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Phosphorus geochemistry in the Luochuan loess section, North China and its paleoclimatic implications

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Abstract

Total P (P_t) on a carbonate-free basis in an entire loess–paleosol sequence and P_t, organic P (P_o) and inorganic P (P_i) in the S_0–L_1–S_1 sequence were investigated in detail with different resolutions for the Luochuan loess section from northern China. P_t content varies between 393 and 786 ppm throughout the loess–paleosol sequence, and is generally higher in the loess than in interstratified paleosols, showing fluctuation cycles of 100 ka in correspondence to loess–paleosol alternations. P_t variations on a carbonate-free basis in the loess–paleosol sequence could indicate variations in atmosphere precipitation resulting in different leaching loss of P from paleosols. P_i has an average value of 499 ppm with a range of 324–560 ppm, accounting for more than 70% of P_t in the S_0–L_1–S_1 sequence, where the minimum of P_i in the Malan loess is higher than the maximum of P_i in S_1. P_o ranges between 59 and 233 ppm with an average of 132 ppm in the S_0–L_1–S_1 sequence. Phosphorus (P) was initially delivered to the Luochuan loess section via influx of aeolian dust from the northern desert and Gobi areas by the East Asian winter monsoon, and then was modified by pedogenesis associated with the East Asian summer monsoon during the last 130 ka. "Preserved P_t" in the loess L_1 is tightly correlated with grain size without leaching loss of P due to enrichment of P in fine-grained fractions, as well as "initial P_t". "Leaching P_t" data show that paleosol S_1 had lost 15–40% of its "initial P_t", and that there was much more precipitation in S_1 than in L_1. P_i subject to slightly weak pedogenesis was completely transformed into P_o without leaching loss of P in loess L_1. By contrast, much P_i disappeared from paleosol S_1 due to strong pedogenesis, partly through leaching and partly through conversion to organic forms during P cycling processes. P_o variation is similar to those of MS and the $<7.8\,\mu m$ fraction in L_1, but contains more information on the East Asian winter monsoon due to weak pedogenesis without leaching of P. P_o in S_1 lower than L_1SS_1 as a consequence of strong decomposition of the organic matter kept constantly in the middle of S_1 where P_i kept at the lowest of 423 ppm, suggesting that there existed a very warm and humid climate related to the enhanced summer monsoon during that period. The mean organic P/ inorganic P ratio (P_o/P_i) is lower in the L_1LL_1 and L_1LL_2 than in the S_0, S_1 and L_1SS_1, indicating that low P_o/P_i ratios coincide with weak weathering-pedogenesis, and higher P_o/P_i ratios correspond to strong weathering-pedogenesis. P_o/P_i ratio can eliminate the effect of grain size on aeolian dust because of chemical uniformity of aeolian dust and enrichment of P_o and P_i in the fine-grained fractions. Thus, P_o/P_i ratio is solely linked to pedogenesis of the Luochuan loess section. Variation in P_o/P_i ratio is similar to those of MS and the Marine Oxygen Isotope composition, indicating the summer monsoon evolution during the last 130 ka and providing the biogeochemical evidence for further understanding the genetic links between the East Asian monsoon and global climate change.

1. Introduction

It is necessary to reconstruct the evolutional history of climate and environment during geological times, not only for better understanding of causes and mechanisms but also to establish reliable forecast models for future climate and environment changes. Over several decades, much research related to paleoclimate and paleoenvironment changes has already been accomplished or is ongoing by scientists from many countries in order to derive much high-resolution information on paleoclimate and paleoenvironment changes from such carriers

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as ice cores, marine sediments, and loess (Liu, 1985; Ruddiman et al., 1986; Muller and MacDonald, 1997; Petit et al., 1999). Chinese loess–paleosol sequences are widely viewed as one of the best records of continental paleoclimatic change (Kukla et al., 1988; An et al., 1991). Many climatic proxy indicators associated with the East Asian monsoon evolution were derived from Chinese loess–paleosol sequences, such as magnetic susceptibility (An et al., 1990; Heller et al., 1993), grain size (Ding et al., 1995; Vandenberghe et al., 1997), geochemistry (Han et al., 1997; Guo et al., 1998; Chen et al., 1999; Ding et al., 2001a, b) and mineralogy (Bronger and Heinkele, 1990; Ji et al., 1999a, b, 2001). However, most of these results mentioned above emphasize the physical and chemical characteristics of the loess and paleosols, but little is known about the biogeochemistry in loess and paleosols until now.

Phosphorus (P) is an essential macronutrient for biological activity, has been observed to limit productivity in a range of ecosystems (Schlesinger, 1997), and is consequently important in the whole biological cycle. Parent materials are the only source of P in soils other than the minimal amount contained in precipitation. P content varies considerably between soils and between horizons in the soils (Cassagne et al., 2000). P forms in soils are closely related to soil acidity (Lindsay and Moreno, 1960; Tyler, 2002). Walker (1965) and Miller et al. (2001) reported that P forms were transformed more commonly and more strongly as precipitation increased. Williams and Walker (1969) summarized changes in the forms of soil P as a result of progressive soil development, and drew a conclusion that primary apatite gradually decreased whereas such secondary phosphate as aluminum phosphate (Al-P), iron phosphate (Fe-P) and occluded phosphate (O-P) increased in content with strengthening of weathering and pedogenesis in soils. Cross and Schlesinger (1995) demonstrated that the relative amount of 'biological' soil P increased with increased weathering at the expense of 'geochemical' soil P. As a result, contents, forms and distributions of P in soils can be regarded as indicators of pedogenetic weathering processes (Smeck, 1973).

Aeolian dust from the northern desert and Gobi areas deposited in the Loess Plateau was modified by weathering and pedogenesis, forming the loess during the glacial period and paleosols during the interglacial period (Liu, 1985). Biogeochemical behavior of P was mainly dominated by weathering and pedogenesis during the formation of the loess and paleosols, largely depending on temperature and precipitation associated with the East Asian monsoon (Yang and Ding, 2001). Therefore, the record of biogeochemical behavior of P preserved in the loess–paleosol sequence can reflect variations in paleoclimate. This paper presents the results of investigation of P geochemistry in the Luochuan loess section, and discusses their significance.

2. Materials and methods

The Luochuan loess section (35°45′N, 109°25′E) in Shaanxi Province, North China, is about 140 m thick, overlying Upper Pliocene red clays (RS). The entire loess–paleosol sequence includes 38 loess and paleosol layers, spanning the time interval from ~2.5 Ma BP to the present (Liu, 1985). The sequence is divided into four units from the base upwards: the Wuchen Loess (WL_4–WS_1); the Lishi Loess (L_{15}-–S_1); the Malan Loess (L_1); and the Holocene Black Loam (S_0). Thirty-eight samples were collected from the loess–paleosol sequence, with one sample from each layer. In addition, one red clay sample was collected from Upper Pliocene red clays. The S_0–L_1–S_1 sequence of the Luochuan loess section, about 12 m thick, consists of the Holocene Black Loam (S_0) developed during the post-glacial period; the Malan Loess (L_1) formed during the last glacial period; and the first paleosol of Lishi Loess (S_1) developed during the last interglacial period. From top to bottom, the sequence spans about 130 kyr (Yang et al., 2000; Porter, 2001). The Malan loess L_1 is rather thick (about 8 m), and can be subdivided into three parts from the top downwards: L_1LL_1 (loess during the last stade of the last glacial period); L_1SS_1 (weakly pedogenic loess during the interstade of the last glacial period); and L_1LL_2 (loess during the early stade of the last glacial period). Ninety-five sub-samples were collected with Quartation from the S_0–L_1–S_1 sequence. The sub-sample interval was 10 cm in S_0, L_1, and at the L_1/S_1boundary, and 30 cm in S_1. After air-drying, all sub-samples were ground in an agate mortar to a size-fraction of <100 mesh, and preserved.

Sequential exaction experiments were performed using the method of Tessier et al. (1979) and Li et al. (1995) with five samples from L_1, S_1, WL_1, WS_3, and red clay, respectively. The result shows that P in the loess, paleosols and red clays was mainly preserved as residual P and organic P without carbonate-bound P, exchangeable P, and water-soluble P (Table 1). Yang et al. (2000) demonstrated that 0.5 M acetic acid could only remove secondary carbonate from bulk samples. Thus, this pretreatment with 0.5 M acetic acid does not result in P loss from bulk samples. The 39 bulk samples from the loess–paleosol sequence were pretreated with 0.5 M acetic acid for 4 h using the method of Yang et al. (2000). Total P (P_t) concentrations were determined by X-ray fluorescence spectrometer (XRF) with analytical uncertainties of $\pm 2\%$. Five grain-size fractions (>45 μm, 45–28 μm, 28–8 μm, 8–2 μm and <2 μm) were separated for two samples from L_1 and S_1, respectively, and were analyzed for P_t content by XRF.

P_t, inorganic P (P_i) and organic P (P_o) of sub-samples in the S_0–L_1–S_1 sequence were analyzed. P_t concentrations were measured after digestion with a $HF + HClO_4 + HNO_3$ mixture followed by colorimetric

Table 1
Results of sequential extraction experiment of P (ppm) in the loess, paleosols and red clay

P (ppm)	L_1	S_1	WL_1	WS_3	Red clay
Water-soluble P	nd	nd	nd	nd	nd
Exchangeable P	nd	nd	nd	nd	nd
Carbonate-bound P	nd	nd	nd	nd	nd
Fe and Mn oxides-bound P	37	32	33	54	60
Organic P	227	114	249	200	160
Residual P	381	476	423	615	416
ΣP	645	622	705	869	636

"nd" denotes that the value is under detected by XRF.

determination of phosphate (Sun and Liu, 1996). P_i was determined by extracting sub-samples with 1 M H_2SO_4 (1:50 soil: solution ratio) for 18 h using the method of Saunders and Williams (1955). P_i concentrations of extracts of sub-samples were measured colorimetrically by molybdate-ascorbic acid procedure of Sun and Liu (1996). P_o concentration was calculated as the difference between P_t and P_i. All sub-samples were analyzed using UV-3000 type spectrophotometer. Data from the samples tested repeatedly ($n = 15$) show a standard deviation for P_t content of ± 2 ppm, and for P_i content of ± 1 ppm. The samples for total iron (Fe_2O_3) concentration determination were dissolved with a $HF + HClO_4 + HNO_3$ mixed solution. The total iron concentrations were measured by AAS with uncertainties of below 5%. Magnetic susceptibility was determined by using a BaringtonMS2 meter with an operating frequency of 0.47 kHz in the City and Environment Department of Peking University, China.

3. Results

3.1. Variations in total P (P_t) of the loess–paleosol sequence

Variations in P_t content of the loess–paleosol sequence are shown in Fig. 1. P_t content varies between 393 and 786 ppm throughout the loess–paleosol sequence. P_t in paleosols ranges from 393 to 611 ppm with an average of 489 ppm. P_t in the loess changes between 568 and 786 ppm with a mean value of 611 ppm. P_t concentration is generally high in the loess and low in paleosols. However, magnetic susceptibility is low in the loess and high in paleosols. Variations of P_t in the loess–paleosol sequence exhibit an opposite oscillation with magnetic susceptibility, and have the fluctuation cycles of 100 ka similar to the alternations of loess and paleosol layers.

3.2. Distribution of P in the S_0–L_1–S_1 sequence

P_t, P_o and P_i concentrations (Table 2) in the S_0–L_1–S_1 sequence are shown in Table 3 and plotted in Fig. 2. P_t in the S_0–L_1–S_1 sequence varies from 450 to 793 ppm with a mean content of 632 ppm. P_t in S_0 changes between 625 and 793 ppm with a mean content of 678 ppm. P_t is the highest in L_1SS_1, intermediate in L_1LL_2, and the lowest in L_1LL_1 within the loess L_1. P_t in S_1 ranges from 450 to 601 ppm with a mean content of 514 ppm, with the maximum in S_1 is lower than the minimum of P_t in S_0 and the Malan loess (L_1). P_t distribution is evidently different between in loess L_1 and in paleosol S_1, which reflects paleoclimate variations over a long time, agreeing with the result obtained by using XRF.

P_i in the S_0–L_1–S_1 sequence varies between 324 and 560 ppm with a mean value of 499 ppm, accounting for more than 70% of P_t. P_i content in the Malan loess changes over a very narrow range, from 493 to 554 ppm with an average of 515 ppm. P_i is the highest in L_1LL_2, intermediate in L_1SS_1, and the lowest in L_1LL_1 within the Malan loess (L_1). P_i in S_1 varies from 324 to 476 ppm with an average of 395 ppm, with the maximum apparently lower than the minimum of P_i in the Malan loess (L_1). The mean value of P_i, as well as the percentage of P_i to P_t, is higher in loess L_1 than in S_0 and S_1. The distribution curve of P_i in S_1 is very similar to that of P_t and there is a good correlation between P_i and P_t, implying that variation of P_t results mainly from variation of P_i in S_1 (Figs. 2 and 3).

P_o in the S_0–L_1–S_1 sequence varies between 59 and 233 ppm with an average of 132 ppm, accounting for less than 30% of P_t. P_o in S_0 varies from 141 to 233 ppm with a mean content of 178 ppm. P_o in the Malan loess ranges between 67 and 233 ppm with a mean value of 128 ppm. P_o in L_1SS_1 is the highest, is intermediate in L_1LL_2, and is the lowest in L_1LL_1 as well as the percentage of P_o to P_t within the loess L_1. P_o in S_1 varies from 59 to 177 ppm with a mean content of 119 ppm, and gradually decreases with increasing depth. As a whole, P_o in S_1 is lower in mean content than in the Malan loess (L_1), whereas the percentage of P_o to P_t in S_1 is higher than those in L_1SS_1, L_1LL_1 and L_1LL_2. The distribution curve of P_o in L_1 is very similar to that of P_t and there is a good correlation between P_o and P_t, indicating that variation of P_t is mainly controlled by variation of P_o in L_1 (Figs. 2 and 4).

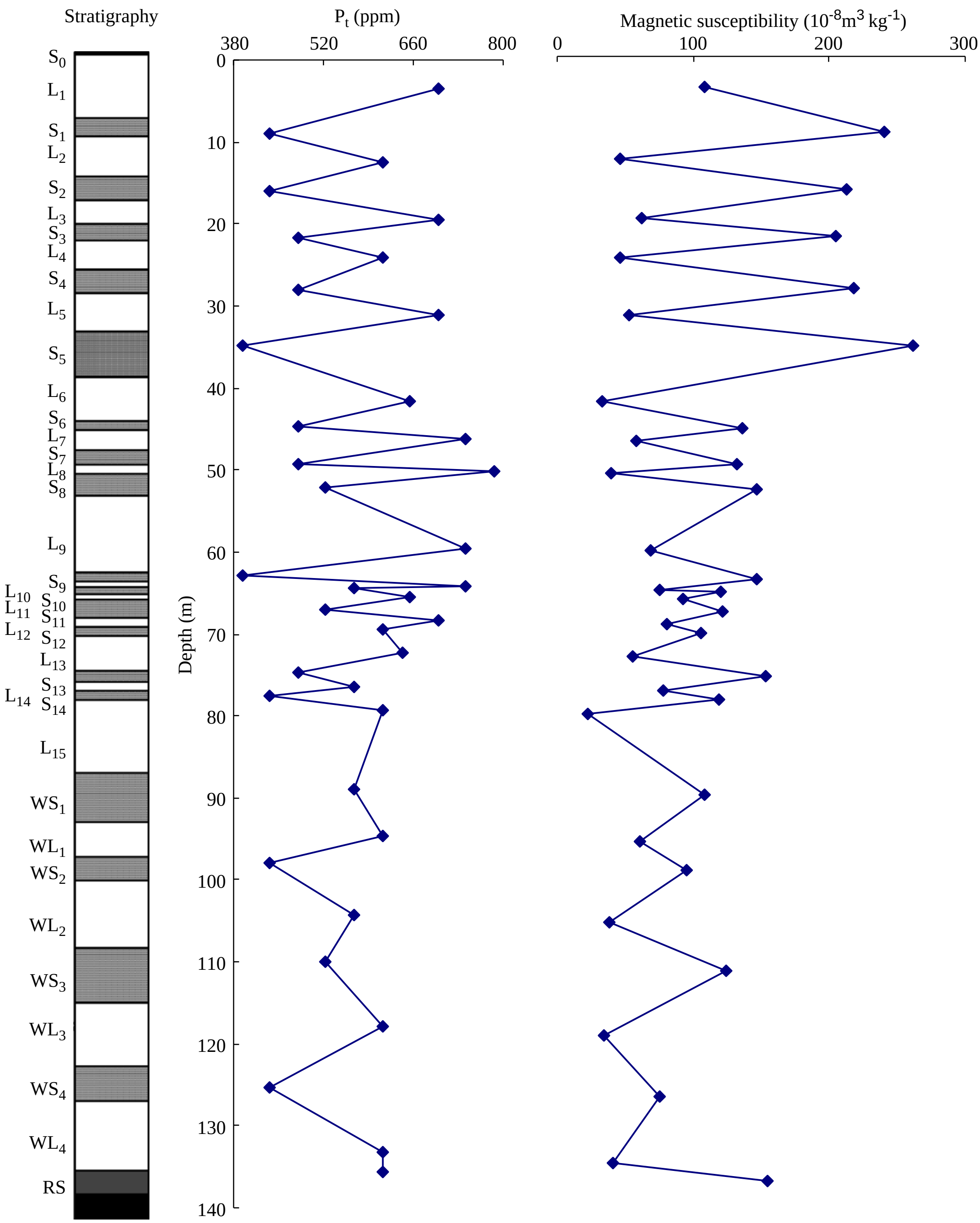

Fig. 1. P_t and magnetic susceptibility in the Luochuan loess–paleosol sequence.

4. Discussion

4.1. Fluctuations of P_t in the loess–paleosol sequence reflect variations in precipitation with a periodic cycle of 100 ka

In general, P does not occur as gaseity (PH_3) under the hypergenic condition (Föllmi, 1996). The Chinese Loess Plateau is of aeolian origin, and has not been invaded by groundwater since the onset of the Quaternary (Guo et al., 2001; Ding et al., 2001b). Therefore, P in the Chinese Loess Plateau solely originated from aeolian dust in the North desert and Gobi areas (Liu, 1985). Aeolian dust would be modified by weathering and pedogenesis as soon as it was accumulated in the Chinese Loess Plateau. Since then, P biological and geochemical processes took place. P cycle in the Chinese Loess Plateau can be simply considered to comprise

Table 2
P contents (ppm) in different layers and sub-layers of the S_0-L_1-S_1 sequence

Stratigraphy	P_t		P_i		P_o	
	Range	Mean	Range	Mean	Range	Mean
S_0(10)	625–793	678	436–560	500	141–233	178
L_1LL_1(21)	641–560	605	493–516	501	67–141	103
L_1SS_1(29)	639–732	660	500–539	512	114–223	148
L_1LL_2(24)	623–702	655	501–554	530	87–179	125
S_1(11)	450–601	514	324–476	395	59–177	119
$\Sigma L+S$(95)	450–793	632	324–560	499	59–233	132
Stratigraphy	P_i/P_t		P_o/P_t		P_o/P_i	
	Range	Mean	Range	Mean	Range	Mean
S_0(10)	0.7–0.78	0.74	0.23–0.3	0.26	0.28–0.43	0.36
L_1LL_1(21)	0.78–0.88	0.83	0.12–0.22	0.17	0.14–0.28	0.21
L_1SS_1(29)	0.7–0.81	0.78	0.18–0.3	0.22	0.22–0.44	0.29
L_1LL_2(24)	0.75–0.85	0.81	0.15–0.25	0.19	0.17–0.34	0.24
S_1(11)	0.71–0.89	0.77	0.11–0.29	0.23	0.13–0.42	0.31
$\Sigma L+S$(95)	0.7–0.89	0.79	0.11–0.3	0.21	0.13–0.44	0.27

Note: The number of samples is shown in bracket.

Table 3
P_t (ppm) in different grain-size fractions of the loess and paleosols

Grain size fractions	>45 μm	45–28 μm	28–8 μm	8–2 μm	<2 μm
P_t in L_1	480	568	786	611	873
P_t in S_1	262	262	262	437	786

three phases (Fig. 5): first, input of P into the Chinese Loess Plateau; second, transformation of P_o and P_i in the loess system; finally, leaching of some P out of the Chinese Loess Plateau.

Although there are many different and contradictory conclusions about the provenance of the Chinese loess (Liu, 1985; Bowler et al., 1987; Liu et al., 1994; Derbyshire et al., 1998; Sun, 2002; Nakano et al., 2004), the viewpoint of the uniformity of its chemical composition has been widely accepted by scientists (Gallet et al., 1996; Jahn et al., 2001). Thus, distribution of P in the loess section observed at present is mainly controlled by two factors: particle sizes of aeolian dust associated with the East Asian winter monsoon (Porter, 2001), and intensity of weathering-pedogenesis related to the East Asian summer monsoon after aeolian dust accumulation in the Chinese Loess Plateau (Porter, 2001). The input of P into the Chinese Loess Plateau is solely related to the particle size of aeolian dust. The second phase of P cycle in the Chinese Loess Plateau resulted from biological and geochemical processes, and the third phase was mainly controlled by atmosphere precipitation.

The investigations on P in modern soils, the loess and paleosols demonstrated that P_t, P_i and P_o are all enriched in the fine-grained fraction (Day et al., 1987; Weber et al., 1998; Makarov et al., 2004; Yokoo et al., 2004). The similar result was obtained as well by the determination of P_t in different grain-size fractions of loess L_1 and paleosol S_1 in the Luochuan loess section (Table 3): whether in loess or in paleosol, P_t tends to be enriched in the fine-grained fractions.

The loess in the Chinese Loess Plateau was accumulated under a climate dominated by a strong winter monsoon during glacial periods, while paleosols were developed under the climate of an intensified summer monsoon during interglacial periods (Liu, 1985; Li et al., 1988; Kukla and An, 1989; An et al., 1991; Rutter and Ding, 1993). Paleosols generally possess higher proportions of fine grain-sizes relative to coarse grain-sizes, and lower median grain sizes reflecting average value of grain-size distribution compared to the loess (Zheng and Whitton, 1994; Lu and An, 1997). So, if the effect of pedogenesis on P is neglected, P_t content of paleosols would be higher than that of the loess, and P would vary only as a function of grain sizes of aeolian dust. Under this circumstance, P in the loess section would be named as "initial P".

However, the results determined in this study show that P_t concentration is higher in the loess than in interstratified paleosols (Fig. 1), opposite to the above results on the assumption of no weathering-pedogenesis. Here, P_t determined in this study, as a mutual result of the effect of particle size sorting and pedogenesis, is called as "preserved P_t", and P leached out of the loess section by water is named as "leached P_t". Thus, there is a simple function about P_t in some loess or paleosol layer: "initial P_t" = "preserved P_t" + "leached P_t". It is

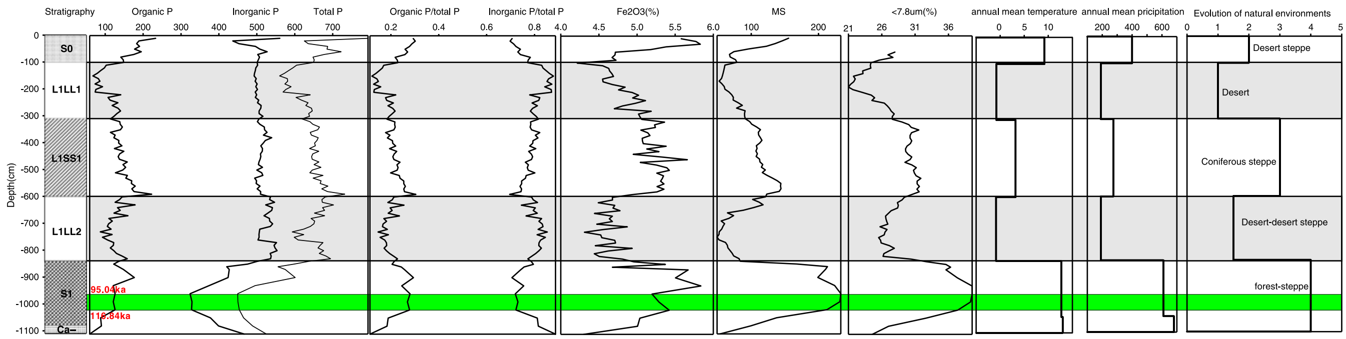

Fig. 2. P content (ppm), P ratios, Fe_2O_3 (%), MS, the <7.8 µm fraction, natural environment evolution, annual mean temperature (°C) and annual mean precipitation (mm) in the Luochuan S_0–L_1–S_1 sequence (Liu, 1985).

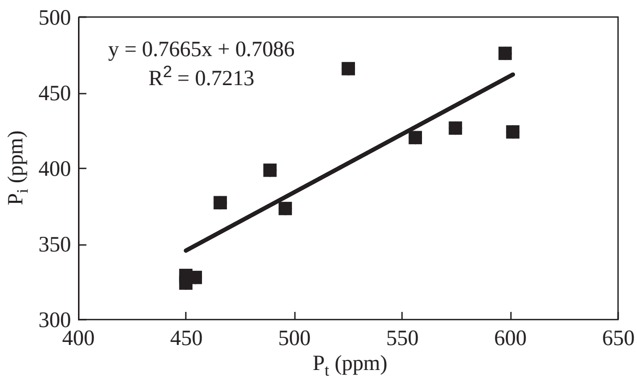

Fig. 3. Correlations between inorganic P (P_i) and total P (P_t) in paleosol S_1.

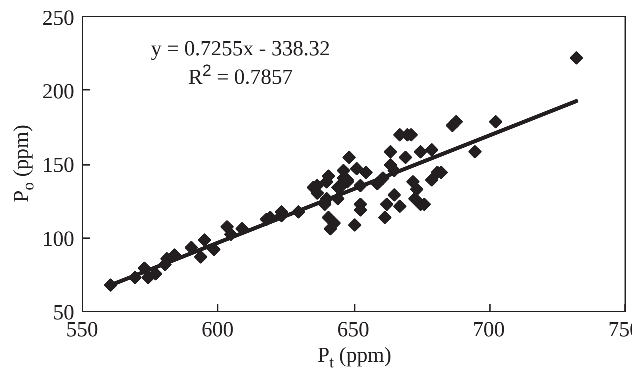

Fig. 4. Correlations between organic P (P_o) and total P (P_t) in loess L_1.

supposed that P_t of the loess overlaying the paleosol approaches initial P_t of the paleosol due to very weak pedogenesis on the loess (Ding et al., 2001a), then another equation is obtained: "leached P_t" (S_n)$\geqslant$"preserved P_t" (L_n)−"preserved P_t" (S_n), ($n = 1, 2, 3\ldots$). The results are calculated as shown in Table 4. Due to different intensity of pedogenesis, leached P_t in different paleosols is different in content. There is the maximum of "leached P_t" in S_5 and S_9, more than 306 and more than 349 ppm, respectively. The minimum of "leached P_t" is more than 87 ppm in S_{12}. Those indicate that pedogenesis had an important role in modifying P concentrations in the loess section, and atmosphere precipitation could be a very key factor to result in leaching of P_t in paleosols. So, variations of P_t in the loess–paleosol sequence with fluctuation cycles of 100 ka can be regarded as a good proxy indicator to reflect variations in precipitation.

4.2. Links between P geochemistry and paleoclimate change during the last 130 ka

Liu (1985) indicated that the natural environments varied from forest-steppe in S_1, to desert and desert-steppe in L_1LL_2, to coniferous steppe in L_1SS_1, then to desert in L_1LL_1 during the last 130 ka (Fig. 2). This result implied occurrence of P biological and geochemical processes whether in loess L_1 formed during the last glacial period or in paleosol S_1 developed during the last interglacial period.

The average temperature and the mean annual precipitation in the Luochuan area were around 0 °C and under 200 mm during the last stade and the early stade of the last glacial period, respectively, and were 3 °C and 260 mm during the interstade of the last glacial period, respectively (Fig. 2) (Liu, 1985). Due to low temperature and little precipitation with a lot of $CaCO_3$ (>10%) in loess L_1 (Liu, 1985), P could not be leached out of loess L_1, and P_t in loess L_1 observed in this study ("preserved P_t") was regarded to approximate "initial P_t", implying that P_t in the loess and paleosols is not sensitive to precipitation variations on less than 10 ka scale. Thus, "preserved P_t" is closely correlated with grain sizes as well as "initial P_t" does (Fig. 6), and is definitely expressed by a linear equation: $y = 8.6308x + 403.02$. The mean temperature was about 12 °C and the annual precipitation ranged from 600 to 750 mm during the last interglacial period (Fig. 2) (Liu, 1985). P in the paleosol S_1 probably suffered from weathering-pedogenesis. If the weathering-pedogenesis is not

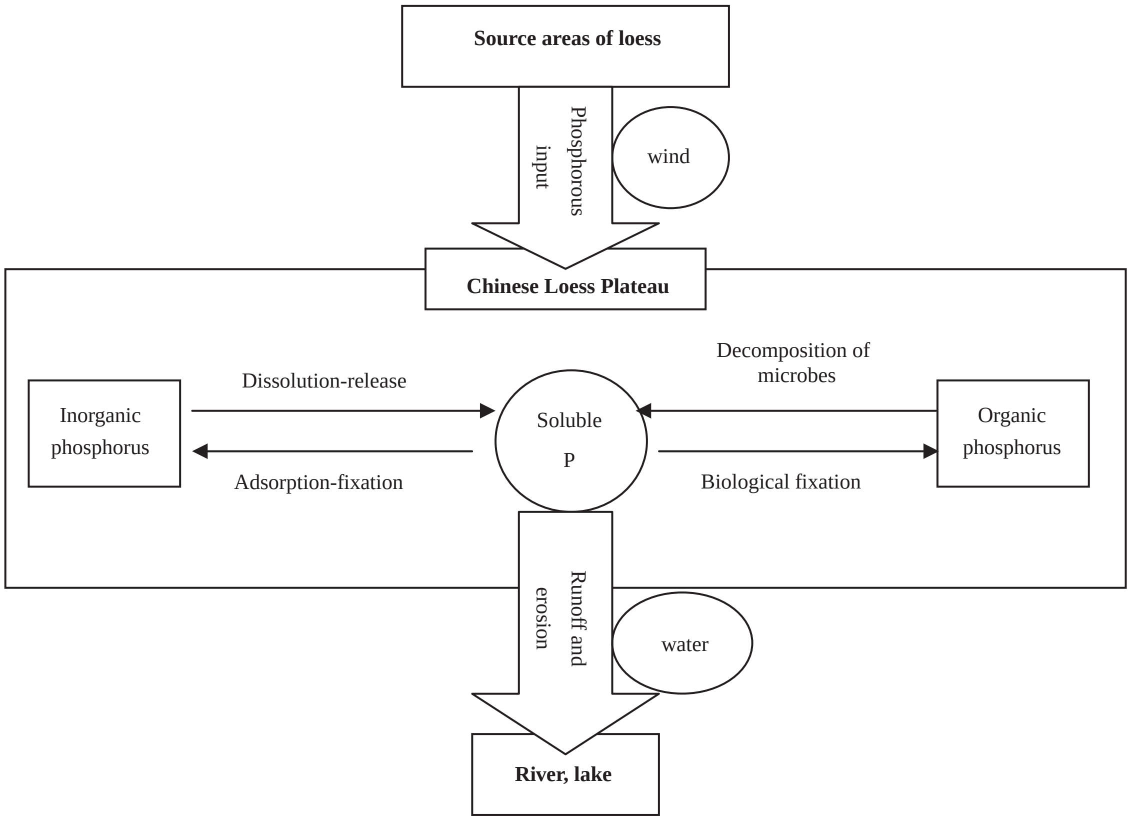

Fig. 5. Sketch map of phosphorous cycle in the Chinese Loess Plateau.

Table 4
Differences of P_t between in the loess and in underlying paleosols calculated as "leached P_t"(ppm) in paleosols by precipitation

Horison	P_t (ppm)	Horison	P_t (ppm)	(L_n-S_n)	(L_n-S_n)/L_n (%)
L1	699	S1	437	262	37.5
L2	611	S2	437	175	28.6
L3	699	S3	480	218	31.3
L4	611	S4	480	131	21.4
L5	699	S5	393	306	43.8
L6	655	S6	480	175	26.7
L7	742	S7	480	262	35.3
L8	786	S8	524	262	33.3
L9	742	S9	393	349	47.1
L10	742	S10	568	175	23.5
L11	655	S11	524	131	20.0
L12	699	S12	611	87	12.5
L13	642	S13	480	162	25.2
L14	568	S14	437	131	23.1
L15	611	WS1	568	44	7.1
WL1	611	WS2	437	175	28.6
WL2	568	WS3	524	44	7.7
WL3	611	WS4	437	175	28.6
WL4	611	Red clay	611	0	0.0

considered as an impact factor on P, the relationship between "initial P_t" and grain sizes in the paleosol S_1 can be also interpreted by the linear equation: $y = 8.6308x + 403.02$, based on analysis of links between grain size and pedogenesis (Rao et al., 2004). Therefore, "Initial P_t" and "leached P_t" are calculated as shown in Table 5. Leached P_t varies from low values to high values, and then to low values as depth of soil increases, as well as leaching ratio of P_t (leached P_t/initial P_t) does. Leached P_t content and P_t leaching ratio reach the maximum values of 273–295 ppm and 38–40%, respectively in the middle of paleosol S_1, indicating that there was a very warm and wet climate

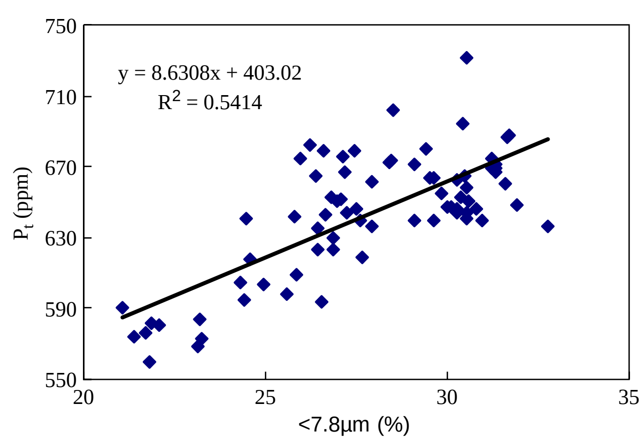

Fig. 6. Correlation between total P (P_t) and the $<7.8\,\mu m$ fraction loess L_1.

Table 5
Measured and calculated values of P_t (ppm) in the paleosol S_1

Sample no.	Depth (cm)	<7.8 μm (%)	Measured P_t	Calculated P_t	Δ (Difference)	Percentage (%)
PTXS1-3	850	35.68	598	711	113	16
PTXS1-4	860	36.39	556	717	161	22
PTXS1-5	870	35.69	575	711	136	19
PTS1-1	900	37.41	601	726	125	17
PTS1-2	930	39.60	495	745	249	33
PTS1-3	960	39.62	450	745	295	40
PTS1-4	990	39.35	450	743	293	39
PTS1-5	1020	37.45	454	726	273	38
PTS1-6	1050	32.65	465	685	220	32
PTS1-7	1080	27.06	489	637	148	23
PTS1-L2	1100	24.62	525	616	90	15

Note: Measured P_t in this study is "preserved P_t" and calculated P_t is "initial P_t". Δ ("Initial P_t"-"preserved P_t") is "leached P_t". Percentage is "leached P_t"/"initial P_t".

with much more precipitation during that period as compared to during other periods.

Chen et al. (1997) and Ding et al. (2001a) considered that weathering-pedogenesis of the loess and paleosols in the Chinese Loess Plateau was relatively weak, and only led to leaching of the diffluent elements such as Ca and Na. As a stable material in the Chinese Loess Plateau, Fe_2O_3 content was generally used as a good proxy indicator reflecting the East Asian summer monsoon variations (Sun et al., 1991). If the impact of weathering-pedogenesis on P_i was not considered or P_i was regarded as a stable element similar to Fe (Fig. 2), the distribution curve of P_i in the loess section would has been similar to that of Fe due to the effect of grain size sorting and leaching of diffluent elements (Ca and Na).

However, P_i observed in this study varies minimally with a range of 493–554 ppm in loess L_1, and changes from 324 to 476 ppm in paleosol S_1 with a deep and wide groove-type curve, not supporting the above hypothesis (Fig. 2). The reasonable interpretation for variations in P_i observed in this study is that weathering-pedogenesis associated with the East Asian summer monsoon affected weakly P_i in loess L_1 where P_i was altered as an increase of P_o without leaching loss of P, whereas strongly in paleosol S_1 where there is strong transformation of P_i and P_o with leaching of much P.

Variations between high and low values of P_o are in accordance with fluctuations between cold and dry climates during the last glacial period. The distribution curve of P_o in loess L_1 exhibits a similar oscillation with those of magnetic susceptibility and the <7.8 μm fraction, and there is a good correlation between P_o and magnetic susceptibility, and the <7.8 μm fraction (Figs. 7 and 8). Variations of P_o in loess L_1 contained mixed signals of both the East Asian winter and summer monsoons. Sparse vegetation was developed by absorbing soluble P in soil solution produced by dissolution of P_i such as apatite under the cold and dry climate that facilitated preservation of organic matter during the last glacial period. Thus, P_o tended to increase in content during the development of the Malan loess without leaching of P_t. However, the increase of P_o through transformation of P_i could be of a small quantity relative to initial P_o carried with aeolian dust from the North desert and Gobi areas in the Chinese Loess

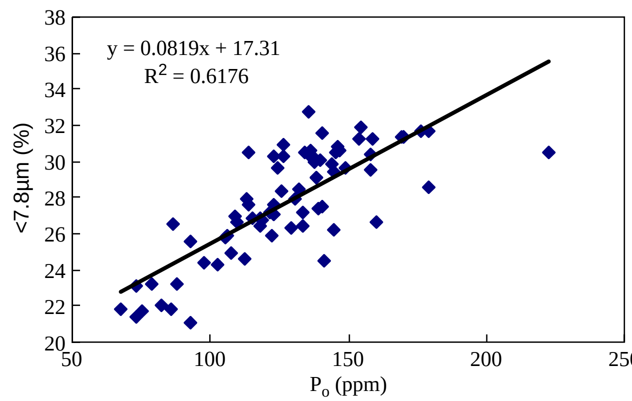

Fig. 7. Correlations between the <7.8 μm fraction and organic P (P_o) loess L_1.

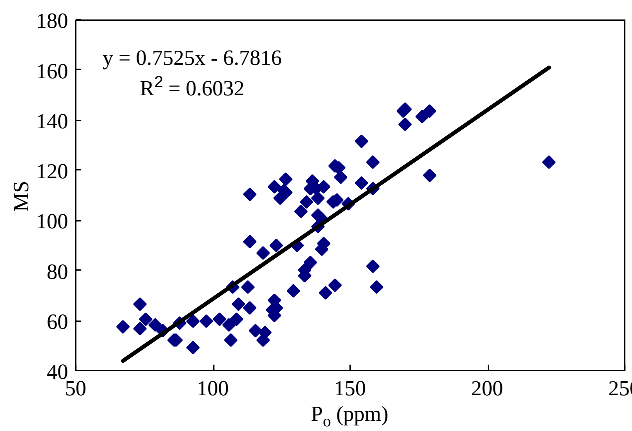

Fig. 8. Correlations between MS and organic P (P_o) in loess L_1.

plateau by the East Asian winter monsoon. So, Variations in P_o of the Malan loess could contain more information on the East Asian winter monsoon than on the summer monsoon. P_o in S_1 is lower than in L_1SS_1, and decreases with the increase of depth, indicating that the intensity of degradation of organic matter in S_1 was higher than in L_1. P_o in the middle of S_1 was constant during the interval of about 20 ka where P_i kept constant at the lowest value of 324 ppm, demonstrating that there was a very warm and humid climate during that time interval under which transformation of P_o and P_i arrived at a quasi-balance state and P_o/P_i ratio was kept constantly at 0.38. Precipitation directly restricted P concentration in solution within the loess and paleosols to change the transformation rate of P_o and P_i into soluble P, as well as the transformation rate of P_o with P_i. More precipitation facilitated the transformation of P_o and P_i into soluble P, and the transformation of P_o with P_i. As a result, more information on the summer monsoon than on the winter monsoon was imprinted on P_o in S_1 although P_o fluctuations in paleosol S_1 could not directly reflect the summer monsoon variations.

Consequently, P cycling was strong under a warm and humid climate, and was weak under a cold and dry climate. Climate change was an important and key driver for P cycling.

4.3. *P_o/P_i ratio is a good proxy indicator for the intensity of pedogenesis*

Fluctuations of P_o/P_i ratio in the S_0–L_1–S_1 sequence are shown in Table 2 and Fig. 9. P_o/P_i ratio varies from 0.14 to 0.44 in the S_0–L_1–S_1 sequence, and is the highest in the paleosol S_0 ranging from 0.28 to 0.43 with an average of 0.36, intermediate in the paleosol S_1 varying between 0.13 and 0.42 with a mean value of 0.31, and is the lowest in the Malan loess (L_1) ranging between 0.14 and 0.44 with an average of 0.25. The spatial distribution curve of P_o/P_i ratio in the S_0–L_1–S_1 sequence exhibits three significant fluctuations between low and high values. Similarly, the spatial distribution curve of P_o/P_i ratio in the Malan loess presents three small fluctuations between low and high values. P_o/P_i ratio is low in the loess L_1LL_1 ranging from 0.14 to 0.28 with an average of 0.21, medium in the loess L_1LL_2 varying between 0.17 to 0.34 with a mean value of 0.24, and high in the weak pedogenic loess L_1SS_1 ranging from 0.22 to 0.44 with an average of 0.29. Therefore, fluctuations of P_o/P_i ratio perfectly parallel climate change in the S_0–L_1–S_1 sequence.

Although P_o and P_i are both correlated with particle sizes of aeolian dust, P_o/P_i ratio can eliminate basically the effect of grain size sorting on itself due to chemical uniformity of aeolian dust entering the Chinese Loess Plateau and enrichment of P_o and P_i in the fine-grained fractions. Thus, input of P ("initial P") is not a main impact factor on the transformation of P_o and P_i. Temperature and precipitation are main indicators for the summer monsoon variations, and both change consistently in the Chinese Loess Plateau. Precipitation controls the amount of leached P out of the Chinese Loess Plateau but also brings about change of P concentration in soil solution to further influence the transformation rate of P into soluble P in soil and the transformation rate of P_o with P_i. Temperature is a very important impact factor on the transformation of P_o and P_i. If the temperature increases on the basis of sufficient precipitation, the activity of microbes will become more active, and the transformation between P_o and P_i, the transformation of P into soluble P in soils would be strengthened. Due to lower temperature and less precipitation, sparse vegetation was developed during the stade time or the glacial period, and P_i was dissolved more slowly with relative less accumulation of organic matter although there is not leaching of P. Thus, the transformation between P_o and P_i was relatively weak with low P_o/P_i ratio (in the L_1LL_1 and L_1LL_2, 0.21 and 0.24, respectively). On the contrary, due to higher temperature and more precipitation, vegetation was well developed during the interstade time and the interglacial period, P_i was dissolved more rapidly with relative more accumulation of the organic matter although there is leaching of much P. So, the transformation between P_o and P_i was strong, with high P_o/P_i ratio (in the S_0, L_1SS_1 and S_1, 0.36, 0.29 and 0.31, respectively).

Consequently, P_o/P_i ratio is high in paleosols with stronger pedogenesis, whereas low in the loess with weaker pedogenesis. These results reveal completely that P_o/P_i ratio is closely related to pedogenic activity in the

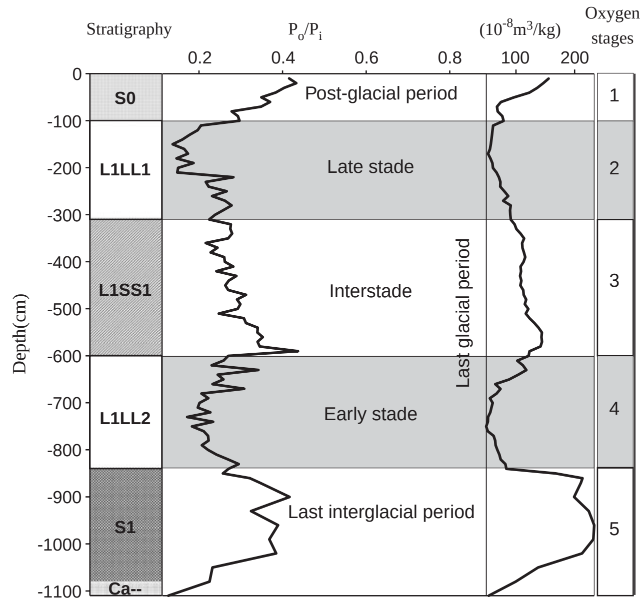

Fig. 9. The P_o/P_i fluctuations and records of magnetic susceptibility (χ) in the Luochuan S_0–L_1–S_1 sequence.

Luochuan loess section, and can be an effective indicator reflecting the intensity of pedogenesis for the loess section.

4.4. Comparison of P_o/P_i ratio with MS and SPECMAP $\delta^{18}O$

Magnetic susceptibility has been considered to be a classical proxy indicator of variations in the summer monsoon strength (An et al., 1991; Heller et al., 1993; Maher et al., 1994), although there are many different viewpoints on origin of magnetic susceptibility (Kukla et al., 1988; Zhou et al., 1990; Kletetschka and Banerjee, 1995). From Figs. 9 and 10, variation in P_o/P_i ratio closely resembles the magnetic susceptibility record in the S_0–L_1–S_1 sequence, implying that P geochemistry is in phase with the East Asian summer monsoon variations and controlled by temperature and precipitation. Furthermore, pedogenic activity recorded by P_o/P_i ratio is an important process of the East Asian summer monsoon variations on the Chinese Loes Plateau. Therefore, P_o/P_i ratio can be regarded as a good proxy indicator reflecting the East Asian summer monsoon evolution.

In addition, time sequences of P_o/P_i ratio in the S_0–L_1–S_1 sequence during the last 130 ka were calculated by using the method proposed by Kukla et al. (1988) (Fig. 11). Variations in P_o/P_i ratio with time are similar to SPECMAP $\delta^{18}O$, representing variations in continental ice volume of the Northern Hemisphere (Liu and Ding, 1990). The two curves of P_o/P_i ratio and SPECMAP $\delta^{18}O$ indicating different climate information oscillates in synchrony during the last 130 ka, which implies existence of a close relationship in driving forces between variations in the continental ice volume and the East Asian summer monsoon evolution.

During the glacial period, the ice fluxes in the Northern Hemisphere expanded, and the East Asian winter monsoon was strengthened, leading to decreased precipitation and declining temperature in the Chinese Loess Plateau, further weakening pedogenesis. On the contrary, during the interglacial period, ice withdrew in

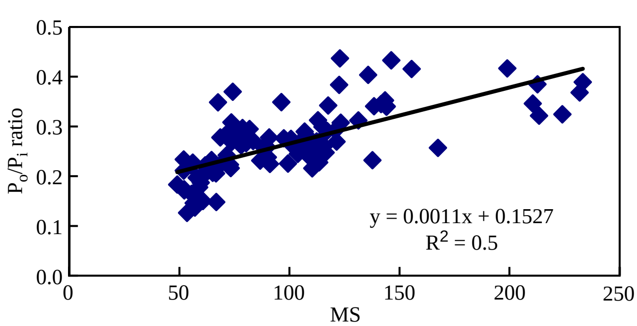

Fig. 10. Correlation between P_o/P_i ratio and magnetic susceptibility (χ) in the Luochuan S_0–L_1–S_1 sequence.

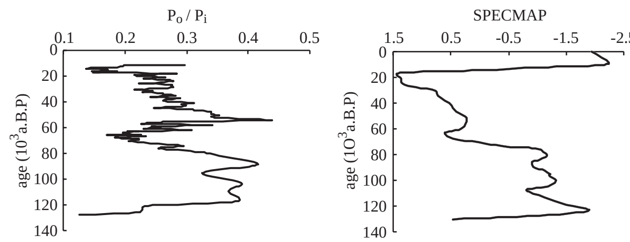

Fig. 11. Matching of the curve of P_o/P_i to marine oxygen-isotope records (SPECMAP$\delta^{18}O$) (Kukla et al., 1988) during the last 130 ka. The formula proposed by Kukla et al. (1988), is used to calculate ages for the Luochuan S_0–L_1–S_1 sequence.

the Northern Hemisphere, and the East Asian summer monsoon circulation was strengthened, resulting in increased precipitation and rising temperatures, further enhancing pedogenesis. The consistency of fluctuations in P_o/P_i ratio with the variation of the marine oxygen isotopic compositions reflecting variations in the continental ice volume in the Northern Hemisphere provides a new biogeochemical evidence for better understanding inner links between the global climate change and the East Asian summer monsoon evolution.

5. Conclusions

P was initially carried into the Chinese Loess Plateau with aeolian dust from the desert and Gobi areas in North China by the East Asian winter monsoon. P was redistributed in the Luochuan loess section due to weathering-pedogenesis after aeolian dust from the desert and Gobi areas was accumulated near Luochuan. P_t content on a carbonate-free basis is obviously higher in the loess than in adjacent paleosols in the Luochuan loess section. Relatively high content of P_t in the loess can be attributed to weak chemical weathering with little precipitation, whereas relatively low content of P_t in paleosols resulted from strong chemical weathering with much precipitation. The fluctuations of P_t in the loess–paleosol sequence indicate evolution of paleoprecipitation associated with the East Asian summer monsoon with a cycle of 100 ka during the Quaternary period. P_o in the S_0–L_1–S_1 sequence varies between 59 and 233 ppm with the average of 132 ppm, accounting for below 30% of P_t. P_i in the S_0–L_1–S_1 sequence varies from 324 to 560 ppm with the mean content of 499 ppm, accounting for above 70% of P_t. Primary P_o and P_i in the Luochuan loess section were partially suffered from pedogenesis during the last 130,000years, and P_o and P_i determined in this study recorded the mixed information of the East Asian summer monsoon and the winter monsoon. A little P_i was transformed into P_o in L_1 without loss of P, and much P_i was leached in S_1. P_o increased as a result of P_i losses, but the majority of P_o

from parent areas was not altered in L_1. Thus, P_o contains more information on the winter monsoon in L_1. However, P_o from parent areas was mostly altered in S_1, thus contains more imprints of the summer monsoon although it could not directly reflect the summer monsoon variations. P_o/P_i ratio with clear climate significance, tightly correlated with pedogenesis of the Luochuan loess section, can be regarded as a good proxy indicator reflecting the East Asian summer monsoon evolution, as new biogeochemical evidences provided for further understanding the inner links between the East Asain monsoon evolution and gobal climate change.

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