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Distribution of Aluminum and Fluoride in Tea Plant and Soil of Tea Garden in Central and Southwest China

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Abstract: The distribution of Al and F contents and the relationship between Al and F in tea plants and soils of 12 tea gardens in Central and Southwest China were investigated from October 31 to November 14, 2006. The results show that there were differences in pH, CEC, the contents of organic matter (OM), Al and F in the different soils of the tea gardens. The Al content ranged from 1196 to 7976mg/kg for old leaf, 370 to 2681mg/kg for young leaf and 285 to 525mg/kg for stem, whereas the content of F ranged from 221 to 1504mg/kg for old leaf, 49 to 602mg/kg for young leaf and 13.5 to 77.5mg/kg for stem. The concentrations of labile Al varied obviously in the different soils, but the distribution law of labile Al content for the same garden was $Al_{exchangeable} \approx Al_{Fe\text{-}Mn\ oxide} > Al_{organic} > Al_{water\text{-}soluble}$. The contents of different labile F fractions varied slightly in the different soils and the different soil layers, though the exchangeable F content was lowest among the labile F in the soils. The concentrations of Al and F in tea plants increased with increasing amount of water-soluble Al or F, especially the amount of water-soluble fractions in the soil layer of 0–20cm. The correlation between Al content and F content in the tea leaf was more significant than that in the tea stem. Furthermore, the correlation between Al content and F content in whole tea plant was strongly significant (r=0.8763, p<0.01, n=36). There were evident tendency that Al concentration increased with the increase of F concentration in different soil layers. The correlation of water-soluble Al with water-soluble F in all soils was also strongly significant (r=0.7029, p<0.01, n=34). The results may provide a proof that Al and F are jointly taken up by tea plants to some extent in natural tea gardens.
Keywords: tea garden soil; tea plant; aluminum; fluoride

1 Introduction

Tea plant (*Camellia sinensis* L.), one of the most popular beverages in the world, has been found to contain higher concentrations of aluminum (Al) and fluoride (F) than many other plants. Therefore, tea would be a potentially important source of dietary Al and F. Aluminum toxicity to human being has been implicated in the pathogenesis of a number of clinical disorders in patients with chronic renal failure on long-term intermittent haemodialysis treatment (Jones and Bennett, 1986; Friga et al., 1997). Moreover, numerous studies have indicated that some neuropathy such as Alzheimer's disease may be related to the high Al content in human brain (Martyn, 1990; McLachlan, 1995; Walton, 2006). Even though some epidemiological reports were contradictory (Forster et al., 1995; Rondeau et al., 2000), there was a mounting scientific evidence suggesting a relationship between the neurotoxicity of Al and the pathogenesis of Alzheimer's disease (Aremu and Meshitsuka, 2006). Unbalance of F in the human body can cause dental caries and fluorosis (Cao et al., 1996; Browne et al., 2005; Cunha-Cruz and Nadanovsky, 2005). Therefore, it will be of theoretical and practical significance to study the Al and F contents in tea plants and factors influencing the uptake of Al and F from soils into tea plants for adjusting the condition of soil in tea garden and decreasing contents of Al and F in tea plants.

Many factors influence the concentrations of Al and F in tea plants such as soil conditions (Xie et al., 2001a; 2001b), variety of tea plants and the leaf ages (Ruan and Wong, 2001; Shu et al., 2003). Moreover, the chemical behavior of Al and F in the ecosystems of soil and tea plant has been paid attention to because aluminum ion

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(Al^{3+}) can be complexed with fluoride ion (F^-) in soil solution of acidic soils (pH＜6.0) (Xie et al., 1999). The complexes such as AlF^{2+}, AlF_2^+, AlF_4^-, AlF_3^0 and Al^{3+} should be the main species of Al and F in soil solution of acidic soils (McLean, 1976; Wenzel and Blum, 1992; Xie et al., 1999). Yamada (1980) found the effect of complexes of F-Al on the Al uptake of tea seedling by the hydro-culture test. Nagata et al. (1993) studied the forms of F and Al in tea plant and indicated the evidence of Al-F complexes in tea tissues. Ruan et al. (2003) examined the characteristic of fluoride uptake by tea plants and the impact of Al. Xie et al. (2001) reported the relationships between Al and F in soil profiles of an abandoned tea plantation in Hong Kong and their uptake by six woody species, but they did not investigate the relationship between Al and F concentrations in soil, and Al and F contents in tea plant. Consequently, up to now, little is known about the joint behavior of F and Al in different soils of tea garden and tea plant tissues. The objective of the present research was to investigate the distribution characteristics of F and Al contents in different soils of tea garden and tea plants in Central and Southwest China.

2 Materials and Methods

2.1 Sampling of soil and plant

To investigate the distribution of Al and F and the relationships between Al and F in tea plants and soils of tea gardens, samples of soils (0–20cm, 20–40cm and 40–60cm), tea leaves with different ages (old leaf and young leaf) and stem (living between old leaf and young leaf) were collected with sampling method of "Z-shaped" from 12 tea gardens located in main tea production areas in Central and Southwest China from October 31 to November 14, 2006. These tea gardens were named SC (Sichuan Agricultural University, Ya'an City, Sichuan Province, 29°58′N, 102°59′E), CQ1, CQ2 and CQ3 (Chongqing Tea Research Institute, Yongchuan District, Chongqing Municipality, 25°53′N, 105°56′E), YN1 and YN2 (Yunnan Agricultural University, Kunming City, Yunnan Province, 25°08′N, 102°45′E), HN1, HN2 and HN3 (Hunan Agricultural University, Changsha City, Hunan Province, 28°11′N, 113°13′E), HB1 (Huazhong Agricultural University, Wuhan City, Hubei Province, 30°36′N, 114°18′E), HB2 and HB3 (Institute of Fruit and Tea, Hubei Academy of Agricultural Sciences, Wuhan City, Hubei Province, 30°36′N, 114°18′E), respectively, which were far away from any possible pollution of Al and F industries and urban centers. The soil samples were air-dried, sifted to pass 2mm sieve and stored in plastic containers. The plant samples were rinsed thoroughly with deionized water, dried in an oven at 50℃, then ground with a mill to pass 2mm sieve and stored in a dessicator.

2.2 Analytical methods

Soil pH and organic matter (OM) content were determined by the methods described by Avery and Bascomb (1974). Cation exchange capacity (CEC) was measured based on the method proposed by Hesse (1971) with sodium (NaAc) used as the exchange ion.

In order to reasonably compare Al-forms with F-forms in the soil extracts under the identical test condition, the various forms of Al and F, i.e., water-soluble Al/F, exchangeable Al/F, Fe-Mn oxide Al/F and organic combined Al/F were obtained with the sequential extraction procedure as follows: deionized water, 1mol/L $MgCl_2$, 0.04mol/L $NH_2OH{\cdot}HCl$ and 0.02mol/L HNO_3+ 30% H_2O_2, respectively (Wu et al., 2002). The total Al contents of soils and plants were digested with a mixture of nitric and perchloric acid (Ramsey et al., 1991) and the concentration of Al in all extracts was determined by ICP-AES at 308.2nm. The total F contents of soils and plants were melted with alkali and the concentration of F in all extracts was determined with F-ion selective electrode (Wu et al., 2003).

3 Results and Discussion

3.1 Characteristics of soils of tea garden

The properties of soils in the tea gardens were showed in Table 1. The total content of Al ranged from 71.4 to 110.0g/kg for the different tea gardens and different soil layers, which was 7.1% higher than the average (71.0g/kg) in the lithosphere (Lindsay, 1979), also higher than that of the soils of 13 tea gardens in East China (Xie et al., 2001a). The total F content ranged from 142 to 526mg/kg for all soil samples, which was similar to that of the soils of 13 tea gardens in East China (Xie et al., 2001b). But most of the F contents in the soils were lower than the F background value (478mg/kg) of soil in China (Jiao et al., 2000). The soil pH increased and the organic matter content of soils decreased with the depth of soil layers for all the tea gardens, however, there were not obvious changing trend for CEC in different soil layers of the same tea gardens. The variation of soil properties among these tea gardens may relate to the different parent material, cultivating ages and manner (Liu and Hu, 2004).

Table 1 Properties of soils in tea gardens

Tea garden	Parent material	Total Al (g/kg)			Total F (mg/kg)		
		0–20cm	20–40cm	40–60cm	0–20cm	20–40cm	40–60cm
SC	Purple rock	75.8	84.9	75.9	328	443	391
CQ1	Yellow sandstone	76.1	76.9	80.2	383	383	462
CQ2	Yellow sandstone	75.5	79.9	71.4	347	387	371
CQ3	Yellow sandstone	77.5	85.3	79.4	514	503	461
YN1	Basic rocks	87.8	110.0	80.5	148	203	162
YN2	Basic rocks	85.1	99.8	90.8	142	180	175
HN1	Quaternary red earth	90.9	91.9	107.7	365	387	405
HN2	Quaternary red earth	103.9	101.4	101.6	416	503	526
HN3	Quaternary red earth	108.3	100.7	85.6	456	461	441
HB1	Middle Pleistocene deposits	88.7	109.5	102.2	406	371	340
HB2	Middle Pleistocene deposits	96.7	100.9	102.5	348	284	262
HB3	Middle Pleistocene deposits	103.6	94.8	93.7	433	455	423

Tea garden	pH (H_2O)			OM (%)			CEC (cmol/L)		
	0–20cm	20–40cm	40–60cm	0–20cm	20–40cm	40–60cm	0–20cm	20–40cm	40–60cm
SC	3.53	4.05	4.78	4.90	3.65	2.99	1.33	0.95	0.61
CQ1	3.80	3.88	3.92	4.49	3.02	2.84	0.63	0.48	0.48
CQ2	3.76	3.74	4.02	4.17	4.46	3.85	0.61	0.87	1.69
CQ3	4.61	4.95	5.48	5.86	3.94	3.60	1.86	2.74	3.35
YN1	5.63	6.87	7.01	8.63	9.37	7.33	4.63	5.39	6.13
YN2	5.75	6.89	6.96	3.89	3.65	3.58	4.40	4.52	5.12
HN1	4.38	4.41	4.52	3.58	3.53	3.41	3.04	3.00	2.21
HN2	4.93	5.25	4.85	6.36	4.83	4.64	3.08	2.74	2.43
HN3	4.36	4.42	4.60	4.40	4.01	2.91	3.59	3.86	3.30
HB1	4.16	4.19	4.04	4.83	3.16	2.23	2.72	2.69	2.22
HB2	4.13	4.35	4.73	4.89	5.09	4.59	2.80	2.82	2.78
HB3	4.26	3.81	3.79	3.63	3.87	3.57	2.46	2.39	2.65

3.2 Distribution of Al and F in tea plants and soils

The contents of Al and F in the tea plants varied obviously for different tea gardens (Table 2). The Al content ranged from 1196 to 7976mg/kg for old leaf, 370 to 2681mg/kg for young leaf and 285 to 525mg/kg for stem, whereas the F contents ranged from 221 to 1504mg/kg for old leaf, 49.0 to 602mg/kg for young leaf and 13.5 to 77.5mg/kg for stem, respectively. The contents of Al and F were closed to those of the tea leaf in East China (Xie et al., 2001a; 2001b). Even though there were great distinctions in the concentrations of Al and F for different tea gardens, the distribution pattern of Al in the tea plants was the same as that of F for all tea gardens, i.e. $Al(F)_{old\ leaf} > Al(F)_{young\ leaf} > Al(F)_{stem}$.

Table 2 Contents of Al and F in tea plants (mg/kg)

Tea garden	Al			F		
	Old leaf	Young leaf	Stem	Old leaf	Young leaf	Stem
SC	7976	2681	525	1504	602	77
CQ1	7448	1526	367	926	163	57
CQ2	6953	1489	420	1209	313	73
CQ3	5944	535	327	443	134	71
YN1	1196	682	349	299	88	26
YN2	2053	564	338	221	49	19
HN1	5395	1067	285	410	145	56
HN2	7008	804	376	381	50	22
HN3	2882	737	367	484	163	36
HB1	5507	918	311	576	127	52
HB2	7055	456	347	702	163	13
HB3	5633	370	345	528	65	21

The contents of labile Al in soils are showed in Table 3. The concentrations of various forms of Al varied in different tea gardens and layer soils, ranging from 0.12 to 14.3mg/kg, 43.0 to 623mg/kg, 230 to 599mg/kg and 4.11 to 49.4mg/kg for water-soluble, exchangeable, Fe-Mn oxide and organic, respectively. No unified variation trend of Al content for the same form was found in different soil layers, but the distribution law of labile Al contents for the same tea gardens was $Al_{exchangeable} \approx Al_{Fe\text{-}Mn\ oxide} > Al_{organic} > Al_{water\text{-}soluble}$. The contents of different labile F, however, varied slightly in different soils and different soil layers, though the exchangeable F content was less than that of the rest labile F (Table 4).

Table 3 Concentration of labile forms of Al in soils (mg/kg)

Tea garden	Water-soluble			Exchangeable			Fe-Mn oxide			Organic		
	0–20cm	20–40cm	40–60cm	0–20cm	20–40cm	40–60cm	0–20cm	20–40cm	40–60cm	0–20cm	20–40cm	40–60cm
SC	14.3	12.9	12.3	490	318	85	370	422	292	31.2	25.4	16.9
CQ1	8.1	10.5	12.1	507	393	351	363	327	455	20.1	10.3	5.2
CQ2	13.9	13.7	4.1	452	422	407	345	363	442	33.6	23.2	19.1
CQ3	4.9	3.2	1.5	193	157	117	396	428	406	49.0	39.2	19.0
YN1	1.3	0.3	0.3	135	43	98	424	305	599	34.0	23.9	11.4
YN2	0.1	—	—	43	—	—	534	—	—	32.5	—	—
HN1	1.4	0.7	0.5	370	400	406	413	490	617	27.2	16.6	13.6
HN2	0.9	0.8	0.1	105	197	213	459	543	470	37.8	4.8	3.4
HN3	0.7	0.3	0.2	418	562	380	381	450	420	16.2	16.9	13.6
HB1	2.1	1.9	0.4	300	206	351	361	336	239	33.3	17.3	4.1
HB2	3.7	1.9	0.5	623	415	143	404	396	327	49.4	38.9	33.8
HB3	0.7	1.3	0.6	127	143	318	265	230	259	16.1	13.6	12.5

Note: "—" means that the soil in the layer was not sampled because the sampling site was located in a new tea garden with a very shallow soil layer

Table 4 Concentration of labile forms of F in soils (mg/kg)

Tea garden	Water-soluble			Exchangeable			Fe-Mn oxide			Organic		
	0–20cm	20–40cm	40–60cm	0–20cm	20–40cm	40–60cm	0–20cm	20–40cm	40–60cm	0–20cm	20–40cm	40–60cm
SC	2.8	3.1	2.5	0.9	1.2	0.9	1.5	4.6	3.1	1.0	1.7	1.3
CQ1	1.9	2.5	1.6	0.6	0.5	0.8	1.6	1.2	1.2	1.1	1.0	1.0
CQ2	2.4	1.3	1.2	0.5	0.4	0.4	1.3	1.2	1.2	1.4	1.7	1.9
CQ3	1.5	1.3	1.1	0.7	0.6	0.6	4.6	2.4	1.8	1.8	1.8	1.4
YN1	1.0	1.2	1.0	0.6	0.7	0.6	6.0	5.8	3.0	2.8	3.4	1.7
YN2	1.3	—	—	0.4	—	—	3.4	—	—	1.7	—	—
HN1	1.2	1.4	1.5	0.8	0.8	1.0	2.6	2.3	2.4	1.5	1.3	1.4
HN2	1.4	1.7	0.9	1.5	1.3	1.3	5.9	13.7	5.3	3.3	4.9	4.0
HN3	1.6	1.1	0.8	0.9	1.1	1.3	3.5	2.8	3.1	2.6	1.8	1.6
HB1	2.6	1.6	1.3	2.1	1.2	1.2	6.0	5.6	3.4	4.8	3.9	2.5
HB2	1.5	1.0	1.8	1.1	0.9	0.5	2.3	2.2	1.5	1.9	1.7	1.5
HB3	1.0	1.1	1.1	0.7	0.9	1.1	3.2	3.2	3.1	1.8	1.9	1.8

Note: "—" means that the soil in the layer was not sampled because the sampling site was located in a new tea garden with a very shallow soil layer

The correlation of the contents of Al and F in tea plants with the water-soluble Al and F in soils were showed in Table 5. The correlation between the concentrations of Al in the tea plants and the amounts of water-soluble Al in different soil layers were significant at P=0.01 (0–20cm), P=0.05 (20–40cm) and P=0.1 (40–60 cm) for old leaf, at P=0.01 (all the soil layers) for young leaf, at P=0.01 (0–20cm and 20–40cm) and P=0.05 (40–60cm) for stem. The significant correlation between the concentrations of F in the tea plants and the amounts of water-soluble F in different soil layers was also obtained (Table 5). The rest labile forms of Al and F in soils, however, were not related significantly with the contents of Al or F in tea plants. The results were in agreement with that reported by LU et al. (2006) and Xie et al. (2001a; 2001b), which further confirmed that the concentrations of Al and F in tea plants could be predicated by the water-soluble Al or F in soils.

Table 5 Correlation of contents of Al and F in tea plants with water-soluble Al and F in soils

Soil profile (cm)	Al			F		
	Old leaf	Young leaf	Stem	Old leaf	Young leaf	Stem
0–20	0.6208**	0.8279***	0.7785***	0.7743***	0.7367***	0.6068**
20–40	0.6019**	0.8192***	0.7388***	0.6762*	0.6841**	0.5606*
40–60	0.5438*	0.8503***	0.6900**	0.7437***	0.7863***	0.7200***

Notes: *significant at P=0.1; **significant at P=0.05; ***significant at P=0.01

3.3 Correlation between Al and F in tea plants and soils

The relationships between Al and F in old leaf, young leaf, stem and in the whole tea plants are shown in Fig. 1 and between water-soluble Al and F in different soil layers and in the whole soil layer are shown in Fig. 2. There were significantly and positively correlations between Al and F in old tea leaf ($P<0.05$) and young leaf ($P<0.01$) while insignificantly in the stem ($P>0.1$) (Fig. 1). The correlation of Al with F in whole tea plant, however, was strongly significant (r=0.8763, $P<0.01$, n=36). There was evidently tendency that water-soluble Al concentrations increased with the increase of water-soluble F concentration in different soil layers (Fig. 2). The correlations between Al and F were significant at $P<0.01$ (r=0.7305) for 0–20cm soil layer and $P<0.05$ for 20–40cm (r=0.6663) and 40–60cm (r=0.7168), respectively. Furthermore, the correlation of water-soluble Al with water-soluble F in 0–60cm soil layer was extremely significant (r=0.7029, $P<0.01$, n=34). Nevertheless, the same relationships between Al and F for the other forms were not observed in the soils.

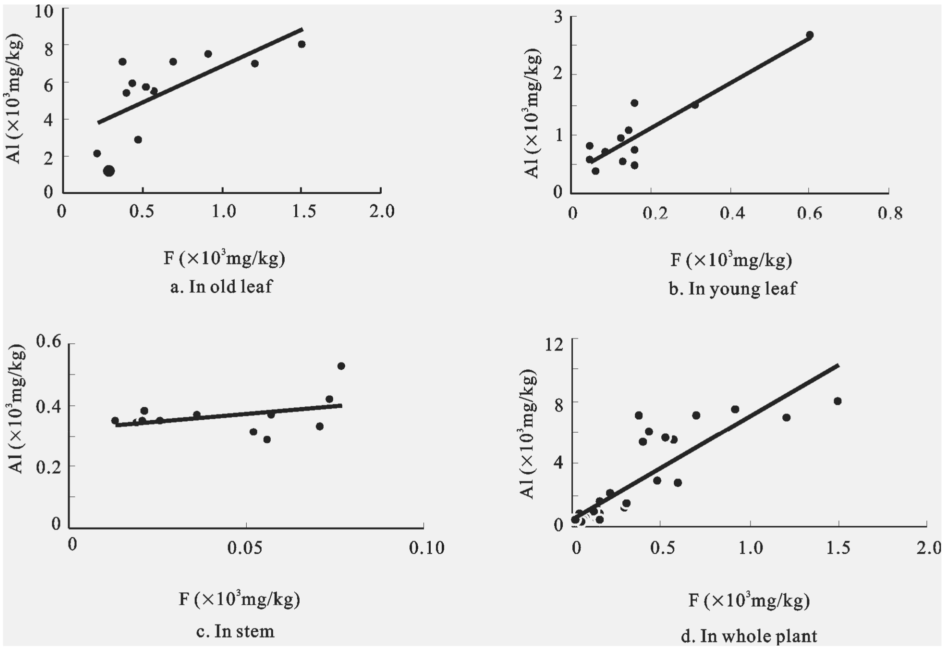

Fig. 1 Correlations between Al and F contents in tea plants

Aluminum ion (Al^{3+}) can be complexed with fluoride ion (F^-) in the forms of AlF^{2+}, AlF_2^+, AlF_4^-, and AlF_3^0 AlF_3^0 and Al^{3+} (McLean, 1976; Wenzel and Blum, 1992; Xie et al., 1999). The relationships between Al and F in tea leaf and in soil solution may demonstrate the joint uptake of Al and F from soil into tea plant to some extent since the existing of Al-F complexes in soil solution (Ding and Huang, 1991; Fung et al., 1999). Xie Z M et al. (2001) also obtained the same result and suggested that the uptake of Al and F of tea leaf were in the form of Al-F complexes. Nagata et al. (1993) found that the form of Al-F in the tea branches was the same as those in the solutions. They observed that Al was in the forms of catechin and F complexes using ^{27}Al and ^{19}F NMR (Nuclear Magnetic Resonance Spectroscopy). The similar relationships between Al and F in other plants were also obtained by Arnesen (1997) and Stevens et al. (1997). But an inconsistent result was reported by Ruan et al. (2003) that the concentrations of F in tea leaf significantly increased with increasing of Al in the uptake solution and soil and adversely the concentrations of Al in tea leaf in solution and soil experiments were not affected by F and Al treatments. This may illustrate that

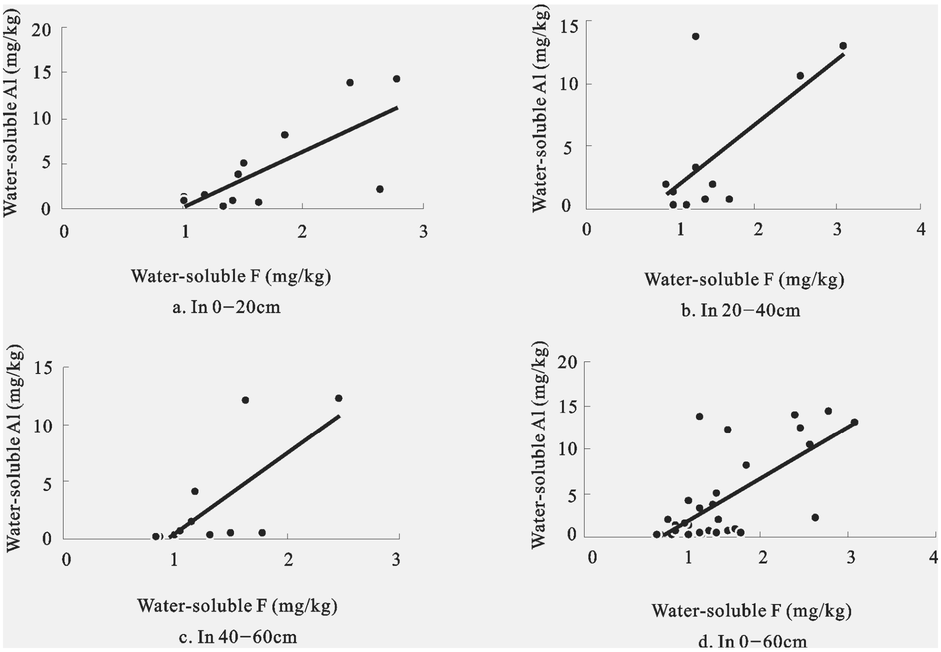

Fig. 2 Correlations between water-soluble Al and F in soil of tea garden

the content of F was more influenced by the complexes of Al-F than that of Al in tea leaves.

4 Conclusions

The soil pH increased and the organic matter content of soils decreased with the depth of soil layers for all the tea gardens. There were not obvious changing trend for CEC in different soil layers of the same tea garden. The contents of Al and F in the tea plants varied obviously for different tea gardens.

The concentrations of labile Al varied obviously in soils of different tea gardens, but the distribution law of labile Al for the same tea garden was $Al_{exchangeable} \approx Al_{Fe\text{-}Mn\ oxide} > Al_{organic} > Al_{water\text{-}soluble}$. The contents of labile F varied slightly in soils of different tea gardens and different soil layers, though the exchangeable F content was lowest among the labile forms of F in soil. The concentrations of Al and F in tea plants increased with increasing of water-soluble Al or F.

The correlations between Al and F in tea leaf were more significant than that in stem, and the correlation of Al with F in whole tea plant was strongly significant. Al concentration evidently increased with the increase of F concentration in different soil layers. The correlation of water-soluble Al with water-soluble F in all soils was also strongly significant. This indicated that there may be cooperation to some extent in the uptake of Al and F from soil into tea plant in tea gardens.

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