



Stable isotopic and elemental characteristics of pale and dark layers in a late Pliocene lignite deposit basin in Yunnan Province, southwestern China: Implications for paleoenvironmental changes

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ABSTRACT

The carbon and nitrogen stable isotopic compositions and elemental characteristics of Pliocene pale and dark lignites in the Jinsuo Basin, Yunnan Province, southwestern China, were investigated. These data were used to develop paleoenvironmental models and to discuss possible geological driving forces for the formation of pale and dark layers.

The $\delta^{13}\text{C}$ values of fossil wood (av. -24.79‰) and fusain samples (av. -24.19‰) are generally higher than the values obtained from bulk pale (av. -27.01‰) and dark (av. -26.38‰) lignite samples. In contrast, the $\delta^{15}\text{N}$ values of fossil wood (av. -0.63‰) and fusain samples (av. 1.29‰) are lower than the average values of bulk pale lignites (av. 2.17‰) and dark lignites (av. 1.79‰). The difference in the carbon isotopic values between pale and dark lignites can be explained by the very high amount of liptinite and strong degradation of woody material by bacterial activity in pale lignites, which resulted in depletion of ^{13}C in pale lignites, and the wet/humid depositional conditions contributed to the relatively heavy stable carbon isotope compositions of dark layers. The higher $\delta^{13}\text{C}$ values in the fossil wood and fusain samples can be explained by the general enrichment of ^{13}C in woody material compared to leaves. The weakly negative correlation between the $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values indicates that, besides bacterial degradation, the liptinite content could also influence the carbon and nitrogen compositions of lignites. The influence of the original plant community (gymnosperms/angiosperms) on the carbon and nitrogen isotopic compositions of bulk lignites is insignificant in the Jinsuo Basin.

The paleoenvironmental model suggests deposition of pale layers under relatively dry, cold conditions supporting the expansion of gymnosperms such as *Pinus yunnanensis*. The slight stepwise uplift of the Tibetan Plateau and the approximate co-occurrence of the more intense Asian monsoon might have supported short periods of a locally dry, cold climate resulting in the lowering of water table which was favorable for the formation of pale layers. The low $\delta^{13}\text{C}$ values in pale layers further support this. The relatively high $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios, higher values of the average detrital/ authigenic index, and high levels of TiO_2 and Zr in pale layers suggest an increased detrital input relative to dark layers. The dark layers, however, formed in a tropical/subtropical warm, humid climate that favored angiosperm dominance of the vegetation. The long-term geologically stable conditions together with the regionally warm, humid climate resulted in a rise in the water table in the basin, which led to the formation of dark layers until the subsequent slight uplift of the Tibetan Plateau. This conclusion is also supported by the high $\delta^{13}\text{C}$ values in dark layers and the thickness differences between pale (thin) and dark (thick) layers and the changes in major elements and minerals changes between the two types of layer.

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1. Introduction

Studies of stable carbon isotope ratios of organic matter from lignites have, in recent years, been more and more used to improve reconstructions of floral assemblages in paleomires and paleoenvironmental changes during deposition of lignites all over the world (Bechtel et al., 2003, 2004, 2008, 2019; Harrington et al., 2005; Holdgate et al., 2009; Kanduć et al., 2012; Korasidis et al., 2016; Stock et al., 2016; Erdenetsogt et al., 2017; Radhwani et al., 2018). The $\delta^{13}\text{C}$ variations of lignites are affected by several factors, such as the degree of degradation of plant tissues, differing lipids contents in extractable organic matter, changes of vegetation in the peat, differences in the processes of carbon cycling and differences in the stable carbon isotopic value of atmospheric CO_2 (Bechtel et al., 2007a, 2008; Diefendorf et al., 2015; Nordt et al., 2016; Radhwani et al., 2018). The stable carbon isotopic compositions of cellulose/fossil wood in lignites have been further linked to climatic changes (e.g. temperature and humidity) because the efficiency of water use by land plants is different (Lipp et al., 1991; Schleser et al., 1999; van Bergen and Poole, 2002; Bechtel et al., 2008; Jahren and Sternberg, 2008). Benner et al. (1987) found that variations of the stable carbon isotope compositions of organic matter are, in addition, dependent on the abundance of decay-resistant macromolecules. The ^{13}C depletion in lignites preferentially results from the progressive decay of cellulose by bacterial activity (Bechtel et al., 2008). Negative relationships between the $\delta^{13}\text{C}$ values and lignite contents of lignites (Bechtel et al., 2002, 2005, 2007a) indicate that the composition of stable carbon isotope in lignite is influenced by the content of lipids in organic matter. The influence of plant community changes (gymnosperms versus angiosperms) on $\delta^{13}\text{C}$ is caused by the approximately 2.5‰ on average higher $\delta^{13}\text{C}$ values of gymnosperms relative to angiosperms (Bechtel et al., 2008; Widodo et al., 2009; Chi et al., 2013) resulting from different leaf morphology as well as carbon assimilation efficiency of different plants (Murray et al., 1998).

In contrast to the large number of studies of the carbon isotopic composition of lignites, lignite nitrogen isotopic compositions have rarely been investigated. A previous study of Pliocene lignites from Slovenia, demonstrated that the $\delta^{15}\text{N}$ values of different lithotypes were influenced by a combination of compositional changes in the parent plant materials and biogeochemical processes during early diagenesis (Kanduć et al., 2005). In contrast, the nitrogen isotopic compositions of organic matter in mature sediments and coals have been studied in more detail. Variations of the nitrogen isotopic signatures have been explained by variations of several parameters, such as climate, source of organic materials, depositional environment, maturity, and degree of decomposition of organic matter (Rigby and Batts, 1986; Quan et al., 2008; Morford et al., 2011). Previous studies suggested that $\delta^{15}\text{N}$ values do not change systematically with increasing maturity of coals/oil shales (Rigby and Batts, 1986; Whiticar, 1996; Ader et al., 1998). Ader et al. (1998) found slightly different $\delta^{15}\text{N}$ values in anthracites from the Pennsylvanian (U.S.A.) and the Bramsche Massif (Germany) and interpreted this as a result of changes in the degree of decomposition of organic matter, plant community, and paleoclimate. The nitrogen content of bedrock sediments and its weathering might also cause alterations in the nitrogen isotopic values in terrestrial ecosystems such as plants and soils (Morford et al., 2011). Furthermore, bacterial alteration of organic matter in woody tissues during early diagenesis and the peat-forming stage could also lead to preferential loss of ^{14}N (Kanduć et al., 2005; Rimmer et al., 2006). Results from laboratory experiments (Lehmann et al., 2002) suggest that the $\delta^{15}\text{N}$ values of organic matter, when decayed under anoxic conditions, decrease to about 3‰ lower values than the original values, whereas the $\delta^{15}\text{N}$ values of organic matter are first increasing, and then decreasing when going through oxic decay (Lehmann et al., 2002). Residual organic matter in alkaline or aerobic environments yields higher nitrogen isotope values than their parent plants (Taylor et al., 1998; Zhu et al., 2000). Moreover, less degradable stem xylem provides higher $\delta^{15}\text{N}$ values than the values

obtained from the degradable leaves (Handley and Raven, 1992).

The Pliocene lignites of the Jinsuo Basin in Yunnan Province, have previously been studied to evaluate the origin of pale and dark layers in these lignites (Ming et al., 1994; Wang et al., 1997; Yu et al., 1997; Liu et al., 2018). The most recent study suggested that pale layers in the lignites are partly composed of gymnosperms and were deposited under dry/aerobic conditions, whereas dark layers are dominated by organic matter from angiosperms and were deposited under prevailing wet/anaerobic conditions (Liu et al., 2018). In this paper, new data on stable carbon and nitrogen isotopes determined from pale and dark lignites and fossil wood samples from the Jinsuo Basin are presented and potential reasons for the observed variations in the carbon and nitrogen isotope compositions are discussed. In addition, the major elements and trace elements in the lignites were measured, to identify the sedimentary sources of the two kinds of lignite. The combined application of stable isotopic and geochemical methods is expected to further improve understanding of the formation of pale and dark lignite layers in the Jinsuo Basin, Yunnan Province, southwestern China.

2. Geological setting

Yunnan Province is geologically located at the junction of the East Asian Monsoon Zone, the Southern Asia–Qinghai/Tibet Plateau Zone, and the Southcentral Tropical Peninsula Monsoon geographical zone (Feng et al., 1998; Xu et al., 2004). Yunnan Province was influenced by the Himalayan movement from the late Eocene to the early Pleistocene (Yunnan BGMR, 1990). The Himalayan orogenic movement resulted in formation of a highly fragmented terrain in Yunnan Province, which was responsible for the variations in temperature and precipitation from the Neogene to the Quaternary (Xu et al., 2004). During the Neogene, the main sedimentary facies in Yunnan Province was lacustrine and several intermountain coal-bearing basins were formed (Yunnan BGMR, 1990).

The Jinsuo Basin is located in the low hilly area of the karst plateau in eastern Yunnan. The basin is located between the East Xiaojiang Fault and the West Xiaojiang Fault (Fig. 1a), and is developed in a north–south direction with an area of about 18 km². During the Himalayan movement, the strata in this area elevated slowly without any intense tectonic activities during the late Pliocene (Yunnan BGMR, 1990). In the Jinsuo Basin, the Ciyang Formation, which belongs to the late Pliocene (Yunnan BGMR, 1990, Wang and Shu, 2004; Hou et al., 2006; Huang et al., 2015), contains four (No. 1 to 4 from top to bottom) lignite seams (Fig. 1b) and the uncomfortable strata underlying the Ciyang Formation consist mainly of a mixture of middle Cambrian calcareous shales of the Douposhi Formation and dolomites of the Shuanglongtan Formation, together with Lower Ordovician shales of the Tangchi Formation and sandstones of the Hongshiya Formation (Yunnan BGMR, 1990; Liu et al., 2018). The stratigraphic column of the Neogene succession in the Jinsuo Basin is shown together with the profile of the studied section in Fig. 1b. The profile of lignite seam No. 3, which was investigated in the present study, is enlarged in Fig. 1b. Further details of the lignite-bearing strata of the studied section were provided by Liu et al. (2018).

3. Samples and analytical methods

A total of 49 samples were collected from the lignite profile in the Jinsuo Basin. The lignite samples are composed of 18 pale lignites, 19 dark lignites, and 2 charcoal samples, marked JS-01 to JS-39 from bottom to top of the succession (Fig. 1b). Bulk pale and dark samples were collected from the continuous sequence of pale and dark layers. Two charcoal samples were collected from the lignite seam, and 10 fossil wood pieces were picked out from both pale and dark layers. Each sample was wrapped in aluminum foil in the field, to minimize contamination and oxidation, and stored in a plastic bag that was immediately sealed. Samples were crushed and ground to pass through a

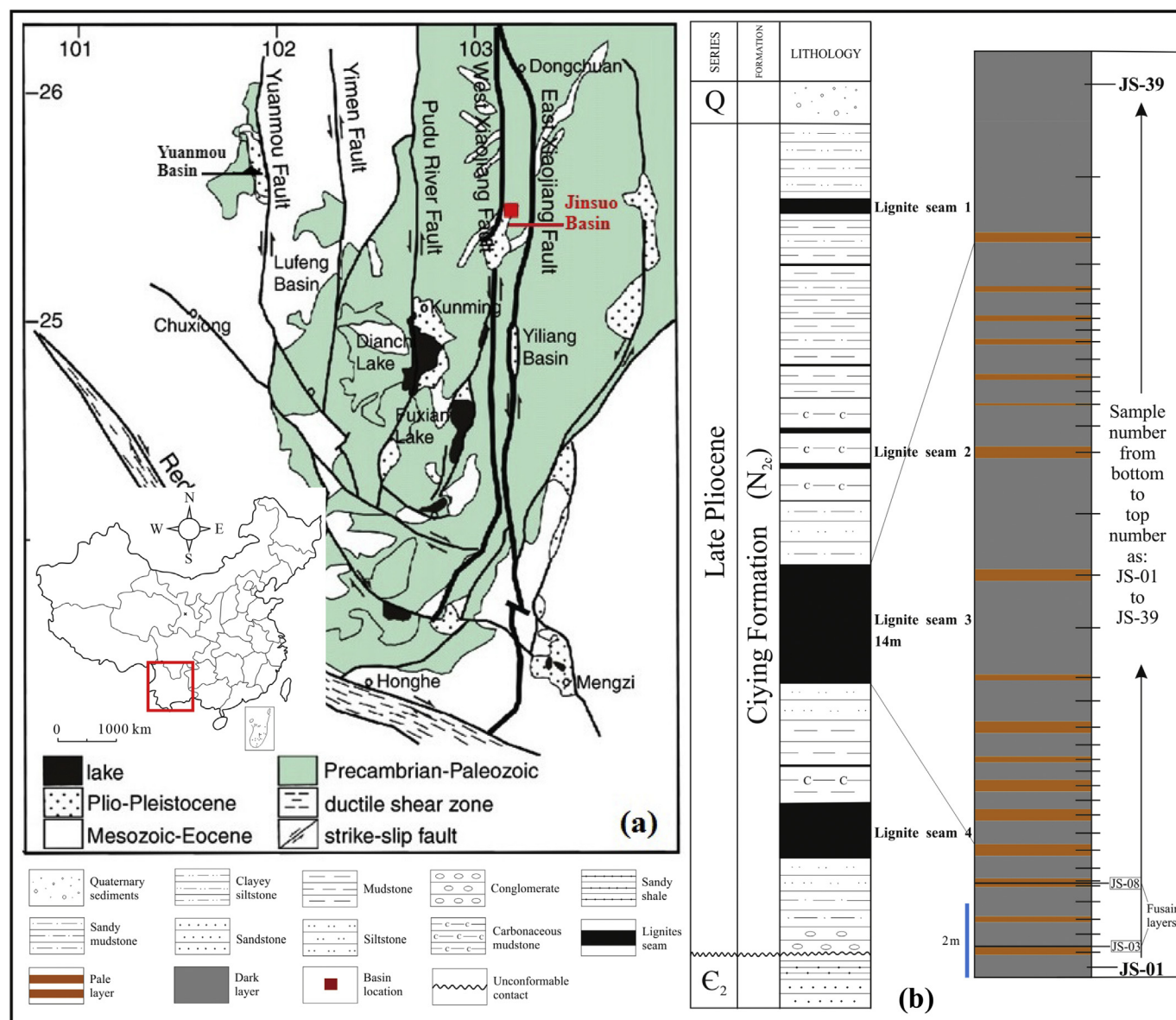


Fig. 1. Geological map and location of the Jinsuo Basin (a), modified from Zhu et al., (2008), along with the stratigraphic column and sample profile of the Jinsuo opencast lignite mine (b). Q = Pleistocene, ϵ_2 = middle Cambrian. Sample JS-03 and JS-08 are fusain samples. Brown color: pale layers; gray color: dark layers. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

200-mesh sieve for X-ray fluorescence (XRF) spectrometry, inductively coupled plasma mass spectrometry (ICP-MS), and stable isotopic composition analyses.

Ash yields of lignite samples were measured following American Society for Testing and Materials (ASTM) Standard D3174-11 (ASTM Standard D3174-11, 2011). The coal petrological composition of the samples has been presented previously by Liu et al. (2018). The random huminite reflectance was measured on ulminite B. However, due to the very low proportion of ulminite in pale layers (Liu et al., 2018), the reflectance of some pale lignite samples could not be determined.

Proportions of major-element oxides in all lignite and fusain samples (on an ash basis; ashing at 815 °C) were analyzed by XRF spectrometry (OXAS, Version 2.2), and proportions of major-element oxides in the whole coal sample were calculated from the ash yield of each sample. The ICP-MS instrument (X series II) was operated in a pulse-counting mode to detect trace elements such as Zr in the lignite samples. The detailed procedure and technical parameters of the XRF and ICP-MS analyses of the samples was described by Zhao et al. (2017) and Zhao et al. (2019), respectively. The sample digestion procedures

for ICP-MS analyses were carried out following the methods described by Dai et al. (2011).

The distribution of minerals in lignite samples were measured with scanning electron microscope (HITACHI, SU8220) coupled with an energy-dispersive X-ray spectrometer (SEM-EDS).

The stable isotopic compositions of organic carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$) in all lignite and fossil wood samples were measured using a FlashEA™ 1112 Elemental Analyzer with a ConFlo III Interface from Thermo Fisher. The detailed procedures followed in for the analyses were described previously by Liu et al. (2019).

4. Results and discussion

4.1. Stable isotopic composition of lignites and fossil wood

4.1.1. Stable carbon isotopic composition of lignite samples

The stable carbon isotope values ($\delta^{13}\text{C}$) and stable nitrogen isotope values ($\delta^{15}\text{N}$) of pale and dark lignite samples and fossil wood and fusain samples from the studied profile in the Jinsuo Basin are shown in

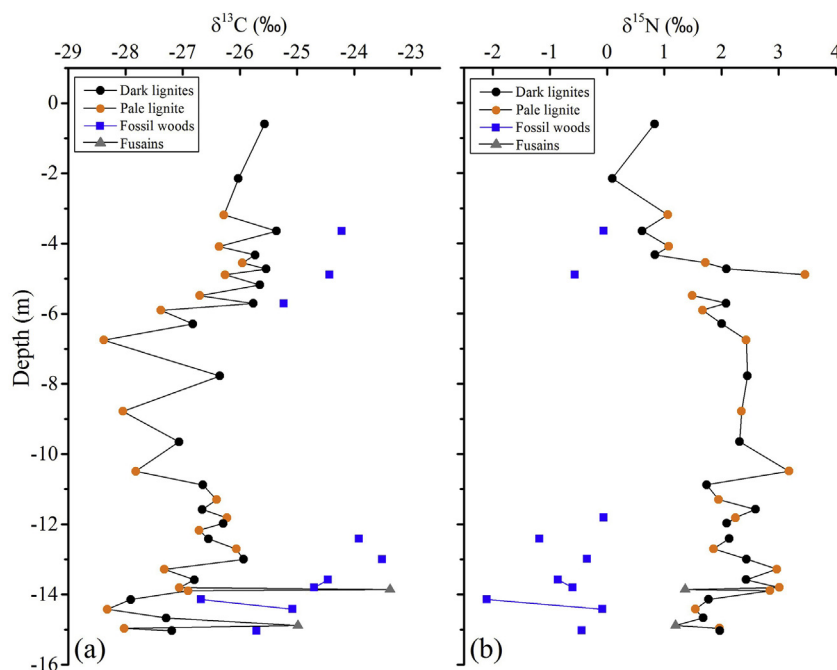


Fig. 2. Stable carbon (a) and nitrogen (b) isotopic composition of samples (pale, dark, fossil wood and fusain samples) collected from the Pliocene lignite profile in the Jinsuo Basin, Yunnan Province, southwestern China.

Fig. 2 and the data are listed in Table 1. The $\delta^{13}\text{C}$ values of all pale and dark bulk lignite samples are between -28.38‰ and -25.36‰ , with an average value of -26.69‰ (Table 1). The two fusain samples yielded much higher values (-24.99‰ for JS-03 and -23.38‰ for JS-08) and were therefore excluded from the calculation of average values. These $\delta^{13}\text{C}$ values of lignites from the Jinsuo Basin exhibit a similar range of values to those of Miocene lignites from the Lower Rhine Basin (-24.5‰ to -26.7‰ ; Lücke et al., 1999), Pliocene lignites from the Velenje Basin (-25.3‰ to -27.0‰ ; Bechtel et al., 2003), and Cenozoic lignites from Central Europe (-27.4‰ to -23.8‰ ; Bechtel et al., 2008). In the Jinsuo Basin, pale lignite samples have $\delta^{13}\text{C}$ values ranging from -28.38‰ to -25.96‰ , average -27.01‰ , and the $\delta^{13}\text{C}$ values of dark lignite samples range from -27.91‰ to -25.36‰ , with an average value of -26.38‰ (Table 1). On average, the $\delta^{13}\text{C}$ values of pale lignites are 0.63‰ lighter than the values obtained from the dark lignites. These results differ from those obtained previously from the M1B seam in the Gippsland Basin, Australia, as pale and light lithotypes show relatively high $\delta^{13}\text{C}$ values (av. -23.75‰ and av. -24.5‰ , respectively) whereas the medium light and medium dark lithotypes possess light carbon isotope compositions (av. -26‰ and av. -26.25‰ , respectively), and the laminated dark and dark lithotypes have intermediate $\delta^{13}\text{C}$ values (av. -25.25‰ and av. -25.5‰ , respectively; Korasidis et al., 2016).

The results are graphically presented in Fig. 2a along the investigated profile. The $\delta^{13}\text{C}$ values of dark lignite samples are generally higher than those of adjacent pale lignite samples. Fig. 2a also shows the outlying heavy $\delta^{13}\text{C}$ values of the fusain samples collected from the lower part of the section. From previous studies (Bechtel et al., 2008), one would expect that the increasing abundance of gymnosperms in the paleomire should result in increasing $\delta^{13}\text{C}$ values of the resulting bulk organic matter, because gymnosperms are generally about 2.5‰ heavier than angiosperms growing in the same mire. However, in the present case of the Jinsuo lignites, the opposite trend is observed. The $\delta^{13}\text{C}$ values of the dark lignites, which are dominated by angiosperms, are on average 0.63‰ heavier than those of pale lignites, which are related to varying extents in being composed of additional gymnosperms, on the basis of the abundance of diterpenoids (Liu et al., 2018). Furthermore, the angiosperm-dominated pale lithotypes in the

Gippsland Basin of Australia generally have heavier carbon isotope compositions than the gymnosperm-dominated dark lithotypes (Holdgate et al., 2016; Korasidis et al., 2016). These differences have been suggested to result from the high levels of aerobic degradation of the peat during the formation of pale/light lithotypes (Korasidis et al., 2016) and the subaqueous depositional environment during formation of the laminated dark layers (Holdgate et al., 2014; Korasidis et al., 2016). In the Jinsuo Basin, no obvious correlation has been observed between the $\delta^{13}\text{C}$ values and the di/(di + triterpenoids) values (the concentration ratios of diterpenoids versus the sum of diterpenoids plus angiosperm-derived triterpenoid hydrocarbons) of lignites (Liu et al., 2018). This is consistent with data from a previous study on the lower Miocene Oberdorf lignite seam of Austria, which also exhibited a lack of correlation between the two parameters (Bechtel et al., 2002). According to those authors, the influence of vegetation changes on the carbon isotopic composition of the lignites could be negligible when the variations of the peat-forming vegetation (gymnosperms vs. angiosperms) are not significant.

In the present study, the correlation coefficient (r) between the $\delta^{13}\text{C}$ values and total organic carbon (TOC) is different between bulk pale and dark samples (excluding fossil wood and fusain) from the Jinsuo Basin. The positive correlation between the $\delta^{13}\text{C}$ values and TOC in dark lignites ($r = 0.62$, Fig. 3a) is much higher than weak negative correlation in pale lignites ($r = -0.34$, Fig. 3b). This finding can probably be explained by the better preservation of organic matter in the dark lignites whereas the organic matter in pale lignites was strongly reworked by microbial activity (Liu et al., 2018), because degradation by bacteria/fungi may result in changes in the stable carbon isotope ratio of the original plant material resulting from differences in $\delta^{13}\text{C}$ values between easily degradable polysaccharides and decay-resistant macromolecules such as lignin (Benner et al., 1987). Bechtel et al. (2008) reported that higher ratios of gymnosperms to angiosperms in the original peat-forming vegetation resulted in a $\delta^{13}\text{C}$ shift to heavier values in Tertiary lignites and brown coals from Central Europe. In the present case of Pliocene lignites from the Jinsuo Basin, this shift is not observed, because the positive shift of $\delta^{13}\text{C}$ values caused by floral changes was presumably overwhelmed by the negative shift caused by increased biodegradation of easily degradable organic

Table 1

Values of stable carbon isotopes ($\delta^{13}\text{C}$) and stable nitrogen isotopes ($\delta^{15}\text{N}$) of lignites and fossil wood, and reflectance (Ro, in %) of lignite from the Jinsuo Basin, Yunnan Province. Total organic carbon (TOC) and total nitrogen (TN) data are from Liu et al. (2018).

Sample	Lithotypes	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	Ro	TOC (wt%)	TN (wt%)	Sample	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$
JS-39	dark	-25.57	0.83	0.281	51.3	1.32	W ⁶ -1	-25.71	-0.44
JS-38	dark	-26.03	0.09	0.262	55.2	1.22	W-5	-25.08	-0.08
JS-37	pale	-26.28	1.06	0.248	52.5	0.76	W-6	-26.68	-2.10
JS-36	dark	-25.36	0.61	0.256	55.8	1.26	W-9	-24.70	-0.60
JS-35	pale	-26.36	1.08	0.258	54.3	1.02	W-10	-24.46	-0.86
JS-34	dark	-25.74	0.84	0.252	55.2	1.09	W-12	-23.51	-0.35
JS-33	pale	-25.96	1.72	0.236	55.2	0.92	W-14	-23.92	-1.18
JS-32	dark	-25.54	2.09	0.249	55.8	1.09	W-28	-25.23	-0.06
JS-31	pale	-26.26	3.46	0.235	53.7	0.81	W-31	-24.43	-0.56
JS-30	dark	-25.65	n.a. ⁷	0.242	47.7	1.11	W-36	-24.22	-0.06
JS-29	pale	-26.71	1.49	0.231	57.0	0.90	AV-W ³	-24.79	-0.63
JS-28	dark	-25.77	2.08	0.282	52.8	1.07	AV-F ⁴	-24.19	1.29
JS-27	pale	-27.38	1.67	0.239	53.4	0.78	AV-L ⁵	-26.69	1.94
JS-26	dark	-26.82	2.01	0.222	48.6	1.01			
JS-25	pale	-28.38	2.43	0.221	62.4	0.68			
JS-24	dark	-26.35	2.46	0.226	54.6	1.28			
JS-23	pale	-28.05	2.35	0.236	54.6	1.07			
JS-22	dark	-27.07	2.32	0.238	46.2	1.36			
JS-21	pale	-27.82	3.18	n.a.	52.2	0.73			
JS-20	dark	-26.65	1.74	0.242	51.0	1.16			
JS-19	pale	-26.41	1.95	n.a.	55.8	1.27			
JS-18	dark	-26.66	2.59	0.248	49.2	1.35			
JS-17	pale	-26.23	2.24	n.a.	43.8	0.94			
JS-16	dark	-26.30	2.09	0.235	51.6	1.16			
JS-15	pale	-26.35	n.a.	n.a.	56.4	0.71			
JS-14	dark	-26.55	2.13	0.241	53.4	1.36			
JS-13	pale	-26.06	1.86	n.a.	42.0	0.77			
JS-12	dark	-25.94	2.44	0.249	51.6	1.29			
JS-11	pale	-27.32	2.97	n.a.	52.8	1.03			
JS-10	dark	-26.80	2.43	0.248	50.4	1.38			
JS-9	pale	-27.06	3.01	n.a.	45.6	0.84			
JS-8	fusain	-23.38	1.37	n.a.	55.8	n.a.			
JS-7	pale	-26.90	2.85	n.a.	49.8	0.95			
JS-6	dark	-27.91	1.77	0.254	45.0	1.19			
JS-5	pale	-28.32	1.54	n.a.	51.0	0.93			
JS-4	dark	-27.29	1.68	0.251	48.6	1.18			
JS-3	fusain	-24.99	1.20	n.a.	52.2	n.a.			
JS-2	pale	-28.02	1.97	0.242	52.2	0.87			
JS-1	floor	-27.19	1.97	0.314	54.3	1.00			
AV-P ¹		-27.01	2.17	0.238	52.5	0.89			
AV-D ²		-26.38	1.79	0.249	51.3	1.21			

¹ AV-P = average of pale lignites

² AV-D = average of dark lignites

³ AV-W = average of fossil woods

⁴ AV-F = average of fusain samples

⁵ AV-L = average of all lignite samples

⁶ W = fossil wood samples

⁷ n.a. = Not analyzed

matter.

Negative deviations of $\delta^{13}\text{C}$ values generally occur in pale lignites (Fig. 2a). Bechtel et al. (2002) found a negative correlation between $\delta^{13}\text{C}$ values and the contents of liptinite macerals in a lower Miocene lignite seam from the Styrian Basin, Austria, because the $\delta^{13}\text{C}$ values of lipids from plants are 5–10 per mil depleted relative to whole plant materials/bulk organic matter (Park and Epstein, 1961; Degens, 1969). Light $\delta^{13}\text{C}$ values are also characteristic of *n*-alkanes, plant/bacteria-derived isoprenoids, and hopanoids (Murray et al., 1998). As there are very high amounts of liptinite macerals and high concentration of *n*-alkanes and hopanoids in the pale lignites of the Jinsuo Basin (Liu et al., 2018), this could be one reason for the depletion in ^{13}C in pale lignites relative to dark lignites. This explanation is also supported by results from previous studies of coal indicating that samples with high lipid contents (liptinites) showed the most depleted carbon isotopic values compared to macerals derived from cellulose and lignin (Rimmer et al., 2006). Furthermore, the poor preservation of fossil woody material and the low content of suberinite and textinite in pale lignites (Liu et al., 2018) might also explain the relatively lower $\delta^{13}\text{C}$ values compared to

the dark lignites, because resistant woody materials such as charcoal, generally yield ^{13}C values that are 1‰–3‰ higher than the values for leaves (Klump et al., 2005; Cernusak et al., 2009). This conclusion is further supported by the previously observed negative relationships between the $\delta^{13}\text{C}$ values of bulk lignite samples and the concentrations of hopanes in the extracts of these samples (Bechtel et al., 2005, 2007b), which has been explained by ^{13}C depletion of lignites by bacterial activity caused by the progressive decay of cellulose (Bechtel et al., 2008). Thus, the intense bacterial activity, along with the high concentration of *n*-alkanes in pale lignites (Liu et al., 2018) probably indicates that higher bacterial activity in pale lignites lowered the $\delta^{13}\text{C}$ values more than the contribution of gymnosperms could increase the values in this case. The dark lignites have relatively heavy carbon isotope compositions, as was also detected in the dark and laminated dark lithotypes in the Gippsland Basin (Holdgate et al., 2014). The higher $\delta^{13}\text{C}$ values in the laminated dark lithotypes were suggested to have been resulted from the wet depositional environment during peat formation, because this wet/sub-aqueous depositional environment produced organic matter with heavy $\delta^{13}\text{C}$ values (discussed by Holdgate

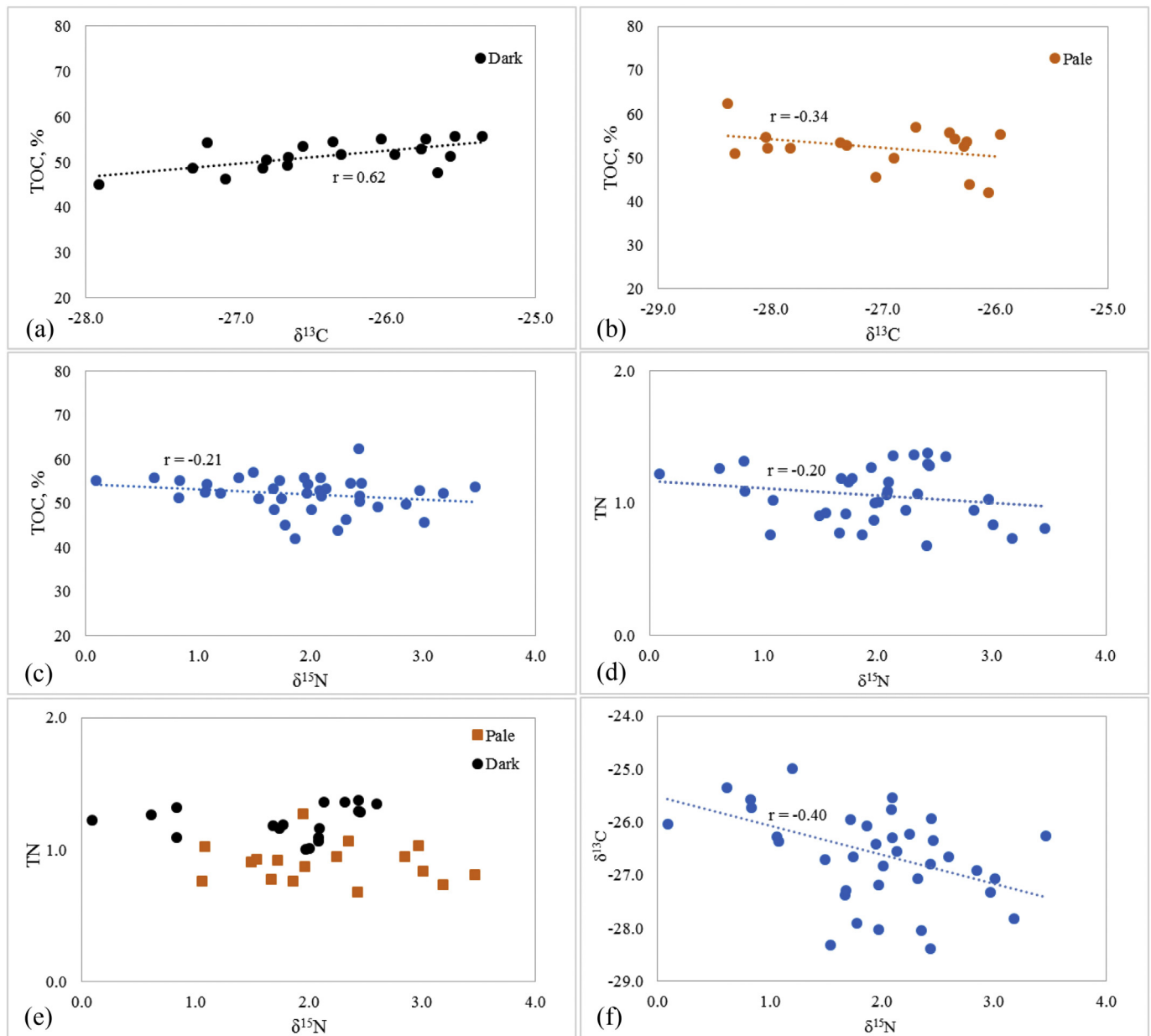


Fig. 3. Correlation between $\delta^{13}\text{C}$ values and TOC of pale (a) and dark (b) samples, together with $\delta^{15}\text{N}$ values versus TOC (c), TN (d and e) and $\delta^{13}\text{C}$ (f) values of all bulk lignite samples from the Jinsuo Basin. TOC and TN data are from Liu et al. (2018).

et al., 2014) as a result of decreasing diffusion of carbon dioxide in plants and water use efficiency in aqueous conditions (Farquhar et al., 1989). The similarly wet depositional conditions (Liu et al., 2018) have contributed to the heavy carbon isotope compositions of dark lignites in the Jinsuo Basin.

4.1.2. Stable carbon isotopic composition of fossil wood samples

The $\delta^{13}\text{C}$ values of 10 fossil wood pieces range from -26.68‰ to -23.51‰ , with a mean value of -24.79‰ (Fig. 2, Table 1). Such values are consistent with previously published data for Neogene fossil wood in lignites from Central Europe (Lücke et al., 1999; Bechtel et al., 2007c, 2008) and modern plants (Smith and Epstein, 1971). The stable carbon isotopic values of fossil wood remains in the Jinsuo Basin are on average higher by 2.22‰ compared to the corresponding bulk lignite material of pale lignites and by 1.59‰ compared to dark lignites (Table 1). Similar positive shifts in $\delta^{13}\text{C}$ values between bulk material of lignites and brown coals and fossil wood collected from the same

lignites have also been reported from the Miocene Lower Rhine Embayment in Germany (Lücke et al., 1999) and the Oligocene Trbovlje lignite in Slovenia (Bechtel et al., 2004). The isotopically lighter values of carbon in bulk lignites compared with values of fossil wood pieces is caused by the higher contribution of organic matter with light isotopic values in the coal matrix (e.g. leaves, resins and barks) (Park and Epstein, 1961) or results from higher microbial activity in the lignite matrix than in the wood fragments. The highest $\delta^{13}\text{C}$ values in the studied section of the Jinsuo Basin were detected in the fusain samples, being 2.83‰ heavier than the average value for pale lignites and 2.20‰ heavier than the average value for dark lignites (Table 1). The average $\delta^{13}\text{C}$ value of fusain samples is only 0.6‰ heavier than the value of fossil wood samples. This shift in $\delta^{13}\text{C}$ values between fusain samples and bulk lignite samples (more than 2‰) is higher than the observed difference between fusain samples and fossil wood pieces (0.6‰). Heavy carbon isotope compositions were also reported previously from the abundant macro- and micro-charcoal lithotypes in

Cenozoic lignite seams from the Gippsland Basin, Australia (Holdgate et al., 2014; Korasidis et al., 2016) with a range of values from -25.50‰ to -25.25‰ (Korasidis et al., 2016). Similar relative $\delta^{13}\text{C}$ enrichments have also been found in high-charcoal soils in Brazil (Gouveia et al., 2002; Pessenda et al., 2004), which was explained by the generally observed 1‰ – 3‰ enrichment in ^{13}C of woody materials, including charcoal, compared to leaves (Nordt et al., 2016, and references therein).

4.1.3. Stable nitrogen isotopic composition

The nitrogen isotope ($\delta^{15}\text{N}$) values of all bulk lignite samples range from 0.09‰ to 3.46‰ , with a mean value of 1.94‰ (Table 1, Fig. 2b). These values are lower than the $\delta^{15}\text{N}$ values (1.8‰ – 4.6‰ ; average 2.99‰) previously reported from Pliocene lignite lithotypes from the Velenje Basin, Slovenia (Kanduč et al., 2012). The $\delta^{15}\text{N}$ values of pale lignites vary from 1.06‰ to 3.46‰ (av. 2.17‰) and the $\delta^{15}\text{N}$ values of dark lignites range from 0.09‰ to 2.59‰ (av. 1.79‰). Although the average $\delta^{15}\text{N}$ value of pale lignites is slightly higher than the average value of dark lignite, the $\delta^{15}\text{N}$ values in the whole profile are relatively consistent (Fig. 2b). The $\delta^{15}\text{N}$ values of the fusain samples are slightly lower than the values obtained from the adjacent bulk material (Table 1, Fig. 2b).

In contrast to the plentiful studies of the nitrogen isotopic composition of modern materials (e.g. plants, soils, and sediments), research reports focusing on the stable nitrogen isotopes of lignites/coal and fossil woods are scarce (Rigby and Batts, 1986; Ader et al., 1998; Kanduč et al., 2005; Rimmer et al., 2006). Stiehl and Lehmann (1980) investigated 18 coal samples of different rank, ranging from brown coal to anthracite, to investigate the influence of the coalification process on $\delta^{15}\text{N}$ values. In their study, the $\delta^{15}\text{N}$ values ranged from 3.5‰ to 6.3‰ and no continuous trend with increasing coalification was observed. This finding is not surprising, as the coal samples originated from different basins and different geological ages. The nitrogen isotopic composition of organic matter can also be altered as a result of chemical exchange with transported nitrogen, which may tend to homogenize the isotopic composition and limit the range of organic and inorganic nitrogen isotopic composition (Schimmelmann and Lis, 2010).

Further studies of coals/anthracites confirmed that the nitrogen isotopic composition is not correlated with either the age or rank of the coal (Rigby and Batts, 1986; Whiticar, 1996; Ader et al., 1998), and the variation of $\delta^{15}\text{N}$ values in coal could result from local variations in the ^{15}N content of the nitrate ions available for the original vascular plant growth (Rigby and Batts, 1986). Moreover, studies on macerals isolated from coals showed that vitrinite (Whiticar, 1996; Rimmer et al., 2006) and liptinites (Rimmer et al., 2006) yielded higher $\delta^{15}\text{N}$ values compared to inertinite because of the preferential loss of ^{14}N of woody tissues, possibly caused by bacterial alternation during peat formation/early diagenesis (Rimmer et al., 2006). In the present study, a very weak negative correlation coefficient ($r = -0.21$) was found between $\delta^{15}\text{N}$ values and TOC (Fig. 3c) which suggests that the nitrogen isotopic compositions of lignites from the Jinsuo Basin are not significantly related to changes in the amount of organic carbon. A weak negative correlation coefficient between $\delta^{15}\text{N}$ values and total nitrogen (TN, data from Liu et al. (2018)) has been found ($r = -0.20$, Fig. 3d), but in the plot of $\delta^{15}\text{N}$ values vs. TN (Fig. 3e) for bulk pale and dark lignite samples, the relatively high $\delta^{15}\text{N}$ values in pale lignites are associated with relatively low TN values, whereas the low $\delta^{15}\text{N}$ values in dark lignites correspond to relatively high TN values. This observation is consistent with results from a previous study showing that a decrease in organic nitrogen during aerobic degradation of organic matter is accompanied by an increase in the $\delta^{15}\text{N}$ values, and a decrease in the stable nitrogen isotope ratio of total nitrogen in anoxic sediments during early diagenesis (Freudenthal et al., 2001). This conclusion was further confirmed by results from petrological and organic geochemical investigations (Liu et al., 2018) indicating that pale layers were formed in a more aerobic sedimentary environment while dark lignites were

deposited under rather anaerobic conditions. Consequently, following the reasoning of Holmes et al. (1999) and Freudenthal et al. (2001), the difference in $\delta^{15}\text{N}$ values between pale and dark lignites could be explained by strong preferential degradation of organic matter containing amino acids as the major N-bearing component in pale lignites under oxic conditions. Amino acids, which are enriched in ^{15}N , have been shown to be preferentially degraded in the water column under relatively anoxic conditions, from analysis of particles collected in sediment traps in the ocean (Nakatsuka et al., 1997). In the present study, relatively higher $\delta^{15}\text{N}$ values correspond to relatively lower $\delta^{13}\text{C}$ values, resulting in a weak negative correlation ($r = -0.4$) of the $\delta^{15}\text{N}$ values with the $\delta^{13}\text{C}$ values (Fig. 3f), which is different from the positive correlation ($R^2 = 0.68$) of $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values in lignite from the Pliocene Velenje Basin, Slovenia (Liu et al., 2019). These observations can be interpreted by a higher degree of organic-matter degradation by aerobic bacteria in the liptinite-rich pale lignites than in liptinite-poor dark lignites formed in anaerobic conditions during peat formation/early diagenesis, which is supported by the organic geochemical results obtained from the same sample set (Liu et al., 2018).

The fossil wood samples have lower $\delta^{15}\text{N}$ values of -2.10‰ to -0.06‰ , with a mean value of -0.63‰ (Table 1, Fig. 2b) compared to bulk lignite samples (av. 1.94‰). No data on the nitrogen isotopic composition of fossil wood have previously been reported. Kanduč et al. (2005) determined the $\delta^{15}\text{N}$ values of recent plant parts around the Velenje Basin and detected low values in the range -3.7‰ to -2.3‰ . These values are much lower than those determined in the bulk lignites in the basin (1.8‰ to 4.6‰). The $\delta^{15}\text{N}$ values in modern higher land plants provide a broad variation of more than 20‰ (Peoples et al., 1991; Craine et al., 2015; Elena et al., 2016) and even in different parts, such as leaves, roots, and stems, of the same plants, the $\delta^{15}\text{N}$ values are different (Kolb and Evans, 2002). This huge variations make it very complicated to compare the $\delta^{15}\text{N}$ values of fossil wood pieces and lignites with values of modern plants. The lower $\delta^{15}\text{N}$ values in recent plants compared with the lignites from the Velenje Basin are, to some extent, consistent with the fact that recent plants are almost always more depleted in ^{15}N than the soils on which they grow (Craine et al., 2015). This is explained by the great isotopic fractionation during solubilization of N in soil and strong mycorrhizal transfer of N and fractionation from soil to plants leading to enrichment of ^{15}N in soils (Craine et al., 2015).

4.2. Chemical composition of lignite samples and implications for paleoenvironment

The levels of major-element oxides and some related parameters for lignites (on a whole-coal basis) in the Jinsuo Basin are given in Table 2. Some of the major-element oxides show differences in abundance between pale and dark layers. The average percentages of SiO_2 (7.22%) and TiO_2 (0.42%) in pale layers are much higher than the proportions determined for dark layers (2.30% SiO_2 and 0.14% TiO_2 ; Table 2). Levels of Al_2O_3 (3.31%) and Fe_2O_3 (0.79%) in pale layers are on average slightly higher than the values of dark layers (2.16% for Al_2O_3 , 0.59% for Fe_2O_3 , Table 2). Other major-element oxides show similar abundance in pale and dark layers. Furthermore, the average Zr content in pale layers (123.5 ppm) is more than three times higher than that in dark layers (36.8 ppm, Table 2).

In the Jinsuo lignite samples, the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio varies from 0.10 to 9.26 (av. 1.93). The average value is higher than the average value of Chinese coals (1.42 ; Dai et al., 2012), and is also much higher than the theoretical value of kaolinite (1.18). However, the ratio differs significantly between pale and dark lignites. The average $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of pale lignite samples (2.98) is much higher than that of dark lignite samples (0.88). The higher $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios in pale lignite samples result from the higher content of quartz observed under the scanning electron microscope (Fig. 4a) compared with dark lignites (Fig. 4b). Vassilev et al. (2010) suggested that high Si/Al ratios and increased ash

Table 2

Major-element oxides (%), Zr (ppm) and related parameters of the lignites from the Jinsuo Basin, Yunnan Province. Data on ash yields are from Liu et al. (2018), on a whole-coal basis.

Sample	SiO ₂	Al ₂ O ₃	CaO	K ₂ O	Na ₂ O	Fe ₂ O ₃	MnO	MgO	TiO ₂	P ₂ O ₅	Zr	Ash (wt%)	DAI ¹	SiO ₂ /Al ₂ O ₃
JS-39	3.33	2.71	1.47	0.14	bd ²	1.60	0.0187	0.66	0.37	0.05	50.8	10.9	1.55	1.23
JS-38	0.32	1.05	1.39	0.01	bd	0.46	0.0063	0.53	0.04	0.02	8.65	4.6	0.48	0.31
JS-37	8.18	2.93	1.32	0.20	0.02	0.51	0.0063	0.62	0.39	0.07	91.4	14.3	4.69	2.79
JS-36	0.47	0.79	1.34	0.01	bd	0.59	0.0065	0.57	0.03	0.03	8.04	4.6	0.39	0.59
JS-35	5.11	1.64	1.16	0.08	0.01	0.42	0.0052	0.54	0.29	0.03	75.8	9.4	3.26	3.12
JS-34	0.10	0.74	1.48	0.00	bd	0.46	0.0066	0.62	0.01	0.02	3.23	4.0	0.28	0.14
JS-33	2.44	0.49	1.67	0.01	bd	0.60	0.0078	0.68	0.10	0.01	137.9	6.8	0.81	5.01
JS-32	0.88	0.44	1.74	0.01	bd	0.53	0.0090	0.65	0.09	0.01	17.8	5.1	0.39	2.00
JS-31	6.14	0.66	1.82	0.02	bd	0.55	0.0081	0.80	0.24	0.02	162.5	11.1	1.79	9.26
JS-30	2.29	2.41	1.68	0.13	bd	0.52	0.0067	0.82	0.11	0.09	22.4	9.6	1.10	0.95
JS-29	3.92	1.39	1.38	0.05	bd	0.51	0.0055	0.60	0.19	0.20	154.4	9.3	1.59	2.81
JS-28	0.12	0.79	1.67	0.01	bd	0.50	0.0074	0.64	0.01	0.02	0.33	4.9	0.24	0.15
JS-27	7.34	1.00	1.81	0.04	bd	0.69	0.0080	0.82	0.25	0.02	76.2	13.3	1.87	7.31
JS-26	0.06	0.62	1.29	0.00	bd	0.31	0.0051	0.54	0.02	0.01	4.91	3.6	0.25	0.10
JS-25	2.73	1.04	1.61	0.02	bd	0.50	0.0071	0.67	0.65	0.03	83.0	8.3	1.17	2.62
JS-24	0.30	0.85	1.89	0.01	bd	0.45	0.0080	0.70	0.06	0.02	10.4	5.2	0.31	0.36
JS-23	2.72	1.09	1.61	0.03	bd	0.98	0.0077	0.65	0.23	0.14	146.9	9.2	0.83	2.49
JS-22	5.44	4.27	1.69	0.28	bd	0.89	0.0116	0.86	0.38	0.03	68.7	15.2	2.22	1.28
JS-21	8.73	3.99	1.52	0.24	bd	1.08	0.0102	0.77	0.32	0.04	84.2	17.9	2.91	2.19
JS-20	3.21	2.74	1.69	0.21	bd	0.72	0.0104	0.77	0.14	0.06	32.1	11.0	1.37	1.17
JS-19	1.43	1.20	1.75	0.05	bd	0.40	0.0102	0.67	0.09	0.07	125.6	6.9	0.69	1.19
JS-18	6.79	5.53	1.73	0.17	bd	0.82	0.0191	0.79	0.30	0.07	182.0	17.7	3.62	1.23
JS-17	10.96	5.63	1.46	0.57	0.02	1.43	0.0108	0.82	0.75	0.05	139.0	22.2	4.32	1.95
JS-16	1.90	1.59	1.76	0.07	bd	0.34	0.0091	0.68	0.10	0.20	28.7	7.4	1.05	1.20
JS-15	3.38	2.21	1.09	0.16	0.01	0.28	0.0057	0.48	0.14	0.08	136.3	7.9	3.16	1.53
JS-14	1.10	1.24	1.31	0.06	bd	0.31	0.0064	0.51	0.06	0.14	16.1	5.2	0.96	0.88
JS-13	34.41	18.86	3.81	1.69	0.05	2.94	0.0252	2.37	2.02	0.22	119.0	67.0	5.97	1.82
JS-12	1.07	2.31	2.12	0.05	bd	0.41	0.0092	0.67	0.09	0.25	19.2	8.4	0.98	0.46
JS-11	5.80	3.73	1.55	0.30	0.01	0.53	0.0083	0.70	0.34	0.33	172.0	13.5	3.53	1.55
JS-10	6.22	4.39	1.16	0.43	0.00	0.72	0.0067	0.70	0.21	0.05	57.5	14.0	4.21	1.42
JS-09	8.05	4.23	1.10	0.48	0.02	0.43	0.0061	0.63	0.30	0.07	88.2	15.5	5.77	1.90
JS-08	3.34	2.46	2.00	0.15	0.00	0.56	0.0097	0.76	0.16	0.36	39.4	10.1	1.77	1.36
JS-07	5.70	3.41	1.50	0.26	0.04	0.88	0.0085	0.67	0.44	0.23	181.2	13.2	3.18	1.67
JS-06	3.33	2.93	1.10	0.16	0.01	1.18	0.0066	0.59	0.25	0.04	57.4	10.0	2.07	1.14
JS-05	6.46	2.48	0.94	0.12	0.02	0.61	0.0054	0.40	0.38	0.07	142.0	11.5	4.80	2.61
JS-04	4.39	3.78	1.02	0.27	bd	0.93	0.0061	0.54	0.40	0.05	87.5	11.6	3.36	1.16
JS-03	3.11	2.19	1.66	0.10	0.01	0.89	0.0087	0.59	0.19	0.06	55.1	9.18	1.61	1.42
JS-02	6.47	3.52	1.42	0.22	0.03	0.81	0.0080	0.56	0.44	0.05	108.0	13.6	3.79	1.84
JS-01	0.91	1.28	1.22	0.04	bd	0.63	0.0052	0.51	0.07	0.03	15.4	5.3	0.78	0.71
AV-P ³	7.22	3.31	1.58	0.25	0.02	0.79	0.0086	0.75	0.42	0.10	123.5	15.1	3.01	2.82
AV-D ⁴	2.30	2.16	1.56	0.11	0.01	0.59	0.0084	0.66	0.14	0.08	36.8	8.4	1.35	0.97

¹DAI = detrital/ authigenic index = (SiO₂ + Al₂O₃ + K₂O + Na₂O + TiO₂)/(Fe₂O₃ + CaO + MgO + SO₃), from Vassilev et al. (2009), ash basis.

²bd = below detection limit.

³AV-P = average of pale lignites.

⁴AV-D = average of dark lignites.

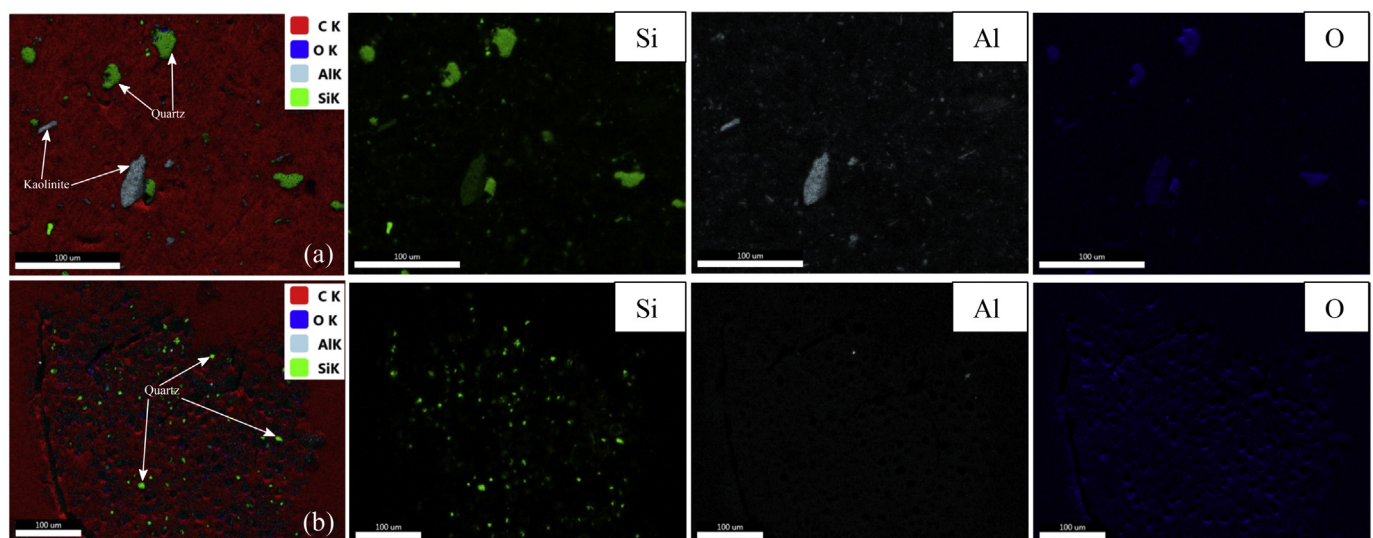


Fig. 4. Elemental distribution maps of pale (a, sample no. JS-02) and dark lignites (b, sample no. JS-26), indicating different distributions of Si, Al and O in the areal scanning by SEM-EDS.

yields of coals indicate either unstable conditions during deposition of the peat from which the coal was generated after diagenesis and/or that the coal originated from peat formed in an intermountain basin. In contrast, low Si/Al ratios and decreased ash yields generally indicate that the peat was formed in stable depositional conditions with a low degree of tectonic movement in a lowland peat-forming environment or was formed from highly weathered products (Vassileva et al., 2010). The higher $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios and higher Zr contents, together with the high ash yield in pale lignites in the present case probably result from the unstable conditions caused by the slight uplift of the Tibetan Plateau during the late Pliocene (Clark et al., 2005; Kou et al., 2006; Sun et al., 2011; Zhang et al., 2012; Su et al., 2013). The intense degradation of organic matter and poor preservation of plant materials in pale lignites further support these results (Moore et al., 1996; Moore and Shearer, 2003; Liu et al., 2018).

According to the elemental distribution map of Si, Al and O, it can be inferred that the distribution and morphology of quartz (Fig. 4) can be attributed to different origins in pale and dark lignites. Large, angular to semi-rounded grains of quartz in pale lignites (Fig. 4a) imply an obvious detrital origin, whereas in dark lignites, small and fine quartz particles fill the cells of organic matter (Fig. 4b). Similar quartz particles associated with organic matter have also been found previously in lignites and coals (Ruppert et al., 1991; Sykes and Lindqvist, 1993; Dai et al., 2014). According to Ward (2002, 2016), such quartz particles are clearly of authigenic origin and were incorporated into organic matter by syn-depositional or early diagenetic processes. Vassileva et al. (2010) suggested that high proportions of quartz and feldspars in coals strongly indicate a considerable influx of detrital materials. The occurrence of detrital and stable Ti-bearing accessory minerals may be used as indicators for previous geotectonic regimes in coal basins associated with washout of minerals from the source areas (Vassilev et al., 2009; Vassileva et al., 2010). The higher average detrital/authigenic index (DAI) value (3.01, Table 2) in pale lignites than in dark lignites further indicates an increased influx of detrital minerals during deposition or early diagenesis of the paleomire (Vassilev and Vassileva, 2007; Vassilev et al., 2009). According to Ward (2016), the origin of authigenic quartz in coal is not always clear because of the independent behavior of other minerals (e.g. clay minerals) during coal formation; similarly, Vassileva et al. (2010) proposed that quartz does not have any significant associations with other minerals in coal. Several hypotheses regarding the origin of authigenic quartz in coal have been made previously: quartz may be released from siliceous phytoliths (Andrejko et al., 1983; Wüst et al., 2008) or it may be leached from the basement rocks or be derived from volcanic ash during peat accumulation (Ward et al., 2001). Dissolved silica could also precipitate to infill plant material and substrates, and the high organic acid concentration in peat may have accelerated the dissolution process (Ruppert et al., 1993). Furthermore, authigenic quartz in coal seams may also have precipitated from hydrothermal fluids (Dai et al., 2014). In the present study, the authigenic quartz (e.g. cell-filling) in dark lignites most likely originated from alteration/dissolution of pre-existing detrital minerals in the peat mire caused by chemical weathering under wet/humid depositional conditions. The different correlations of SiO_2 versus Al_2O_3 and K_2O (Fig. 5a and b) between the dark and pale lignites suggests that the silica in the two kinds of lignite had different origins. The low correlations of SiO_2 versus Al_2O_3 and K_2O indicate that the silica in pale lignites occurs mostly as independent quartz, with small amounts in other silicates (e.g. feldspar and clay minerals). In contrast, the high correlation of SiO_2 versus Al_2O_3 and K_2O in dark lignites suggests that the silica is largely associated with Al and K minerals such as K-feldspar, illite, and/or kaolinite, and the anaerobic and wet conditions which yield organic acid probably promoted the dissolve of silica and the deposition of small silica particles associated with organic materials in dark layers.

4.3. Paleoenvironmental model for pale and dark layers

The suggested model for the paleoenvironment during formation of pale and dark lignite layers in the Jinsuo Basin is shown in Fig. 6. The model is based on field observations, the petrographic and organic geochemical characteristics of the lignite samples (Liu et al., 2018), regional geological and climate changes (An et al., 2001; Xu et al., 2004; Kou et al., 2006; Dupont-Nivet et al., 2007; Chang et al., 2010; Xie et al., 2012; Sun et al., 2015), paleobotany (Wu et al., 1987; Xu et al., 2004), and stable isotopic and mineralogical data.

Previous coal petrological and organic geochemical data suggest that the thick dark layers in the Jinsuo Basin lignites were formed under wet, humid conditions with the paleomire containing almost entirely angiosperm plants (Liu et al., 2018). The regional paleoclimate reconstruction further supports warmer and more moist climatic conditions during the Pliocene in Yunnan Province compared to today (Xu et al., 2004; Sun et al., 2011; Yao et al., 2012; Su et al., 2013). The vegetation in central Yunnan Province during the late Pliocene was mainly composed of the angiosperms *Cyclobalanopsis glaucoides* and *Castanopsis orthacantha* and the gymnosperm *Pinus yunnanensis* (Wu et al., 1987; Xu et al., 2004), with variations in the vertical vegetation zonation depending on the altitude of the particular region (Kou et al., 2006; Xu et al., 2008; Sun et al., 2011; Huang et al., 2016).

Fig. 6a shows the model for paleoenvironmental conditions and vegetation composition during formation of the dark layers in the Jinsuo Basin. Angiosperm trees (such as *Cyclobalanopsis* and *Castanopsis*) dominated the plant community, in association with some shrubby and herbaceous plants. This reconstruction is consistent with results from the previous biomarker analysis (Liu et al., 2018). The occurrence of gymnosperm trees of the Pinaceae family, such as *Pinus yunnanensis*, was restricted to the eastern and western mountains because of the higher altitude and lower ambient temperature. During the warm periods, intensive rainfall events within the basin resulted in formation of a paleomire with a high degree of gelification of organic material, a high proportion of macerals of the huminite group, a relatively high ground water table, and good preservation of plant tissues (Moore and Shearer, 2003; Liu et al., 2018). The wet/humid depositional conditions in the peat probably also generated relatively heavy carbon isotope compositions in the dark layers. Moreover, during the formation of dark layers, thick peat horizons accumulated under stable conditions, and contained larger amounts of authigenic minerals (e.g. cell-filling quartz) compared to pale layers.

The model of the formation of thin pale layers in the Jinsuo Basin lignites is shown in Fig. 6b. The pale layers were generated under less wet and more oxic conditions compared to the dark layers (Liu et al., 2018). The less wet and more oxic conditions were probably caused by fluctuations of the water table during peat deposition (Moore and Shearer, 2003; Liu et al., 2018), which might have been caused by the slight uplift of the Tibetan Plateau together with the co-occurrence of a more intensive Asian monsoon. Compared to the warmer climate during the formation of dark layers, the thin pale layers were formed during cooling periods associated with less precipitation. As shown in Fig. 6b, during the dry and cooler periods, the abundance of angiosperm plants (such as *Cyclobalanopsis* and *Castanopsis*) decreased whereas those of gymnosperm and shrubby plants increased. The contribution of gymnosperms (*Pinus yunnanensis*) to the peat-forming vegetation increased, forced by climate cooling and the increasing altitude resulting from uplift of the Tibetan Plateau. The contemporaneous enhancement of the Asian monsoon led to less rain in the basin and increasing activity of aerobic bacteria in the mire, which enhanced the decomposition of organic matter. The change of in the plant community (gymnosperms vs. angiosperms) in dry/wet conditions for pale/dark layers in the Jinsuo Basin is consistent with observations from a previous study showing that the changes in floral composition result from changes in substrate wetness during peatland aggradation (Korasiadis et al., 2017a). The lower water table further supports the increased

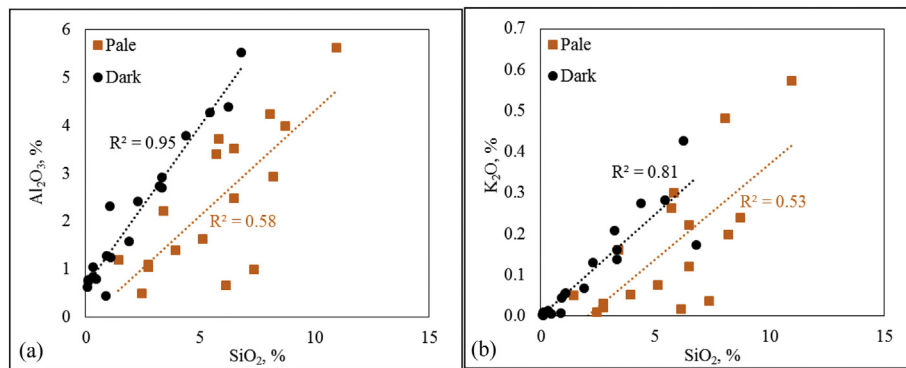


Fig. 5. Correlation diagrams of (a) SiO_2 versus Al_2O_3 , (b) SiO_2 versus K_2O in pale and dark lignites from the Jinsuo Basin.

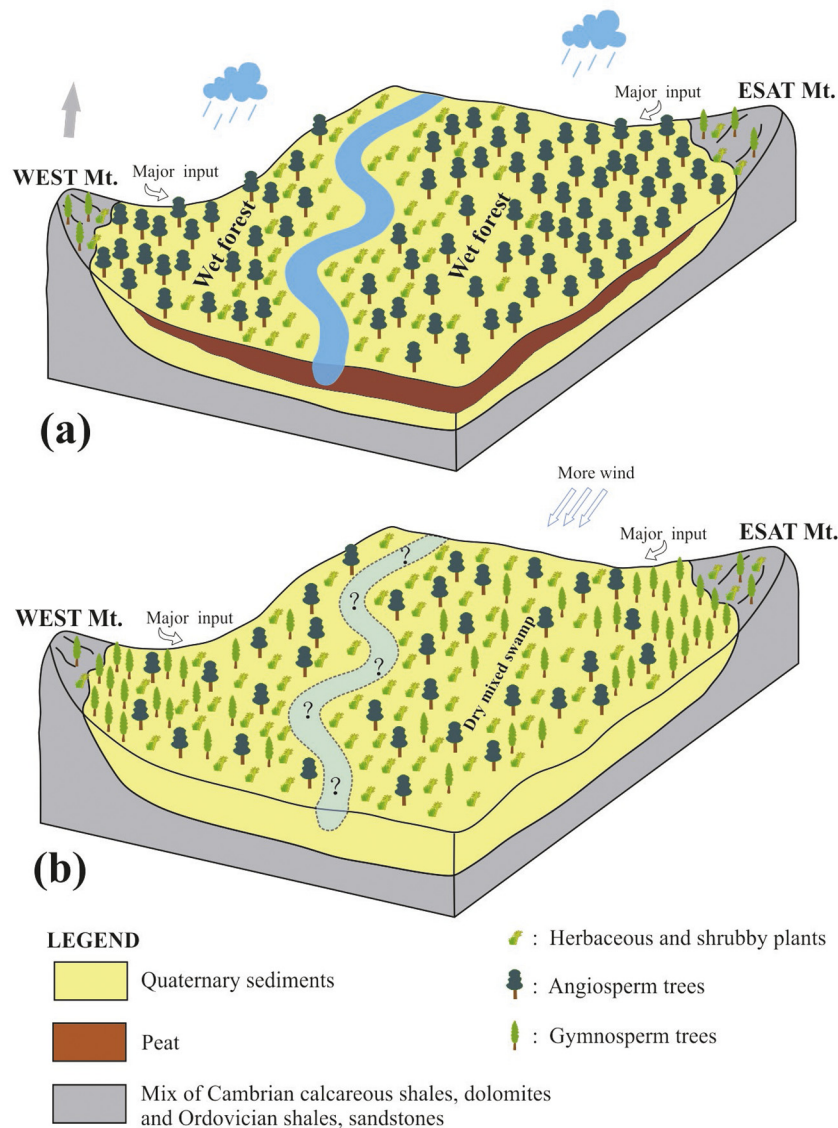


Fig. 6. Conceptual models of the depositional paleoenvironment for the formation of dark (a) and pale (b) layers in the Jinsuo lignite deposit basin, Yunnan Province, China.

degradation of organic matter in the pale layers (Moore and Shearer, 2003; Korasidis et al., 2017a). The increased decomposition of organic matter and the strong influence of bacterial activity in pale layers in the Jinsuo Basin (Liu et al., 2018) might be the reason why pale layers are very thin compared to the dark layers and could also have caused the

light stable carbon isotope compositions in the pale layers. This is consistent with the observation of residual thin/pale lithotypes in the Gippsland Basin, Australia, which also experienced intense weathering and aerobic degradation (Korasidis et al., 2017b). Moreover, the step-wise slight uplift of the surrounding mountains and the increasing Asian

monsoon probably resulted in an increasing influx of detrital minerals (e.g. quartz), TiO_2 , and Zr (Table 2) into the paleomire during formation of the pale layers. This conclusion is also supported by the high DAI values in pale samples.

During formation of pale layers (Fig. 6b), the conditions were different from those during the formation of dark layers. One possible scenario is that intermittent strengthening of the Asian monsoon occurred as a result of the phased uplift of the Tibetan Plateau associated with global cooling (An et al., 1999, 2001; Li et al., 2014). In a previous study of the Cenozoic plant diversity in Yunnan, changes in the plant composition and vegetation types were suggested to be related to the altitude changes from north to south and associated global cooling (Huang et al., 2016). On the basis of results obtained from the investigation of the late Pliocene paleoflora of the Longling lignite mine (western Yunnan), Xu et al. (2004) reported five major cycles of warming and cooling during that time. Furthermore, the Cenozoic monsoon evolution (Clift et al., 2008, 2010) and the approximate co-occurrence of the phased uplift of the Tibetan Plateau with stepwise strengthening of the Asian monsoon from the late Miocene (An et al., 2001) is well established. In general, the interaction between monsoon intensity and global climate change is well known (Kutzbach and Street-Perrott, 1985; Licht et al., 2014; Sun et al., 2015). Previous studies also suggested that stepwise enhanced monsoon activity and drying of the Asian interior were associated with episodic tectonic uplift of the Tibetan Plateau from the Miocene on (~8 Ma). Moreover, global cooling in the Pliocene at ~3.2 Ma (Li et al., 2014), and growth of the Tibetan Plateau together with contemporaneous enhancement of the monsoon further accelerated the Miocene–Pliocene global cooling (Sun et al., 2008; Ruddiman, 2013; Li et al., 2014; An et al., 2015). The combined influences of uplift of the Tibetan Plateau, intense Asian monsoon, and global cooling on the local geological climate of the studied area during the late Pliocene might ultimately have caused formation of pale layers. However, the formation of periodic pale and dark layers cannot be directly related to uplift and fall of the Tibetan Plateau, because there is no evidence for a fall of the Tibetan Plateau during the late Pliocene. The pale layers probably formed following periods of uplift of the Tibetan Plateau that led to an intense monsoon and locally cold climate and a lower groundwater level in the basin. Considering the relatively warm, humid tropic/subtropical climate in Yunnan Province from the Miocene to the late Pliocene and the long-term stability of the geological conditions (Yunnan BGMR, 1990; Xiao et al., 2010; Xie et al., 2012; Zhang et al., 2012; Su et al., 2013), the dark layers formed gradually following pale layers during periods in which the local climate became warmer and more humid between two uplift steps of the Tibetan Plateau. This interpretation is also supported by the thicknesses of pale and dark layers. The thin pale layers (5–25 cm thick) were probably formed during periods of short-term cold, dry climate following each period of uplift of the Tibetan Plateau. In contrast, the thick dark layers (tens to hundreds of centimeters thick) most likely formed in long-term stable conditions with a warm, humid tropical/subtropical climate during periods of little/no uplift activity of the Tibetan Plateau.

5. Conclusions

New data on the carbon and nitrogen stable isotopic compositions and the distributions of major elements in pale and dark lignites in the Jinsuo Basin, Yunnan Province, southwest China, were used to reconstruct the paleoenvironmental conditions in the paleomire and the driving forces for the formation of pale and dark layers.

Pale lignites generally have lower $\delta^{13}\text{C}$ values (av. -27.01‰) compared to dark lignites (av. -26.38‰), and the $\delta^{13}\text{C}$ values of fossil wood (av. -24.79‰) are generally higher than those of bulk lignite. Maximum $\delta^{13}\text{C}$ values were detected in two fusain samples (av. -24.19‰). The very high amount of liptinite together with the strong degradation of plant materials by bacterial activity in pale lignites (Liu

et al., 2018) could have had a marked influence on $\delta^{13}\text{C}$ values and resulted in lower ^{13}C values in pale lignites compared to dark lignites. The relatively high $\delta^{15}\text{N}$ values in pale lignites (av. 2.17‰) are related to the low TN values (av. 0.89). In dark lignites, $\delta^{15}\text{N}$ values are lower (av. 1.79‰) and correspond to higher TN values (av. 1.21). This difference might be caused by the greater loss of organic nitrogen during aerobic degradation of organic matter in pale lignites associated with preferential degradation of the ^{14}N isotope. The weak negative correlation between the $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values indicates that some factors (e.g. liptinite content and/or bacterial degradation) influenced both the stable carbon and the nitrogen compositions of lignites. The light stable carbon isotope compositions in pale layers were generated by strong degradation of organic matter by bacteria and the higher liptinite content of pale layers, whereas the heavy stable carbon isotope composition resulted from the wet/humid depositional environment.

The driving force for the formation of pale layers might be the stepwise uplift of the Tibetan Plateau and the approximate co-occurrence of the intensive Asian monsoon, which resulted in a locally cold, dry climate in the study area. These locally cold/dry and unstable conditions caused a low/fluctuating water table in the basin which facilitated the formation of pale layers. The lower $\delta^{13}\text{C}$ values in pale layers support this results. The relatively high $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratios, DAI values, and high levels of TiO_2 and Zr in pale layers suggest increased detrital input during peat deposition. Gymnosperms (such as *Pinus yunnanensis*) become more abundant in the basin during these short periods. In contrast, the dark layers formed during long-term stable geological conditions with a regionally tropical/subtropical warm, humid climate generating a relatively high water table which is favor of the formation of dark layers and the predominance of angiosperms in the basin. Changes in the composition of major elements and the variations in thickness of pale and dark layers further support this conclusion.

Author statement

Bangjun Liu: PhD student, carried out most of the analyses and wrote the manuscript with support of W.P.

Cunliang Zhao: Data of major elements were determined under his supervision.

Jens Fiebig: Stable carbon and nitrogen isotopes were determined under his supervision.

Achim Bechtel: Helped to interpret the stable carbon and nitrogen isotopic data.

Yuzhuang Sun: Helped to interpret the data of major element.

ilhelm Püttmann: Supervision of the PhD project of Bangjun Liu, wrote parts of the manuscript together with Bangjun Liu.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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