Front. Earth Sci. China 2009, 3(3): 266–272
DOI 10.1007/s11707-009-0037-1

RESEARCH ARTICLE

CPI values of terrestrial higher plant-derived long-chain *n*-alkanes: a potential paleoclimatic proxy

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Abstract Carbon Preference Index (CPI values) of higher plant-derived long-chain *n*-alkanes extracted from 62 surface soil samples in eastern China exhibited a specific pattern of variations, namely gradual increase with the increasing latitudes. Such regular variations existed in both forest soil and grassland soil. Our data implied that CPI values of higher plant-derived long-chain *n*-alkanes had a certain connection with climatic conditions, and such a connection was not influenced by vegetation types. Together with previous data from marine sediments, loess/paleosol sequences, tertiary red clay and modern plants, our observation made us conclude that CPI values of higher plant-derived long-chain *n*-alkanes may be used as an excellent proxy for paleoclimatic studies.

Keywords long-chain *n*-alkanes, CPI values, paleoclimatic proxy

1 Introduction

With strong anti-diagenesis ability, relative stable chemical property and naturally high content in sediments (Laboratory of Organic Geochemistry and Sedimentation, Institute of Geochemistry, Chinese Academy of Sciences, 1982), *n*-alkanes have been gaining increasing concern in biogeochemical studies for past global change, especially long-chain *n*-alkanes with significant odd-to-even carbon number predominance (Street-Perrott et al., 1997; Huang et al., 2001 ; Schefuß et al., 2005; Rao et al, 2008). Previous investigations have demonstrated that terrestrial higher plants produced plentiful long-chain *n*-alkanes with significant odd-to-even carbon number predominance (Eglinton and Hamilton, 1967; Rieley et al., 1991a). Long-chain *n*-alkanes with such characteristics have been generally thought to be mainly derived from higher plants. Therefore, they have great implications in paleovegetation and paleoenvironmental studies (Freeman et al., 1990; Rieley et al., 1991b).

At present, studies on higher plant-derived long-chain *n*-alkanes of past global change focused mainly on three aspects: 1) Analyses of molecular distribution of long-chain *n*-alkanes with significant odd-to-even carbon number predominance have been employed to reflect the process of plaeovegetational evolution (Wang et al., 2004; Liang et al, 2005; Yang et al., 2006; Zhong et al., 2007; Liu et al., 2008; Yang et al., 2008). 2) Compound-specific carbon isotopic composition of long-chain *n*-alkanes with such characteristics has been used to reconstruct past change of relative contribution of C_3/C_4 plants to source vegetation (Street-Perrott et al., 1997; Brincat et al., 2000; Huang et al., 2001; Zhang et al., 2003). 3) Compound-specific hydrogen isotopic composition of long-chain *n*-alkanes with such characteristics has been investigated for paleoenvironmental reconstruction (Bi et al., 2005; Krull et al., 2006; Pagani et al., 2006; Smith and Freeman, 2006; Jia et al., 2008), such as paleohydrological changes (Schefuß et al., 2005).

Carbon Preference Index (CPI values) is an important parameter for the description of the molecular distribution characteristics (odd-to-even carbon number predominance) of long-chain *n*-alkanes. CPI values reflect the differences of the relative concentrations between odd-carbon-number and even-carbon-number long-chain *n*-alkanes in a certain carbon number range. More significant odd-to-even carbon number predominance, larger the differences, higher the CPI values, and vice versa. Until now, CPI values of long-chain *n*-alkanes have been widely reported in red earth (Liang et al., 2005), lacustrine sediments (Brincat et al., 2000; Ficken et al., 1998; Huang

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Received December 8, 2008; accepted April 14, 2009

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et al., 1999), marine sediments (Ishiwatari et al., 1994; Yamada and Ishiwatari, 1999), loess/paleosol sequences (Wang et al., 2004; Xie et al., 2004; Liu et al., 2005), Tertiary red clay (Liu et al., 2008) and peat bog (Yang et al., 2008). However, modern process studies on CPI values of long-chain *n*-alkanes aimed at their climatic and environmental implication were rare. This paper studied CPI values of long-chain *n*-alkanes from the surface soils under different vegetation types along latitudinal gradient in eastern China. Our objective was to investigate if there are certain variation pattern along this latitudinal gradient and their climatic and environmental indicative significances.

2 Materials and methods

62 surface soil samples were collected from eastern China, which were distributed in 11 provinces from Hainan to Heilongjiang ranging ca. 18°N to 50°N (Fig. 1). The vegetation types in the study region vary from tropic rainforest in the southern part of Hainan to cold-temperate coniferous and deciduous forest in the northeastern China. The whole study region is influenced by monsoon climate, with most precipitation falling in summers when the temperature is high. Both the mean annual precipitation (MAP, ca. 500–2500 mm) and mean annual temperature (MAT, ca. 0°C–26°C) are obviously latitude-depend, namely both of them gradually decreased northwards in the study region.

Most soil samples were collected from surface layer of 2 cm of the weathering crust of basalt or other seeable bedrocks and all the sampling sites were carefully chosen far away from the cropland and cities to avoid strong human disturbances. Usually one to three surface soil samples were collected from different physiographic locations or under different vegetation types for a single sampling site (Fig. 1). Field investigation and sampling were carried out from September to November 2005.

All the samples were analyzed at the State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences. After removal of modern rootlets and gravels by sieving, fine-ground subsamples of ~10 g were dipped in methylene chloride (CH_2Cl_2) for ca. 2 hours and then extracted ultrasonically. Each sample was extracted three times, ca. 10 minutes for each time. The extracts were concentrated by rotor-evaporator, and then the total lipid extracts were separated by silica gel flash-column chromatography. Elution with hexane gave the aliphatic hydrocarbon fraction, containing the long-chain *n*-alkanes. The hexane fractions were analyzed using an HP 6890 gas chromatograph (GC) equipped with an HP-5 MS fused silica capillary column (30 m×0.32 mm×0.25 μm). The oven temperature program was 80°C (2 min) to 220°C (hold 2 min) at 10 °C/min, and then to 290°C (hold 15 min) at 3 °C/min. A set of *n*-alkanes (n-C_{12}, n-C_{14}, n-C_{16}, n-C_{18}, n-C_{20}, n-C_{22}, n-C_{25}, n-C_{28}, n-C_{30} and n-C_{32}) from the University of Indiana (Indiana STD) were used as the reference material and the components were GC analyzed daily with the same measuring machine and procedure of samples. The carbon numbers of the extracted *n*-alkanes were determined by comparison with the retention times of the reference material (Fig. 2), and then the relative abundances of the extracted *n*-alkanes homologues were determined by their peak areas. The CPI values of the extracted long-chain *n*-alkanes (n-C_{22} to n-C_{34}) were calculated using a modified formula of Cranwell (1984) and Ratnayake et al. (2006) as following:

$$\mathrm{CPI} = 0.5 \times \left[\frac{C_{23}+C_{25}+C_{27}+C_{29}+C_{31}+C_{33}}{C_{24}+C_{26}+C_{28}+C_{30}+C_{32}+C_{34}} + \frac{C_{23}+C_{25}+C_{27}+C_{29}+C_{31}+C_{33}}{C_{22}+C_{24}+C_{26}+C_{28}+C_{30}+C_{32}}\right]$$

3 Results and discussion

Carbon numbers of extracted *n*-alkanes for most samples ranged from n-C_{14} to n-C_{35}. The total extracted *n*-alkanes had a bimodal distribution characteristics, namely short-chain *n*-alkanes were dominated by n-C_{17} or n-C_{19} homologues and long-chain *n*-alkanes were dominated by n-C_{27}, n-C_{29} or n-C_{31} homologues. The significantly low concentrations of short-chain *n*-alkanes relative to long-chain *n*-alkanes (Fig. 2) indicate that the *n*-alkanes extracted from these surface soils were dominated by long-chain homologues. Obviously short-chain *n*-alkanes had no significant odd-to-even carbon number predominance, however, long-chain *n*-alkanes had such characteristics (Fig. 2).

CPI values of long-chain *n*-alkanes extracted from all the surface soils ranged from ca. 3 to 12.8 with an average value of ca. 6.3. Previous studies demonstrated that *n*-alkanes from low organism (such as, bacteria and algae) were short-chain homologues without significant odd-to-even carbon number predominance, and their carbon numbers ranged from n-C_{15} to n-C_{20} mainly with a unimodal distribution characteristics which were dominated by n-C_{17} or n-C_{19} homologues (Cranwell, 1981; Cranwell et al., 1987; Meyers and Ishiwatari, 1993). Terrestrial higher plants produced plentiful long-chain *n*-alkanes with significant odd-to-even carbon number predominance that usually dominated by n-C_{27}, n-C_{29} or n-C_{31} homologues, and their CPI values normally larger than 5 (Eglinton and Hamilton, 1967; Rieley et al., 1991a). The molecular distribution characteristics of the *n*-alkanes extracted from our surface soils indicated that the long-chain fractions were largely derived from terrestrial higher plants and not significantly altered by diagenesis.

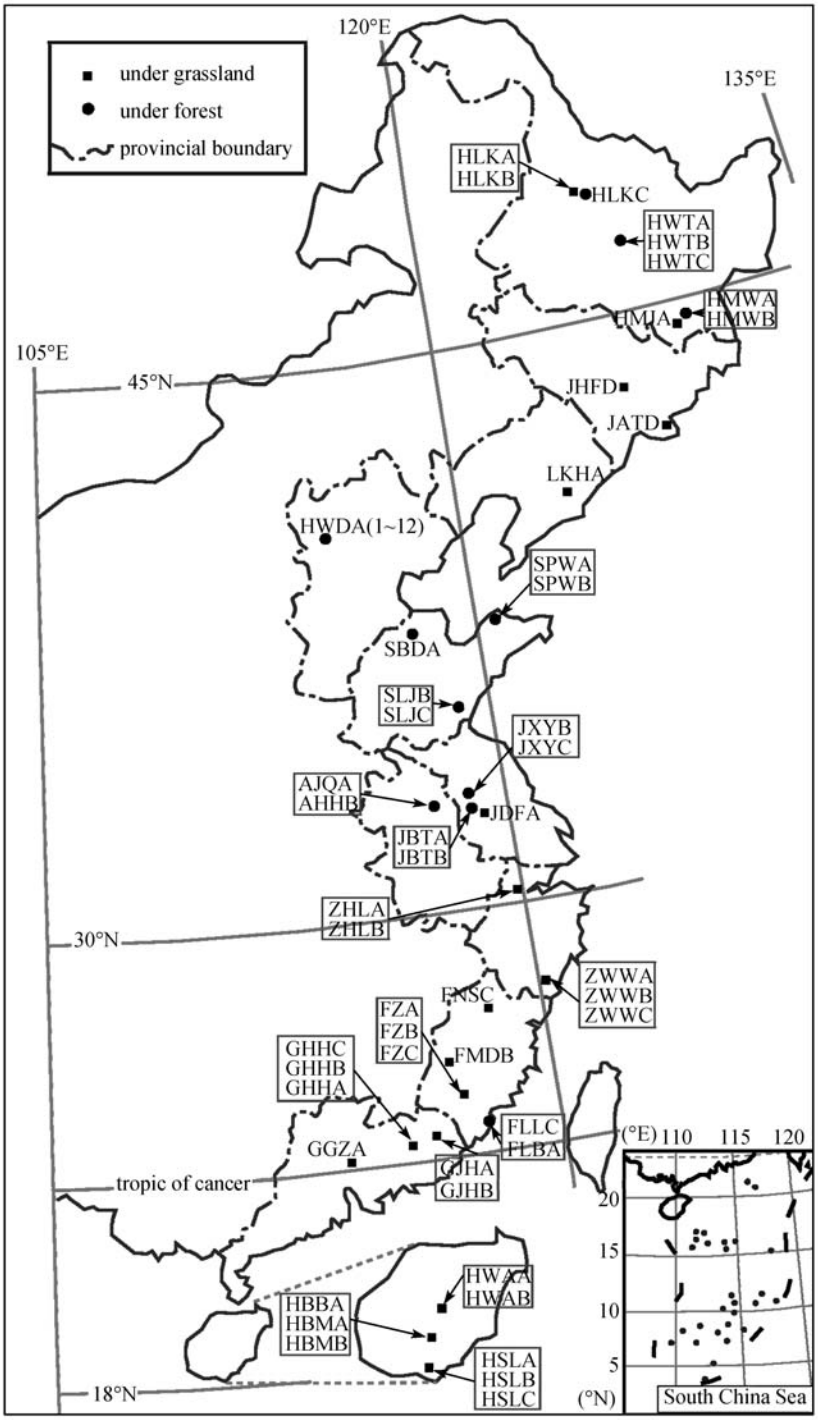


Fig. 1 Sketch map showing the study region and locations of the sampling sites

CPI values of surface soils under grasslands ranged from 3.39 to 10.1 with an average value of ca. 7, and those under forests ranged from 3.04 to 12.75 with an average value of ca. 5.5, respectively. Though the average CPI value of surface soils under grasslands was larger than that under forests, the numerical range of CPI values under grasslands fell into same zone as those under forests (Fig. 3(a)). This means CPI values of higher plant-derived long-chain *n*-alkanes had less possibility to be treated as paleovegetational type diagnostic discriminator.

Both the CPI values of surface soils under forests and grasslands were increased northwardly (Fig. 3(a)), though only the correlation between CPI values under forests and latitudes was significant ($n = 31$, $R^2 = 0.647$, $p < 0.001$ for

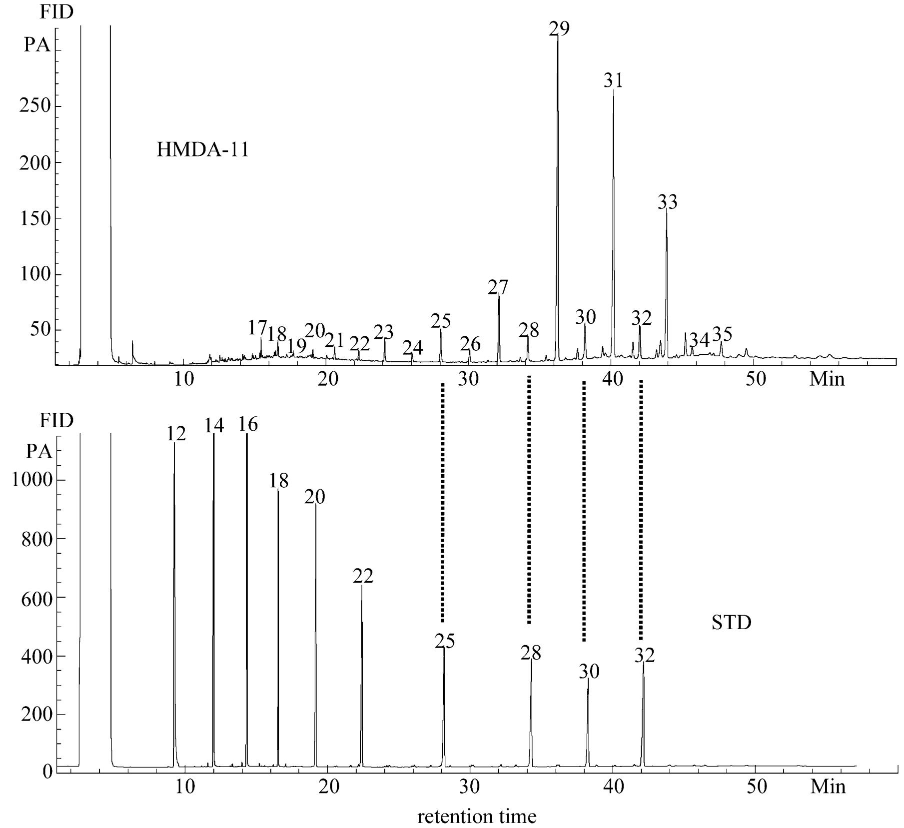


Fig. 2 Retention times of n-alkanes extracted from surface soil sample (HMDA-11, Fig. 1) comparison with the reference material (Indiana STD)

surface soils under forests; $n = 31$, $R^2 = 0.177$ for surface soils under grasslands). As a whole, CPI values of long-chain *n*-alkanes extracted from surface soils in eastern China displayed a generally increasing trend from low to high latitudes ($n = 62$, $R^2 = 0.518$, $p < 0.001$, Fig. 3(b)). The significant correlation between surface soil long-chain *n*-alkanes CPI values and latitudes may imply their environmental significance. We speculated that latitudinal climatic variation may have significant influence on physiological process of the terrestrial higher plants, namely, more odd carbon number long-chain *n*-alkanes be produced relative to even carbon number homologues under cold and dry climatic conditions. Studies on the long-chain *n*-alkanes extracted from plant leaf samples from 10 broad-leaf plant species growing at tropical (6°S, Bidadari Island, Indonesia), subtropical (27°N, Chichi-Jima Is, western North Pacific), and temperate (43°N, Sapporo, Japan) regions, found that CPI values in the subtropical and temperate plants ranged from 9.5 to 28.7 with an average of 23.7 and median of 24.0 whereas the *n*-alkanes in the tropical plant leaves showed lower CPI values (around 5.3) (Kawamura et al., 2003). More efforts are needed to discover the real causes of this phenomenon.

Previously reported data from two Japan sea marine sediment cores (KH-79-3, C-3 and L-3) (Ishiwatari et al., 1994; Yamada and Ishiwatari, 1999) indicated that the variation of CPI values (n-C_{23}–n-C_{35} *n*-alkanes) was in consistent with glacial/interglacial cycles, with lower CPI values occurring in warmer and wetter interglacial periods (Fig. 4(a)). CPI values (n-C_{22}–n-C_{35} *n*-alkanes) from loess/paleosol sequences of Yuanbao section, Linxia Basin, western Chinese Loess Plateau demonstrated that CPI values of paleosol layers representing warmer and wetter climatic conditions (MIS1, MIS3 and MIS5) were lower than that of loess layers representing colder and drier climatic conditions (MIS2 and MIS4) (Xie et al., 2004). Similar variation characteristics of CPI values came from Luochuan and Xunyi sections (n-C_{24}–n-C_{34} *n*-alkanes) in eastern Chinese Loess Plateau (Zhang et al., 2006) and Xifeng section (n-C_{22}–n-C_{32} *n*-alkanes) in central Chinese Loess Plateau (Liu et al., 2005) (Fig. 4(b)). Average CPI value (n-C_{22}–n-C_{32} *n*-alkanes) of 24 samples from the Holocene paleosol (S_0) and the last glacial loess (L_1) of Xifeng section in central Chinese Loess Plateau was ca. 6.1 (Liu et al., 2005) (Fig. 4(b)). However, average CPI values (n-C_{22}–n-C_{32} *n*-alkanes) of 18 samples from the last interglacial paleosol (S_1) of Yuanbao section, Linxia Basin, western Chinese Loess Plateau was 7.3 (Wang et al., 2004). Due to the data from Xifeng section, which included higher CPI values of the last glacial loess layer

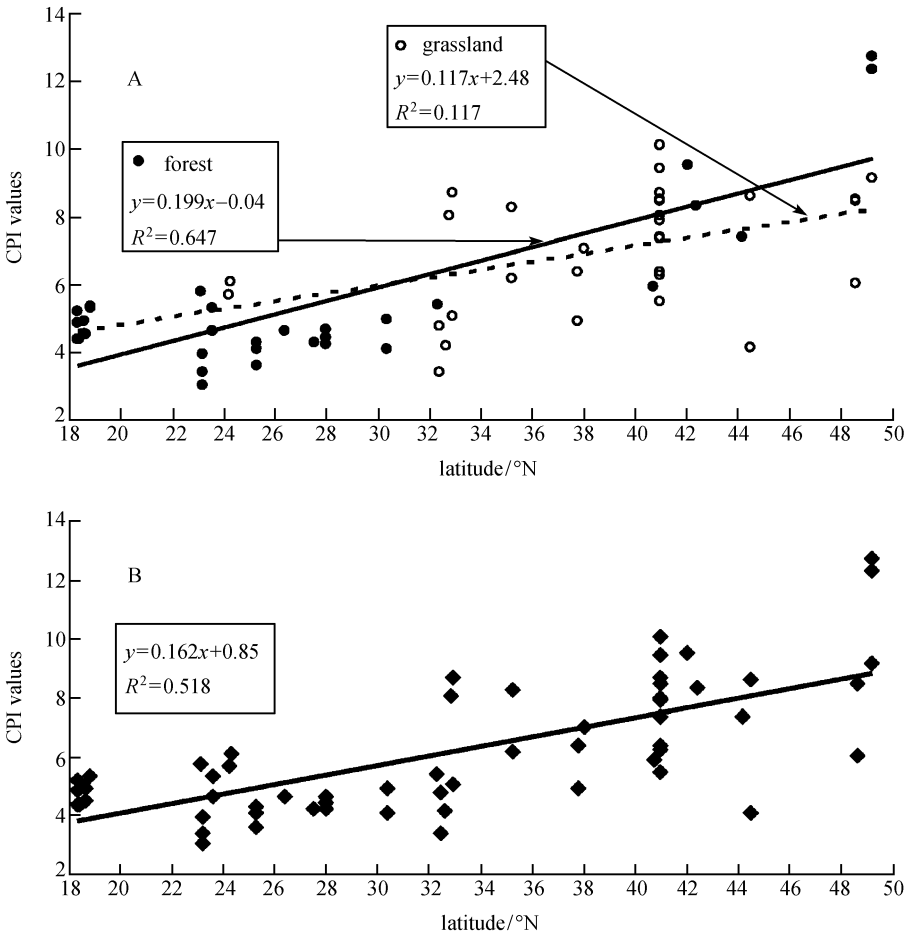
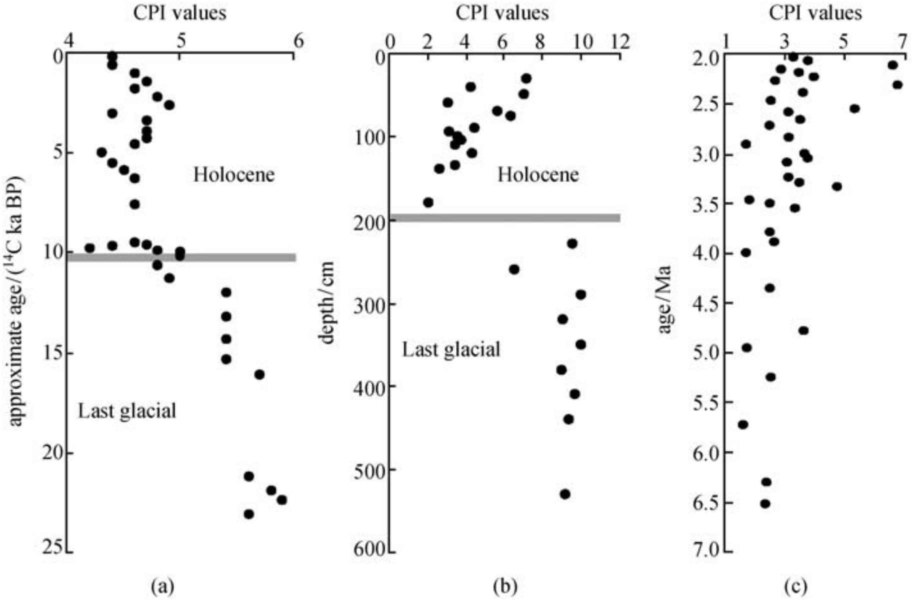


Fig. 3 Spatial variation of CPI values of long-chain *n*-alkanes (*n*-C_{22} to *n*-C_{34}) extracted from surface soils in eastern China. (a)surface soils underlying grassland and forest respectively; (b) all surface soils in east China



(a) (b) (c)

Fig. 4 (a) CPI values of *n*-C_{23}–*n*-C_{35} *n*-alkanes from Japan sea marine sediment core (KH-79-3, L-3) (Yamada and Ishiwatari, 1999); (b) CPI values of *n*-C_{22}–*n*-C_{32} *n*-alkanes from Xifeng loess/paleosol sequences in central Chinese Loess Plateau (Liu et al., 2005); (c) CPI values of *n*-C_{22}–*n*-C_{34} *n*-alkanes from Xifeng Tertiary red clay sequences in central Chinese Loess Plateau (Liu et al., 2008).

(L_1), the average CPI value of the Holocene with warmer and wetter conditions was approximately 4.2(Liu et al., 2005) (Fig. 4(b)). If we assume the climatic conditions of the Holocene and the last interglacial was similar in a certain place, we can conclud that CPI values of long-chain *n*-alkanes were lower in eastern Chinese Loess Plateau (ca. 4.2 in S_0 of Xifeng) under warmer and wetter climatic conditions and higher in the western Chinese Loess Plateau (ca. 7.3 in S_1 of Yuanbao) under colder and drier climatic conditions. Obviously, previously reported data from the Chinese Loess Plateau suggested that CPI values of long-chain *n*-alkanes were lower under warmer and wetter climate and higher under colder and drier climate in both temporal and spatial sequences. Recently reported data from Tertiary red clay (Zhaojiachuan section in Xifeng area) showed that CPI values (n-C_{22}–n-C_{34} *n*-alkanes) had an increasing trend from ca. 7 Ma to 2 Ma (Liu et al., 2008) (Fig. 4(c)), with the same temporal sequence, regional climate became more and more cold and dry (Sun et al., 1998; 2001).

In eastern China, CPI values of terrestrial higher plant-derived long-chain *n*-alkanes extracted from surface soils underlying both grasslands and forests increased from low latitudes to high latitudes. Such variation characteristics was apparently in consistent with previously reported paleo-data from marine sediments (Ishiwatari et al., 1994; Yamada and Ishiwatari, 1999), loess/paleosol sequences (Wang et al., 2004; Xie et al., 2004; Liu et al., 2005) and Tertiary red clay (Liu et al., 2008). This consistency indicated that sedimentary CPI values of higher plant-derived long-chain *n*-alkanes had a certain relationship with climatic conditions, and this relationship was independent of vegetational types.

4 Conclusions

GC analyses has been conducted on 62 surface soil samples underlying different vegetational types in eastern China spanning from 18°N to 50°N. Molecular distribution characteristics of long-chain *n*-alkanes extracted from those surface soils demonstrate their terrestrial higher plant origin and they are not significantly altered by diagenesis. CPI values of those long-chain *n*-alkanes (n-C_{22} to n-C_{34}), serving as a parameter of odd-to-even carbon number predominance, increased northwardly with the decreasing temperature and precipitation. Such variation pattern existed in surface soils underlying both grasslands and forests, suggesting that the relationship between CPI values of surface soil long-chain *n*-alkanes and latitudinal climatic conditions was independent of vegetational types. CPI value of terrestrial higher plant-derived long-chain *n*-alkanes can potentially used as an excellent paleoclimatic indicator.

Acknowledgements This research was supported by the Knowledge Innovation Program of Chinese Academy of Sciences (No. KZCX3-SW-152), the National Natural Science Foundation of China (Grant No. 40672121), China Postdoctoral Science Foundation (No. 20080430792) and the NSFC National Innovative Research Team Project (No. 40721061). The authors were grateful to Dr. HAN Jiangwei and Mr. JIA Wanglu for assistance in the field and lab analysis. Thanks also go to the reviewers and editor who help improve this manuscript.

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