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A Carbon Isotopic Stratigraphic Pattern of the Late Palaeozoic Coals in the North China Platform and Its Palaeoclimatic Implications

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Abstract This paper gives the stable carbon isotopic data in coals from the Late Namurian to Kazanian stages in the Serteng Mt., Xishan and Huainan coalfields of the North China Platform. Its stratigraphic pattern shows that several isotopic shifts are apparent, and the large $\delta^{13}C$ negative shifts (approximately 2.5 to 3.0 ‰) occurred during the Stephanian, Artinskian and Kazanian are observed in three Permo-Carboniferous coalfields. Those negative shifts are neither related to the coal rank and coal macerals, nor caused by the variety of peat-forming plants. The general decrease in the $\delta^{13}C$ values of the Stephanian, Artinskian and Kazanian coals is consistent with an overall decrease in the $\delta^{13}C$ values of ambient atmospheric CO_2 and/or a relative increase in atmospheric P_{CO_2} during the coal-forming periods. Therefore the authors postulate that the oxidation of peat, and the $\delta^{13}C$-depleted CO_2 flux into the atmosphere during the above stages may have contributed to coeval palaeoclimatic warming by way of the greenhouse effect.

Key words: carbon isotope, coal, palaeoclimate, Late Palaeozoic, the North China Platform

1 Introduction

The increase of CO_2 and other greenhouse gases such as CH_4 in the atmosphere is an important cause for palaeoclimatic warming in geological history, and also a major parameter for making GCM simulations of palaeoclimate (Chandler et al., 1992). The increase of CO_2 and sharp reduction of biomass during the mass extinction in the terminal Late Palaeozoic have been proved by the negative shift of the $\delta^{13}C$ value of kerogen from marine shales in Canada (Wang K. et al., 1994). The carbon isotopic measurement of the Late Permian *Diictodon* (a mammal-like reptile) teeth by Tackoray and others (1990) shows an reduction of its $\delta^{13}C$ value by 6 ‰ in the Tatarian. As the variation in $\delta^{13}C$ of animals generally reflect their food sources (Faure, 1977), the sharp reduction of the $\delta^{13}C$ value for *Diictodon* teeth in the Tatarian should coincide with that of the plants they ate. If this inference holds water, coal, as the "compacted body" and burial form of plants, can better record the biological $\delta^{13}C$ variations.

The study of carbon isotope composition in coals of China has made great achievements (Duan, 1995; Xu and Shen, 1991), and has been used to determine the sources and the carbon isotopic differences of organic macerals in coals as well as the geochemical behaviours of organic matter in coalification. Based on the carbon isotope data of the Late Palaeozoic coals in the North China Platform, this paper attempts to establish a stratigraphic pattern and further discusses its palaeoclimatic implications.

2 Carbon Isotopic Stratigraphy of the Late Palaeozoic Coals in the North China Platform

2.1 Sampling coal beds and correlation of coal-bearing strata

The samples are collected from the Late Palaeozoic coal beds in the northern (Serteng Mountain) and southeastern (Huainan Coalfield) parts of the North China platform (Fig. 1). The coal beds sampled include 2 beds of the Lower Shetai Formation, 4 of the Upper Shetai Formation, and 14 of the Shuanmazhuang For-

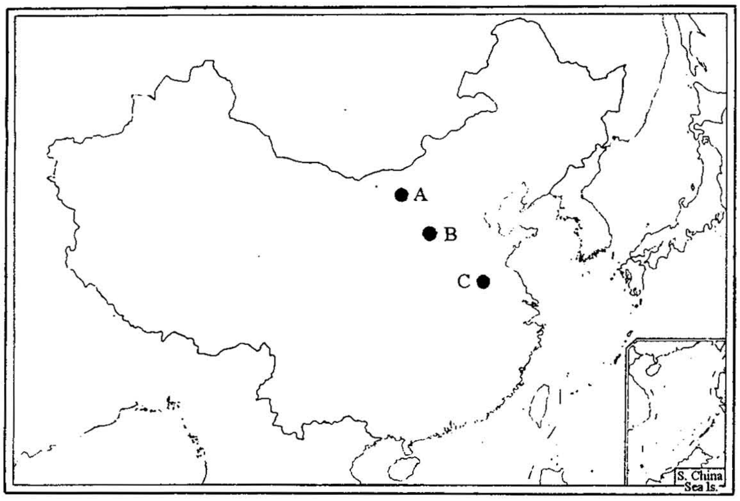

Fig. 1. Sampling locations.

A–Serteng Mt. coalfield, Inner Mongolia; B–Xishan coalfield, Taiyuan, Shanxi Province; C–Huainan coalfield, Anhui Province.

mation in the Serteng Mountain area; and 3 of the Shanxi Formation, 7 of the Lower Shihezi Formation and 5 of the Upper Shihezi Formation in the Huainan coalfield. Besides, data of 17 coal beds of the Benxi, Taiyuan and Shanxi formations in Xishan (the West Hills) coalfield, Taiyuan are also quoted (Institute of Geological Exploration, CCMRI et al., 1987; Zhuang, 1987).

The correlation of coal beds of the Permo-Carboniferous coalfields in the North China Platform is a very complicated problem, but the chronostratigraphic correlations of the formations they belong to have been largely determined. For convenience, the "IUGS 1989 Global Stratigraphic Chart" (Cowie and Bassett, 1989) is used in this paper for a brief illustration of the above issue.

According to recent studies, the Upper and Lower Shetai formations represent the Namurian B-C and the Westphalian deposits respectively (Zhang, 1997); the Shuanmazhuang Formation corresponds only to the lower Taiyuan Formation (Jinci Member) of the Xishan coalfield, Taiyuan, and may be correlated with the Stephanian (Wu, 1995; Zhang, 1987, 1989, 1997); the upper Taiyuan Formation of Central Shanxi (Mao'ergou and Qiligou members) is representative of the Asselian deposits; the Shanxi Formation corresponds to the Sakmarian to early Artinskian (Shen, 1995), the Taiyuan and Shanxi formations in Huainan are not strictly isochronous with those bearing the same names in Central Shanxi (Shang, 1997); and the Lower and Upper Shihezi formations roughly correspond to the late Artinskian to the Kazanian. Fig. 2 (left part) shows the chronostratigraphic correlations of the lithostrati-graphic units of the above-mentioned three places. It not only gives out basic time constraints for the various coal beds, but also provides information for constructing a carbon isotopic stratigraphic pattern of the Late Palaeozoic coals in the North China Platform.

2.2 Results of analysis and negative shift of the $\delta^{13}C$ value

Carbon isotope data for major coal beds in the Serteng Mountain, West Hills of Taiyuan and Huainan Coalfield are given in Table 1. It should be noted that the $\delta^{13}C$ values given in the table are mean values as 2–5 samples are taken for most of the coal beds.

Coal beds of the Serteng Mountain consist of highly-coalified bituminous and meagre coals (R_{max} > 1.58), those of the West Hills consist of bituminous coal and anthracite (R_{max}: 1.12 – 2.46%), while those in the Huainan Coalfield consist of gas coal, fat coal and primary coking coal (Rmax: 0.688 – 1.046%). As can be seen from Table 1, the $\delta^{13}C$ values for coal beds of various areas range between –26.20 ‰ (Coal bed 25 in Huainan) and –22.10 ‰ (coalbed 31 ofthe Lower Shetai Formation in the Serteng Mountain), averaging –24.16 ‰, which is very close to the average $\delta^{13}C$ value (–24.40 ‰) of coals of various ages in China, but quite different from those of Permian coals of the Gondwanaland (Faure et al., 1995) and the algal coals of China (Duan, 1995) (Table 2).

As is mentioned before, the $\delta^{13}C$ values of the Late Palaeozoic coals in the North China Platform analyzed cover all the geological time intervals from the late Namurian to the Kazanian. Fig. 2 shows their stratigraphic pattern on the basis of correlations of the coal-bearing strata. In the figure, obvious negative shifts (2.5 – 3.0 ‰) of the $\delta^{13}C$ values in the Stephanian, Artinskian and Kazanian stages can be

Table 1 The $\delta^{13}C$ values for some Permo-Carboniferous coals in the North China Platform (PDB, ‰)

Serteng Mountain[1)]			Taiyuan, Shanxi[2)]			Huainan, Anhui		
Formation	Coal bed	$\delta^{13}C$	Formation	Coal bed	$\delta^{13}C$	Formation	Coal bed	$\delta^{13}C$
	161	-23.00		1	-25.30		25	-26.20
	159	-23.72		2	-23.85	Upper	20	-25.62
	157	-24.87	Shanxi	3	-23.05	Shihezi	17	-25.45
	155	-25.20		4	-23.45		16	-25.52
	149	-25.47		5	-23.20		13	-24.40
	146	-25.31		6	-23.20		9	-24.82
Shuangma-	143	-25.47		7	-22.70		8	-23.15
zhuang	414	-25.93		8	-22.40		7-2	-23.51
	139	-25.46	Taiyuan	8	-22.25	Lower	7-1	-22.45
	135	-25.82		9	-22.45	Shihezi	6	-22.12
	130	-25.48		Coal streak	-22.80		5	-23.25
	123	-25.61		Coal streak	-24.10		4	-23.10
	105	-25.34		10	-24.00			
	93	-25.84						
				11	-25.30	Shanxi	3	-25.95
Upper	83	-24.91	Benxi	Thin coal bed	-24.50		2	-22.55
Shetai	81	-24.50		ditto	-24.15		1	-23.51
	75	-24.62		ditto	-24.60			
	42	-23.51						
Lower	31	-22.10						-23.51
Shetai	25	-22.30						

Notes: 1) There are no serial numbers or names for coal beds in the Serteng Mountain area, therefore the numbers of the profiles are used in the table (Zhang, 1997).

2) The $\delta^{13}C$ values in coal for the West Hills are quoted from the Institute of Geological Exploration, CCMRI et al., 1987 and Zhuang, 1987, which were measured by the Xi'an Branch, CCMRI.

seen for three coalfields which are several hundreds of kilometers apart. Similar phenomena can be found in South Africa (Faure et al., 1995) and Australia (Compston, 1960). According to studies of coal chemistry and coal petrography (Institute of Geological Exploration, 1987; Duan, 1995), the sampling beds all consist of humic coal. Humic coal does not result in fractionation of carbon isotopes because of coalification or a raise in coal rank (Duan, 1995; Faure, 1977; Faure et al., 1995), therefore the influence of macerals and sources of coals on the carbon isotopic composition of humic coal must be determined before we make explanations of the negative shifts of the $\delta^{13}C$ values ocurring in the above-mentioned geological ages.

3 Discussion

3.1 Macerals and carbon isotopes of coals

Some researchers (Duan, 1995) consider that the discrepancy in carbon isotopic compositions of coals is mainly constrained by organic macerals: carbon isotopes of the exinite are the lightest, those of the inertinite the heaviest, and those of the vitrinite lie in between. That is to say, if the coal has a higher content of exinite, its carbon isotopes will become lighter, reflecting a negative correlation between them.

In the macerals of major coal beds in the West Hills and Huainan, the content of exinites varies between 7.6% to 29%, that of inertinites 0–34.9%, and that of vitrinites 48.6–88.0% (Table 3). But in the diagram showing the relationship between exinites and the $\delta^{13}C$ values (Fig. 3), such a negative correlation is not shown. This indicates that the variations in carbon isotopic composition under study may not be related with the content of exinites.

We have noticed that the $\delta^{13}C$ values of sapropelic coal formed by algae are relatively low, generally ranging from −27.0 ‰ to −35.0 ‰, averaging −32.0 ‰ (Duan, 1995), quite different from those of humic coal. This may be related with isotopic fractionation of organic matter in the sapropelic coal during its early diagenesis, which resulted in removal of heavy carbon

Table 2 Carbon isotopic compositions of coals in some selected areas

Country or area (coalfield)	Age	$\delta^{13}C$ (PDB) /‰			Data source
		Maximum	Minimum	Average	
Serteng Mt., Inner Mongolia	Carboniferous	-22.10	-25.84	-24.72	This paper
West Hills, Shanxi	Permo-Carboniferous	-22.25	-25.30	-23.60	ditto
Huainan, Anhui	Permian	-22.12	-26.20	-24.05	ditto
Pingsu, Shanxi	Permo-Carboniferous	-22.59	-23.46	-23.16	IGE, CCMRI, 1987
Lingwu, Ningxia	Jurassic	-23.65	-24.56	-24.15	Qian et al., 1994
Shuicheng, Guizhou (algal coal)	Late Permian			-30.80	Duan, 1995
Fushun, Liaoning (jet)	Tertiary			-26.40	ditto
China (humic coal included)	All ages	-19.90	30.80-	-24.40	ditto
Europe and America	ditto	-21.10	-27.40	-24.70	Silverman et al., 1959
South Africa	Permian	-21.20	-23.80	-22.70	Faure et al., 1995
South Africa	Triassic	-24.00	-24.40	-24.30	ditto

Table 3 Maceral contents in the coal beds of the Xishan and Huainan coafields

Area	Coal bed	Maceral / %		
		Vitrinite	Inertinite	Exinite
Xishan, Taiyuan	1	81.03	1.61	16.97
	2	72.91	1.16	25.12
	3	70.27	0.59	29.00
	6	86.27	2.26	11.47
	7	79.22	1.85	18.72
	8	78.85	1.84	19.82
	10	88.00	0.00	12.00
	11	86.00	0.00	14.00
Huainan	25	62.10	19.20	18.70
	20	63.50	14.70	21.80
	17	74.40	13.40	12.20
	13	62.90	21.40	15.70
	8	57.80	34.60	7.60
	7-1	52.10	33.40	14.50
	6	48.60	34.90	16.50
	5	76.80	12.40	10.80
	1	65.00	24.10	10.90

isotopes so that the solid organic matter became richer in ^{12}C. From the viewpoint of coal petrology, the fact that the contents of alginite consisting of lower organisms such as aquatic algae and plankton as well as bacteria, and bitumenite comprise more than 90% of the sapropelic coal indicates that these two components are the major factors causing the carbon isotopes of coals to get lighter. The exinites of humic coal consist of sporinite, cutinite, suberinite, periblinite, resinite, alginite, bituminite, exsudatinite, fluorinite and liptodetrinite. In the exinites of humic coal studied in this paper, there is only a minor amount of alginite and bituminite, which will not affect the carbon isotopic composition of the coals.

3.2 Sources (coal-forming plants) and carbon isotopes in coals

There are three approaches for plants to fix carbon through photosynthesis (Faure, 1977; Smith et al., 1971). The first is the C_3 (Calvin-Bensopl) circulation, most of terrestrial vascular plants use this approach, the $\delta^{13}C$ values range from −22 ‰ to −31 ‰. The second one is the C_4 (Hatch-Slack) circulation, in which ^{13}C is relatively enriched, and the $\delta^{13}C$ values vary between −10 ‰ and −16 ‰. Desert plants and saline plants fall in this category. The third, the CAM (Crassulocean Acid Metabolism) circulation is a mixture of the above two circulations. Aquatic algae and lichen belong to this category, the $\delta^{13}C$ values of which lie between the former two approaches (−12 ‰ to −23 ‰). Judging from the carbon isotopic compositions of the Late Palaeozoic humic coals in the North China Platform (Table 1), their sources are apparently the vascular plants.

Generally, the same plants growing in the same environment will have close $\delta^{13}C$ values, whereas different categories of plants growing in different environments will obviously differ from each other (Faure, 1977). The Permo-Carboniferous coals in the North China Platform were formed in peat swamp environments of a tropical rainforest climate (Zhang, 1991). Based on botanical studies of permineralized peat (coal balls) (Tian et al., 1995), the coal-forming plants include lycopods, sphenopsids, ferns, seed-ferns and cordaites, for which more than 30 fossil species have been determined. However, quantitative analyses reveal that the tree-like lycopods (including *Stigmaria, Lepidodendron, Lepidophylloides* and reproduction organs)

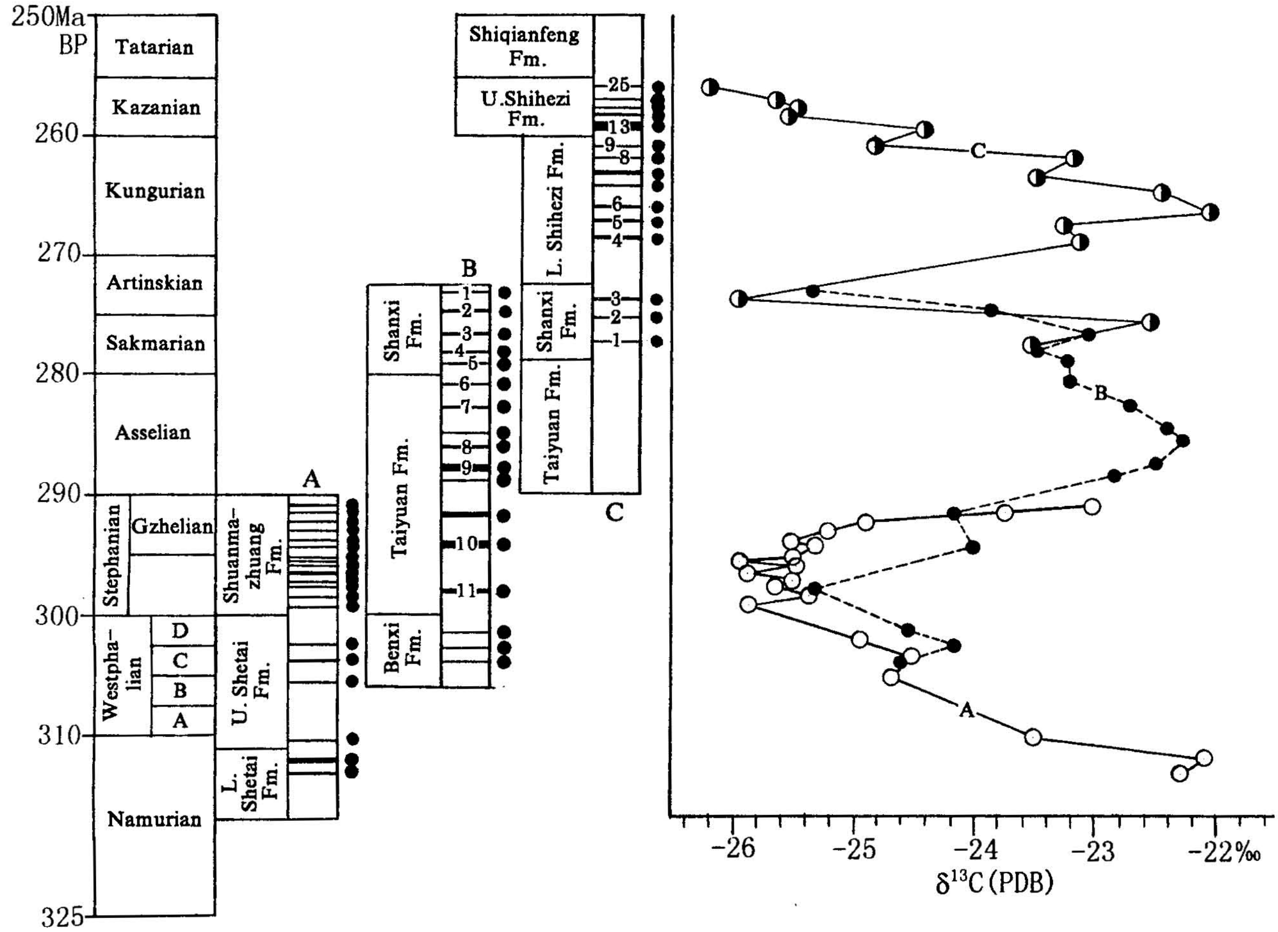

Fig. 2. Stratigraphic pattern of $\delta^{13}C$ for Permo-Carboniferous coals in the North China Platform.

A–Serteng Mt. coalfield; B–Xishan coalfield; C. Huainan coalfield.

comprises 68–95% of the coal-forming materials (Shang, 1997; Zhang, 1990), in which the root-form called *Stigmaria* occupies 60% of the lycopods (Zhang, 1990).

The tree-like lycopodiales show features adaptive to peat-swamp or half-aquatic environments (Frankenberg et al., 1969; DiMichele, 1981). Therefore, as the source material for primary peat, they naturally became the prevailing coal-forming plants. On the other hand, the coal-forming swamp environment (water-abundant, nutrient-poor and acidic) (Stach et al., 1982; Teichmuller, 1989) was a relatively stable one. Therefore, when the Permo-Carboniferous non-peat-swamp vegetation in the North China Platform was forced to reform, among which the tree-like lycopods began to decrease since the Artinskian stage and gradually went to extinction (Shen, 1995; Wang Z.,1989), the peat-swamp environment served as a "nature reserve" for these plants to survive.

The above analysis indicates that the Permo-Carboniferous coal-forming plants in the North China Platform did not show obvious changes, and they were by no means the cause for changes in the $\delta^{13}C$ values of the coals. It can thus be inferred that the $\delta^{13}C$ values for the Palaeozoic humic coal in the North China Platform represent the carbon isotopes of the CO_2 recorded by the tree-like lycopods.

4 Palaeoclimatic implications

As has been illustrated, the $\delta^{13}C$ values showed an overall decrease of 2.5–3 ‰ in coals of the North

China Platform during the Stephanian, Artinskian and Kazanian stages. The main factor controlling the ^{13}C depletion of plants is the photosynthesis (Smith et al., 1971), and the Late Palaeozoic coal-forming plants in the North China Platform fixed the atmospheric CO_2 only by the C_3 circulation. The main factors decreasing the $\delta^{13}C$ values of the C_3 plants include declining irradiance, low nutritent, low temperature, high P_{CO2} (Faure et al., 1995) and the $\delta^{13}C$ values of ambient CO_2 (O'leary, 1989), whereas that increasing the $\delta^{13}C$ values is osmoitic stress (increase of salinity in water) and increased aridity (Tieszen, 1991). According to Faure et al. (1995), the general decrease in the $\delta^{13}C$ values in coal is consistent with an overall decrease in the $\delta^{13}C$ values of ambient atmospheric CO_2 and/or relative increase in atmospheric P_{CO2} during the coal-forming period. Variations in carbon isotopic composition of coal, organic matter and carbonate may be explained by some large-scale processes that affected the carbon storage of the earth. Continent-continent collision played a leading role in the global carbon cycle (Beck et al., 1995; Raymo, 1994). The Carboniferous and Permian were the most active geological periods for convergence, collision and matching of continent blocks in the globe (Veevers, 1994): The Laurussia land, which was formed by the collision of Laurentia, Baltica, Avalonia and England at the end of Devonian, collided and matched with the Siberian plate and Gondwanaland along the Alleghanian-Veriscan-Ural mountain chain during the Late Carboniferous, forming a mountain range at a scale of the Himalayas near the equator (Rowley et al., 1985). Afterwards, the Khazakstan plate, Tarim massif and North-China plate collided with the south margin of the Siberian plate separately in the terminal Late Carboniferous, Early Permian and early Late Permian. During the late Late Permian or the early Early Triassic the various major massifs joined together to form Pangea. In the Late Permian compression activities were rather intensive along the margin of Pangea, especially in the Samfrau orogenic belt on the margin of the Gondwanaland, mainly between 247 and 260 Ma (Halbich et al., 1983; Mpodozis et al, 1992). The above continent-continent collisions and compression activities resulted in the following consequences: the central (the craton) part of the various continents uplifted, and the peat fields were oxidized; the active and passive continental margins were deformed, causing exhumation and oxidation of organic carbon of marine deposits; methane in the coal-bearing foreland basins was discharged; and finally a great amount of ^{13}C-depleted CO_2 was escaped into the atmosphere, which caused an obvious reduction of the $\delta^{13}C$ values in the biosphere (Faure et al., 1995). Therefore, the negative shifts of $\delta^{13}C$ values of coals in the North China Platform during the Stephanian, Artinskian and Kazanian stages may be indicative of the high concentrations of CO_2 and CH_4 (Zhang et al., 1997) in the atmosphere.

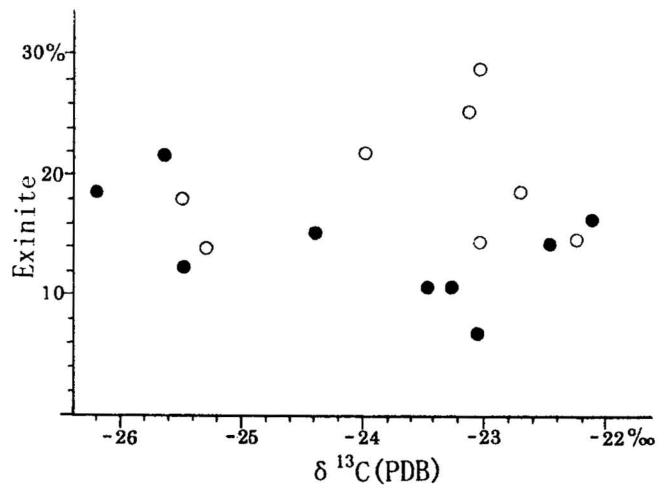

Fig. 3. Relationship between exinites and the $\delta^{13}C$ values in major coal beds of the Xishan coalfield (circles) and Huainan coalfield (dots).

The palaeoclimatic warming by way of the greenhouse effect related to the high concentrations of CO_2 and CH_4 in the atmosphere is an important subject in the study of the palaeoecosystem in geological times. Palaeobotanists have confirmed that the transformation of vegetation in the North China Platform during the Stephanian, or the occurrence of "new peaks" of Acticulatae and Filicinae at the boundary between the Benxi and Taiyuan formations was closely related with the climatic warming event of that time (Wang Z., 1989; Zhang et al., 1997). As Phillips (1984) and Durante (1995) pointed out, the palaeoclimatic warming in the Stephanian was worldwide. The negative shift (c. 3 ‰) of $\delta^{13}C$ values of the North China coals in the Stephanian may serve as new evidence for this warming event.

There are not many carbon isotope data available related to the North China coals in the Artinskian are available. Only the $\delta^{13}C$ values of two coal seams show a negative shift of about 2.5 ‰, which is of little palaeoclimatic significance. However, it can be verified from the relationship between the stomatal index of the plant epidermis and the atmospheric CO_2 content. As is well known, the stomatal index of plant leaves decreases rapidly with increasing atmospheric CO_2 concentration (Wagner et al., 1996; Woodward, 1987). An SEM study of *Lepidodendron*, a major coal-forming plant in central Shanxi, shows that in the vicinity of the No. 1 coal of the Shanxi Formation (the Artinskian Stage), the epidermal cells of leaf cashion are large (10-20 μm) and the stomatal index is low; whereas in the vicinity of a coal seam of the lower Shihezi Formation (Kungurian Stage), the cells are smaller (8-10 μm) and the stomatal index is 2.4 times of the former. The distinct variation in the two shows that the coal-forming plants were affected by the obvious increase of the atmospheric CO_2 content in the Artinskian.

The negative shift of $\delta^{13}C$ values in Kazanian coals of the North China Platform (Fig. 2) is similar to that in South Africa (Faure, 1995) and Canada (Wang, 1994). It is an indicator of the sudden increase of atmospheric CO_2 content, the palaeoclimatic warming, and the deterioration of the palaeoenvironment prior to the mass extinction (Erwin, 1994) or the global coal hiatus (Faure, 1995; Retallach et al., 1996) at the end of the Permian. As for the geological periods of relative increases in the $\delta^{13}C$ values between the negative shifts (Fig.2), they may be attributed to increasing absorption of CO_2 by oceans due to the global cooling, or to the increase of biomass stimulated by the enhanced atmospheric CO_2 content.

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