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Relationship of Fluoride Content with Physico-Chemical and Chemical Properties of Soil

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Abstract: *This paper analyses fluoride content in 82 soil samples and correlated with physico-chemical and chemical properties of soil. The results indicated that, available F concentration in soils positively correlated with pH and total F content of the soil whereas the correlation coefficient between available F and EC was negative. The relationship between fluoride and CEC, fluoride and P_2O_5 was negative whereas the correlation coefficient between fluoride and other ions is very poor during both seasons. Increasing fluoride content in soil decreases the available P_2O_5 content of the soil. The reduction of P_2O_5 content in soil due to F addition can be attributed to formation of insoluble phosphorus compounds in the soils. Fluoride is negatively correlated with clay percent of soils and positively correlated with sand percent of soils during both seasons but no significant correlation was observed. Clay, soil pH and P_2O_5 in soil control fluoride content in the soil solution. Soil with high pH and low clay and P_2O_5 favours greater solubilization of the fluoride bearing minerals leads to leaching of fluoride which results increase in the concentration of soil fluoride.*

Keywords: *Nalgonda district, fluoride, soil, Physico-chemical and chemical properties.*

I. Introduction

The mobility of fluoride in soils depends on a number of factors, including the initial fluoride concentration, solubility of mineral phases, type of soil, pH values in the soil and other dissolved components in the soil solution.

These factors determine whether or not fluorideis precipitated, sorbs to mineral surfaces or stays in solu tion either as a free or complexes species. The study and monitoring of fluorine and fluorides has been of interest to scientists for a long time. This is because of the reactivity and ready availability of fluorine in many forms. The main natural source of inorganic fluorides in soil is the parent rock. It is the 13th most abundant element of the earth's crust representing about 0.3 g kg^{-1}. It occurs naturally in the combined state as fluorite (fluorospar), apatite, fluorapatite, topaz and cryolite (Rakshit, 2004).

The world average of fluoride concentration in granitic rocks was found to be 810 ppm (Wedepohl 1969), while fluoride content of granitic rocks from Nalgonda district was found to be in the range of 325 to 3,200 ppm, with a mean of 1,440 ppm (Rao *et al*., 1993). Thus, the granitic rocks of Nalgonda possess the highest fluoride content than in any other parts of the world. Assessment of the granite gneisses from Nalgonda showed the presence of fluoride containing minerals such as fluorite (0–3.3%), biotite (0.1–1.7%), and hornblende (0.1–1.1%) (Reddy *et al*., 2009). The clay content and pH of soil are primarily responsible for the origin and/or retention of F in soils. F in soil is primarily associated with the soil colloid or clay (Omueti and Jones, 1977). Correlation coefficients between fluorine content (total and available) and some important chemical and physical soil properties were studied.

II. Materials And Methods

A survey was carried out in Ramannapet, Narkatpalli and Aatmakoor mandals of Nalgonda district by covering all the villages, with a view to assess characterization of soil during two seasons *i.e*. *kharif (*2012) and *rabi* (2012-13). As per the objectives laid down for this study, from each village one soil sample was collected with the help of a handheld Global Positioning System (GPS). Totally 82 benchmark sites were fixed depending on the number of villages in three mandals for collection of soil samples at each site in each season. The samples were collected at 0-15 cm depth by adopting the standard procedures of soil sample collection. The collected soil samples were air dried, gently pounded in wooden mortar and pestle, sieved (2 mm sieve) and preserved in polythene bags for further analysis.

The samples were analyzed for pH, EC and OC as per the standard procedure outlined by Jackson (1973). Available nitrogen was estimated by alkaline potassium permanganate method (Subbaiah and Asija, 1956). Available phosphorus content was estimated by Olsen's reagent as described by Olsen *et al*. (1956) and determined by ascorbic acid method (Watanabe and Olsen, 1965), available potassium was extracted by using

neutral normal ammonium acetate (Jackson, 1973) and determined by flame photometer. Available micronutrients (DTPA extractable) Fe, Mn, Cu and Zn were analyzed by DTPA method using atomic absorption spectrophotometer (Lindsay and Norvell, 1978). The total fluoride was analyzed by NaOH fusion technique using Fluoride Ion Selective Electrode (FISE) method given by McQuaker and Gurney (1977). Available fluoride in soil samples analyzed by Potentiometric method (McQuaker and Gurney, 1977).

III. Results And Discussion

3.1 Available Fluoride and Total Fluoride

The mean values of available F present in the soil samples of Ramannapet, Narkatpalli and Aatmakoor mandals were 1.17, 1.18 and 1.04 mg kg^{-1}, respectively in *kharif* season and 1.41, 1.66 and 1.54 mg kg^{-1}, respectively in *rabi* season (Table 1 and 2). All the values obtained were well within the desirable range of 2.57 to 16.44 mg kg^{-1} soil F. All the values obtained are well within the range of 2.57 to 16.44 mg kg^{-1} soil leachable F stipulated by WHO, 1984. The soil leachable F is the main factor which determines the availability of F for plant absorption. Similarly, F content in soil between 0.02 and 1.00 mg kg^{-1} as reported by Davidson (1983) and between 0.075 and 0.200 mg kg^{-1} as obtained by Okibe *et al*. (2010).

The average content of total F distributed in soils of Ramannapet, Narkatpalli and Aatmakoor mandals were 279, 310 and 291 mg kg^{-1}, respectively in *kharif* season and 289, 338 and 314 mg kg^{-1}, respectively in *rabi* season. Nearly 6% of soil samples in both *kharif* and *rabi* seasons recorded higher total F content than the usual range and remaining samples were within its normal range. Total F of normal soils is usually in the range from 150 to 400 mg kg^{-1} as prescribed by Newman (1984). Nearly 6% of soil samples recorded higher than the usual range and remaining 94% of samples were within its normal range indicates its geo-chemical origin without any form of artificial contamination. So, a danger from F accumulation in plants and its toxicity to human and animals are not to be expected. The results are in conformity with the findings of Jakovljevic *et al.,* 2002.

The percentage of available F from its total F content ranges from 0.10 to 0.82. The available F content was very low (0.26 to 2.64 mg kg^{-1} soil), being less than 1% from its total amount, so it could be concluded that there was no danger from F accumulation in the plants. The content of available F in the soil samples is very low indicating that major part of deposited F had transformed itself in to insoluble compounds like CaF_2 (Blagojevic *et al*. 2002). These results are in conformation with the findings of Jakovljevic *et al.* (2002). Unfortunately there is no Indian standard available prescribing a limit to the F in soil and biological tissue. Although around 95% of the F deposited in soil surface is not available for plant uptake because of its conversion to unavailable form.

3.2 Physico-Chemical and Chemical Properties of Soil

The results indicated that the soils are neutral to alkaline in reaction, non saline in nature, low to medium in organic carbon (OC) content and cation exchange capacity (CEC). The texture of the soils collected in different villages of Ramannapet, Narkatpalli and Aatmakoor mandals varied as sandy loam, sandy clay and sandy clay loam, respectively (Table 1 and 2). The information regarding contents of available N, P_2O_5 and K_2O content in both the *kharif* and *rabi* seasons were categorized from very low to medium. With respect to available Zn, 36 (44%) samples in *kharif* and 46 (56%) samples in *rabi* were deficient, while remaining samples were found above critical limit (0.6 mg kg^{-1}) of available Zn. The results on available Fe revealed that, 44% samples in *kharif* and 27% samples in *rabi* were below critical limit (<4.5 mg kg^{-1}) of available Fe. The available Mn, Cu content of soils in both the *kharif* and *rabi* seasons are found to be above critical limit. Considering the critical limit for available B as 0.45 mg kg^{-1}, about 75% samples in *kharif* and about 32% samples in *rabi* were found to be deficient. The results revealed that, all the heavy metals viz., Cd, Cr, Ni, Pb and Co analyzed during *kharif* and *rabi* seasons were within the permissible limits.

Table 1. Range and mean values of physico-chemical and chemical characteristics of the soil samples collected from different mandals of Nalgonda district (*Kharif*-2012).

Characteristics	Ramannapet		Narkatpalli		Aatmakoor	
	Range	Mean	Range	Mean	Range	Mean
Sand (%)	46.0-66.6	56.8	44.0-70.8	57.0	47.5-68.5	55.7
Silt (%)	7.4-20.5	13.1	7.5-17.8	12.3	8.2-22.5	14.3
Clay (%)	19.8-41.4	30.1	12.8-41.4	30.8	11.2-40.8	30.0
pH	7.14-8.64	7.97	7.25-8.52	7.76	6.98-8.52	7.77
E.C (dS m^{-1})	0.14-0.29	0.21	0.12-0.32	0.20	0.11-0.32	0.21
CEC (c mol(p^+) kg^{-1})	9.86-22.31	15.18	8.98-24.64	15.32	9.25-21.23	15.40
OC (%)	0.23-1.04	0.50	0.27-0.70	0.48	0.24-0.65	0.40
N (kg ha^{-1})	110-289	191	144-296	203	116-278	189
P_2O_5 (kg ha^{-1})	8.55-32.12	19.49	9.36-25.36	19.50	8.26-35.21	20.55
K_2O (kg ha^{-1})	125-312	229	158-325	255	160-320	248
S (mg kg^{-1})	5.78-28.21	12.73	5.98-25.56	13.23	5.58-23.4	11.30

Available F (ppm)	0.41-2.32	1.17	0.26-2.64	1.18	0.29-2.02	1.04
Total F (ppm)	178-451	279	195-481	310	128-456	291
Zn (mg kg^{-1})	0.23-2.17	0.68	0.22-2.02	0.84	0.17-6.58	1.06
Mn (mg kg^{-1})	2.56-17.68	8.39	2.42-14.56	8.18	2.09-21.04	8.48
Fe (mg kg^{-1})	2.52-13.56	6.99	2.02-18.69	8.28	1.98-23.78	7.08
Cu (mg kg^{-1})	0.24-2.99	1.06	0.24-3.12	1.13	0.34-2.58	1.10
B (mg kg^{-1})	0.18-0.89	0.53	0.22-1.23	0.64	0.18-1.18	0.51
Cd (mg kg^{-1})	0-0.21	0.05	0-0.24	0.07	0-0.32	0.09
Cr (mg kg^{-1})	0-0.18	0.03	0-0.09	0.02	0-0.08	0.02
Ni (mg kg^{-1})	0.02-0.32	0.20	0.09-0.38	0.20	0.08-0.45	0.20
Pb (mg kg^{-1})	0.25-1.4	0.71	0-1.44	0.42	0.08-0.89	0.39
Co (mg kg^{-1})	0.02-1.28	0.28	0.07-1.2	0.49	0.05-1.28	0.43

Table 2. Range and mean values of physico-chemical and chemical characteristics of the soil samples collected from different mandals of Nalgonda district (*Rabi* 2012-13).

Characteristics	**Ramannapet**		**Narkatpalli**		**Aatmakoor**	
	Range	**Mean**	**Range**	**Mean**	**Range**	**Mean**
pH	7.18-8.65	8.08	7.26-8.66	7.95	7.20-8.64	7.97
E.C (dS m^{-1})	0.15-0.98	0.49	0.14-0.62	0.30	0.19-0.81	0.39
OC (%)	0.23-0.71	0.47	0.25-0.70	0.46	0.16-0.65	0.39
CEC (c mol(p^{+}) kg^{-1})	7.82-21.58	13.45	8.52-23.86	14.15	6.58-16.58	13.60
N (kg ha^{-1})	153-364	218	159-314	215	156-314	216
P_2O_5 (kg ha^{-1})	12.21-32.65	21.86	8.03-35.42	16.86	8.66-21.53	13.84
K_2O (kg ha^{-1})	171-312	260	159-325	254	185-348	263
S (mg kg^{-1})	4.58-34.00	11.39	5.50-29.50	10.99	6.50-23.65	12.41
Available F (ppm)	0.77-2.39	1.41	0.53-2.64	1.66	0.68-2.63	1.54
Total F (ppm)	186-456	289	246-485	338	164-458	314
Zn (mg kg^{-1})	0.24-2.14	0.66	0.29-1.25	0.70	0.27-1.60	0.67
Mn (mg kg^{-1})	2.61-29.55	8.62	4.02-39.72	12.63	1.35-26.88	9.52
Fe (mg kg^{-1})	2.22-25.23	8.58	2.53-16.08	7.84	2.82-16.20	8.71
Cu (mg kg^{-1})	0.18-2.71	0.82	0.18-1.44	0.73	0.16-1.53	0.64
B (mg kg^{-1})	0.35-0.55	0.42	0.12-0.75	0.36	0.15-0.61	0.30
Cd (mg kg^{-1})	0-0.47	0.07	0-0.32	0.04	0-0.12	0.03
Cr (mg kg^{-1})	0-0.3	0.09	0-0.89	0.20	0-0.75	0.22
Ni (mg kg^{-1})	0.24-1.2	0.71	0.04-1.23	0.45	0.25-2.57	0.95
Pb (mg kg^{-1})	0.24-3.01	1.12	0.12-1.25	0.64	0.28-6.24	3.07
Co (mg kg^{-1})	0.12-1.02	0.35	0.12-0.65	0.30	0.15-1.21	0.48

3.3 Correlation between Fluoride Content and Other Constituents of Soil

Simple correlation co-efficient have been worked out between the available (water soluble) fluoride and other properties of soils. The 'r' values are presented in the Table 3. The positive correlation of available fluoride with soil pH, suggesting that the pH of the soil is more important in determining the concentration of fluoride, in agreement with earlier observation made by Blagojevic *et al*. (2002).

The available fluoride concentration in soils negatively correlated with EC, CEC and P_2O_5 content of soil whereas the correlation coefficient between fluoride and other ions is very poor during both seasons. Increasing fluoride content in soil decreases the available P_2O_5 content of the soil. The reduction of P_2O_5 content in soil due to F addition can be attributed to formation of insoluble phosphorus compounds in the soils. Positive correlation coefficients between available fluoride and the content of some micronutrients metals (Cu, Mn and Zn) were also found, which indicated their mutual geochemical origin. Similar results reported by Jakovljevic *et a*l, (2002).

The relationship between available fluoride and total fluoride was positive but no significant correlation was observed. The available fluoride content was very low (< 3 mg kg^{-1}), being mostly less than 2% from its total amount, so it could be concluded that there was no danger from fluorine accumulation in the plants. Statistically significant correlation coefficient between total and available fluorine contents was not obtained. Similar results reported by Jakovljevic *et a*l, (2002).

It is interesting to note that, fluoride is negatively correlated with clay percent of soils and positively correlated with sand percent of soils during both seasons but no significant correlation was observed. Clay, soil pH and P_2O_5 in soil control fluoride content in the soil solution. Most of the fluoride in the soil is insoluble and, therefore, less available to plants. However, high soil fluoride concentrations or clay can increase fluoride levels in soil solution, increasing uptake via the plant root.

Table 3: Correlation coefficients (r) between available fluoride content and other chemical constituents of soils

S. No	Correlation Among	r value in					
		Ramannapet		Narkatpalli		Aatmakoor	
		Khari	*Rabi*	*Kharif*	*Rabi*	*Kharif*	*Rabi*
1	Available Fluoride vs pH	**0.68	*0.382	*0.398	*0.464	*0.451	*0.439
2	Available Fluoride vs EC	-0.289	-0.105	-0.259	-0.213	-0.140	-0.032
3	Available Fluoride vs CEC	-0.415	-0.241	-0.353	-0.384	-0.525	-0.353
4	Available Fluoride vs Available P_2O_5	-0.394	-0.423	-0.562	-0.694	-0.366	-0.407
5	Available Fluoride vs Available K_2O	-0.014	0.101	0.074	0.041	0.019	-0.077
6	Available Fluoride vs Available Cu	-0.046	0.056	-0.020	0.164	0.336	0.265
7	Available Fluoride vs Available Mn	0.049	-0.029	0.202	0.186	-0.105	0.131
8	Available Fluoride vs Available Iron	-0.167	-0.108	-0.033	-0.091	0.089	-0.256
9	Available Fluoride vs Available Zinc	-0.049	-0.122	-0.157	-0.039	0.177	0.111
10	Available Fluoride vs Total fluoride	0.276	0.349	0.340	0.271	0.381	0.253
		*5 % (0.389)					
		**1 % (0.499)					

IV. Conclusions

A significant positive correlation coefficient (r) values were observed between fluoride and pH, while fluoride was negatively related with soluble salt content. Soil with low clay and P_2O_5 favors greater solubilization of the fluoride bearing minerals leads to leaching of fluoride which results increase in the concentration of soil fluoride. Clay, soil pH and P_2O_5 in soil control fluoride content in the soil solution.

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