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Paleoceanographic Indicators for Early Cambrian Black Shales from the Yangtze Platform, South China: Evidence from Biomarkers and Carbon Isotopes

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Abstract: The lower Cambrian Niutitang Formation, a widespread black shale deposition, is of geological interest because of its polymetallic beds, Cambrian explosion, depositional ages, dramatic environmental changes and so on. Previous study focused mainly on inorganic geochemistry and few studies have investigated the organic fractions of upper Neoproterozoic-lower Cambrian strata in South China. Here we report a study of biomarkers plus organic carbon isotopes for black shales from Ganziping, Hunan Province (China). All the saturated hydrocarbon fractions have a unimodal distribution of *n*-alkanes, a high content of short-chain alkanes and maximize at C_{19} or C_{20} (C_{23} for sample Gzh00-1). The C_{27}/C_{29} sterane ratio ranges from 0.77 to 1.20 and 4-methylsteranes are in low abundance. These parameters indicate that algae and bacteria are the important primary producers. Furthermore, biomarker maturity proxies show the samples to be higher maturity. The low Pr/Ph values (<0.7) suggest that the samples were deposited under anoxic conditions and, likely, under stratified water columns. In addition, 25-norhopanes and gammacerane are present as diagnostic indicators of normal marine salinity and dysoxic to anoxic conditions. During the Early Tommotian, known to coincide with a transgression event, small shelly fossils increased in abundance and diversity. Moreover, positive $\delta^{13}C_{org}$ excursions close to 1.4‰ occur at the base of the Tommotian stage. In summary, the Early Cambrian black shales were deposited under dramatic paleoenvironmental changes, including oceanic anoxia, higher primary productivity and sea-level rise.

Key words: black shales, biomarkers, carbon isotope, paleoceanography, Early Cambrian, South China

1 Introduction

Marine black shales of the lower Cambrian Niutitang Formation in South China have been discussed for many years with respect to the origin of the extreme enrichment of Mo-Ni-Pt group elements (PGEs; Lehmann et al., 2003; Jiang et al., 2007; Wille et al., 2008). At the Precambrian/Cambrian (PC/C) transition (ca. 542±0.3 Ma ago; Amthor et al., 2003), deposition of the polymetallic sulfide sediments may have been accompanied by global events such as the Neoproterozoic Snowball Earth and the Cambrian Explosion (Jiang et al., 2007). Accordingly, during the time of the Neoproterozoic Snowball Earth, an episodic decrease in greenhouse gas may have occurred through the effect of erosion and weathering, which could also have led to ice ages and mass extinctions (Maruyama and Santosh, 2008); however, the Cambrian Explosion was an explosive radiation of different metazoan phyla (Zhao et al., 1999; Budd, 2003; Ishikawa et al., 2008). In the light of trace element, rare earth element (REE; Zhou and Jiang, 2009), isotope (C, S, Sr, Se, Os, Mo; Goldberg et al., 2007;

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Jiang et al., 2007; Ishikawa et al., 2008; Wille et al., 2008; Chen et al., 2009; Wen et al., 2009; Wen and Carignan, 2011) and bio-evolution [arthropods, sponges, small shell fossils (SSFs), echinoderms; Clausen et al., 2010; Li et al., 2011] studies, these global events implied the dramatic environmental changes (i.e., oceanic anoxic, increasing productivity, global cooling and subsequent sea-level fall with methane release).

As mentioned above, the PC/C transition was a key period for understanding one of the most important changes in the Earth's history. Extensive research on the remarkable biological, oceanic and geochemical changes during this transition, infer that the changes in environmental conditions were a possible explanation for events such as the Neoproterozoic Snowball Earth, Cambrian Explosion, Tommotian Oceanic Anoxic Events. The trigger for these environmental transformations remains, however, unclear and the subject of debate (Wille et al., 2008; Derry, 2010). Few studies have been carried out on molecular indicators of palaeoclimate, palaeoenvironment and palaeoecology, so the aim of the present study was to determine biomarker distributions with respect to organic matter (OM) input and Paleoceanographic conditions for lower Cambrian strata in South China.

The lower Cambrian black shales in the Guizhou, Hunan and Yunnan provinces, e.g. those of the Yangtze platform, South China were deposited under shallow-water continental margins to anoxic slope-basinal conditions (Jiang et al., 2007; Goldberg et al., 2007). Accordingly, the Huangjiawan (Zuiyi, Guizhou province) and Ganziping sections (Dayong, Hunan province) have been studied in detail because of extreme enrichment of Mo-Ni-PGEs. In addition, the interval is thought to be one of the most significant periods in animal evolution whereby soft-bodied Ediacaran biota declined (Narbonne, 2005) and Cambrian-type small shell fossils (SSFs) increased rapidly (Budd, 2003). Previous studies have focused mainly on isotopic dating (Mao et al., 2002; Amthor et al., 2003; Jiang et al., 2009), strata correlation (Zhu et al., 2003; Peng et al., 2011), bio-evolution (Li et al., 2007; Zhu et al., 2007) and models (Goldberg et al., 2007). Although the inorganic geochemistry of the strata has been intensively studied, few studies have investigated the geochemistry of the organic fractions of upper Neoproterozoic-lower Cambrian strata in South China. Because these strata were deposited under anoxic conditions, they contain sufficient OM for biomarker analysis. We report first on the biomarkers of lower Cambrian black shales of the Niutitang Formation in the Ganziping section (Hunan province) and then discuss the OM origin and maturity, and redox conditions, which also provide an insight into the mechanisms of black shales formations and the paleoceanography.

2 Geological Background

During the late Neoproterozoic to early Cambrian transition, the Yangtze platform evolved from a rift basin to a passive continental margin basin (Wang and Li, 2003) with palaeoenvironmental changes from shallow-water carbonate platform to black chert-shale margin zone and deeper slope-to-basin setting (Zhu et al., 2003; Fig. 1). The basal Cambrian of the platform is characterized by a diachronous, transgressive black shale succession. The chronostratigraphic subdivision and correlation of late Neoproterozoic to Early Cambrian strata are difficult because there are few reliable radiometric ages available for South China (Steiner et al., 2005). In view of the Concordia U-Pb zircon age of 542±0.3 Ma for the PC/C boundary in Oman (Amthor et al., 2003), the zircon ages afforded for a volcanic ash bed in the lowermost black shale sequence of the Niutitang Formation in Guizhou Province (South China; Jiang et al., 2009) and at Ganziping (Hunan; Chen et al., 2009) were 532.3±0.7 Ma and 536.3±5.5 Ma respectively. Therefore, an international and regional correlation is based mainly on bio- and litho-stratigraphic evidences. Biostratigraphically, the occurrence of *Cloudian* reflects a terminal Neoproterozoic age, followed by the appearance of Cambrian SSFs (Steiner et al., 2005; Goldberg et al., 2005).

Our study area is paleogeographically located along southern marginal zone of the Yangtze platform, near Dayong (or Zhangjiajie), western Hunan (Fig. 1). The Ganziping section is composed of Dengying, Niutitang and Balang Formations of lower Cambrian from lower to upper, accordingly, the regional stratigraphy is subdivided into the Ediacaran (Neoproterozoic), Meishucunian, Qiongzhusian and Canglangpuian (Lower Cambrian), or is correlated with the Siberian Precambrian to Lower Cambrian subdivisions: Ediacaran, Nemakit-Daldynian, Tommotian, Atdabanian and Botomian (Steiner et al., 2005). The depositional environment of the Dengying Formation is interpreted as a widespread prograding platform comprising thick successions of dolostone and limestone; however, the Niutitang Formation often condensed in deeper water of further transgressive flooding, led to extended black shale deposition (Fig. 2). The Niutitang Formation rests disconformably on dolostone of the Neoproterozoic Dengying Formation. In addition, the whole stratigraphic sequence is characterized by different lithological types: grey-white heavy bedded dolostones with good intercrystalline pores, grey-black middle bedded silicalites interbedding with thinly carbonaceous shale, black heavy bedded carbonaceous shales with a conformable Mo-Ni-PGE polymetallic sulfide horizon, laminated coal stone with lenticular bodies, and the carbonaceous shales with four

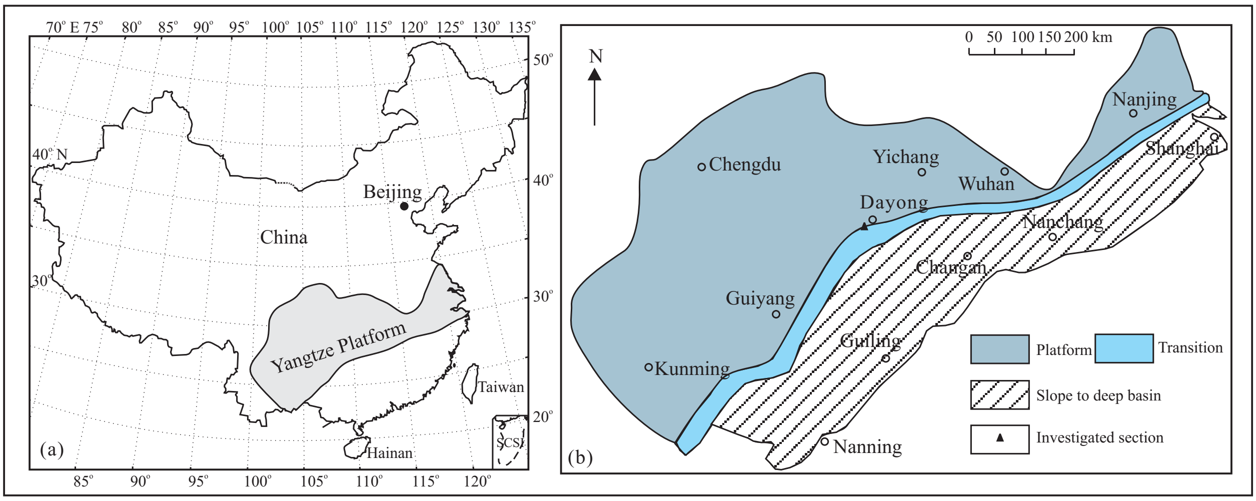

Fig. 1. (a) A map showing location of the Yangtze Platform. (b) Investigated section and depositional environments of the platform, ranging from carbonate platform to transition, then slope to deep basin from west to east. The Ganziping section is located in the transition belt (modified from Steiner, 2001; Chen et al., 2009).

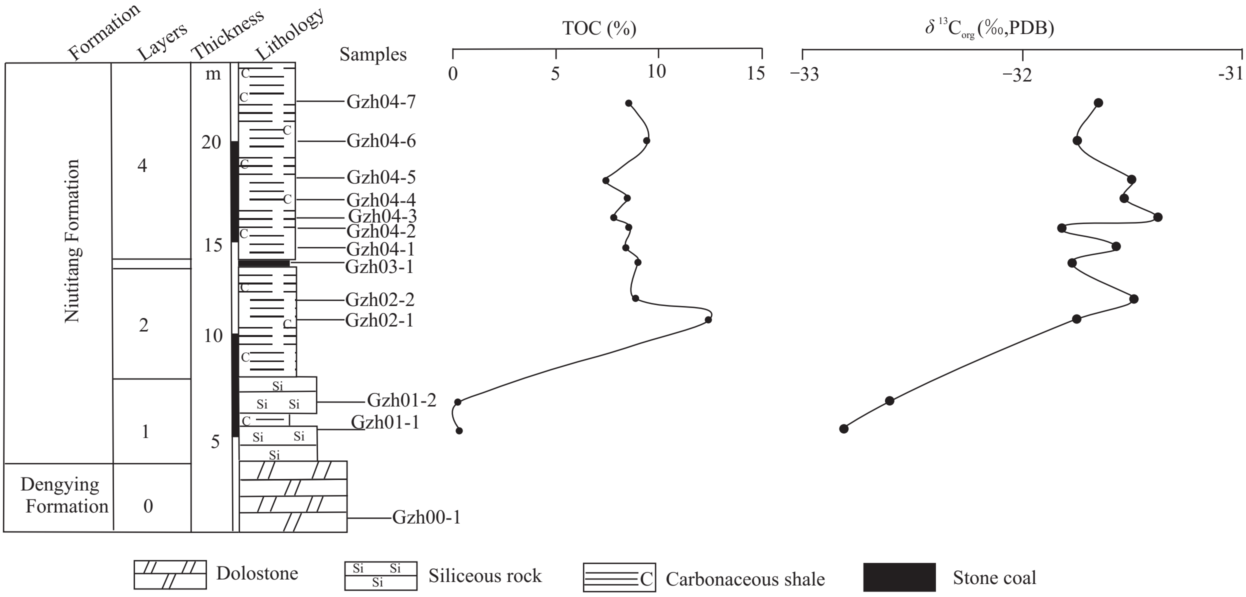

Fig. 2. Lithostratigraphy of Niutitang Formation showing stratigraphic variation in TOC and $\delta^{13}C_{org}$ of the black shales at the Ganziping section, Hunan Province.

heavily-latimated cycles (Fig. 2). Nevertheless, the uppermost part had been observed at the Ganziping section, Dayong (Hunan province) and the lowermost Cambrian successions, consisting of carbonates, cherts and phosphatic horizons, are often missing from the basinal part of the Yangtze platform (Zhu et al., 2003; Goldberg et al., 2007).

3 Analytical Methods

3.1 Samples

All 13 samples were collected from five horizons (Gzh00-1 to Gzh04-7) of the Dengying Formation to Niutitang Formation, Ganziping section of Dayong (Hunan province, China). The section is ca. 24 m thick and had been studied bed by bed with vertical sample spacing of 1.85m on average (Fig. 2); 7 samples were used for biomarkers. The samples were first washed thoroughly with dichloromethane (DCM) to remove possible contamination from the surface and ground to <200 mesh. All experiments were carried out at State Key Laboratory of Organic Geochemistry, Guanghzou Institute of Geochemistry, Chinese Academy of Sciences, China.

3.2 Total organic carbon (TOC) and stable carbon isotopes

The TOC content was measured by way of combustion with a Vario EL Ш elemental analyzer after dissolution of the carbonate with 1M HCl. For organic carbon isotopes values ($\delta^{13}C_{org}$) determination, the sediment samples were treated with HCl/HF to remove carbonate and silicates, cleaned with de-ionized water until neutral pH. They were then extracted with benzene/acetone (2:5 v/v) for 72 h to remove dissolved OM. The $\delta^{13}C_{org}$ measurements on the resulting kerogen were made with an EA-Finnigan Delta Plus XL mass spectrometer and given relative to the Peedee Belemnite (PDB) standard; the standard deviation being better than ± 0.2‰.

3.3 Biomarkers

Each sample was reacted with DCM for 72 h in a Soxlet apparatus. The solvent was removed by means of rotary evaporation and the residue dissolved in cyclohexane. Asphaltenes were removed by centrifugal precipitation with chilled *n*-heptane (at least 40x v/v). Aliquots of the maltenes were separated by way of silica gel chromatography using hexane to elute the saturated hydrocarbons, 4:1 hexeane: DCM for the aromatic hydrocarbons and 1:2 DCM: MeOH for the polar fraction. Activated Cu was added to the saturate fraction to remove elemental sulfur and the fraction further separated into straight chain and branched/cyclic hydrocarbons using 5Å molecular sieve (Merck, Germany).

Gas chromatography mass spectrometry (GC-MS) was performed using a Micromass Platform II mass spectrometer equipped with a HP 6890 gas chromatograph. For the saturated hydrocarbons, a 30m HP-5 fused silica column (0.25mm i.d., 0.25 μm film thickness) was used with He as the carrier gas. The oven temperature programme was: 65°C (1 min) to 290°C (held 30 min) at 3 °C/min. The transfer line temperature was 250°C and the ion source temperature 200°C. The ion source was operated in the electron impact mode at 70 eV.

4 Results

4.1 TOC and $\delta^{13}C_{org}$

The TOC and $\delta^{13}C_{org}$ values for the Ganziping section (Dayong) are presented in Table 1 and Fig. 2. TOC exhibits a sharp fluctuation between 0.25% and 12.31% around an average value of 7.41%, and the $\delta^{13}C_{org}$ values fluctuate from −31.82‰ to −31.38‰ PDB with a mean value of −31.80‰ PDB. The TOC and $\delta^{13}C_{org}$ values of Cambrian black shales at Ganziping section are higher than those at the Huangjiawan section (Chen et al., 2006). In the lower part of the Niutitang Formation, the lowest $\delta^{13}C_{org}$ values of silicites display a range between −32.82‰ and −32.60‰ followed by a rapid shift to more positive values of −31.49‰ at 12 m with a positive excursion of 1.4‰ (Fig. 2). The middle part (between 12 m and 16 m) is characterized by carbonaceous shales interbedded with stone coal with distinct fluctuations of $\delta^{13}C_{org}$ values showing two fall-rise cycles. At ca. 15 m, the $\delta^{13}C_{org}$ values reach the maximum (−31.38‰) for the whole section, and are then stable above 16 m from −31.80‰ to −31.51‰.

4.2 Molecular composition

Several biomarkers, including *n*-alkanes, acyclic isoprenoids, terpenoids and steroids were detected in all samples (Figs. 3, 4, 5 and Table 2).

4.2.1 *n*-Alkanes and acyclic isoprenoids

The samples are generally dominated by C_{14} to C_{31} *n*-alkanes maximising at C_{19} or C_{20} in the Niutitang Formation and C_{23} in the Dengying Formation, respectively (Fig. 3a). There is a marked presence of short chain *n*-alkanes with essentially no odd/even predominance (OEP; 0.94 to 1.06) and a carbon preference index (CPI) of 0.97 to 1.15 (Table 2). In addition, the stratigraphic variations in nC_{17}/nC_{31}, nC_{21}^{-}/nC_{22}^{+} and $(nC_{21}+nC_{20})/(nC_{28}+nC_{29})$ alkanes are shown in Table 2, indicating the predominance of the light hydrocarbons.

The acyclic isoprenoids, dominated by pristane (Pr) and phytane (Ph) were detected in all samples (Fig. 3). Pr/Ph ranges from 0.33 to 0.65, Pr/nC_{17} from 0.37 to 0.57, and Ph/nC_{18} from 0.65 to 0.84 (Table 2).

Table 1 Basic geochemical data of the early Cambrian black shales at Ganziping, Hunan Province

Sample	Horizon (m)	Formation	Lithology	TOC (%)	$\delta^{13}C_{org}$ (‰)	Element content of kerogen (%)			Chloroform asphalt A (mg)
						N	C	H	
Gzh04-7	22.27	Niutitang	Carbonaceous shale	8.55	−31.65	1.06	49.85	1.28	2.5
Gzh04-6	20.27		Carbonaceous shale	9.47	−31.75	1.37	55.45	1.50	2.0
Gzh04-5	18.27		Carbonaceous shale	7.56	−31.51	1.14	36.65	0.95	1.6
Gzh04-4	17.27		Carbonaceous shale	8.38	−31.53	1.19	45.45	1.18	3.4
Gzh04-3	16.27		Carbonaceous shale	7.78	−31.38	2.16	40.45	1.12	1.7
Gzh04-2	15.77		Carbonaceous shale	8.45	−31.82	1.36	60.25	1.56	2.8
Gzh04-1	14.77		Carbonaceous shale	8.39	−31.57	1.15	38.95	1.03	2.5
Gzh03-1	13.94		Stone coal	8.95	−31.77	1.35	63.7	1.57	2.2
Gzh02-2	12.04		Carbonaceous shale	8.81	−31.49	1.12	49.25	1.40	1.7
Gzh02-1	11.04		Carbonaceous shale	12.31	−31.75	1.28	55.7	1.41	3.3
Gzh01-2	6.82		Siliceous rock	0.27	−32.60	1.43	66.6	1.78	1.3
Gzh01-1	5.35		Siliceous rock	0.29	−32.82	1.50	70.1	1.96	1.2
Gzh00-1	0.76	Dengying	Dolostone	N.d[a]	N.d[a]	N.d[a]	N.d[a]	N.d[a]	1.4

N.d[a]: no data.

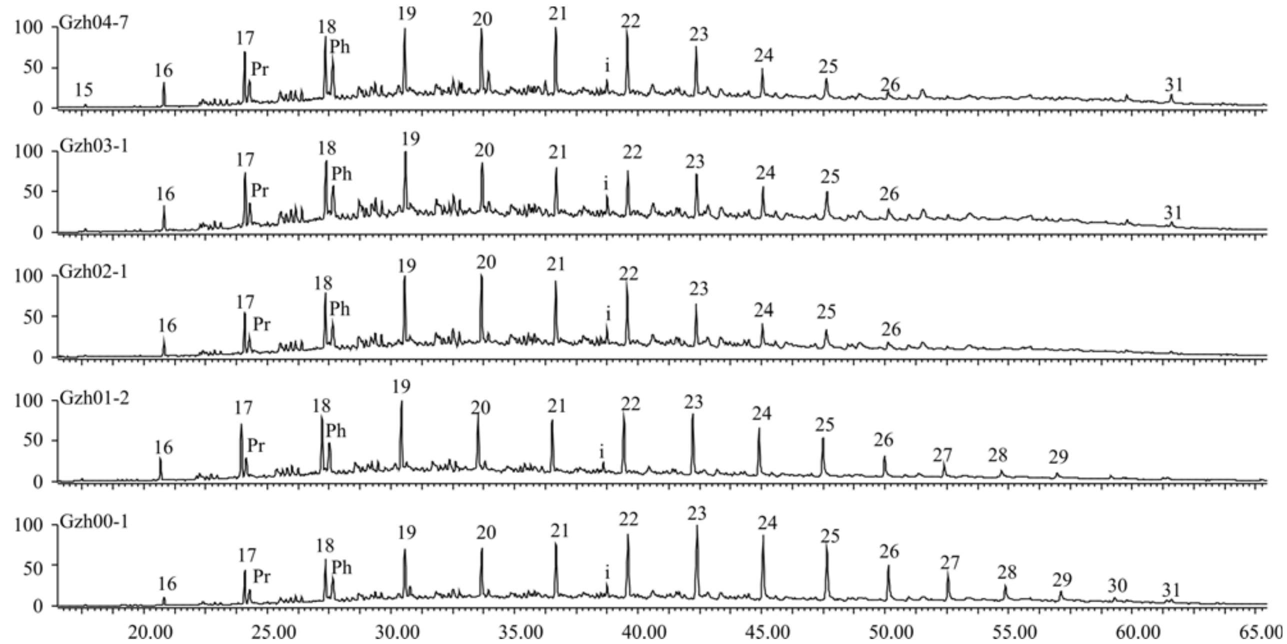

Fig. 3. Total ion current (TIC) showing relative abundance of *n*-alkanes and regular isoprenoids of the black shales in the study area. Numbers above symbols denote carbon number.

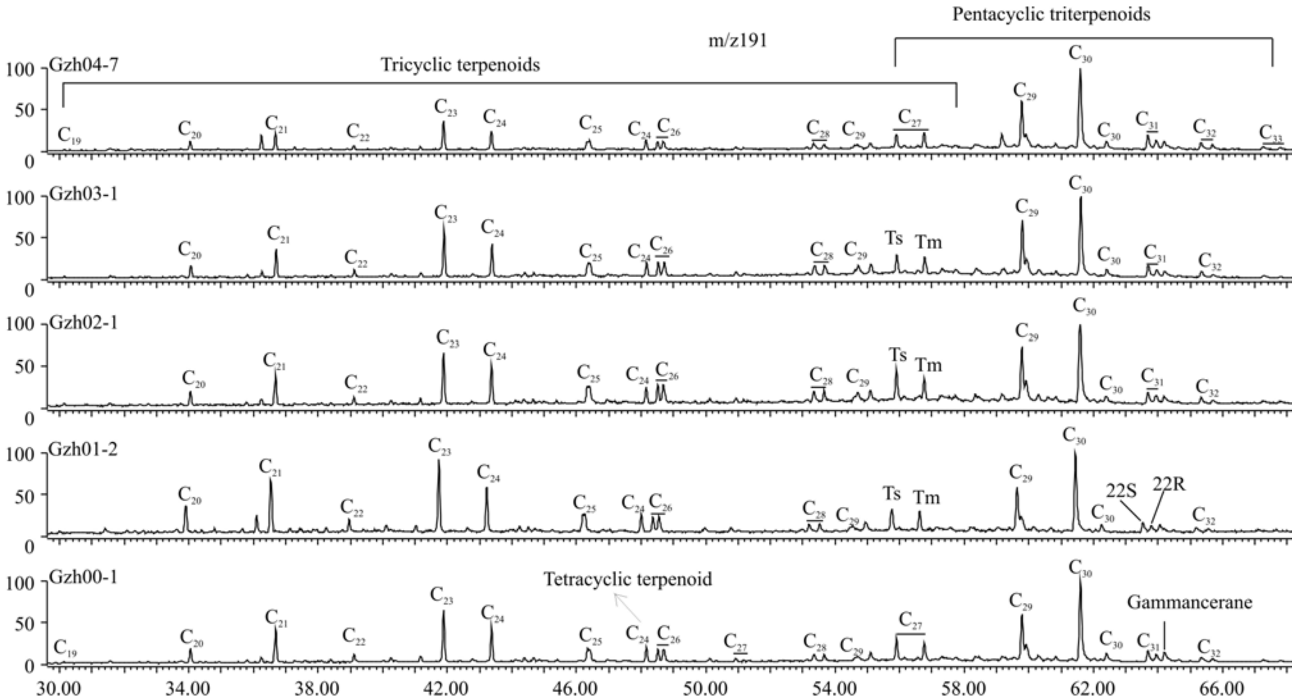

Fig. 4. The *m/z* 191 chromatograms showing relative abundance of terpenoids in the representative samples.
Ts, 18α(H)-22, 29, 30-trisnorneohopane; Tm, 17α(H)-22, 29, 30-trisnorneohopane. The 22S and 22R epimers are shown for C_{31}-homohopanes.

4.2.2 Terpenoids

The *m/z* 191 chromatograms for the Neoproterozoic to Early Cambrian samples are shown in Fig. 4. The most abundant hopanes were C_{29} and C_{30} through the whole strata, but the C_{27} hopanes (Ts, 18α-22, 29, 30-trinorneohopane and Tm, 17α-22, 29, 30-trinorhopane) were subordinate. C_{31} to C_{33} homohopanes were also detected, although in low abundance. Gammacerane occurred in lower abundance, and oleanane and lupane were not detected in any sample. In contrast, tricyclic terpanes were represented by a series from C_{19}-C_{30}, and only C_{24} was present for tetracyclic terpanes. No methyl hopanoids, which originate from cyanobacteria, were detected in *m/z* 205 chromatograms, although shales from offshore West Africa (upper two panels), and Green River shale (U.S.A) and Serpiano shale (Switzerland) show the

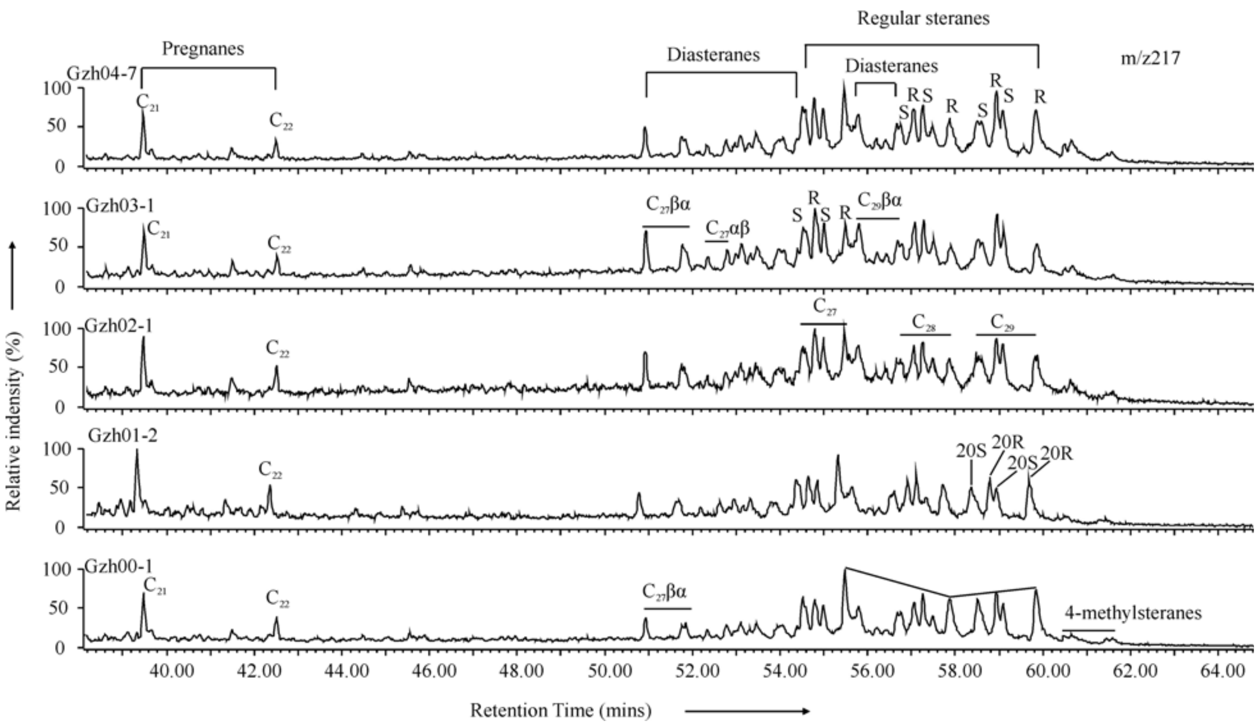

Fig. 5. The *m/z* 217 chromatograms showing relative abundance of steroids in the representative samples.
The 20S and 20R epimers are shown for regular C_{29} steranes.

2α- and 3β-methylhopanes derived (Farrimond et al., 2004) from distinct bacterial sources: cyanobacteria (2-methyl) and methanotrophic bacteria (3-methyl). The 25-norhopanoids, however, occurred in *m/z* 177 mass chromatograms. Without exception, some classic 25-norhopanes were recognized, such as 17α, 21β-25, 30-dinorhopane, 17β, 21α-25, 30-dinormoretane, 17α, 21β-25-norhopane, and 17β, 21α-25-normoretane. Blanc and Connan (1992) regarded the 25-norhopanes as a palaeobiodegradation indicator in apparently not-biodegraded oil. Furthermore, 25-norhopanes appear to be diagnostic of specific environmental conditions (marine and lacustrine source rocks, dysoxic and not very hypersaline).

4.2.3 Steroids

Besides the regular steranes (C_{27}-C_{29}) and diasteranes (C_{27}-C_{29}), series of 4-methyl steranes (C_{28}-C_{30}) and abundant pregnanes (C_{21}-C_{22}) were detected in the *m/z* 217 chromatograms (Fig. 5). Furthermore, the regular steranes show a V-type distribution with variable values from 0.77 to 1.20 for the $\Sigma C_{27}/\Sigma C_{29}$ ratio, and the dia-ΣC_{27}/ regular steranes ΣC_{27} ratio changes between 0.26 and 0.39 through the section.

5 Discussion

Bulk geochemical parameters, including total organic carbon (TOC), carbon isotopes ($\delta^{13}C_{org}$), and parameters based on normal and branched alkanes, terpenoids and steroids are given in Table 1 and Table 2; then these parameters are considered in terms of OM origin and maturity, redox conditions in the ocean and Paleoceanographic implications.

5.1 OM origin

n-Alkanes occur widely in crude oils and sediments and the distributions often provides information relating to OM origin (Peters et al., 2005; Romero-Sarmiento et al., 2011). Accordingly, the short chain *n*-alkanes may indicate an algal/bacterial contribution and the long chain ones could derive from land plants (Peters et al., 2005). Our Early Cambrian samples are characterized by a marked predominance of short chain *n*-alkanes with a maximum at C_{19} or C_{20}, and nC_{17}/nC_{31} ration values between 1.93 and 20.11 (Table 2), representing the algal/bacterial input. In contrast, the lowermost Cambrian Kuanchuanpu Formation in Shaanxi Province has an *n*-alkane peak near C_{24}, indicating an abundance of algae and eukaryotic phytoplankton that fueled a diverse ecosystem, including cnidarians and SSFs (Kunimitsu et al, 2009). Based on OEP (0.94–1.06) and CPI (0.97–1.15) close to 1.0, all the samples showed no distinct odd/even predominance. Wang (1990) suggested the *n*-alkanes without an odd/even predominance indicated two origins for the OM, one bacteria/ microbial waxes, the other land plant waxes

Table 2 Biomarker parameters based on *n*-alkanes, acyclic isoprenoids, terpenoids and steroids

Sample	n-alkanes					Isoprenoids			Steranes					Hopanes		
	Main peak	OEP[a]	CPI[b]	nC_{17}/nC_{31}	nC_{21}^{-}/nC_{22}^{+}	Pr/Ph	Pr/nC_{17}	Ph/nC_{18}	Pregnane/ C_{29}-20R	Dia-ΣC_{27}/ Reg-ΣC_{27}	Reg-ΣC_{27} /ΣC_{29}	$C_{29}\alpha\alpha\alpha$20S/ (20S+20R)	$C_{29}\alpha\beta\beta$/ ($\alpha\alpha\alpha+\alpha\beta\beta$)	$C_{31}\alpha\beta$22S/ (22S+22R)	Ts/ (Tm+Ts)	$\gamma/\alpha\beta C_{30}H$[c]
Gzh04-7	nC_{20}	1.05	1.07	1.93	1.12	0.65	0.54	0.77	0.43	0.27	1.06	0.44	0.42	0.56	0.47	0.17
Gzh04-4	nC_{19}	0.94	0.97	20.11	3.41	0.44	0.42	0.65	1.06	0.39	0.77	0.57	0.52	0.55	0.58	0.11
Gzh04-1	nC_{19}	1.05	1.04	5.55	1.49	0.33	0.37	0.72	0.45	0.29	0.93	0.48	0.45	0.57	0.56	0.16
Gzh03-1	nC_{19}	1.03	1.14	3.32	0.94	0.35	0.46	0.84	0.30	0.26	0.92	0.54	0.43	0.57	0.54	0.13
Gzh02-1	nC_{20}	1.04	1.15	4.48	1.38	0.54	0.57	0.67	0.62	0.36	0.79	0.55	0.46	0.54	0.55	0.17
Gzh01-2	nC_{19}	1.03	1.10	5.63	1.04	0.64	0.39	0.66	1.08	0.33	1.20	0.45	0.40	0.56	0.56	0.14
Gzh00-1	nC_{23}	1.06	1.11	4.04	0.53	0.65	0.50	0.71	0.58	0.30	0.96	0.41	0.32	0.55	0.54	0.17

[a] $(C_{21}+6C_{23}+C_{25})/(4C_{22}+4C_{24})$; [b] $[2(C_{23}+C_{25}+C_{27}+C_{29})]/[C_{22}+2(C_{24}+C_{26}+C_{28})+C_{30}]$; [c] γ, gammacerane.

through serious biodegradation. However, there were no land plants in the Cambrian, so we may conclude that algae/bacteria were the predominant primary producers during deposition of the uppermost Dengying to lower Niutitang Formation.

In addition, distribution of hopanes was similar to the Early Cambrian black shales from Zunyi, Guizhou Province (Chen et al., 2006) and the Late Devonian marly shales from Kowala (Poland; Marynowski et al., 2011), with dominant C_{30}-17α, 21β-hopane and a gradual decrease in the abundance of homohopane epimers (C_{31}S+R homologues dominating) with increasing carbon number (Fig. 4). All samples contained minor amounts of gammacerane (Fig. 4), as indicated by gammacerane/C_{30}-17α-hopane values from 0.11 to 0.17 (Table 2). High concentrations of tricyclic terpanes with C_{19} to C_{30} carbons were present, maximising at C_{23} terpanes, but we detected no components indicating cyanobacteria, such as methyl hopanoids (Summons et al., 1999). Kunimitsu et al. (2009) suggested it is not necessary to indicate an absence of cyanobacteria because algal components in general have a greater preservation potential than cyanobacteria. It is noteworthy that not all cyanobacteria biosynthesize methylhopanoids (Brocks et al., 2005). On the basis of previous results for black shales of the Lower Cambrian Niutitang Formation from Zunyi, Guizhou province, the Mo-Ni-PGE mineralized body is interpreted as representing a remnant of phosphate-rich and sulfide-rich sub-aquatic hardground supplied with OM derived from plankton and benthic communities as well as with algal/microbial oncolite-like bodies that originated in a wave-agitated, shallow-water, nearshore environment (Křibek et al., 2007). The above terpenoids characteristics of all our samples are therefore consistent with an aquatic organism source.

With respect to regular steranes, one of the most distinctive characteristics is the predominance of C_{29} steranes (Fig. 5), constituting on average 39.3% of the C_{27}-C_{29} steranes (C_{27}/C_{27}-C_{29} ratio 36.8% and 23.8% for C_{28}). However, although the abundance of C_{29} steranes was comparable to or higher than that of C_{27} steranes ($\Sigma C_{27}/\Sigma C_{29}$ between 0.77 and 1.20), the origin is not necessarily connected with terrestrial OM. C_{29} steranes occur in marine sedimentary rocks (Peters et al., 2005; Volkman, 2005), deriving from photosynthetic algae cell membranes. Kelly et al. (2011) reported hydrocarbon biomarkers from Neoproterozoic to Lower Cambrian oils from eastern Siberia with C_{27}/C_{29} sterane values averaging ca. 0.2 and also suggested photosynthetic algae as important contributors to the organic matter. Moreover, 4-methylsteranes have been identified in all samples (Fig. 5) in low abundance, being related to methanotrophic bacteria (Schouten et al., 2000) and/or algae (Peters et al., 2005; Kelly et al., 2011).

5.2 Maturity

During diagenesis the original biological configuration of some biomarkers evolves to thermally more stable stereochemistry. Thus, ratios of isomers give an insight into the level of maturity (Hermann et al., 2011). Ts/(Ts+Tm) values are commonly used to evaluate maturity, with higher values indicating higher maturity (Seifert and Moldowan, 1978). However, there is a strong sedimentary facies control on this ratio, so it is most reliable when oils or sedimentary rocks of similar sources are compared. Table 2 showed Ts/(Ts+Tm) values fluctuating between 0.47 and 0.58, with an average of 0.54. Further biomarker maturity parameters are 22S/(22S+22R) $C_{31}\alpha\beta$-homohopane, 20S/(20S+20R) ααα and 20R αββ/(ααα+αββ) C_{29} steranes (Seifert and Moldowan, 1978; Peters et al., 2005). The homohopane 22S/(22S+22R) values in this study are all >0.55 (except for sample Gzh02-1 of 0.54) and the sterane 20S/(20S+20R) values are all between 0.41 and 0.57 (Table 2), consistent with a higher level of thermal maturity. In addition, the contents of chloroform extracts show lower values from 1.2 mg to 3.4 mg (Table 1), which indicates the higher thermal evolution.

5.3 Sedimentary environments

The low Pr/Ph ratio values suggest an origin from sediments deposited under anoxic conditions and, likely, under stratified water columns (ten Haven et al., 1987). Ten Haven et al. (1987) noted that the utility of this indicator is compromised by several factors, and challenged the assumption that both compounds are derived from a common source. In our samples, the OM was derived from a single source: aquatic organisms. Empirical evidence suggests that a Pr/Ph value <0.8 is diagnostic for anoxic environments, as commonly encountered in strongly stratified water columns. Values >1 suggests slightly more oxygenated environments, while values >3 are generally observed for settings where terrigenous OM is transported and deposited in oxygenated waters (Peters et al., 2005). As shown in Fig. 3 and Table 2, the values are significantly lower, between 0.33 and 0.65 for all samples, suggesting that Early Cambrian black shales were deposited in anoxic and stratified water columns conditions.

Gammacerane is formed from tetrahymanol through dehydration-reduction and/or sulfurisation/desulfurisation (ten Haven et al., 1987), tetrahymanol being synthesized by bacteriovorous ciliates found at the chemocline. Relatively high gammacerane abundances are typically interpreted as evidence of a stratified anoxic water column (ten Haven et al., 1987), which can be associated with hypersaline conditions. For our samples, the gammacerne/hopane values were around 0.11 to 0.17 which would be consistent with black shale deposition at normal marine salinity. In addition, 25-norhopanes, distributed through the whole section, have been suggested as being diagnostic of specific environmental conditions (marine and lacustrine source rocks, dysoxic and not very hypersaline; Blanc and Connan, 1992).

With respect to steranes, the C_{27} of diasteranes to regular steranes ratio, when high, suggests a higher pH, lower Eh depositional environment (Peters et al., 2005). The values range between 0.26 and 0.39 with an average of 0.31 (Table 2), whereas the pregnane/C_{29}-20R regular sterane values are all around 0.30 to 1.08. Superficially, this suggests dysoxic to anoxic, normal saline conditions.

5.4 Paleoceanographic implications

The lower Cambrian black shales in South China have long been regarded as deposited in an anoxic slope-basinal environment (Goldberg et al., 2007; Wille et al., 2008; Jiang et al., 2009; Zhou and Jiang, 2009). During this time, the Yangtze Platform was located within a tropical-subtropical latitude (Kirschvink, 1992). Starting in the Early Cambrian a worldwide transgression regionally called "Niutitang/Badaowan Event" (Steiner, 2001), has been postulated together with a greenhouse type climate (Brasier, 1992; Wang et al., 1998). The proposed bottom water anoxia and greenhouse climate are apparently contrary to the in situ preservation of large benthic sponge body fossils (Steiner et al., 1993), SSFs (Steiner et al., 2005) and $\delta^{13}C_{org}$ (Brasier, 1992; Goldberg et al., 2007; Chen et al., 2009; Jiang et al., 2011). Several rapid $\delta^{13}C$ excursions of carbonates and OC during the Lower Cambrian likely reflect changes in OC burial, possibly linked directly to variations in primary productivity (Goldberg et al., 2007).

In the Niutitang Formation, the TOC and $\delta^{13}C_{org}$ values of silicites are the lowest at 0.25% and −32.82‰, respectively, followed by a rapid increase to values as high as 12.31% and −31.49‰ in the laminated and carbonaceous shales (Fig. 2 and Table 1). Accordingly, Chen et al. (2009) researched in detail the variation in $\delta^{13}C_{org}$ across the PC-C succession at Ganziping, Dayong, Hunan Province, and identified three negative (N1, N2 and N3) and two positive (P1 and P2) excursions, which likely correspond to those in western Hubei (Ishikawa et al., 2008), northeastern Yunnan (Zhou et al., 1997), northern Siberia (Kaufman et al., 1996) and Morocco (Magaritz et al., 1991). Our $\delta^{13}C_{org}$ curve displays a positive excursion of 1.4‰, probably consistent with P2, which was interpreted as possible CO_2 recycling, high primary productivity, sea level rise and so on. Kunimistu et al. (2009) interpreted the increased primary productivity and settling of organisms to the sea-floor as likely resulting in reducing conditions at the sediment-water interface, causing high TOC values and low Pr/Ph values. The Early Cambrian Niutitang Formation afforded an abundant and diverse sponge assemblage (Zhao et al., 1999), the occurrence of the trilobites *Zhenbaspis*, *Runnania*, *Tsunyidiscus* and *Mianxiandiscus* (Zhou and Jiang, 2009) and the SSFs (Budd, 2003; Steiner et al., 2007), implying, in general, dramatic environmental changes (Zhu et al., 2007; Ishikawa et al., 2008). Therefore, high primary productivity during this time is likely to have been responsible for selective uptake of ^{12}C from the ocean DIC (dissolved inorganic carbon) pool, resulting in high $\delta^{13}C_{org}$ values (Ishikawa et al., 2008). High productivity could have gradually led to bottom water anoxia, caused by the aerobic degradation of OM during its passage to the sea floor (Goldberg et al., 2007; Wille et al., 2008; Jiang et al., 2009). The relatively positive excursion is probably caused by an enhanced rate of OC burial. Furthermore, the depositional period of the lower Niutitang Formation coincides with a transgression event leading to extended black shale deposition from shallower to deeper parts of the

Yangtze Platform (Brasier, 1992; Zhu et al., 2003; Goldberg et al., 2007). This event was accompanied by enhanced upwelling (Li et al., 1999), which may have brought dissolved nitrogen species and phosphate to the surface and increased primary productivity (Yang et al., 2007). Trappe (1998) indicated that the formation of phosphatic rocks, such as those in the lowermost Cambrian (Nemakit-Daldynian) on the Yangtze platform, was favoured in suboxic/anoxic conditions.

Based on the above discussion, sea level rise and deposition of carbonate and phosphorites occurred on the shelf in Nemakit-Daldynian, with cherts and phosphate nodules in the basin (Goldberg et al., 2007) resulting from anoxic to dysoxic conditions with moving levels of the chemocline. During the Lower Tommotian, further sea level rise led to the deposition of black shales in shallow and deep regions of the Yangtze Platform. In contrast, OC and $\delta^{13}C_{org}$ values were relatively higher under the chemocline rise and widespread anoxic conditions. Taking into account the biomarkers, low Pr/Ph values, gammacerane, 25-norhopanes and methylsteranes indicated the deposition of early Cambrian black shales was under anoxic, stratified water column conditions accompanied by anaerobic bacteria, including methanotrophs and sulfate-reducing bacteria. Furthermore, the Mo isotopes of the Niutitang Formation black shales can provide insight into the redox condition of the ocean in relation to intense upwelling of H_2S-rich deep ocean water (Wille et al., 2008).

6 Conclusions

A new organic geochemical basis has been established for the Early Cambrian black shales in Hunan Province, South China. Biomarker parameters can provide insight into OM origin and maturity, redox conditions and palaeoenvironmental changes. In accord with the biomarker composition, the *n*-alkanes are characterized by a unimodal pattern with a predominance of lower carbon numbers and no distinct OEP or CPI, indicating that algae and bacteria are the important primary producers. Furthermore, the low Pr/Ph (<0.8) values and 25-norhopanes and gammcerane occurrence provide information to the effect that the OM was preserved under dysoxic to anoxic conditions, with normal marine salinity and a stratified water column. In general, Ts/(Ts+Tm), αβ-homohopane 22S/(22S+22R) and C_{29} ααα sterane 20S/(20S+20R) values can be used to assess maturity. Our samples show higher levels of maturity through the whole section. However, the triggers for the widespread black shale deposition and Cambrian explosion remain unclear and subject to intense debate although there is much geological and geochemical evidence. Using organic carbon isotopes and biomarker proxies, we found that positive carbon isotope excursions close to 1.4 ‰ are consistent with CO_2 recycling, high primary productivity and sea level rise. The increased primary productivity likely resulted in reducing conditions at the sediment -water interface, and caused high TOC values and low Pr/Ph values.

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