APPLICATION OF GIS AND GEOSTATISTICS TO CHARACTERIZE SPATIAL VARIATION OF SOIL FLUORIDE ON HANG-JIA-HU PLAIN, CHINA

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Abstract: Spatial variability of soil fluoride in the plough layer（0~20cm）of paddy soil from Hang-Jia-Hu Plain of Zhejiang Province in China was studied using geostatistical analysis and GIS technique. The results of Semivariograms analysis showed that two forms of soil fluoride were correlated in a given spatial range, and total fluoride (T-F) was controlled by intrinsic factors of parent material, relief and soil type, whereas water-soluble fluoride (Ws-F) was greatly affected by extrinsic factors such as fertilization and soil management. Kriging method was applied to estimate the unobserved points and their distribution maps were obtained, which indicated that the concentrations of soil T-F and Ws-F had a close relationship with parent material, pH value, organic matter, cation exchange capacity content and soil texture. The main contents distribution of T-F and Ws-F were 200~300mg kg^{-1}, 0.5~1.0mg kg^{-1} in the studied area, respectively. And what is more, the range of T-F contents in soil was as low as less than 100 mg kg^{-1} in Yu-hang area accounting for 23.7% area scale. The range of fluoride contents in the soils from central and eastern parts of Hang-Jia-Hu Plain was higher than that from the western part. The accumulation of fluoride contents in soil was lower in the whole studied area , suggesting that local fluoride epidemic such as dental caries due to lack of fluoride should be prevented by using fluoride-containing toothpaste.

Key words: Fluoride, Geostatistics, GIS, Spatial variation, Kriging method

1. INTRODUCTION

Fluoride (F) is regarded as an essential trace element, primarily because of its benefits to dental health and its suggested role in maintaining the integrity of bone (Underwood and Mertz 1987; Wheeler and Fell 1983). A small amount of fluoride is beneficial in the prevention of dental caries. It has also been used to treat osteoporosis (Fung *et al*. 1999). However, excessive fluoride is built up in the apatite ctystals in teeth and bones and reduces their solubility (Fejerskov *et al*. 1994; Fung *et al*. 1999). Traditionally, excessive fluoride has been connected with high intake of fluoride through drinking water and food (Marian *et al*. 1997; Singh and Dass 1993), but water and food take up fluoride from soil and accumulate it in human body finally though food web (Marian *et al*. 1997), unbalance of fluoride in the human body can cause diseases of teeth and bones (Fung *et al*. 1999; Xie *et al*. 2001). Therefore, increasing attention should be paid to soil fluoride quality.

In recent years, geostatistics has been proved as a successful method to study distributions of soil heavy metals (Atteia *et al*. 1994; Steiger *et al*. 1996; White *et al*. 1997; Yu *et al*. 2001; Romic and Romic 2003) and soil nutrient (Tsegaye and Robert 1998; Fisher *et al*. 1998; Cahn *et al*. 1994). However, most of the previous geostatistical studies were focused on data at small scale (Wang 1999; Goovaerts 1999). With the development of Geographical Information System (GIS), GIS can integrate attribute data with geographical data of system variables, which makes the application of geostatistics technique for large spatial scale more convenient (Steiger 1996; Bai *et al*. 1999; Mendonca Santos *et al*. 2000). Geostatistics and GIS are becoming indispensable in characterizing and summarizing spatial information in large regions to provide quantitative support to decision and policy making for soil, agricultural and natural resources management (Wang 1999; Guo *et al*. 2000; Liu *et al*. 2003).

However, the papers on soil fluoride was less relatively, and that according to local fluoride epidemic has evolved in response to high soil concentrations of fluoride in contaminated sites (Horner and Bell 1995), so previous study had stressed on the fluoride contents in contaminated soil too and there is minimum information on spatial distributions of soil fluoride in paddy fields, and less information in a large scale (Geeson *et al*. 1998; Li *et al*. 2004). In this paper, we applied geostatistics combined with GIS to (1) analyze the spatial dependency and explain the variation mechanism of soil fluoride in the paddy soils; (2) map the spatial distribution of soil fluoride in the soil; (3) provide information for environmental monitoring and evaluation in Hang-Jia-Hu Plain.

2. MATERIALS AND METHODS

2.1 Study area

Hang-Jia-Hu Plain is in the center of Hangzhou-Jiaxing-Huzhou in the North of Zhejiang Province in the southeast of China, including Jiaxing, Pinghu, Tongxiang, Haining, Jiashan, Haiyan, Hangzhou, Yuhang, Deqing, Changxing, Anji, Huzhou and part of Lin'an, 13 regions altogether. It boders the Hangzhou Gulf, a part of the East China Sea (Fig. 1). It is an coastal and lacustrine alluvial plain with an altitude of 3~7m above sea level. The climate of the area is subtropical humid monsoonal climate and has abundant rain capacity, the average rang of temperature and rainfall density are 16~19 ℃ and 1200~1300mm every year, respectively. It is densely dotted with drainage ditches that form a network waterway. Rice (Oryza Satiya) has been dominant crop in the studied area, a large part of the area has acidic paddy soil.

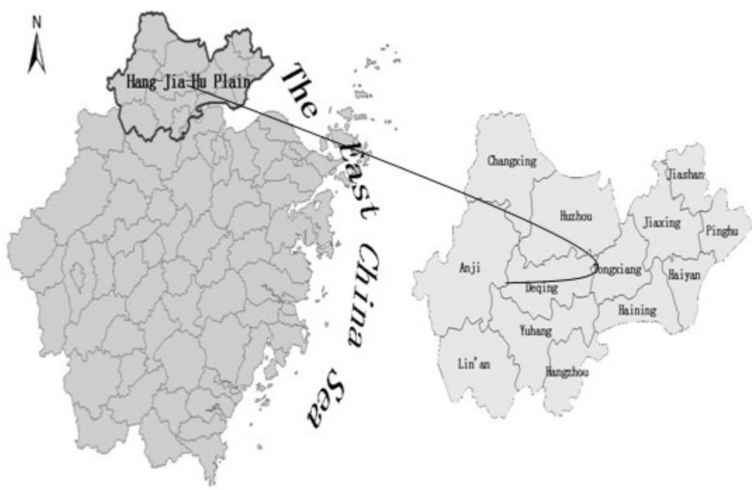

Fig. 1. Location of the study area

2.2 Soil sampling and analysis

Soil samples were taken from over 460 locations within Hang-Jia-Hu Plain in April 2000. Sampling points are presented in Fig. 2. Because there are more low mountains and hills in Anji, Lin'an and Deqing region in the western of Hang-Jia-Hu Plain, the sampling points were sparse comparatively.

Fig. 2. Distribution of sampling sites in Hang-Jia-Hu Plain

Some characteristics of the soils are presented in Table 1. Pipette method was used to determine the particle composition according to the International System. Soil pH value, organic matter (OM) and cation exchange capacity (CEC) were tested according to the conventional methods (Nanjing Agricultural University 1981).

Table 1. Basic properties of the soil samples in Hang-Jia-Hu Plain

	pH (H_2O)	OM (g kg^{-1})	CEC (cmol kg^{-1})	Particle composition (%)		
				<0.002mm	0.002~0.05mm	0.05~2mm
Range	4.1 ~ 8.3	10.9 ~ 61.4	5.3~ 24.8	4.9 ~ 23.5	36.4 ~ 80.2	5.1 ~ 51.4
Mean	5.8	34.0	14.8	14.2	69.0	16.7
S.D.	0.7	9.1	3.9	2.8	7.0	7.6
CV%	12.0	26.7	26.3	19.9	10.1	45.2

S.D., standard deviation; CV, coefficient of variation

For total fluoride (T-F) analysis, direct determination of total fluoride in samples was made using a NaOH fusion-selective ion electrode technique (Baker 1972; Villa 1979). 0.25 g of sample were passed through a 100-mesh sieve and put into a 50-ml nickel crucible 3.0 ml of 16.75 mol/l NaOH solution, then placed in an oven at 150℃ for 1 h until dry. The crucible with sample was then placed in a Muffle furnace. The temperature was raised to 600℃. The samples were fused after 30 min at this temperature. After the samples had been removed from the muffle furnace and cooled, 5 ml of de-ionised water was added and then heated slightly to facilitate the dissolution of the fused soil with sodium hydroxide. Then, 4 ml of concentrated HCl were added slowly, with stirring, to adjust pH to 8~9 (checked with pH test paper). The cooled dissolved sample was transferred to a 50-ml volumetric flask, diluted with distilled water to volume, and then filtered through dry filter paper. The filtrate was used for the determination of fluoride. A reagent blank was produced.

Water-soluble fluoride (Ws-F) was extracted by ratio of 1:5 soil to water. Ten grams of soil passed through a 60-mesh sieve and 50-ml distilled water were placed in 60-mesh sieve and 50-ml distilled water were placed in polyethylene bottles, shaken for 0.5 h on an end-over-end shaker, then centrifuged. Then fluoride levels were measured by ion-specific electrode potentiometer (Xie *et al.* 2003).

2.3 Data analysis

Distribution of soil fluoride element were characterized using the Kolmogrov-Smirnov (K-S) test for goodness-of-fit (Sokal and Rohlf 1981) to ensure that the distribution could be validly applied to data sets. Descriptive statistics, including the range, mean, standard deviation (SD) and coefficient of variation (CV), were determined for each set of data using the statistical analysis system (SPSS) and correlation analysis was conducted.

Geostatistics were used to estimate and map soils in unsampled areas (Goovaerts 1999). Among the geostatistical techniques, Kriging is a linear interpolation procedure that provides a best linear unbiased estimation for quantities that vary in space. The procedure provides estimates at unsampled sites. Kriging's estimates are calculated as weighted sums of the adjacent sampled concentrations. That is, if data appear to be highly continuous in space, the points closer to those estimated receive higher weights than those farther away (Cressie 1990).

In this study, spatial patterns of soil fluoride element were determined using the geostatistical analysis. Semivariograms were developed to evaluate the degree of spatial continuity of soil fluoride element among data points and to establish a range of spatial dependence for each soil soil fluoride element using GS+3.1 software. Information generated through variogram was used to calculate sample-weighted factors for spatial interpolation by a Kriging procedure (Isaaks and Srivastava 1989) using Arc/Info8.1 and Arcview3.2 software based on GIS technique from ESRI company.

3. RESULTS AND DISCUSION

3.1 Summary statistics

Fig. 3 displays the histograms (a) on the orginal scales and (b) as common logarithms ($\log_{10}$) of T-F and Ws-F. The distributions of T-F and Ws-F had long upper tails, and there are several data that might be considered as outliers. Some transformation was desirable for further analysis. Taking logarithms achieved approximate symmetry (Fig. 3(b)) and allowed a confident comparison of mean value for different forms of land use. It also brought the apparent outliers of T-F and Ws-F within the distributions and showed that they should not be treated as exceptional. The statistical results using the Kolmogrov-Smirnov (K-S) test indicated that the soil T-F was more nearly normally distributed than logarithm transformed, but the distribution of Ws-F remained more strongly peaked than normal (leptokurtic). Taking logarithms for Ws-F brought the skewness to only 0.23 and the kurtosis was –0.14. Clearly, its distribution is close to logarithm normal distribution.

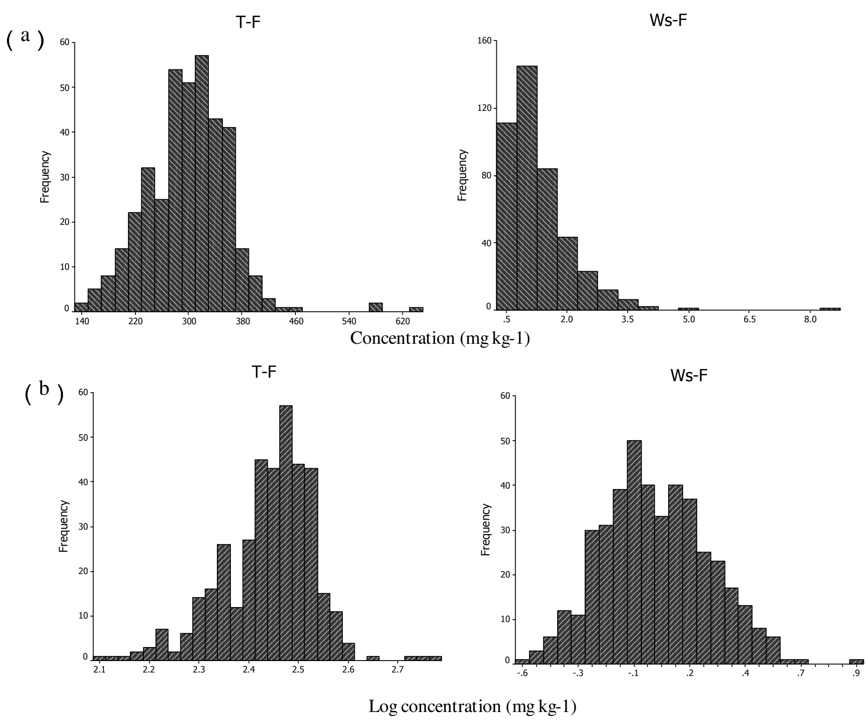

Fig. 3. Histograms (a) on original scales and (b) after transforming to logarithms

3.2 Geostatistical analysis

The concentrations of T-F and Ws-F were transformed to standard normal deviates by Hermite polomomaials, as described above. For T-F and Ws-F all the data were included. Fig. 4 presented the semivariogram and fitted models for fluoride element. The attributes of the semivariograms for soil fluoride were summarized in Table 2.

Table 2. Best-fitted semivariogram models of soil F and corresponding parameters

F Element	Model	C_0	C_0+C_1	Range（km）	$C_0/(C_0+C_1)$	R^2
T-F	Gaussian	2200	9510	632.7	23.1%	0.995
Ws-F	Exponential	0.178	0.357	42.9	49.9%	0.960

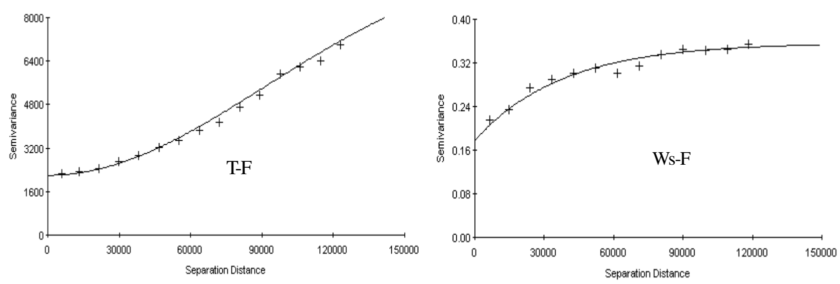

Fig. 4. Experimental semivariograms of soil fluoride element with fitted models

Nugget variance (C_0) represents the experimental error and field variation within the minimum sampling spacing. The Nug/Sill ratio ($C_0/(C_0+C_1)$) can be regarded as a criterion to classify the spatial dependence of soil properties. If the ratio is less than 25%, the variable has strong spatial dependence; between 25% and 75%, the variable has moderate spatial dependence; and greater than 75%, the variable shows only weak spatial dependence (Chien *et al*. 1997). The spatial variability of soil properties may be affected by intrinsic (soil formation factors, such as soil parent materials) and extrinsic factors (soil management practices, such as fertilization). Usually, strong spatial dependence of soil properties can be attributed to intrinsic factors, and weak spatial dependence can be attributed to extrinsic factors (Cambardella *et al*. 1994). Range is the distance over which spatial dependence. Regression coefficient (R^2) provides an indication of how the model fits the variogram data. The higher the regression coefficient, the better the model fits (Hu *et al*. 2004).

The semivariograms results suggested that the semivariagrams of T-F was well fitted for the gaussian model, while semivariagrams for logarithm conversion value of soil Ws-F was well fitted for exponential model. And their Nug/Sill ratios were 23.1% and 49.9%, respectively. The results suggested that T-F had strong spatial dependence, its spatial variabilities were mainly controlled by intrinsic factors such as parent material, relieves and soil types; Ws-F had moderate spatial dependence, mainly controlled by intrinsic factors and extrinsic factors. The ranges for T-F and Ws-F were 632.7km and 42.9km, respectively, indicating T-F in soil was mainly affected by parent material. The R^2 about T-F and Ws-F in soil were both over 0.9, indicating the selective model better fitted.

3.3 Spatial distributions

Fig.5 shows the spatial patterns of T-F and Ws-F in soil generated from their semivariagrams. In order to know easily the distribution of soil T-F and Ws-F in Hang-Jia-Hu Plain, according to a guideline in practical level of soil and the area proportion were analysed (Table 3). To understand the effect of Parent material and soil property on T-F and Ws-F content, main parent materials with corresponding average soil property and soil F contents in 13 regions of Hang-Jia-Hu Plain were listed (Table 4), the correlativity between them and soil properties was analyzed (Table 5 and Table 6).

Fig. 5. Filled contour maps produced by ordinary Kriging of T-F and Ws-F in soil

Table 3. The guideline and the area ratio of T-F and Ws-F produced by ordinary Kriging

T-F			Ws-F		
Guideline	Area (km^2)	Ratio (%)	Guideline	Area (km^2)	Ratio (%)
< 100	1019.0	8.1	< 0.5	213.8	1.7
100-200	1959.0	15.6	0.5-1	6475.5	51.4
200-250	3606.3	28.6	1-1.5	4419.4	35.1
250-300	2902.6	23	1.5-2	1319.5	10.5
300-400	3108.4	24.7	2-2.5	167.1	1.3
> 400			> 2.5		

Table 4. Main parent material and corresponding average soil property and soil F contents in 13 regions of Hang-Jia-Hu Plain

Code	Region	Parent material	pH (H_2O)	OM (g kg^{-1})	CEC (cmol kg^{-1})	Particle composition (%)			T-F	Ws-F
						<0.002mm	0.002~0.05mm	0.05~2mm		
1	Jiaxing	River deposit, lake warp	5.80	36.92	18.65	14.58	70.32	15.19	332.94	1.07
2	Pinghu	lake deposit, offing deposit	5.61	35.06	18.01	14.55	73.07	12.38	329.05	1.33
3	Tongxiang	Fluvio-marine deposit, River deposit	6.32	28.44	15.67	12.85	72.77	14.36	338.51	1.78
4	Haining	River deposit, marine deposit	6.05	24.01	14.04	13.63	73.74	12.64	294.16	1.50
5	Jiashan	River deposit, lake warp	5.72	37.08	18.33	14.61	67.37	18.04	338.92	1.41
6	Haiyan	Ancient lake warp, fluvio-marine deposit	5.52	35.27	15.69	14.32	72.61	13.07	298.07	1.49
7	Hangzhou	Old-river alluvium, shallow-sea alluvium	6.07	32.80	11.83	12.19	68.26	19.55	259.81	1.64
8	Yuhang	Red slope deposit, transported redeposit	5.64	33.83	12.29	14.56	63.98	21.46	255.34	1.08
9	Lin'an	Diluvial alluvium	5.84	33.87	10.22	17.18	56.72	26.13	316.32	0.88
10	Deqing	Lagoonal lake wrap	5.67	34.52	14.41	15.23	69.31	15.45	288.00	0.92
11	Changxing	Lagoonal lake wrap	5.41	31.05	13.24	15.84	65.06	19.14	223.06	0.74
12	Anji	Yellow and red soil redidual deposit	5.19	33.42	9.20	15.81	63.26	20.93	211.97	0.57
13	Huzhou	Lagoonal lake wrap, Fluvio-marine deposit	6.03	40.58	14.63	13.77	70.97	15.26	277.44	1.36

Table 5. Correlation coefficients among pH, organic matter, CEC, clay and F elements

	pH(H_2O)	OM (g kg^{-1})	CEC (cmol kg^{-1})	Particle composition (%) <0.002mm	0.002~0.05mm	0.05~2mm	T-F (mg kg^{-1})	Ws-F (mg kg^{-1})
pH	1							
OM	0.019	1						
CEC	0.113^{*}	0.388^{**}	1					
<0.002mm	-0.140^{**}	0.359^{**}	0.359	1				
0.002~0.05mm	0.142^{**}	-0.142^{**}	0.241	0.023	1			
0.05~2mm	-0.079	-0.003	-0.355	-0.396^{**}	-0.927^{**}	1		
T-F	0.193^{**}	0.186^{**}	0.580	0.274^{**}	0.054	-0.160^{**}	1	
Ws-F	0.612^{**}	-0.050	0.070	-0.234^{**}	0.159^{**}	-0.060	0.169^{**}	1

$^{*}p$ <0.05; $^{**}p$ <0.01

Table 6. Stepwise regression of F elements in tested soil in Hang-Jia-Hu Plain

Regression equation of T-F and Ws-F	Parameters of regression equation		
	Multiple correlation coefficient	F value	Significant level
$Y_{T\text{-}F} = 26.584 + 19.253\ X_1 - 6.473X_2 + 8.372\ X_3 + 3.989\ X_4$	0.661	52.37	$p < 0.05$
$Y_{Ws\text{-}F} = -2.551 + 0.661\ X_1 - 0.049X_4 + 0.010\ X_5$	0.628	86.26	$p < 0.05$

X_1, pH value; X_2, organic matter; X_3, CEC content; X_4, clay fraction (<0.002mm); X_5, silt fraction (0.002~0.05mm); X6, sand fraction (0.05~2mm)

The content distribution of T-F in the plough layer（0~20cm）of paddy soils in Hang-Jia-Hu Plain was the eastern part > central part > western part. As a whole, however, the concentration of soil T-F in Hang-Jia-Hu Plain was comparatively lower, had 2978 km^2 area accounting for 23.7% in the studied area and was lower than the fluoride content in soil which is 200mg kg^{-1} on average in the world (China Environmental Monitoring General Station, 1990). The main range of T-F content was 200~300mg kg^{-1} accounting for 51.6% in the studied area and lower than the fluoride content in soil which is 478mg kg^{-1} on average in China (China Environmental Monitoring General Station, 1990). The average range of soil T-F concentration in Anji, Deqing, Hangzhou zone was 100~200mg kg^{-1}, especially the range of T-F contents in soil was as low as less than 100 mg kg^{-1} in Yu-hang area. The range of T-F contents in soil in Jiaxing, Jiashan, Pinghu zone and part area of Lin'an was exceed 300mg kg^{-1}. The characteristic distribution of soil T-F was mainly associated with each parent material distribution in the 13 regions of Hang-Jia-Hu Plain, the contents of soil T-F developing from river deposit and lake warp exceeded others, yellow and red soil redidual deposit had the least soil T-F contents. And the distribution rule of soil T-F was as same as pH value, organic matter and clay fraction (<0.002mm), which was in positive correlation with them, and stepwise regression of soil T-F indicating the concentration of soil T-F was mainly affected by pH value and CEC contents. The studied results were the same as the previous studied papers. Although fertilization and irrigation can also add up soil fluoride contents, the less effect on it for the less source.

The content distribution of Ws-F in the plough layer（0~20cm）of paddy soils in Hang-Jia-Hu Plain was the central part > eastern part > western part. The main range of Ws-F content was 0.5~1.0mg kg^{-1} in the whole studied zone accounting for 51.4% and centralized in the western area. The average range of soil Ws-F concentration in part area of Anji and Lin'an was less than the water-soluble fluoride content in uncontaminated soil surface layer which is 0.5mg kg^{-1} on average in the world (China Environmental Monitoring General Station, 1990) and only had 213.8km^2 area, where had the lowest pH value and CEC content in the whole area. The content of Ws-

F was comparatively higher in the central and eastern zone, especially in part area of Tongxiang and Hangzhou, the range of Ws-F contents in soil was 2.0~2.5mg kg^{-1}, where had the highest pH value and the lowest organic matter in the whole zone. Which was mainly connected with different parent material distribution in this 13 region of Hang-Jia-Hu Plain too. And the content distribution rule of Ws-F was as same as that of pH value and CEC content and which was in positive correlation with them, in contrast to that of organic matter and clay fraction (<0.002mm) which was in negative correlation with them, and stepwise regression of soil Ws-F indicating the concentration of soil Ws-F was mainly affected by pH value.

The results of this study confirm previous reports indicating that the fluoride concentration has connected with a number of factors such as soil type, parent material, pH value and farm management. Further research should be carried out to study the relationship between the fluoride level of the soil and the fluoride content of agricultural products, et al., and, likewise, the relationship between the content of the water and food and the bioavailability of soil fluoride should be elucidated.

4. CONCLUSIONS

1. The distribution of T-F contents in the plough layer（0~20cm）of paddy soils was normally distributed, and Ws-F was fitted for logarithm normal distribution from Hang-Jia-Hu Plain in China. The semivariagrams of T-F was well fitted for the gaussian model, while semivariagrams for logarithm conversion value of soil Ws-F was well fitted for exponential model, respectively. And their Nug/Sill ratios were 23.1% and 49.9%, respectively. The results suggested that T-F had strong spatial dependence, its spatial variability was mainly controlled by intrinsic factors such as parent material, relives and soil types; Ws-F had moderate spatial dependence, mainly controlled by intrinsic factors and extrinsic factors. The order of range was T-F > Ws-F, indicating T-F in soil had correlation among the whole area and which was mainly affected by parent material.

2. Spatial distribution of the main contents of soil T-F and Ws-F were among 200~300mg kg^{-1} and 0.5~1.0mg kg^{-1}, respectively in the studied area. And what is more, the range of T-F contents in soil was as low as less than 100 mg kg^{-1} in Yu-hang area accounting for 23.7% area scale. The range of fluoride contents in the soils from central and eastern parts of Hang-Jia-Hu Plain was higher than that from the western part. The

soil fluoride concentrations had a close relationship with soil parent material, pH value, organic matter, CEC and soil texture.

3. The accumulation of fluoride contents in soil was lower in the whole studied area , suggesting that local fluoride epidemic such as dental caries due to lack of fluoride will occur easily in Hang-Jia-Hu Plain in China, so where should be prevented by using fluoride-containing toothpaste and other effective measure.

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