental and reproductive capabilities in sensitive plants.[8] Plant leaves consist of an external layer, the cuticle,

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which is discontinuous and contains cracks and perforations (stomata). So leaf structure also decides the penetration and biological action of accumulated F. Highly susceptible gladiolus plants may become chlorotic with 20 ppm F,[9] while cotton plants may appear healthy with 4,000 ppm F.[9] The severity of F toxicity in rice plants varies with the distance from the smelter plant. Besides the amount of F entering inside the plant, the factors of duration of exposure, growth stage, variety, and stress, along with other factors such as the local environment (precipitation, wind, temperature, and humidity), soil texture, and structure may increase the prevalence of F toxicity.[10] This problem is frequent in the cropped area near the aluminium smelter of Hirakud in the West Central Table Land Zone of Odisha. The crop field around the smelter comes under the double cropping pattern of rice-rice and is subjected to total loss of crop, usually during *kharif* (the monsoon season), due to F toxicity.

The present study was an attempt to determine the concentration levels of F in the leaves and grains of rice crop, soil, and water samples collected at various distances from the F emitting aluminium smelter in Odisha, India and to assess the effect on growth and yield loss in rice plants growing in the periphery of this F emitting factory.

MATERIALS AND METHODS

Study site: The study was conducted in the farmers' fields of Hirakud at the base of the famous Hirakud reservoir, Odisha, India. The aluminium smelter, a unit of Hindalco, has been operating at Hirakud town in Sambalpur district of Odisha since 1958 producing aluminium. It is located 20 km away from the Regional Research and Technology Transfer Station, Chiplima, of Odisha University of Agriculture and Technology under West Central Table Land agro climatic zone of Odisha at 20.21º N, 80.55º E and at an elevation of 178.8 m. The area is canal irrigated and the predominant cropping pattern is rice-rice.

Sample collection: The soil, plant, and water samples were collected from a randomly selected direction (East) at 1.0, 1.5, 2, 2.5, and 3 km distance from the aluminium smelter. At each distance, 5 sites were randomly selected and rice plant samples were collected from each site. Similarly from the vicinity of same sites soil samples were collected at 0 – 20 cm and 20 – 40 cm depth layers. The soil and plant samples were sealed in clean polythene bags. Water samples were also collected from the rice field in plastic bottles from these sites. All the samples were transported to the laboratory at the Regional Research and Technology Transfer Station, Chiplima, for analysis.

Fluoride concentration level: Initially, the weight of all the leaf samples was measured, and they were then dried at 70ºC for 48 hr and ground into fine powder. The F analysis was done by the acid digestion method.[11] For the measurement, a 10 mL sample extract was mixed with TISAB (total ionic strength adjustment buffer) III and then measured using an ion selective electrode (Orion 9609BNWP) coupled to an analyzer (Orion, USA). For the analysis of the soil samples, the samples were air dried and crushed and then the gravel was picked out. The F content was determined after the samples were decomposed by fusion with NaOH in Ni crucibles, at 600ºC for 30 min. These fused samples were then dissolved with deionized water and mixed with TISAB buffer solution to determine the F content using the ion selective

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electrode method.[12] Fluoride in water was analyzed by an ion-analyzer (Orion, USA). For the rice grain samples, the analysis was performed after appropriate digestion and determined by the ion selective electrode method in a similar way to that used for the soil samples.

Climatic data: The climatic data used for this study included the observation of the temperature and the precipitation taken from the meteorological observatory, situated at the Regional Research and Technology Transfer Station, Chiplima. The climate is

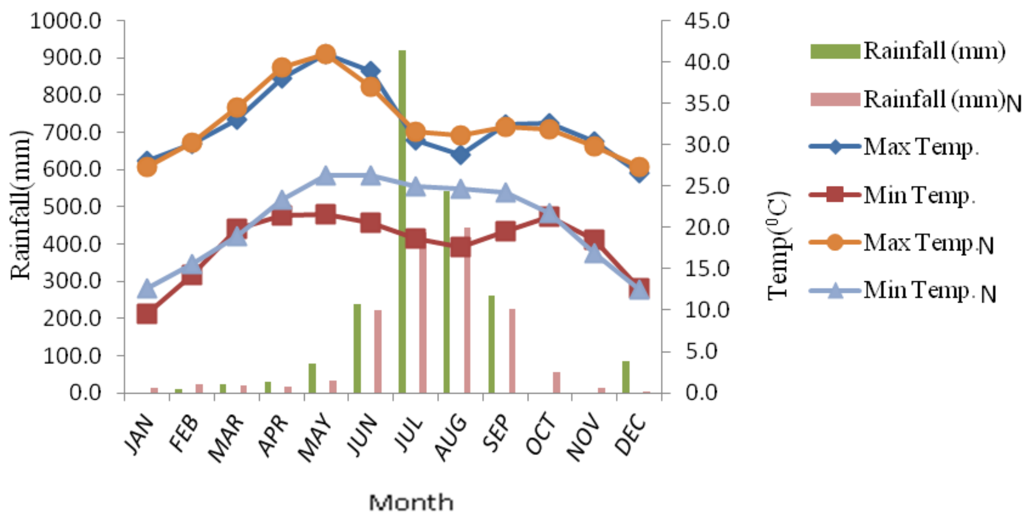

Figure 1. Weather parameters (total monthly rain fall and mean monthly maximum and minimum temperature) in the year 2018 and normal (N) (average of 90 years) of Regional Research and Technology Transfer Station, Chiplima, Sambalpur, Odisha, India.

The summer (March to June) is dry and hot, with scorching sun and strong westerly winds during the day. The dry summer is followed by the warm and moist rainy season of about 4 months (July to October), as the monsoon usually brings rain by the end of June. The total annual rainfall for the study year 2018 was 2,192.2 mm against a normal rain fall of 1,495.7 mm. The rainy season is the one with the maximum biological activity, when the farmers are busy in raising *kharif* rice in the major portion of their fields. This is followed by the winter season, which in this region normally extends from November to February. The temperature begins to fall from early November and the coldest period lies between December and January. The winter days are sunny but cool and the nights are cold and clear.

Yield attributes and yield: Panicle length was measured from the nodal base to the tip of the panicle. The above ground parts of 5 plants under observation in each plot were oven dried at 60ºC until they attained a constant weight to find out the dry matter/plant weight (g) at the physiological maturity stage. At harvest, the plants were threshed to separate the grains from the straw and, after proper drying in the sun, the grain yield was measured in g/plant. Each seed lot was harvested individually and a thousand seeds were randomly collected, properly dried (14%

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moisture), and then weighed in an electrical balance. The well filled grain was determined using a 1.20 specific gravity salt solution.[13] The leaf chlorophyll content was estimated by measuring leaf greenness using a portable SPAD (soil plant analysis development) -502 chlorophyll meter (Minolta Camera Co. Ltd. Japan).[14]

Statistical analysis: Two way analysis of variance was performed to find out if the grain parameters measured were significantly affected by the distance between the sampling locations and the fluoride pollution source.[15]

RESULTS

F concentration in the samples: The F concentrations in the samples together with the distance from aluminium smelter are shown in Table 1.

Table 1. Assessment of fluoride (F) concentrations (ppm) in rice leaf, grain, soil, and water at various distances from the aluminium smelter. Values are mean±standard deviation (SD)

Distance from smelter (km)	F concentration (ppm) in rice leaf (Mean±SD)	F concentration (ppm) in soil (Mean±SD)		F concentration (ppm) in water (Mean±SD)	F concentration (ppm) in grain (Mean±SD)
		Upper layer	Deeper layer		
1.0	1123.4±663.8	203.1±5.9	167.2±9.1	3.7±0.9	1.4±0.9
1.5	821.0±580.6	183.5±1.5	149.7±1.8	2.3±0.8	1.2±0.7
2.0	372.0±158.1	157.7±0.7	121.9±5.2	1.7±0.6	0.8±0.5
2.5	95.8±28.6	97.7±0.8	63.8±1.1	0.8±0.4	0.5±0.3
3.0	25.4±2.7	83.3±0.7	49.3±0.7	0.6±0.3	0.3±0.1
p value	<0.001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*

*Significant at p=0.05

The F concentration in the leaves of the rice plants varied between 25.4±2.7 and 1123.4±663.8 ppm with a significantly higher mean concentration of F (1123.4±663.8 ppm) at the place nearest (1 km) to the smelter. The F concentration in the leaves of the rice plants showed an inverse relationship to the distance from the smelter. With respect to the soil layers, it was observed that the mean F concentration was significantly highest at the same point, 1 km from the smelter, for both the upper soil layer (203.1±5.9 ppm) and the deeper layer (167.2±9.1 ppm). Moreover, the upper soil layer revealed a significantly higher mean F concentration than the deeper layers at all the distances. The F concentrations in the water and grain samples, at the various distances from the smelter, were found to be in the range of 0.6±0.3 to 3.7±0.9 ppm and 0.3±0.1 to 1.4±0.9 ppm, respectively.

Prevalence of F toxicity effect on growth and yield of crop: Plant height was significantly affected by F toxicity (Table 2). The lowest plant height (97.7 cm) was recorded in the crop raised nearest to the smelter (1 km), while it increased with increasing the distance from the smelter. There was little effect of F toxicity on plant

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height (109 cm) at 2.5 km from the source as this was statistically at par with that of the distance at 3 km (112.7 cm).

Table 2. Growth, chlorophyll content, yield attributes, and yield of rice in fluoride (F) toxicity plot (TDM = total dry matter production; NS = not significant; DAT = days after transplant; SPAD = soil plant analysis development; CD = confidence distribution)

Distance from source (km)	Plant height (cm)	TDM (g/hill)	Chlorophyll content at 50 DAT (SPAD)	Panicles /plant	Panicle length (cm)	Grains /panicle	1,000 seed wt (g)	Grain yield (g/hill)
1.0	97.7	30.0	3.5	3.7	17.4	122.0	19.5	14.4
1.5	100.0	33.9	4.9	4.8	22.6	143.0	20.5	23.3
2.0	104.0	39.2	6.3	5.6	22.8	153.0	20.6	26.0
2.5	109.0	41.3	10.9	5.7	25.6	165.0	20.9	29.5
3.0	112.7	41.9	12.6	6.4	26.5	178.0	21.4	31.1
CD (p=0.05)	4.1	1.8	2.8	NS	2.8	24.0	0.5	2.7

Dry matter production increased with increasing distance from the smelter. The highest dry matter production (41.9 g/hill) occurred at a distance of 3 km from the smelter followed by 2.5, 2, 1.5, and 1 km (Table 2). The plants situated after 2.5 km from the source were not affected by F toxicity and the dry matter produced at 2.5 km (41.3 g/hill) was statistically at par with that at 3 km.

Chlorophyll content (SPAD value) differed significantly as the distance from the smelter varied. It was less (3.5) in the rice plants situated nearer to the smelter within 1 km, while it was highest (12.6) in the plants at 3 km distance. The plants were greener and grew normally as the distance from the smelter increased.

Panicle length, grains/panicle, and the 1,000 seed weight differed significantly with distance up to 2.5km from the smelter but the difference was not significant between 2.5 and 3 km distance.

The effects on the growth and yield parameters were clearly reflected in the grain yield and it also followed the same trend giving the highest yield at 3 km (31.1 g/hill), which was statistically at par with that of 2.5 km (29.5 g/hill). Grain yields at all other locations were significantly lower with the lowest yield (14.4 g/hill) at the nearest point of 1 km. The yield reduction varied significantly, with distances up to 2 km from the source, with a range of 16.4 to 53.7%.

DISCUSSION

Leaf injury: Leaf injury due to F toxicity in the rice plant was observed in the month of July, 2018, when the rice crop was at the early vegetative stage and exposed to extreme monsoon showers. The initial symptom, giving a clear symptom of F toxicity, was the development of chlorosis at the tips with an elongation of this along

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the margin of the leaves and extensions inwards toward the midrib. Later on, the leaf tip turned to necrotic with a burnt margin (Figures 2-4).

Figure 2. Industrial fluoride (F) toxicity affected rice plant showing leaf necrosis with a burnt margin.

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Figure 3. Stunted rice plants due to industrial fluoride (F) toxicity.

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A B C D

Figure 4. Industrial fluoride (F) toxicity in rice plant leaves. A. healthy leaf; B. chlorosis in the leaf; C. tip burning in leaf; D. burning of tip and side margin of leaf.

Similar observations have been made by Weinstein and Davison in forests, crops, and natural vegetation.[26] Accumulation of high levels of F in vegetation is commonly observed in areas polluted by air borne F. Emissions from the aluminium industry contain both particulate and gaseous forms of fluoride. The particulate ones may vary in size and chemical composition. They may contain fluorapatite ($Ca_5FO_{12}P_3$), cryolite (Na_3AlF_6), or fluorite (CaF_2). These particles are deposited on leaf surfaces and their penetration into the plants depends on their solubility. Predominant among the gaseous forms is hydrogen fluoride (HF). It has the greatest effect on vegetation and is closely followed by silicon tetrafluoride (SiF_4), fluosilicic acid (H_2SiF_6), and free fluorine (F_2).[17] Both gaseous and particulate pollution can dissolve on contact with water vapour or rain drops and get inside the plants, chiefly through the stomata of the leaves. The fluorine which penetrates into the plant tissue affects its metabolism in a number of ways.[18] In the present study, the F content was measured in the leaves of rice plants collected from the study site. Significantly the highest mean concentration of F in dry matter (1123.4±663.8 ppm) was found at the place nearest to the smelter (1 km), which was higher than the threshold limit of 30 ppm.[19] The lowest values (25.4±2.7 ppm) was recorded at a distance of 3 km from the smelter, which was within the threshold limit. It has been confirmed that the vegetation in the vicinity of the factory accumulates large quantities of F with variable specific symptoms of toxicity and the F concentration decreases with increasing distance from the pollution source.[20]

Prevalence of F content in soil, water, and grain samples: The concentration of F in the soils collected from the different sampling sites situated at variable distances from the smelter showed an inverse relation with the distance from the source. The high concentration of F in the soil nearer to the smelter might be due to an industrial operation emitting F.[21] With respect to soil layers, it was observed that the mean F concentration was significantly higher in the areas near to the smelter in both the upper soil layers (203.1±5.9 ppm) and the deeper soil layers (167.2±9.1 ppm) than the respective F concentrations at the distant locations. The upper soil layer recorded a higher mean F concentration than the deeper soil layers at all the distances. The same result was also observed in the study conducted in the Hirakud industrial areas of Odisha.[22] F accumulates in the top soil layers because it is retained by the iron, aluminium hydroxide, oxide, and silicate compounds in the soil.[23] A high soil F concentration affects the fertility and structure of the soil by preventing the decomposition of organic substances,[24] due to a decrease in the activity of microorganisms.[25]

The range of the F concentration of the surface water at various distances from the smelter was 0.6±0.3 to 3.7±0.9 ppm. At distance of 1 to 2 km from the smelter, it was higher (1.7±0.6 to 3.7±0.9 ppm) than the threshold value (1.5 ppm), whereas it was within the threshold (0.6±0.3 to 0.8±0.4 ppm) at a distance above 2.5 km from the factory site. The high concentration of F in surface water might be due to the industrial emission of F and its eutrophication in water.[26,27] The F concentrations in the rice grains varied from 0.3±0.1 to 1.4±0.9 ppm. It was lower than threshold limit of 1 ppm after a distance of 2 km from the smelter.[28]

Grain yield: Plants growing nearer to the smelter (1 km) were exposed more to the F toxicity, resulting in reduced vegetative growth (30 g/hill) and were less green, i.e., a lower chlorophyll content (SPAD value of 3.5). Therefore, the yield of the plants suffering from F toxicity was reduced due to less photosynthesis.[29] The grain yield was significantly reduced by 53.7% in rice raised nearer to the smelter than that of 3 km away (31.1 g/hill). The reduction in grain yield was due to a decrease in the yield attributes, such as panicle length (17.4 cm), grains/panicle (122), and the 1,000 seed weight (19.5 g).[28]

Climate: Precipitation frequency and intensity has a very significant role in the emission, distribution and effects of pollution on nearby aluminium smelter. It was common observation that the rice crop was not affected during *rabi* (January to May) due to less rain falling during this period. The *kharif* crop (July to November) was affected more and the crop was severely affected during extreme weather conditions, i.e., continuous drizzling rain after heavy rainfall during the crop growth period, as happened in the month of July 2018 (Figure 4).

CONCLUSIONS

It can be concluded that the F emission from aluminium smelters has a great impact on the growth and yield of the rice plants in the surrounding areas. More foliar injury and poor plant growth were recorded in the rice crops situated within a 2.5 km radius of an aluminium smelter. Earlier investigations from the same area reported the prevalence of F toxicity in the soil, water, and plant samples.[22] The increase in the prevalence of F toxicity and the yield loss found in the present study might be due to a gradual increase in the F concentration in the environment and to an extreme weather event due to climate change. From the health point of view, the present findings are important and highly significant. The consumption of surface water and agricultural products from the peripheral areas of an aluminium smelter causes fluorosis in both cattle and human beings.[30] The results of the present investigation add a significant insight into the effects of industrial F intoxication.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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