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Differentiation characteristics and source analysis of heavy metals in typical brown soil under different vegetation

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Abstract: Anomalous enrichment of soil elements (especially heavy metals) has aroused popular attention in China. In order to discuss distribution characteristics and analyze sources of elements in brown soil, field investigation and sample collection were carried out under different vegetation (cherry, apple, bamboos and pine) in Qixia, a typical apple production base in China. Element contents, pH, electrical conductivity (EC) and magnetic susceptibility (MS) were tested. Results showed that element concentrations were about roughly 2.48 times as China's background values, while significantly lower than the class ii of National soil Environment Quality Standard (Ni excepted). Meanwhile, vertical distribution and accumulation characteristics of elements in typical brown soil were significantly different under different vegetation. In detail, elements (Zn excepted) of Pine soil accumulated in surface, while they (Cd, Arsenic excepted) increased with depth under other vegetation. Moreover, pH and EC changed like elements, while MS was exactly opposite. It was found that those differences above were mainly caused by human activities (such as improper use of fertilizer, pesticide and inadequate use of organic fertilizer, etc.). Additionally, differences in composition and decomposition rate of vegetation litter also resulted in vertical differentiations of soil elements under different vegetation.

1 Introduction

Recently, with rapid development of urbanization, industrialization and agricultural modernization, human activities are still causing soil element enrichment [19]. These anomalous enrichment of soil elements (especially heavy metals) has aroused scholars' attentions [16, 21], since they are invisible, hysteretic, cumulative, nondegradable and charactered with poor mobility, a long residence time and difficult to restore [9, 20]. Moreover, due to bioconcentration and magnification (Manuel et al. 2016), they not only reduce crop yields and agricultural produce quality, but also endanger human health or cause nuisance events [20], such as Itai-itai Disease etc. Researches show that soil physicochemical properties could be changed with residues decomposition and/or secretion during physiological activities of plants [9, 23]. And there are great differences in physiological activities and residue decomposition of different plant types [9, 10, 23, 26]. These all have important impacts on chemical species, activity, migration and transformation of soil elements [17]. Research also demonstrate that many methods and techniques could be successfully applied for element source analysis [18, 32], such as concentration ratio, enrichment factor, multivariate statistical analysis, chemical speciation analysis, element association, Scanning Proton Microprobe (SPM), isotope tracing technique [19, 29, 31], etc. Especially, element sources could be traced by analyzing relationships of soil magnetic susceptibility and element contents [4, 8]. However, in Jiaodong orchards, affected by extremely fierce agricultural activities, characteristics of element enrichment and vertical differentiation under different vegetation were scarcely reported. Meanwhile, element source analysis still needs to be further modified.

This study aimed to discuss distribution and vertical differentiation characteristics of soil elements, pH, electrical conductivity (EC) and magnetic susceptibility (MS) under different vegetation (such as cherry, apple, bamboos and pine). Meanwhile, element sources were successfully analyzed.

2 Materials and Methods

Qixia, praised as "China's first Apple city", is located in center of Jiaodong Peninsula and northeast of Shandong Province, with an area of 2016 km^2 and population of 66 million. Belonging to warm temperate monsoon type semi humid climate, it enjoys sufficient sunshine and four clear seasons, with annual average temperature of 11.3℃ and annual rainfall of 650 mm or so. The stratums such as Lower Proterozoic Jingshan group, Shangyuan Penglai group, Mesozoic Laiyang group and Cenozoic Quaternary, are all exposed except for Paleozoic. Brown Soil covers nearly 70% of farmland area. And orchards occupy an area of 650 thousand acres, with annual fruit production up to 1 million and 200 thousand tons.

Surface soil and profile samples under well-developed vegetation (Cherry, Apple, Bamboos and Pine) were collected in a typical orchard, south of Phoenix Village, Qixia city, Shandong Province, in September 2016 (Fig.1 and Table 1). At each site, five surface (0-20cm) samples were collected by five-spot-sampling method under the same vegetation. Meanwhile, soil profile was excavated at appropriate place, and three samples at each intervals (0-10/10-20/20-35cm) based on natural soil stratification. Samples were collected according to norms and standards (NY/T 1121.1-2006).

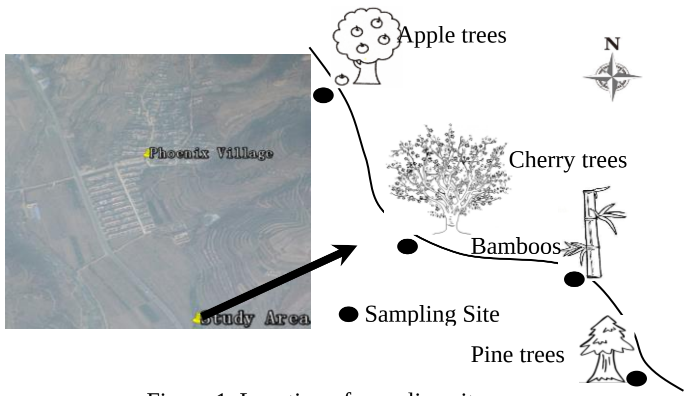

Figure 1. Location of sampling sites

Table 1. Basic information of sampling sites

Sampling	Vegetation	Geography coordinate	
FHZ-1	Cherry trees	N37°14′22.10″	E121°08′27.24″
FHZ-2	Apple trees	N37°14′22.73″	E121°08′27.24″
FHZ-3	Bamboos	N37°14′22.55″	E121°08′27.62″
FHZ-4	Pine trees	N37°14′22.88″	E121°08′28.02″

Samples were air dried and sieved (2 mm), then were ground to fine particles (<0.074 mm) prior for chemical analysis. Prior to element determination, samples were digested using an $HCl+HNO_3+HClO_4+HF$ method. Pb, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As and Cd contents were determined with an inductively coupled plasma mass spectrometer (ICP-MS, Thermo Electron Corporation, Element X Series). Quality assurance and quality control were estimated with the blank and duplicate samples and Certified Reference Materials (GBW07401, GBW07403) approved by the General Administration of Quality Supervision, Inspection and Quarantine of China. The accuracies met the qualification of China Geological Survey (China … 2005). Generally, the relative standard deviations (RSDs) for Certified Reference Materials were less than 3.0%.

Soil pH and EC were analyzed in a suspension of 1:2.5 soil to water ratio (w/v) using pH meter and conductivity meter. The error of pH or EC was less than 2%, using triplicate measurements.

MS was measured at two different frequencies (470 Hz, χ_{LF}; 4700 Hz, χ_{HF}) using a Bartington MS2 dual frequency sensor. Each sample was measured three times in order to check reproducibility and to avoid measurement errors. The error of the susceptibility measurements was less than 3%. Frequency-dependent susceptibility (χ_{FD}) of soils was then calculated and expressed as a percentage: $\chi_{FD}\% = (\chi_{LF} - \chi_{HF})/\chi_{LF} \times 100\%$.

All the statistical analyses were performed by using STATISTICA 6.0 and Micro Excel 2003 for windows. And CorelDraw 9.0 was used to draw the sample map.

3 Results and Discussion

3.1 *element contents in surface soils*

Results showed that surface soil element contents were relatively different under different vegetation (Table 2). Cd and Pb concentrations ranked from high to low as: under Pine, Apple, Cherry and Bamboos. For V, Cr, Mn, Fe, Ni and Zn, contents from high to low followed by under Cherry, Apple, Pine and Bamboos. However, Co concentrations decreased as under Cherry, Pine, Apple and Bamboos. Similar to Co, descending order of Cu and Arsenic contents were under Cherry, Pine, Bamboos and Apple. In general, compared with other vegetation, majority soil elements were higher under Apple, while lower under Bamboos. And the former was 1.14-2.35 times of the latter. Finally, compared with background values in Shandong province and China (Table 2), element concentrations were about 2.48 times higher. Nevertheless, they were lower than class ii of National soil Environment Quality Standard, except for Ni (1.17 times higher).

3.2 *Vertical differentiation of soil element contents*

Fig. 2 showed that contents of Pb, Cr, V, Mn, Fe, Ni, Co and Cu increased with soil depth under Cherry. Meanwhile, Cd concentration increased first and then decreased (lowest in the surface). By contrast, Zn and Arsenic contents decreased first and then increased (highest at the bottom). As to soils under bamboos, concentrations of Cr, V, Mn, Fe, Ni, Cu and Zn increased with profile depth. Simultaneously, Pb, Cd and Arsenic contents raised first and deceased later (lowest in surface). On the contrary, Co level increased after decreased (highest at bottom). For Apple soils, V, Mn, Fe and Ni contents, increased with the increment of soil depth. Moreover, together with Cd and Arsenic (lowest in surface), Zn content rose first and then declined (lowest at bottom). Conversely, content levels of Pb, Cr, Co and Cu decreased and then increased. Finally, Cu and Cd concentrations early dropped and then ascended. In addition, metal contents of Pb, Cr, V, Mn, Fe, Ni, Co and Arsenic increased after decreasing (highest in surface). Although, decreased and then increased, Zn content was highest at bottom. Based on these above, conclusion could be drawn that vertical differentiations of soil elements differed under four vegetation. On the whole, element contents basically showed an increasing trend with profile depth under Cherry and Bamboos, similar to those under Apple except for Cd and Arsenic. Instead, elements under Pine showed an obvious "surface accumulation". Namely, vast majority of element contents basically decreased with profile depth, except for Zn.

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Table 2. Contents of soil heavy metals under different vegetation （mean ± SD, mg kg^{-1} dw）

Sample (N=5)	Pb	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Cd
FHZ-1 (surface)	39.0 ±3.7	204.4 ±15.1	187.6 ±8.7	1182.8 ±71.7	70091.9 ±1059.8	32.5 ±4.0	92.3± 7.2	67.7 ±3.4	183.4± 5.2	23.2± 2.2	0.46± 0.23
FHZ-2 (surface)	40.0 ±4.6	186.5 ±22.5	141.0 ±1.7	1043.9 ±21.5	61159.4 ±33.5	25.7 ±2.2	67.9± 1.5	82.0 ±3.3	165.2± 2.8	29.8± 0.7	0.57± 0.05
FHZ-3 (surface)	35.1 ±3.8	120.6 ±8.1	78.6± 12	719.4± 55.4	41480.4 ±17.9	17.4 ±0.9	41.1± 8.4	35.0 ±1.5	126.7± 1.9	17.7± 0.7	0.39± 0.09
FHZ-4 (surface)	40.2 ±3.9	184.6 ±8.9	132.0 ±0.9	953.9± 20.5	59545.6 ±95.2	27.3 ±3.6	62.8± 2.8	42.3 ±2.9	140.6± 5.0	20.2± 1.8	0.60± 0.21
A layer in Shandong province	25.8	80.1	66.0	644.0	13600	272	25.8	24.0	63.5	9.3	0.08
Background value1 in China	23.6	-	53.9	-	-	-	23.4	20.0	74.2	9.2	0.07
Background value2 in China	24.5	-	64.3	-	-	-	24.4	22.3	6.5	8.9	0.08
A layer in China	26.0	82.4	61.0	583.0	29400	12.7	26.9	22.6	74.2	11.2	0.10
A layer of Brown earth in China	25.1	84.2	64.5	618.0	28900	40.8	26.5	22.4	68.5	10.8	0.09
A layer of Cinnamon in China	21.3	82.6	64.8	633.0	31100	47.5	30.7	24.3	74.1	11.6	0.10
A layer of Grey cinnamon in China	21.2	70.4	65.1	643.0	31800	20.7	36.3	23.6	73.9	11.4	0.14
A layer of Dark brown in China	23.9	75.7	54.9	1109.0	32100	48.5	23.1	17.8	86.0	6.1	0.10
class ii of National soil Environment Quality Standard	300	-	-	-	-		50	100	250	25	0.60

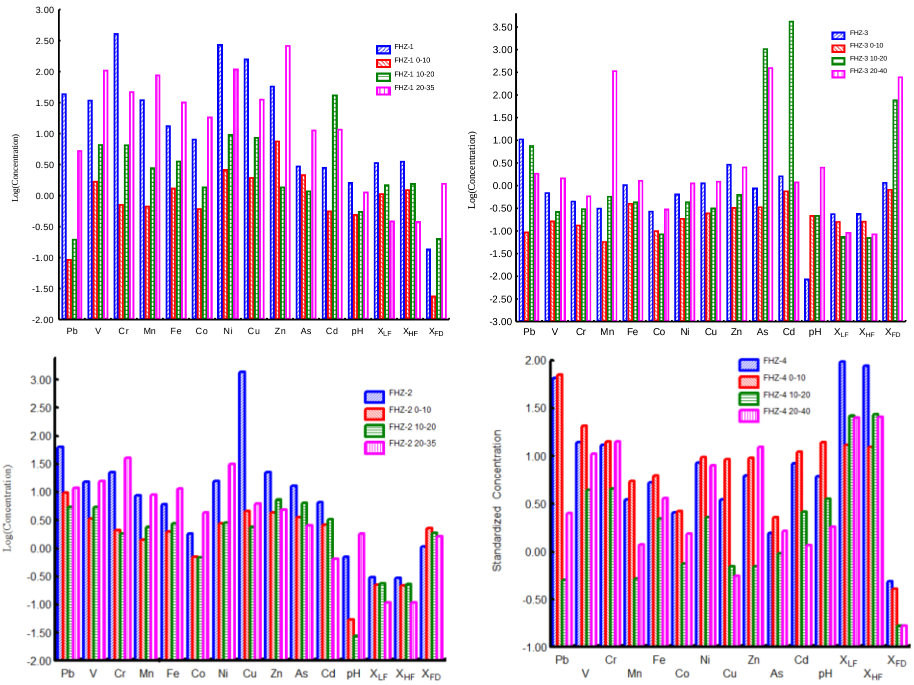

Figure 2. Vertical distributions of soil elements under different vegetation

3.3 *Variation characteristics of soil pH and EC*

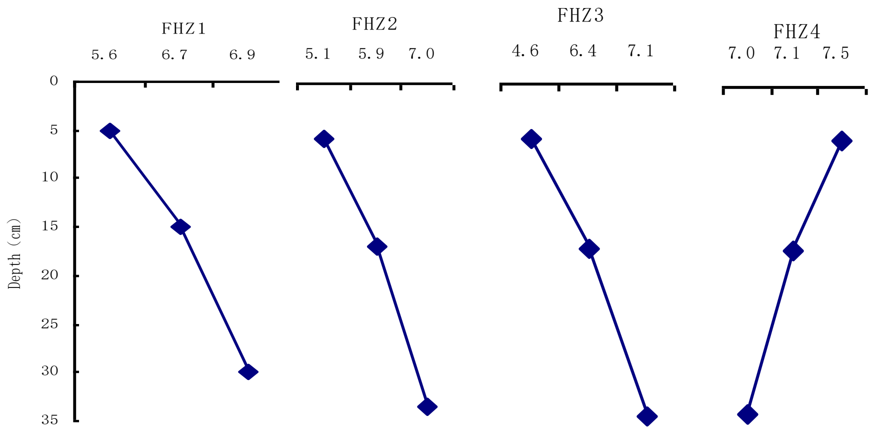

Figure 3. Vertical distribution of soil pH under different vegetation

Figure 3 showed that soil pH increased with profile depth under Cherry, Apple, and Bamboos. Nevertheless, it declined under Pine. Meanwhile, it was showed in Figure 4 that although increased after decreasing with soil depth under Cherry and Bamboos, EC values had a general increasing trend. In addition, though declining after increasing under Apple, they also had a general increasing trend. In contrast to Cherry, Apple and Bamboos, EC values declined with depth under Pine.

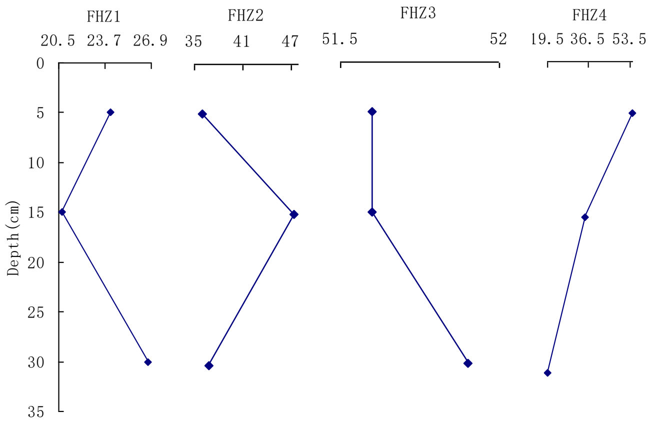

Figure 4. Vertical distribution of EC under different vegetation

3.4 *Variation characteristics of soil MS*

It was worthwhile pointing out that low frequency MSs, especially for those under Cherry, Apple and Bamboos, were lower than those in Xi'an [6], Lanzhou [27], Xuzhou [28], Luoyang [14], with mean values of 154×10^{-8}, 219×10^{-8}, 234×10^{-8}, 215×10^{-8} m^3 kg^{-1}, respectively. Simultaneously, they were lower than that of Isfahan city (74.34×10-8 m^3 kg^{-1}) in Iran [11], except for under Pine.

According to Figure 5, although increased first and then decreased with soil depth under Cherry and Apple, low frequency MSs had a general decreasing trend. In addition, though increasing after decreasing under Bamboos, they also had a general decreasing trend. Instead, though declining after increasing, low frequency MSs increased with soil depth under Pine.

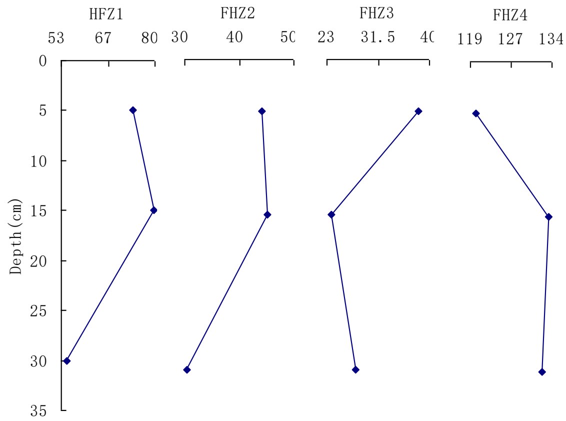

Figure 5. Vertical distribution of low frequency MSs under different vegetation

3.5 *Element source analysis based on enrichment*

Pollution index (PI) of chemical compositions, not only can be used for quantitative assessment of pollution degree, but also can be applied effectively to distinguish their natural and anthropogenic sources [5, 32]. Generally, if PI value higher than 1, it refers to pollution has been caused by human activity. Otherwise, chemical compositions mainly come from natural process.

PI values under different vegetation calculated based on background values of Shandong province, were listed in Table 3. Ranging from 1.12 to 7.10, they were more than 1, except for Co. This indicated that all examined elements except for Co, were probably generated from anthropogenic sources.

Table 3. Pollution Index (PI) of soil elements

	Pb	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Cd
FHZ1	1.51	2.55	2.84	1.84	5.15	0.12	3.58	2.82	2.89	2.49	5.46
FHZ2	1.55	2.33	2.14	1.62	4.50	0.09	2.63	3.42	2.60	3.20	6.73
FHZ3	1.36	1.51	1.19	1.12	3.05	0.06	1.59	1.46	2.00	1.90	4.62
FHZ4	1.56	2.30	2.00	1.48	4.38	0.10	2.43	1.76	2.21	2.17	7.10

Note：PI=Ci/C_{ib}, C_i is element content; C_{ib} is the background value of this element.

3.6 *Element source analysis based on magnetism*

Researches [1, 3] indicated that there may be a linear relationship between element content and MS [11, 13]. However, some researchers believed it was a positive correlation [8, 12, 14, 22]. On one hand, spherical magnetic particles generated by human activities (industrial production, fossil fuel combustion, iron and steel smelting, cement manufacturing, transportation, exhaust emissions, tire erosion, etc.), lead to significant enhancement of soil magnetism when they entered into soil through atmospheric dry and wet deposition, waste dumped, etc [2, 27]. On the other hand, magnetic particles and associated metal elements produced by different human activities are different. Moreover, some researchers found that MS was negatively correlated with frequency dependent susceptibility in human polluted soils [28]. In addition, researchers believed that elements negatively or poor correlated with frequency dependent susceptibility were mainly generated by human activities, while those positively correlated were produced by soil parent materials [27].

Correlation coefficients between element content and low frequency MS under different vegetation were listed in Table 4. Results showed that there was an obviously negative correlation between them. Thus, based on previous understanding of relations between element concentration and MS or frequency dependent susceptibility, combined with field survey data, authors believed that soil elements under different vegetation in this study were mainly generated by human activities, especially by massive application of chemical fertilizer and pesticide, as well as insufficient application of organic fertilizer, and so on.

3.7 *Responses on changes of soil pH and electrical conductivity to human activities*

Generally, content, distribution, migration and transformation, accumulation of heavy metals are closely related to soil physical and chemical properties such as pH, electrical conductivity and so on [5, 17]. As one of the important physical and chemical properties, pH greatly affects other properties together with the migration and transformation of chemical components [17, 24]. As is known, under natural conditions, soil pH values are mainly influenced by soil formation factors such as parent materials, climate, vegetation, terrain, etc [24]. Meanwhile, they will change during the geological cycle and the biological cycle [32]. Nevertheless, compared with other soil formation factors, human activities are more likely to affect and change soil pH values [5]. Previous research suggested that soil pH values could be greatly changed by agricultural production activities such as massive application of chemical fertilizer and pesticide, as well as insufficient application of organic fertilizer [28], etc. It was a good example that soil acidification was serious in orchard planting base in Jiaodong Peninsula, where the pH values were 4.69 and 4.22 in Qixia city and Zhaoyuan city respectively.

Mean value of soil pH in this study is 5.12 under cherry trees, apple trees and bamboos. It was an obvious acidification. Researchers believed that such surface acidification also were related to irrational use of chemical fertilizers, pesticides and organic fertilizers, as well as other improper agricultural measures [28]. Based on field investigation, authors believed that surface acidifications under vegetation in this study were caused by reasons as follows. First of all, a large number of ions would be introduced into soil massive during massive application of chemical fertilizer. And then, buffer capacity would be led to decrease by insufficient application of organic fertilizer. Furthermore, exchangeable base cations would be leached during flood irrigation. Beyond those above, to some extent, surface acidification was also related to both natural process (such as non equilibrium absorption of plants to nutrient particles, metabolism of soil microbes and plant roots, etc.) and acid deposition caused by environmental pollution [5, 28].

3.8 *Possible reasons for different distribution characteristics of elements*

As is known, content, distribution and accumulation of soil elements should be roughly the same when geological and soil forming conditions are similar [5]. However, they will be greatly affected by biological absorption selectivity and enrichment ability differentiations of different vegetation [9, 10, 23]. Although geological and soil forming conditions are similar, vertical migration of soil elements could be affected greatly by vegetation of their different absorption, accumulation, resistance, enrichment ability [9, 26], together with physical and chemical properties such as pH, EC, redox potential, cation exchange capacity and so on [24].

As mentioned earlier, there were obvious differences of soil element content vertical differentiations under different vegetation in this study. In details, all elements but Cd and Arsenic increased with soil depth under Cherry, Apple and Bamboos, while they (Zn excepted) exhibited characteristics of surface accumulation. Based on actual situation, authors believed that the reasons could be as follows. On one hand, dissolution of insoluble matters can be enhanced to improve the chemical processes of the rhizosphere by small molecular substances such as organic acids secreted into soils by

Table 4. Correlation coefficients between element content and low frequency magnetic susceptibility

	Pb	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Cd
FHZ1	-0.9149	-0.8430	-0.7003	-0.8639	-0.8519	-0.8933	-0.8297	-0.7134	-0.9964	-0.9998	0.0056
FHZ2	-0.7431	-0.9135	-0.9998	-0.9377	-0.9697	-0.9986	-0.9973	-0.7915	-0.3498	0.8136	-0.9976
FHZ3	-0.9982	-0.4343	-0.7403	-0.4843	-0.3402	-0.1222	-0.6701	-0.3722	-0.5405	-0.9805	-0.7493
FHZ4	-0.8698	-0.5677	-0.963	-0.9123	-0.8643	-0.6654	-0.9889	-0.4923	-0.8288	-0.9035	-0.9698

Note: P<0.01.

root system [16, 23]. As is known, content and composition of root exudates differ with vegetation types, especially for pine. This may lead to different desorption rates of soil elements, especially for heavy metals [23]. On the other hand, litter composition and decomposition rate ranges a lot for different vegetation types, especially for pine. And chemical properties (eg. pH and contents of organic matter, N, P, K, etc.) changed with them [25], will deeply affect soil element content and vertical differentiation [14]. Finally, it also may be related to the differences among human activity types, properties and intensity under different vegetation.

4 Conclusions

1. Except for Ni, soil element contents were about twice as China's background values, while obviously lower than class ii of National soil Environment Quality Standard.

2. Elements (Zn excepted) of Pine soil accumulated in surface, while they (Cd, Arsenic excepted) increased with depth under other vegetation.

3. EC and pH changed like elements, but MS was exactly opposite. These may be related not only to differences of physiological activities of plant roots, but also to differences caused by different composition and decomposition rate of litter, even to differences among human activity types, properties and intensity under different vegetation.

4. Accumulation characteristics and vertical differentiations of element, pH, EC, especially for MS, together with PI, all indicated that physical-chemical properties changed with human activities, like improper use of fertilizer, pesticide and inadequate use of organic fertilizer, etc.

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