

Relationships between humic substance-bound mercury contents and soil properties in subtropical zone

YU Gui-fen^{1*}, WU Hong-tao², JIANG Xin¹, HE Wen-xiang¹, QING Chang-le²

(1. Institute of Soil Science, Chinese Academy of Sciences, Nanjing 210008, China. E-mail: gfyu@issas.ac.cn; 2. College of Environmental Sciences, Southwest Agricultural University, Chongqing 400716, China)

Abstract: The bioavailability of humic substance-bound mercury (HS-Hg) has been established, while the distribution of HS-Hg in soils in relation to soil properties remains obscure. Path analysis and principal component analysis were employed in present study to investigate how soil factors influence the contents of HS-Hg in soils. Results showed that HS-Hg ranged from 0.0192 to 0.2051 mg/kg in soils. The two fractions existed in soils as humic acid-bound mercury (HA-Hg) > fulvic acid-bound mercury (FA-Hg) and the ratio of HA-Hg/FA-Hg was 1.61 on the average. Soil organic carbon (OC) and HS favorably determined soil HS-Hg and the two fractions. The mercury source forming HS-Hg derived from soil total mercury and HS-Hg. FA-Hg and HA-Hg served as mercury source for each other. In acidic soils, FA-Hg and HA-Hg consistently rose with the increase of OC, and generally HA-Hg increased more dramatically. Soils with lower pH and lighter texture contained more HS-Hg, particularly fraction of FA-Hg. Among all influencing factors, organic material source showed the strongest effect, followed by other soil properties and soil mercury source.

Keywords: humic acid-bound Hg; fulvic acid-bound Hg; soil properties

Introduction

Mercury (Hg) as a global environmental contaminant has been recognized to represent both a potential and actual threat to human health (WHO, 1976), since its bioaccumulation was established in 1953 when the Minamata disease broke out (Fujiki and Tajima, 1992). Mercury occurs usually in the environment in various species including inorganic and organic Hg (Morita *et al.*, 1998). Speciation determines toxicity, transport pathways and residue time of the metal in different compartments of the environment (Lindqvist *et al.*, 1991; Claudia, 1996). Species and the subsequent availability of Hg depend on some ambient parameters, of which humic substance is the key factor (Sjöblom *et al.*, 2000).

Due to specific coordination chemistry, Hg has high affinity for humic substances including fulvic acid (FA) and humic acid (HA) which bear a large number of functional moieties such as carboxylic, phenolic and alcoholic groups. The binding between Hg and humic substance is so strong that the speciation and contents of Hg are largely dominated by humic complexes (Hintelmann *et al.*, 1997; Meili *et al.*, 1997). The free aqueous Hg could be immobilized by humic substance (Driscoll *et al.*, 1995; Watras *et al.*, 1998; Sjöblom *et al.*, 2000; Navas and Machin, 2002). Hg has been suggested to bind humic substance exclusively via S-containing ligands through inner- as well as outer-sphere complexation (Kinniburgh *et al.*, 1996; Wallschläger *et al.*, 1998). In this sense, the distribution, transport and fate of Hg largely rely on environmental humic substance or organic carbon (OC) contents either in aquatic system

or in soils (Mierle and Ingram, 1991; Melamed *et al.*, 2000; Wu *et al.*, 2001; Navas and Machin, 2002). Humic substance-bound Hg (HS-Hg) especially fraction of fulvic acid-bound Hg (FA-Hg) in the runoff water leached from forest soil was the important Hg source to fish in the forest lakes remote from local sources of Hg pollution in Canada, the United States and Sweden (Hånkanson *et al.*, 1988; Lathrop *et al.*, 1989). Hg in soil is one of the most important sources of Hg in terrestrial system for the food chain (Kabata-Pendias and Pendias, 1984; Qing and Mou, 1993). In soils, strong correlation between soil OM and Hg have been reported (Roulet and Lucotte, 1995). Water soluble and exchangeable Hg is apparently available Hg among all Hg species. Furthermore, the mobility and phyto-availability of HS-Hg have also been demonstrated by pot experiment (Yu *et al.*, 2004).

Although abundant information is available on the interaction of humic substance and Hg and the influence of humic substance on Hg mobility and toxicity, few of the studies provided the direct evidence of HS-Hg bioavailability in soils for terrestrial plant. None of the studies reported the relationship between HS-Hg and soils properties; this relationship, thus, merits clarification.

Simple correlation and multiple regression are usually employed to study the relationship between two variables. However, both methods have their own disadvantages which impede the proper estimation of the effect of a single factor. On the contrast, path analysis could partition correlation into direct and indirect effect and distinguish between correlation and causation (Afifi and Clark, 1984). It has thus been

used extensively in agronomic studies (Basta *et al.*, 1993; Krishnasamy and Mathan, 2001; Garcia del Moral *et al.*, 2003).

Therefore, the objective of the present study is to investigate how soil properties determine or influence the contents of HS-Hg and the two fractions, i.e., FA-Hg and HA-Hg, by using path analysis and principal component analysis.

1 Materials and methods

1.1 Soils sampled

Fourteen soils were collected from territory of Chongqing in Sichuan Provinces, southwest China. Soils in this study represent the typical soils in subtropical region originated from diverse climate and vegetation. The soils could be classified as five soil orders and sub-classified as eight soil types (Table 1). Soils were sampled from the surface horizon (A horizon or plow horizon), air dried and ground to pass a 0.149-mm and 1-mm sieves, respectively, stored in bottles for use.

1.2 Analytical methods

Soil samples were characterized for pH, cation exchange capacity (CEC) and base saturation percentage (BSP), contents of CaCO₃, organic carbon (OC), and particle size distribution by routine methods. Soil pH was measured in 1:1 soil/H₂O suspension (Li, 1982). Exchangeable cations (Ca²⁺, Mg²⁺, K⁺, and Na⁺) for CEC were extracted with 1 mol/L ammonium acetate solution, buffered to pH 7.0, concentrations being determined by atomic absorption spectrometry (AAS). Exchangeable acidity was determined using 1 mol/L KCl replacing solution and titrated to a phenolphthalein endpoint. BSP was calculated as the percentage of exchangeable cations to total exchangeable ions, i.e., sum of exchangeable cations and acidity (Sparks, 1982). CaCO₃ was determined by chloride acid titration based on the quantity of CO₂ production (Li, 1982). OC was determined by acid dichromate digestion according to Li (1982). Particle size distribution was determined by the hydrometer method (Black, 1965). The obtained basic properties of the soils are listed in Table 1.

Two quality-controlling samples, calcareous

Table 1 Classification and general properties of soils

Soil	Classification		pH ^a	CaCO ₃ , g/kg	OC ^b , g/kg	CEC ^c , cmol/kg	BSP ^d , %	Particle size distribution ^e , %		
	Soil order	Soil type						Clay	Silt	Sand
Humidified soil	Inceptisol	Conifer soil	4.05	0	61.00	43.93	38.8	13	32	55
Meadow soil	Inceptisol	Meadow soil	4.85	0	31.80	40.82	41.4	35	38	27
Black calcareous soil	Mollisol	Calcareous soil	7.73	12.4	74.40	38.52	95.5	16	26	58
Bushy yellow brown soil	Alfisol	Yellow brown soil	4.04	0	144.00	45.57	20.7	11	38	51
Yellow brown soil	Alfisol	Yellow brown soil	4.73	0	76.20	42.72	35.4	25	32	43
Fiery stone yellow mud	Ultisol	Yellow soil	8.13	22.5	15.00	29.05	99.2	26	46	28
Cold sand yellow mud	Ultisol	Yellow soil	4.44	0	10.20	18.80	24.4	21	22	57
Calcareous yellow soil	Ultisol	Yellow soil	7.96	26.0	8.78	29.16	94.7	36	36	28
Red soil	Ultisol	Red soil	4.62	0	7.85	18.75	10.3	45	34	21
Fiery-red brown purple mud	Entisol	Purple soil	4.65	0	9.30	21.62	8.5	47	32	21
Neutral purple soil	Entisol	Purple soil	6.61	5.4	9.19	27.38	70.3	18	30	52
Coarse dark purple mud	Entisol	Purple soil	7.81	6.0	8.49	33.30	95.8	36	24	40
Red brown purple mud	Entisol	Purple soil	7.85	89.3	5.02	30.52	100.0	16	36	48
Alluvial soil	Entisol	Alluvial soil	7.98	12.3	6.86	30.21	95.1	10	22	68

Notes: ^a Soil pH was measured in 1 : 1 soil/H₂O suspension; ^b organic C; ^c cation exchange capacity; ^d base saturation percentage; ^e clay (<2 μm), silt (2---20 μm), sand (>20 μm)

yellow soil and yellow brown soil at distinct contents of organic carbon, i.e., 11.7 and 70.8 g/kg, respectively, provided by Chongqing Key Lab for Agricultural Resources and Environment, were set up to guarantee the analytical quality of humic substance-bound mercury (HS-Hg). Based on the background value, precision and accuracy (recovery) of the quality-controlling samples, the analytical quality of all the samples were monitored for the contents of organic carbon, humic substance (HS) and

its two fractions, i.e., humic acid (HA) and fulvic acid (FA), HS-Hg and its two fractions, i.e., humic acid-bound mercury (HA-Hg) and fulvic acid-bound mercury (FA-Hg) by the method of Qing (1986). The data of quality control were obtained by the following procedure: the background value, content of background plus standard and recovery were collected by at least 24 times of batch-wise repetitive determination; the mean value (μ) and standard deviation (SD) were calculated after the abnormal

values were deleted in terms of reliability (Chen and Liu, 1998); $\mu \pm 2SD$ was set as alert lines and $\mu \pm 3SD$ as control lines.

HS and the two fractions HA and FA were analyzed by the classical fractionation procedure for humic substance including extraction, acidification, separation and purification (Stevenson, 1994). HS-Hg was determined by extraction of humic substance, followed by determination of Hg. HA-Hg and FA-Hg were separated for HA and FA using the method of Stevenson (1994), followed by determination of Hg. Soil total Hg was extracted by acid digestion with $H_2SO_4-HNO_3-KMnO_4$. All extracted Hg was determined by YYG-2 cold vapor atomic fluorescence detector (CVAFS, Sweden).

1.3 Statistical analyses

Statistical analyses were performed using software package SPSS 10.0. The effects of soil properties on the contents of HS-Hg was evaluated by statistical technique of path analysis (Chen and Liu, 1998), through which both direct and indirect effects of various soil properties parameters could be achieved, indicating the relative strength of causal relationships (Loehlin, 1987). Direct effects are referred to as direct path coefficients and are standardized partial regression coefficients (Basta *et al.*, 1993). Principal component analysis (PCA) was also utilized to screen out the principal soil influencing factors (Pei, 1990). PCA transforms a number of (possibly) correlated variables into a (smaller) number of uncorrelated variables called principal components. The first principal component accounts for as much of the variability in the data as possible, and each succeeding component accounts for as much of the remaining variability as possible.

2 Results and discussion

2.1 Contents of humic substance-bound mercury in soils

The contents of HS-Hg ranged from 0.0192 to 0.2051 $\mu\text{g/g}$ (data not shown), accounting for 15.4% to 83.1% of soil total mercury, which is by far higher than that of free ionic Hg as reported (Wu *et al.*, 2001), and proven to be available to plants (Yu *et al.*, 2004). In this sense, HS-Hg was a very important integral part of active Hg in soils and not negligible. Generally, content of humic acid-bound mercury (HA-Hg) was higher than that of fulvic acid-bound mercury (FA-Hg) with the mean ratio of HA-Hg to FA-Hg of 1.61 (data not shown). The contents of HS-Hg and the two fractions followed the sequence of Inceptisol > Mollisol > Alfisol > Ultisol > Entisol in terms of soil order, which suggested that not only the natural soil-forming process but also anthropogenic activities influence the formation of HS-Hg.

2.2 Relationships between humic substance-bound mercury and soil properties

HS-Hg derives from soil organic carbon and Hg in soils. Therefore, soil humic substance and the two fractions, FA and HA, as well as total Hg, serving as material sources were also quantified in this study. The organic carbon, humic substance (measured by carbon) and total Hg contents were tested to be 33.3 g/kg, 12.0 g/kg and 0.168 mg/kg, respectively on the average (data not shown).

HS-Hg was significantly correlated with soil organic carbon, humic substance and soil total Hg with the correlation coefficients reaching 0.675, 0.726 and 0.687, respectively (Table 2). In present study, the variation coefficients, calculated via dividing the standard deviation by the mean value of the samples, were comparably high for organic carbon (1.22) and humic substances (1.17), indicating our samples were diverse in organic matter. However, the variation was not accordingly high for HS-Hg, but relatively low, with variation coefficient only 0.675. This could be attributable to the narrow range of Hg contents of the samples, seen from the lower variation coefficient 0.517. Under contemporary natural surroundings with non-point source pollution, mercury in agricultural soils dominantly originates from atmospheric mercury and precipitation of suspended particulate Hg, resulting in the homogeneous dispersion of mercury in air. Hence, the range of total mercury in soils was not as wide as that of humic substance in soils.

Soil total mercury was clearly mercury source for forming HS-Hg. Besides, the two fractions of HS-Hg, i.e., FA-Hg and HA-Hg served as mercury source for each other, illustrated by correlation coefficients in Table 2.

Besides the effects of carbon and Hg source, the diversity of genetic background arousing from soil properties also influenced the contents of HS-Hg formed. HS-Hg was positively correlated with soil CEC and negatively correlated with soil clay content, with correlation coefficient 0.653 and -0.576, respectively (Table 2). It could be due to the fact that the soil with higher CEC could provide more active adsorption sites for mercury. Clayey soils with high fine particle contain low content of HS-Hg, which was probably because clayey soils usually with high pH is not favorable to the formation of HS.

2.3 Relationship between fulvic acid-bound mercury and soil properties

Among all organic matter sources, only organic carbon (OC) and HS-C contents were significantly correlated with FA-Hg, with correlation coefficient 0.593 and 0.600, respectively (Table 2). Soil total mercury and HS-Hg were positively correlated with FA-Hg, with simple correlation coefficient 0.650 and 0.921, respectively. It suggested that soil mercury

Table 2 Simple correlation coefficients (*R* values) of HS-Hg and the two fractions with soil properties (*n*=14)

R	pH	CaCO ₃ , g/kg	OC, g/kg	T-Hg ^a , mg/kg	CEC, cmol/ kg	BSP, %	Clay, %	Sand, %	HS-C ^b , g/kg	FA-C ^c , g/kg	HA-C ^d , g/kg	HS-Hg, mg/kg	FA-Hg, mg/kg	HA-Hg, mg/kg
pH	1.000	0.496	-0.413	0.025	-0.139	0.959**	-0.124	0.225	-0.452	-0.497	-0.506	-0.391	-0.268	-0.426
CaCO ₃		1.000	-0.252	-0.144	0.093	0.507	-0.206	0.095	-0.289	-0.252	-0.258	-0.334	-0.223	-0.358
OC			1.000	0.235	0.784**	-0.279	-0.452	0.358	0.985**	0.973	0.971	0.675**	0.593*	0.676**
T-Hg				1.000	-0.270	0.139	-0.286	0.249	0.264	0.126	0.167	0.687**	0.650*	0.668**
CEC					1.000	-0.080	-0.473	0.461	0.824**	0.790**	0.787**	0.653*	0.449	0.711**
BSP						1.000	-0.307	0.374	-0.295	-0.364	-0.367	-0.191	-0.117	-0.215
Clay							1.000	-0.894**	-0.453	-0.379	-0.422	-0.576*	-0.625*	-0.525
Sand								1.000	0.383	0.268	0.335	0.504	0.539*	0.463
HS-C									1.000	0.956**	0.963**	0.726**	0.600*	0.746**
FA-C										1.000	0.991**	0.597*	0.481	0.619*
HA-C											1.000	0.633*	0.514	0.655
HS-Hg												1.000	0.921**	0.985**
FA-Hg													1.000	0.839**
HA-Hg														1.000

Notes: * $p < 0.05$, ** $p < 0.01$; ^a total mercury; ^b humic substance carbon; ^c fulvic acid carbon; ^d humic acid carbon

could be bound to FA upon being incorporated with humic substance.

FA-Hg was not significantly correlated with soil pH, for whole soil samples (Table 2). However, if soil samples were analyzed by considering as acidic soil (pH < 6.5) and non acidic soil, i.e., neutral and calcareous soils (pH ≥ 6.5), separately, soil pH showed adverse effect on soil FA-Hg in acidic soil with partial regression coefficient -0.990, on extremely significant level.

FA-Hg was positively correlated with sand content and negatively correlated with clay content, as indicated in correlation coefficients 0.539 and -0.625, respectively (Table 2). Among all soil properties, clay exhibited the highest negative influence, direct effect (-0.586) being higher than indirect effect (-0.153 and 0.114), followed by positive effects of soil total mercury with direct path coefficient 0.533 (Table 3).

Table 3 Path coefficient matrix of soil properties to FA-Hg

Path coefficient	Total mercury→ FA-Hg	BSP→ FA-Hg	Clay→FA-Hg
Total mercury	0.533	-0.052	0.168
BSP	0.074	-0.371	0.180
Clay	-0.153	0.114	-0.586

Note: BSP. Base saturation percentage

For one thing, it implies that clay particles compete for mercury with fulvic acid; for another thing, it demonstrates that soil with lighter texture (lower clay content) facilitates the formation of

FA-Hg. Basically, in sandy soil with very good permeability, bases tend to leach, which promotes the development of soil acidic circumstance and aerobic conditions. Under this condition, FA-Hg thus appreciably produced and accumulated (Stevenson, 1994). In acidic soil with rich organic matter, the more acidic the soil is, the more FA-Hg is.

The effect of base saturation percentage (BSP) primarily manifests on two aspects. BSP posed adverse effect on FA-Hg. The effect was particularly significant on neutral and calcareous soils, with correlation coefficient -0.994 (Equation (1)). On the other hand, FA-Hg and humic acid appear to counteract each other on neutral and calcareous soils.

The relationship between soil properties and FA-Hg on neutral and calcareous soils by using partial correlation analysis could be expressed as the following optimal equation:

$$\text{FA-Hg} = 0.107 + 0.0052 (\text{OC}) - 0.008 (\text{BSP}) - 0.005 (\text{Clay}) - 0.084(\text{HA}) \quad (1)$$

where $R^2 = 0.996$, $R(\text{OC}) = 0.986^{**}$, $R(\text{HA}) = -0.967^*$, $R(\text{BSP}) = -0.994^{**}$, $R(\text{clay}) = -0.984^{**}$.

It indicated that OC and HA had positive and negative effect on FA-Hg, respectively. Furthermore, the effect of HA via OC (1.586) was higher than its direct negative effect (-1.066) as shown in Table 4.

Although in acidic soils, HA and FA-Hg had the same changing trend, HA and FA-Hg had relationship of ebb and flow on neutral and calcareous soils, where HA inclined to accumulation but FA decreased. The increase of total OC provided good conditions for the

Table 4 Path coefficient matrix of soil properties to FA-Hg on neutral and calcareous soils

Path coefficient	OC→ FA-Hg	BSP→ FA-Hg	Clay→ FA-Hg	HA→ FA-Hg
OC	1.641	-0.069	0.080	-1.030
BSP	0.190	-0.592	-0.064	-0.136
Clay	-0.315	-0.091	-0.418	0.304
HA	1.586	-0.076	0.119	-1.066

formation of FA-Hg. However, if only HA fraction increased, FA-Hg declined instead, as HA and FA showed antagonistic effect. Therefore, the relationship between HA and FA was not regular coexistence and accordance but dependent on the actual conditions. The intrinsic mechanism deserves to be studied.

The optimal equation could be developed by partial regression analysis on FA-Hg and soil properties, taking the whole soils into consideration.

$$\text{FA-Hg} = 0.0413 + 0.1008(\text{Total Hg}) - 0.0016(\text{BSP}) - 0.00077(\text{Clay}) \quad (2)$$

where $R^2 = 0.756$, $n = 14$. $R(\text{Total Hg}) = 0.719^{**}$, $R(\text{clay}) = -0.737^{**}$, $R(\text{BSP}) = -0.581^*$.

Among all the significant influencing factors, total mercury showed the most significant positive effect, followed by the negative effect of clay, finally the negative effect of BSP.

2.4 Relationships between humic acid-bound mercury and soil properties

HA-Hg was positively correlated with organic carbon, humic substance and the two fractions FA and HA, with correlation coefficients 0.676, 0.746, 0.619 and 0.655, respectively (Table 2). HA-Hg was positively correlated with soil total mercury, HS-Hg and FA-Hg, revealing that all types of mercury resources was in favor for the formation of HA-Hg.

As for the influence of soil properties, soil CEC had positive effect on HA-Hg. Soil pH, as the strongest influencing factor (-1.668) for the formation of HA-Hg (Table 5), showed negative effect with partial regression coefficient -0.765. It lied on the fact that soils with low pH were usually Alfisols, where the acidity was derived from acidic humic substance and organic matter, FA and HA were abundant, promoting the formation of HA-Hg. BSP positively affected HA-Hg with partial regression coefficient 0.719 and showed the highest direct positive effect (1.399). Soils with high BSP theoretically had relatively high calcium and magnesium ions which is good for HA to be stored in soils, HA-Hg thus accumulated.

Taking neutral and calcareous soils separately into consideration, the optimal equation of soil properties to HA-Hg could be expressed as follows:

$$\text{HA-Hg} = 0.0541 + 0.0261(\text{OC}) - 0.4494(\text{FA}) \quad (3)$$

where $R^2 = 0.933$. HA-Hg was positively correlated with OC and negatively correlated with FA with partial regression coefficient 0.921 and -0.896, respectively.

Table 5 Path coefficient of soil properties to HA-Hg Path coefficient

Path coefficient	pH→ HA-Hg	OC→ HA-Hg	Hg→ HA-Hg	BSP→ HA-Hg
pH	-1.668	0.112	0.011	1.342
OC	0.689	0.2709	0.106	-0.390
Hg	-0.042	0.064	0.453	0.194
BSP	-1.600	-0.076	0.063	1.399

FA had the highest negative effect on HA-Hg as shown in the direct path coefficient -3.998 (Table 6), indicating HA-Hg had relationship of ebb and flow with FA on neutral and calcareous soils, which was similar to the relationship between FA-Hg and HA.

Soil CEC posed significant favorable effect on HA-Hg with simple correlation coefficient 0.711 (Table 2). However, CEC did not show any drastic effect on FA-Hg, indicating that the positive effect of CEC on HS-Hg was achieved via the effect on fraction HA-Hg.

Table 6 Path coefficient matrix of soil properties to HA-Hg on neutral and calcareous soils

Path coefficient	OC→HA-Hg	Clay→HA-Hg	FA→HA-Hg
OC	4.688	0.051	-3.964
Clay	-0.901	-0.263	0.772
FA	4.648	0.051	-3.998

By using partial regression analysis, the relationship between soil influencing factors and HA-Hg could be:

$$\text{HA-Hg} = 0.139 - 0.0352(\text{pH}) + 0.1906(\text{Total Hg}) + 0.0014(\text{BSP}) + 0.0278(\text{CEC}) \quad (4)$$

where $R^2 = 0.898$, $n = 14$. $R(\text{pH}) = -0.765^{**}$, $R(\text{Total Hg}) = 0.789^{**}$, $R(\text{BSP}) = 0.719^*$, $R(\text{CEC}) = 0.763^{**}$.

2.5 Principal soil factors influencing the contents of humic substance-bound mercury

Three principals were selected, based on the principle that accumulative contribution rate of eigenvalue should be higher than 85%. The contribution rate of the selected factors reached 85.3% (Table 7), indicating they could represent the predominant information of all factors.

Further analyses showed that the first principal (F1) was negative load arising from soil OC, FA, HA, HA-Hg and FA-Hg (Table 8), representative of humic substance-bound mercury and organic material sources in soils. The second principal (F2) was positive load of soil pH, BSP and sand, when soil HS-Hg and humic substance keep unchanged, and it

represented soil acidity-basicity and texture properties. The third principal (F3) reflected the accumulation of soil mercury when the above mentioned two principals remained unchanged.

Table 7 The eigenvalues from principal component analysis

Principal	Eigenvalue	Contribution rate, %	Accumulative contribution rate, %
F1	5.56	46.5	46.5
F2	2.87	23.9	70.4
F3	1.79	14.9	85.3

Table 8 Normalized eigenvectors from principal component analysis

Factor	F1	F2	F3	F4	F5	F12
pH	0.2090	0.4613	-0.0595	0.1699	0.4201	-0.1484
CaCO ₃	0.1166	0.3365	-0.3822	0.3194	-0.6531	0.1945
OC	-0.3950	-0.0435	-0.1622	0.1226	0.2479	-0.2437
Total Hg	-0.1674	0.1725	0.5189	0.5609	-0.0503	-0.0199
CEC	-0.2987	0.0754	-0.4868	0.2134	-0.1038	-0.3246
BSP	0.1325	0.5191	-0.0432	0.1817	0.3755	0.1501
Clay	0.2548	-0.3974	0.0030	0.3941	0.1991	0.1999
Sand	-0.2149	0.4119	0.0417	-0.5286	0.0291	0.1890
FA	-0.3842	-0.1081	-0.2369	0.1082	0.2147	0.7849
HA	-0.3942	-0.0911	-0.2077	0.0649	0.1803	-0.2376
FA-Hg	-0.3262	0.1348	0.3421	0.0033	-0.2356	0.0606
HA-Hg	-0.3616	0.0384	0.3130	0.1009	-0.0816	-0.0250

by soil condition and soil mercury source. Of material sources, organic material preceded mercury source, which suggest that inborn material humic substance was essential premise, compared to extraneous mercury source.

3 Conclusions

Humic substance-bound mercury (HS-Hg) and the two fractions FA-Hg and HA-Hg in soils were favorably determined by forming sources including soil OC, HS, HA, FA and total mercury. Soil properties including soil pH, texture and CEC posed dominating effect on HS-Hg. In acidic soils, the lower soil pH or the lighter soil texture, the higher soil FA-Hg is. The higher soil CEC was, the higher HS-Hg is, particularly the faction of HA-Hg. Since low soil pH, light texture and high CEC was in favor of the bioavailability of soil HS-Hg (Yu *et al.*, 2004), more attention should be paid on the harm of soil mercury to human body through food chain. In acidic soils, FA-Hg and HA-Hg consistently rose with the increase of OC, and generally HA-Hg increased more drastically.

References:

Afifi A A, Clark V, 1984. Path analysis [M]. In: Computer-aided

To sum up, HS-Hg and its two fractions in soils, was dominantly influenced by material base including OM, HA and FA, less influenced by other soil properties including soil acidity-basicity and texture properties, finally by soil mercury material base. The data confirmed the soil factors influencing the distribution of HS-Hg, in good agreement with the result of path analysis. Moreover, it revealed the primary-secondary relationship of primary influencing factors. Among all the influencing factors, organic material source showed the strongest effect, followed

multivariate analysis. Belmont, CT: Lifetime Learning Publishing Publication, 235—237.

Basta N T, Pantone J D, Tabatabai A M, 1993. Path analysis of heavy metals adsorption by soil [J]. *Agronomy Journal*, 85: 1054—1057.

Black A C, 1965. *Methods of soil analysis, Part 1* [M]. Madison WI: American Society of Agronomy.

Chen Y C, Li Z Q, 1998. *Environmental mathematics analysis* [M]. Chongqing, China: Southwest Normal University Press. 102—105.

Claudia C, 1996. Mercury speciation in contaminated soils by thermal release analysis[J]. *Water Air and Soil Pollution*, 89: 399—416.

Driscoll C T, Blette V, Yan C *et al.*, 1995. The role of dissolved organic carbon in the chemistry and bioavailability of mercury in remote Adirondack lakes [J]. *Water Air and Soil Pollution*, 80: 919—930.

Fujiki M, Tajima S, 1992. The pollution of Minamata Bay by mercury [J]. *Water Science Technology*, 25(11): 133—140.

Garcia del Moral L F, Rharraabti Y, Villages D *et al.*, 2003. Evaluation of grain yield and its components in durum wheat under Mediterranean conditions: An ontogenic approach[J]. *Agronomy Journal*, 95: 266—274.

Häkanson L, Nilsson Å, Andersson T, 1988. Mercury in fish in Swedish lakes[J]. *Environmental Pollution*, 49: 145—162.

Hintelmann H, Welbourn P M, Evans R D, 1997. Measurement of complexation of methylmercury (II) compounds by freshwater humic substances using equilibrium dialysis [J]. *Environ Sci and Technol*, 31: 489—495.

Kabata-Pendias A, Pendias H, 1984. *Trace elements in soils and plants* [M]. Boca, Raton, Florida: CRC Press Inc. 315.

Kinniburgh D G, Milne C J, Benedetti M F *et al.*, 1996. Metal ion binding by humic acid: application of the NICA-Donna model[J].

- Environ Sci and Technol, 30(5): 1687—1698.
- Krishnasamy R, Mathan K K, 2001. Path coefficient analysis of zinc and boron adsorption in soils[J]. *Communications in Soil Science and Plant Analysis*, 32: 465—475.
- Lathrop R C, Noonan K C, Gueuther P M *et al.*, 1989. Mercury levels in walleyes from Wisconsin lakes of different water and sediment chemistry characteristic [R]. Wisconsin Dept of Natural Resources: Madison, WI. Technical Bulletin. 163: 41.
- Li Y K, 1982. Methods of soil and agro-chemistry routine analysis[M]. Beijing: Agriculture Press. 457.
- Lindqvist O, Johnson K, Aastrup M *et al.*, 1991. Mercury in the Swedish environment: recent research on causes, consequences and corrective methods [J]. *Water Air and Soil Pollution*, 55: 1-261.
- Loehlin J C, 1987. Path models in factor, path, and structural analysis [M]. In: *Latent variable models: An introduction to factor, path, and structural analysis*. NJ: E. Erlbaum Assoc., Hillsdale. 1—37.
- Melamed R, Trigueiro F E, Villas Bôas R C, 2000. The effect of humic acid on mercury solubility and complexation [J]. *Applied Organometallic Chemistry*, 14(9): 473 — 476.
- Meili M, 1997. Mercury in lakes and rivers [M]. In: *Mercury and its effect on environment and biology. Metal ions in biological systems*(Siegel A, ed.). New York: Marcel Dekker Inc. Vol. 34. 21—51.
- Mierle G, Ingram R, 1991. The role of humic substances in the mobilization of mercury from watersheds [J]. *Water Air and Soil Pollution*, 56: 349—357.
- Morita M, Yoshinaga J, Edmonds J S, 1998. The determination of mercury species in environmental and biological samples[J]. *Pure Applied Chemistry*, 70: 1585—1615.
- Navas A, Machin J, 2002. Spatial distribution of heavy metals and arsenic in soils of Aragon (northeast Spain): controlling factors and environmental implications [J]. *Applied Geochemistry*, 17: 961—973.
- Pei X D, 1990. Multiple component statistical analysis and application [M]. Beijing: Beijing Agricultural University Press.
- Qing C, 1986. Background contents analysis of 11 elements in Chongqing purple soils and quality control [J]. *Science and Technology of Southwest Agricultural University*, 2(48): 12—26.
- Qing C L, Mou S S, 1993. Preventing Hg transference from soil to terrestrial food chain[J]. *Pedosphere*, 3: 67—73.
- Roulet L, Lucotte M, 1995. Geochemistry of Hg in pristine and flooded ferrallitic soils of a tropical rain forest in French Guiana, South America[J]. *Water Air and Soil Pollution*, 80: 1079—1088.
- Sjöblom Å, Meili M, Sundborn M, 2000. The influence of humic substances on the speciation and bioavailability of dissolved mercury and methylmercury, measured as uptake by *Chaoborus* larvae and loss by volatilization [J]. *The Science of the Total Environment*, 261: 115—124.
- Sparks L D, Page L A, Helmke A P *et al.*, 1982. Methods of soil analysis [M]. 2nd ed. Madison, WI: Agronomy No. 9, American Society of Agronomy.
- Stevenson F J, 1994. Humus chemistry, genesis, composition, reactions [M]. 2nd ed. New York: John Wiley & Sons.
- Wallschläger D, Desai M V M, Spengler M, 1998. How humic substances dominate mercury geochemistry in contaminated floodplain soils and sediments [J]. *Journal of Environmental Quality*, 27(5): 1044—1054.
- Watras C J, Back R C, Halvorsen S *et al.*, 1998. Bioaccumulation of mercury in pelagic freshwater food webs [J]. *The Science of the Total Environment*, 219: 183—208.
- WHO, 1976. Environmental health criteria 1: Mercury [S]. Geneva: World Health Organization, 1—132.
- Wu H T, Yu G F, Qing C L *et al.*, 2001. Relationship between soil properties and different fractions of soil Hg [J]. *Pedosphere*, 11 (3): 257—262.
- Yu G, Wu H, Qing C *et al.*, 2004. Bioavailability of humic substance-bound mercury to lettuce and its relationship with soil properties [J]. *Communications in Soil Science and Plant Analysis*, 35(7/8): 1123—1139.

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