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To cite this article: Min Ding *et al* 2019 *IOP Conf. Ser.: Earth Environ. Sci.* **237** 032052

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Distribution of Trace Elements in Holocene Loess-paleosol Sequence and Environmental Change in Lower Reaches of The Yellow River

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Abstract. Based on field investigation, samples in loess-paleosol sequence profile were collected systematically at Longshan site in Shangdong province in lower reaches of The Yellow River. Trace elements(Mn, Cu, Ba, Ni, Pb, Rb, Sr, Ti, V and Zn) were measured and analyzed systematically by X-Ray and compared with climate substitution index(grain size, Rb/Sr, Ba/Sr) .The results as follows:(1) According to the significant correlation between the content of Ti, Ba, Cu, Ni, V, Zn, Rb, Pb ,Sr in the LS loess-soil sequence and the index of soil strength (soil grain size, Rb/Sr, Ba/Sr), it can be concluded that the migration and change of trace elements in Holocene period were mainly affected by the environmental evolution of this period. Mn is insensitive to climate change in this profile. The content of Pb in the surface layer of soil is the highest, which may be caused by human activities. (2) The contents of Ti, Ba, Cu, Ni, V, Zn and Rb in the paleosol(S₀) were accumulated accompanied by the clay(< 5μm) increase while the coarse silt(10~50μm) reduced, suggesting the intensified pedogenic modification to the accumulated dust when it was humid climate. It implied that the climate was warm and humid during the mid-Holocene optimum. (3) Content of Ti, Ba, Cu, Ni, V, Zn, Rb and clay(< 5μm)in loess layer (L_t, L₁, L₀) was lower and coarse silt (10-50μm) content was higher when compared with the paleosol layer (S₀), suggesting that the intensity of soil formation was weak and aeolian sand activity was frequent. It further indicated that the paleoenvironment was cold and dry with frequent dust storms during the periods of loess formation .

1.Introduction

The migration, enrichment and dispersion of elements in the epigenetic environment are deeply affected by the geographical environment. The predecessors used the chemical parameters and elemental characteristics of trace elements to reveal the material source, soil weathering , and environmental changes of the loess-paleosol sequence in the Loess Plateau and its surrounding areas, having achieved fruitful results[1-8].However, there are few studies on the trace elements in the loess-paleosol sequence in the lower Yellow River, and the longitudinal distribution and time succession of trace elements are still lack of systematic research. The existing research in this area is concentrated on the modern topsoil, which involves the presence of some sensitive elements, spatial distribution and pollution assessment [9-11]. Therefore, it is especially important to carry out high-resolution system research on trace elements at the Holocene LS section in the central Shandong province. It has important scientific



significance to understand the mechanism of soil evolution and the response of trace elements to environmental evolution, and to objectively evaluate the modern heavy metal pollution in this area.

2. Material and Methods

2.1 Material

The study area is located at the central Shandong province in lower reaches of the Yellow River (Figure.1). To the Northwest of this region is the plain, bordering the Yellow River, to the north is the Yellow River delta, and to the northeast is the Bohai Bay. The region has a warm temperate and semi-humid monsoon climate with distinct season, prevailing northerly wind in winter and southerly wind in summer, respectively. Annual temperature and precipitation is about average 13.5 °C and 700 mm. Thick loess deposits in central Shandong Mountains mainly spread in the east-west direction along the northern piedmont regions of the central Shandong Mountains.

Longshan section (hereinafter referred to as LS) lies in Longshan Town (117°21'46.2"E, 36°43'59.4"N). The exposed depth of LS section is about 4m (bottom not seen), the elevation is 56m. 120 samples were obtained continuously from upside to downside with an interval of 2.5cm. The 3m depth of loess section is sampled. After the field profile morphological characteristics observation and ^{14}C dating (^{14}C data will be published elsewhere), it is determined that the section has formed since the last Glacial, we can divide the LS profile into the following layers: L_1 (190cm below) sediments formed in the last glacial period, it is yellow, clay silt. L_t (190-160cm deep) is the transition layer, which formed in the early Holocene, it is grey yellow; S_0 (160-50cm deep) formed in Mid-Holocene, it is brown, clay soil, and is prismatic structure; L_0 (50-0cm deep), is the modern dust accumulation layer which formed in the late Holocene, it is orange and sandy soil [12-13].

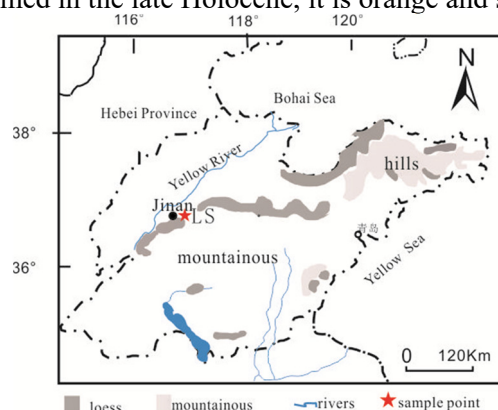


Figure 1. Geographical locations of the study area and the LS profile in Shandong province, China.

2.2 Methods

The PW2403 X-Ray fluorescence spectrometer produced by Panalytical Company of the Netherlands is used for trace element analysis. The sample was ground to a size less than 74 μm , and the sample was prepared by YY-60 sample-pressing machine. The relative error RSD of measured and reference values is less than 10% when the national standard soil reference materials GSS-1 and GSD-12 are added to the measurement process for error control.

The particle size analysis of the sample was carried out by a Mastersizer-2000 laser particle size analyzer manufactured by Malvern, UK. The pretreatment was carried out by adding 10% H_2O_2 and 10% HCl to completely remove the organic matter and the secondary carbonate to fully disperse the clay particles in the sample. The measurement range is 0.03~1000 μm , and the relative error is less than 4%.

3.Results

3.1 Grain- Size and Rb/Sr and Ba/Sr and Analysis at LS Profile

The grain size characteristics of the sediments record the environmental characteristics and some characteristics of the source area during the deposition. In the aeolian sediments with uniform provenances, the grain size mainly reveals the characteristics of wind power and the soil-forming environment. Coarse silt (10-50 μm) is the main aeolian dust group. It has been widely used as an environmental substitute index to reveal the size of winter monsoon in loess-paleosol study on the Loess Plateau [8,14]. Clay (< 5 μm) mainly reveals the change of soil-forming environment, which is proportional to the intensity of soil formation. LS profile in the study area is also a typical aeolian deposit, so the environmental significance of grain size parameters is consistent with the environmental plateau. The grain- Size results of LS profiles are shown in Fig. 2. The variation trend of clay content (< 5 μm) is consistent with that of fine silt (5-10 μm), and that of coarse silt (10-50 μm) is consistent with that of sand content. The content of coarse silt (10-50 μm) is the main grain group, which varies from 33.0% to 58.5%, and the content of Paleosol (S_0) is the lowest (mean 43.6%). This indicates that the winter monsoon is the weakest, the moving force is reduced, and the sedimentary grain size becomes finer. In the loess layer (L_1 , L_t , L_0), the content of coarse silt (10-50 μm) increases significantly, with an average of 44.8%, 49.7% and 45.6%, respectively, indicating that the winter monsoon is stronger at that time. Strength, moving ability increased significantly. The content of clay (< 5 μm) in the paleosol layer (S_0) is the highest, indicating that the weathering intensity is the highest, the climate is the warmest and humid, the content in the loess layer is low, the weathering conditions are poor, and the climate is relatively colder and drier, which is consistent with the significance revealed by the coarse silt (10-50 μm).

The values of Ba/Sr and Rb/Sr reflect the leaching degree of active elements in the process of weathering and soil formation. The greater the leaching intensity, the greater the values of Ba/Sr and Rb/Sr, which are proportional to the precipitation in the local climate [1,3]. It can be seen that the change trend of Ba/Sr and Rb/Sr values is identical (correlation coefficient is 0.988) in Table 1, and the ratio of Ba/Sr and Rb/Sr is the highest in the paleosol layer (Table 2, Figure 2), which is 4.31 and 1.00, respectively. This indicates that the climate is humid, precipitation is abundant, and soluble elements such as Sr are leached out in large quantities, resulting in the increase of Ba/Sr, Rb/Sr ratios, and the decrease of Ba/Sr and Rb/Sr ratios in the loess layer (L_1 , L_t , L_0). The Rb/Sr ratios were 0.57, 0.60 and 0.72, indicating that the leaching intensity was weakened, and the precipitation was reduced.

The experimental results show that (Table 1 and Figure 2), the grain size curves in LS profiles are closely related to the elemental ratio (Ba/Sr, Rb/Sr) curves. The clay content (<5 μm) shows a synchronous and symmetrical change trend with Ba/Sr and Rb/Sr, and the correlation coefficients are 0.580 and 0.59, respectively. The coarse silt (10-50 μm) shows a reverse synchronous change with Ba/Sr and Rb/Sr, with a very significant negative correlation. The correlation coefficients were - 0.715 and - 0.749, respectively. The results of grain size and element ratio show that the weathering degree of Paleosol layer (S_0) is significantly higher than that of Malan loess layer (L_1) and modern loess layer (L_0). Clay content, Ba/Sr, Rb/Sr ratio are the highest in paleosols, while coarse silt (10-50 μm) is the lowest and weathering degree is strong. This indicates that the soil in this area has a good soil-forming environment, strong weathering and soil-forming effect, warm and humid climate and more precipitation in the middle Holocene. Clay content, Ba/Sr, Rb/Sr ratio and loess layer (L_1 , L_t , L_0) are relatively low. The high content of coarse silt, relatively weak weathering and soil formation, and large accumulation of loess indicate that the period was strongly influenced by winter monsoon, with less precipitation and more aeolian sand activities.

3.2 Trace elements and Analysis at LS Profile

The geochemical behavior of trace elements in the supergene environment is directly related to the nature of the elements, and deeply influenced by the sedimentary environment. In this paper, the

elements Mn, Ba, Ti, Cu, Ni, Pb, V, Zn, Rb and Sr in the LS section in central Shandong province were selected for analysis (Table 2, Figure 3).

The distribution regularities of Ti, Ba, Cu, Ni, V, Zn and Rb in the profile are similar. It is shown as unimodal distribution. The content of them in paleosol S_0 is the highest, followed by that in L_t (except Ti, L_0 is slightly lower than S_0), while the content of them in loess layer MS and L_0 and L_1 is similarly lower. The correlation coefficients of Ti, Ba, Cu, Ni, V, Zn and Rb were significantly positively correlated, and with clay ($< 5 \mu\text{m}$), Ba/Sr, Rb/Sr.

Ti content is the highest in the paleosol (S_0), the average is 4201.52 mg/kg, the lowest in the L_1 layer, the average is 3894.09 mg/kg, and the coefficient of variation in this profile is small. Ti is mainly concentrated in rutile and ilmenite, the content of which is very stable and difficult to migrate in the supergene environment [7,14]. The high content of S_0 in LS profile is due to the leaching of soluble elements.

Table 1. Trace element content, Ba/Sr, Rb/Sr and particle size correlation coefficient at LS section in central Shandong Province.

	Mn	Ti	Ba	Cu	Ni	Pb	V	Zn	Rb	Sr	Ba/Sr	Rb/Sr	X1	X2
Mn	1													
Ti	.031	1												
Ba	.477**	.707**	1											
Cu	.072	.855**	.828**	1										
Ni	.334**	.688**	.893**	.857**	1									
Pb	-.122	-.051	-.473*	-.306*	-.564*	1								
V	.273**	.638**	.836**	.850**	.945**	-.522*	1							
Zn	.249**	.849**	.794**	.858**	.873**	-.242*	.848**	1						
Rb	.144	.823**	.884**	.936**	.921**	-.520*	.868**	.881**	1					
Sr	.065	-.776*	-.747*	-.809*	-.690*	.422**	-.604*	-.675*	-.882*	1				
Ba/Sr	.115	.780**	.894**	.879**	.832**	-.521*	.761**	.755**	.947**	-.958*	1			
Rb/Sr	.040	.796**	.853**	.903**	.850**	-.541*	.785**	.786**	.973**	-.957*	.988**	1		
X1	-.231*	.399**	.425**	.553**	.396**	-.344*	.413**	.249**	.516**	-.560*	.580**	.590**	1	
X2	-.204*	-.714*	-.770*	-.872*	-.853*	.382**	-.894*	-.822*	-.838*	.580**	-.715*	-.749*	-.715*	1

X1 shows the content of clay ($< 5 \mu\text{m}$); X2 shows the content of coarse silt ($10\sim 50 \mu\text{m}$); * represents significance is 0.05, ** represents significance is 0.01, and the others untitled show their differences are not significant.

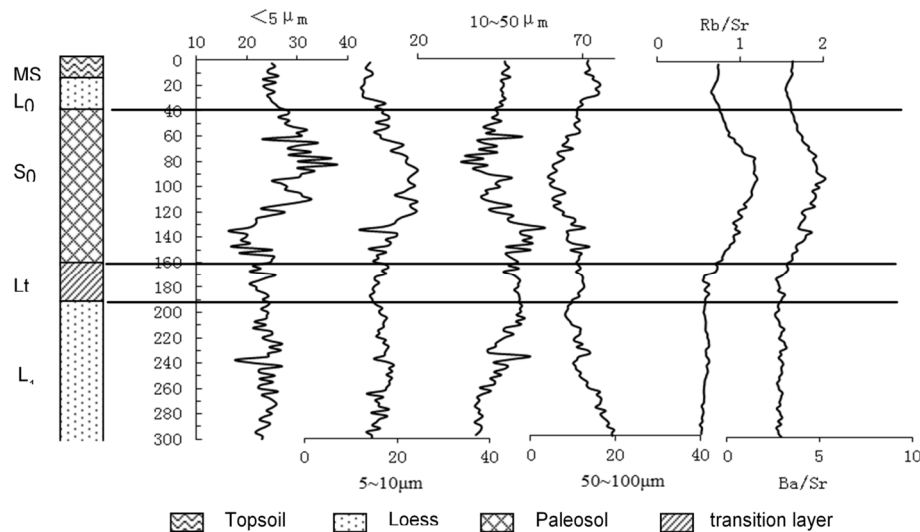


Figure 2. The curves of Grain size and Rb/Sr, Ba/Sr at LS profile in central Shandong.

Cu, Ni, V and Zn are all iron group elements and belong to transition metals. The reason why S₀ content in ancient soil was the highest was when a large number of secondary clay particles were formed in this layer due to the influence of biogeochemistry. Cu is the most abundant element in the ancient soil layer S₀ with a large variation range, and the least abundant element in the loess layer with an average value of 27.47mg/kg. The content in the transition layer Lt is between the ancient soil and the loess layer. The maximum Ni value appeared in S₀, which was up to 53.8mg/kg, with a very large change range. The average content of Lt in the transition layer was up to 38.23mg/kg, which was second only to ancient soil S₀. The contents of element V and Zn were the highest in S₀, with an average value of 98.32 mg/kg and 75.81 mg/kg, respectively. The contents of element L₁ in the loess layer were the lowest, with 86.78 mg/kg and 59.85 mg/kg, respectively.

Rb, Ba and Sr belong to alkali metals and alkaline earth metals. But the chemistry is different. Sr is a typical dispersed element, which is mainly distributed in the form of homogeneity in nature in the form of calcite containing Ca and other coarse-grained minerals, and its properties is similar to Ca. The ionic radius of element Rb (0.147nm) is not much different from that of element K (0.133nm), so it is often dispersed in the minerals containing K, such as mica, biotite and potassium feldspar. Element enrichment characteristics of Ba elements are similar to those of Rb elements. Studies have shown that under the same chemical weathering, minerals rich in Rb and Ba are more stable than minerals rich in Sr [15-17]. Some scholars have also found that the concentrations of Rb and Ba are significantly positively correlated to the microns with particle diameters less than 10 μm, while the concentrations of Ca and Sr are significantly positively correlated to the microns with particle diameters ranging from 10 ~50 μm [3]. It is consistent with the conclusion of this paper. Therefore, Rb and Ba are enriched in fine particles, while Sr is mainly dispersed in the coarse lattice, making the change trend of Rb and Ba opposite to Sr. The content of Ba element reached the maximum in paleosol S₀, up to 654.40mg/kg, with an average value of 586.34mg/kg, followed by the content of Lt in the transition layer, with an average value of 541.4mg/kg, with the minimum content in the loess layer. The maximum value of Rb element is as high as 148.60mg/kg, which appears in paleosol S₀ with a small change range and the least content in loess L₁ with an average value of 105.03mg/kg. Sr was the most abundant in L₁ and the least abundant in S₀, with an average value of 184.65 mg/kg and 137.43 mg/kg, respectively.

Table 2. Trace elements LS profile in Shandong province (mg/kg)

Soil stratigraphy/(mg/kg)					Change range	Coefficient of variation
TS and L ₀ (n=18)	S ₀ (n=46)	L _t (n=15)	L ₁ (n=44)	/(mg/kg)		%
Mn	607.87	734.44	781.76	717.90	452.8~1525.1	24.66

Ti	4140.44	4201.52	4028.67	3894.09	3744.1~4331.2	3.90
Ba	535.56	586.34	541.40	525.39	473.5~654.40	6.68
Cu	30.46	35.83	30.59	27.47	22.3~40.9	14.22
Pb	17.44	13.17	17.27	15.71	0.9~19.1	15.17
Ni	33.54	43.10	38.23	33.83	28.6~53.8	12.80
Rb	112.94	135.21	113.26	105.03	91.1~148.6	14.25
Sr	158.17	137.43	183.33	184.65	123~200.7	3.90
V	84.51	98.32	93.23	86.78	77.1~113.3	8.72
Zn	67.70	75.81	69.47	59.85	47.3~82.4	13.02

The variation trend of element Pb is opposite to that of element Ti, Ba, Cu, Ni, V, Zn, Rb, and is significantly correlated with clay, coarse silt, Ba/Sr, Rb/Sr, which indicates that the distribution of Pb in this section is also controlled by environment. The content of Pb is the most in modern soil MS and L₀, and the smallest in paleosol S₀, with an average value of 21.17 mg/kg. The variation range is very large, especially in surface soil, which may be related to environmental pollution. It is mainly related to the impact of human activities since the late Holocene, the impact of human activities is mainly related to the large-scale use of chemical fertilizers and pesticides, industrial "three wastes" emissions, metal mining and smelting, coal and petroleum combustion and automobile exhaust emissions, which may bring heavy metal pollution such as Pb.

The content of Mn varied from 452.8 mg/kg to 1525.1 mg/kg, the highest in Lt, with an average content of 81.76 mg/kg, and the lowest in MS and L₀ layers, with an average content of 607.87 mg/kg. It has weak correlation with grain size index and climate substitution index such as Ba/Sr and Rb/Sr, may be insensitive to environmental change.

The content of Ti, Ba, Cu, Ni, V, Zn and Rb elements in palaeosol(S₀) was higher than those in Loess layers. These elements were positively correlated with soil clay (< 5 μ m), Rb/Sr and Ba/Sr, and negatively correlated with sand grains (10-50 μ m). The trend of change of Pb and Sr was opposite to that of Ti, Ba, Cu, Ni, V, Zn and Rb, and was negatively correlated with soil clay (< 5 μ m), Rb/Sr and Ba/Sr. These elements are sensitive to environmental change and can indicate the change of sedimentary environment. The correlation between Mn and other indicators is slightly poor, and the environmental sensitivity is insufficient.

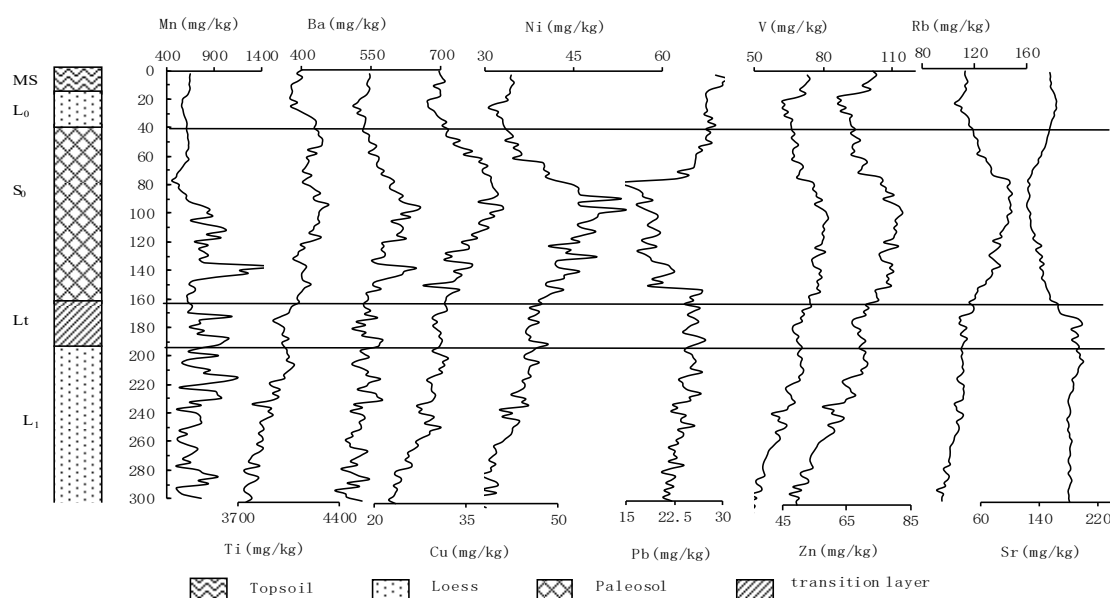


Figure 3: Trace Element Change Curve of LS Profile in Central Shandong

4. Discussion

During the last glacial period, the contents of Ti, Ba, Cu, Ni, V, Zn, Rb, Rb/Sr, Ba/Sr, clay ($<5\ \mu\text{m}$), fine silt ($5\text{--}10\ \mu\text{m}$) in loess L_1 ($>190\text{cm}$) were lower. Sand grains ($10\text{--}50\ \mu\text{m}$) and ($50\text{--}100\ \mu\text{m}$) were the highest compared with other layers. It is indicated that the soil formation was weaker, and aeolian activity was frequent. Winter winds were stronger in this period, the climate was cold and dry.

In the early Holocene, the contents of Mn, Ti, Ba, Cu, Ni, V, Zn and Rb in the transition layer L_t ($160\text{cm}\text{--}190\text{cm}$) began to increase, the active elements of Pb and Sr began to leach out, the clay content increased, and the content of coarse silt decreased. The reason for this phenomenon is that the influence of winter monsoon in this geological period is weaker than that in the last glacial period, the pedogenic process gradually strengthened and the climate began to improve.

In the middle Holocene, the contents of Ti, Ba, Cu, Ni, V, Zn and Rb in the paleosol S_0 ($45\text{cm}\text{--}160\text{cm}$) all reached the maximum, the clay content was the highest and the coarse silt content was the lowest. It implied that the climate was warm and humid during this period, and biochemical chemistry was strong, forming a large number of secondary clay minerals. Trace elements such as Ti, Ba, Cu, Ni, V, Zn, and Rb were adsorbed together with organic colloids, oxides, and clay particles in the soil, resulting in an increase in content. At the same time, the elements of Pb and Sr are more active, easy to leach, dissolve and migrate under warm and humid climate conditions, and the content of Pb and Sr decreases, which further increases the content of other stable elements. It indicated that the summer monsoon was strong and it was the warmest and wettest in this geological period compared with the others.

From the late Holocene to the present, the content of trace elements Ti, Ba, Cu, Ni, V, Zn, Rb and the clay ($<5\ \mu\text{m}$) in modern topsoil ($0\text{--}45\text{cm}$), began to decrease, and the coarse silt content increased. This suggested that the degree of soil formation was weakened at this stage and the climate was deteriorating. The relative enrichment of Pb elements may be related to agricultural development and industrial pollution.

5. Conclusions

(1) According to the significant correlation between the content of Ti, Ba, Cu, Ni, V, Zn, Rb, Pb, Sr in the LS loess-soil sequence and the index of soil strength (soil grain size, Rb/Sr, Ba/Sr), it can be concluded that the migration and change of trace elements in Holocene period were mainly affected by the environmental evolution of this period. Mn is insensitive to climate change in this profile. The content of Pb in the surface layer of soil is the highest, which may be caused by human activities.

(2) The contents of Ti, Ba, Cu, Ni, V, Zn and Rb in the paleosol (S_0) were accumulated accompanied by the clay ($<5\ \mu\text{m}$) increase while the coarse silt ($10\text{--}50\ \mu\text{m}$) reduced, suggesting the intensified pedogenic modification to the accumulated dust when it was humid climate. It implied that the climate was warm and humid during the mid-Holocene optimum.

(3) Content of Ti, Ba, Cu, Ni, V, Zn, Rb and clay ($<5\ \mu\text{m}$) in loess layer (L_t , L_1 , L_0) was lower and coarse silt ($10\text{--}50\ \mu\text{m}$) content was higher compared with the paleosol layer (S_0), suggesting that the intensity of soil formation was weak, aeolian sand activity was frequent, and element activity was reduced. It further indicated that the paleoenvironment was cold and dry with frequent dust storms during the periods of loess formation.

Acknowledgements

This study was supported by National Natural Science Foundation of China (41402319, 41472313, 41771218, 41602353, 41877442), National Key Laboratory of Loess and Quaternary Geology Open Fund (SKLLQG1002), Taishan University Talent Research Fund (Y-01-2016001). Min Ding and Shuzhen Peng both are corresponding authors.

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