



# 1 Orbital-scale climate dynamics impacts on Gzhelian peatland wildfire activity in

### 2 the Ordos Basin

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### 14 Abstract

15 The Carboniferous, an important coal-forming period in geological history, was characterized by extensive vegetation and high oxygen levels. Numerous wildfire evidence suggests that high 16 frequency of wildfire occurred at that time, specifically in peatlands. However, the control 17 18 mechanisms for changes in wildfire activity in peatlands during this period are still not clearly understood. In this study, evidence from the Gzhelian in the Ordos Basin, such as the 19 inertinite/vitrinite (I/V) ratio, indicated the existence of different frequencies of wildfire activity at 20 that time. The CaO/MgO and CaO/MgO • Al<sub>2</sub>O<sub>3</sub> climate indicators revealed that high-frequency 21 22 wildfires mainly occur in warm and humid climates. Based on former age constraints, we deduced 23 that orbital cycles (long eccentricity) controlled the climate influence on peatland wildfires during https://doi.org/10.5194/cp-2024-42 Preprint. Discussion started: 20 June 2024 © Author(s) 2024. CC BY 4.0 License.





the Gzhelian. When eccentricity was high, abundant sunshine and frequent rainfall led to warmer 24 25 and more humid peatlands. The latter environments were more favourable for vegetation 26 development, leading to increased fuel loads, which in turn led to more frequent wildfires. 27 Moreover, the Gzhelian global wildfire records, showed that evidence of wildfire during this period was mainly located in areas with abundant tropical vegetation, supporting the view that wildfire 28 29 activity during this period was mainly controlled by the fuel loads. Although Hg could be produced 30 by peatland wildfires, but our results show that Hg was mainly from frequent volcanic activity 31 during this period.

Keywords: Wildfire; Inertinite; Gzhelian; Hg; Long eccentricity

### 1. Introduction

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34 In recent years, information about wildfires preserved in coal seams has been exposed (Robson et al., 2015; Hou et al., 2022; Zhang et al., 2022). The macerals in the pyrogenic inertinite (fossil 35 charcoal in coal), such as fusinite, semifusinite, and inertodetrinite, have been widely used as direct 36 37 evidence to demonstrate the occurrence of paleo-wildfires (Scott, 2010, 2022; Jasper et al., 2013; 38 Belcher and Hudspith, 2017; Uhl et al., 2022; Shen et al., 2023). The Carboniferous through 39 Permian were important coal-forming periods in geologic history where large deposits of coal developed (Berner, 2003). Although wildfires of the period have been somewhat summarized by 40 41 previous authors (e.g., Scott, 2000; Glasspool and Scott, 2010; Glasspool et al. 2015; Jasper et al. 42 2021), there is a dearth of high-resolution wildfire records from the period. Glasspool et al. (2015) 43 concluded from the analysis of the inertinite in coal that the frequency of wildfires during the Late Paleozoic was mainly due to the higher oxygen content in the atmosphere at that time. However, 44 45 more detailed controlled factors for Carboniferous through Permian wildfires need to be improved.

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47 changes in orbital cycle forcing climate change, which in turn influences wildfire activity (Verardo 48 and Ruddiman, 1996; Thevenon et al., 2004; Hao et al., 2020; Shi et al., 2020; Cheng et al., 2022). 49 Daniau et al. (2013) found that the past 170 kyr of grassland wildfire activity in southern Africa has 50 been controlled by precession. Zhang et al. (2020a, 2023) found that wildfire activity during the Early to Middle Jurassic was influenced by changes in long eccentricity (405 kyr) and the 51 precession cycle. The orbital period (eccentricity, obliquity, and precession) forces climate change, 52 53 thus affecting the amount and flammability of vegetation fuels to control wildfire activity (Zhou et 54 al., 2023). The study of whether wildfire activity in Carboniferous to Permian peatlands was controlled by changes in orbital cycles is unclear. Late Carboniferous Gzhelian global wildfire 55 records indicate that the wildfire records were all distributed in tropical climate zones. Tropical 56 57 climate zones have abundant fuel accumulation and orbital cycle forcing wildfire activity may be 58 more pronounced (DiMichele, 2014). As the largest Late Carboniferous to Early Permian peat basin in China, the Ordos Basin 59 60 contained a large amount of coal seams, which were preserved with numerous volcanic ashes and 61 wildfire products in the coal seams (Wang, 2017; Xu et al., 2020; Zhao et al., 2023; Zhang et al., 62 2023a, 2023b). In this paper, the focus is on the No. 9 coal of the Yaogou Coal Mine in the Jungar Coalfield of the Ordos torder to understand the occurrence of the Late Carboniferous wildfires and 63 64 the variation of burning temperatures. Employing the clear age constraints of the No.9 coal seam, 65 the possible role of long eccentricity orbital cycle changes in driving wildfires by means of wildfire frequency and climate indicators at that time can be explored (Zhang et al., 2023a). Volcanic 66 activity, frequent wildfires, and other causes can lead to Hg enrichment, and this study also explores 67 68 the relationship between wildfire activity, volcanic activity, and Hg at that time, using changes in 69 Hg content in No. 9 coal.





# 2. Geological setting

71 The Ordos Basin, located at the western margin of the North China Craton, is the second 72 largest terrestrial sedimentary basin in China (Ao et al., 2012; Zhang et al., 2023a). The Jungar 73 Coalfield is in the southwestern part of the Inner Mongolia Autonomous Region of China and in the northeastern part of the Ordos Basin, which contains about 26.8 Gt of coal reserves, which is one of 74 the richest coal reserves in northern China (Dai et al., 2006,2008,2012; Li et al., 2016). The 75 76 coalfield contains coal-bearing sequences of the Carboniferous and Permian systems (Wang et al., 77 2011). 78 This study was conducted in the Yaogou Mine, in the northeastern part of the Jungar Coal 79 Field (Fig. 1b). The coal-bearing sequences in the Yaogou Mine include the Benxi Formation, 80 Taiyuan Formation, and Shanxi Formation, with a total thickness of 84~200 m (Zhang et al., 2023a). The Benxi Formation, with a total thickness of 84~200 m (Zhang et al., 2023a), lies unconformably 81 82 on the Middle Ordovician Majiagou Formation, with a thickness of about 17~28 m. The Taiyuan 83 Formation is underlying by the Benxi Formation. The Taiyuan Formation, with a thickness of about 84 31-75 m (average 58 m), conformably overlies the Benxi Formation. The Taiyuan Formation 85 consists mainly of grey and gray-white quartz sandstone, siltstone, mudstone, and five coal beds marked from top to base as No. 6 to No. 10 (Fig. 1c) (Zhang et al., 2023a). Zhang et al. (2023a) 86 87 determined by U-Pb zircon from altered volcanic ashes at the top and bottom of No. 9 coal that the Taiyuan Formation in this study area belongs to the Gzhelian stage of the Late Pennsylvanian. The 88 Shanxi Formation with a thickness of about 35-97 m is mainly terrigenous coal-bearing clastic 89 90 rocks, dominated by sandstones and coal seams (Zhang et al., 2023a).



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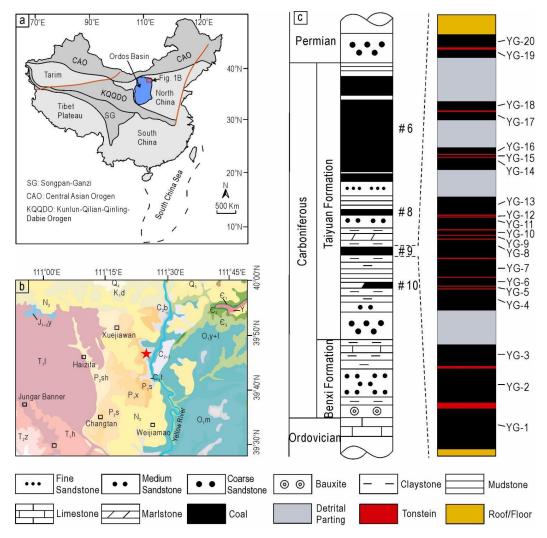


Fig. 1 Comprehensive geologic map of the Yaogou coal mine in the Ordos Basin, northern China. (a) Map showing the location of the Ordos Basin and the study area. (b) Geologic map of the study area showing the location of the study area (red stars). (c) Sedimentary sequence of the Jungar Coal Field and columnar map of the No. 9 coal seam. The No. 9 coal occurs in the Taiyuan Formation. Q4-Holocene, Q3-Pleistocene, N2-Pliocene, K1d-Dongsheng Formation (Fm.), J1+2y-Yan'an Fm., T2z-Zhifang Fm., T1h-Heshanggou Fm., T1l-Liujiagou Fm., P2sh-Shiqianfeng Fm., P2s-upper Shihezi Fm., P1x-lower Shihezi Fm., P1s-Shanxi Fm., C3t-Taiyuan Fm., C2b-Benxi Fm., O1m-Majiagou Fm., O1y+l-Yeli and Liangjiashan Formations. Figure modified from Zhang et al. (2023a).





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# 3. Material and methods

### 3.1 Sampling

In this study, a total of 20 coal samples were collected from the exposed face of the No. 9 coal at the Yaogou Mine, with a cumulative thickness of the profile of about 6.1 m. To minimize contamination and oxidation, the samples were all immediately stored in plastic bags. From bottom to top, all coal samples were marked YG-1 to YG-20 (Fig. 1c).

### 3.2 Analytical method

The analytical methods used in this study to demonstrate wildfire characteristics included petrographic analysis, coal rock micro component identification, fusinite reflectance measurements, and scanning electron microscopy (SEM).

The collected fossil charcoal fragments were crushed to a < 20 top size and the crushed samples were embedded in epoxy resins. The reflectance of the fusinite was measured by the oil immersion at room temperature and determined using an MSP UV-VIS2000 spectrophotometer (Petersen and Lindström, 2012; Xu et al., 2020). To estimate the wildfire burning temperatures represented by the experimental samples, calculations were based on the correlation proposed by previous works:

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$$T=184.1+117.76\times\%R_o$$
, (1)

with R<sub>o</sub> represents the reflectance of the inertinite (Jones, 1997; Petersen and Lindström, 2012).

118 Based on the model of oxygen content proposed by Glasspool (2010, 2015), the oxygen content in

the atmosphere was predicted for the period of this study:

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$$I=(0.5-0.5\cos[\pi (O-O_{min})/(O_{max}-O_{min})])^n,$$
 (2)

with I as the average inertinite content, O as the oxygen content,  $O_{max}$  as the oxygen content when

the inertinite content reached 100%, and O<sub>min</sub> as the oxygen content when was no inertinite.





The microstructure of fossilized charcoal samples was observed by SEM to make a clearer observation of fossilized charcoal samples. The samples were observed under vacuum conditions using a 10 kV accelerating voltage, standard beam light, and a secondary electron probe. All the above experiments were completed at the Shandong Provincial Key Laboratory of Depositional Mineralisation and Sedimentary Minerals, Shandong University of Science and Technology.

To determine the coal rock components, the coal samples taken were measured according to the National Standard GB/T8899-2013. The reflectivity of the vitrinite group of the coal samples was measured according to the National Standard GB/T 6948-2008. To improve the quantification of charcoal abundance in each single sample, the ratio between inertinite maceral and vitrinite macerals (I/V), was calculated to determine the frequency of wildfires (Zhang et al., 2022). All of the latter experiments were completed in Xi'an Coal Science and Technology Ltd. All samples were analyzed for Total Organic Carbon (TOC) according to the National Standard GB/T19145-2003. All samples were analyzed for Hg concentration according to the National Standard GB/T 22105.1-2008. All above experiments were completed at the Beijing Qingchen Huanyu Petroleum Geological Technology Co. Ltd.

### 3.3 Data Synthesis

To build a reliable database of the Gzhelian wildfire evidence, data was compiled from peer-reviewed journal publications following the program of Lu (2021) and Lv (2024). Personal data and unpublished materials were not included in our study. We used the keywords 'Gzhelian wildfire', 'charcoal', 'inertinite', 'fusain', 'fusinite', and/or 'PAHs' to search papers in Google Scholar, Web of Science, ScienceDirect, and JSTOR. Literature searches were conducted by the end of April 2024.

To minimize the possibility of low credible/reliable wildfire data, the data published in the





original article was double-checked and evaluated before inclusion in our database. The evidence 146 147 for fossil charcoal in the database is based on the standardized guidelines in Scott (2000,2010,2020). 148 The distinction between sources of PAHs (pyrogenic and petrogenic) was made according to Yunker 149 et al. (2002), and only records of pyrogenic PAHs were included. For example, Hower et al. (2022) had two inertinite data that did not have details of where samples were taken, so they were 150 discarded. The inertinite in Presswood et al. (2016) was due to magmatic intrusion, so it was 151 152 discarded. 153 Three types of Gzhelian wildfire evidence were included in the database: charcoal type I (pyrogenic inertinite macerals from coals), charcoal type II (fossil charcoal from clastic sediments), 154 155 and pyrogenic PAHs. The location (e.g., country, state/province, city, latitude, longitude, etc.), 156 palaeogeographical localities (e.g., palaeocontinents, palaeolatitude, and palaeolongitude), 157 stratigraphic unit (e.g., formation, group), and rock type (e.g., coal, sandstone, mudstone) were integrated for each wildfire record. When multiple studies reported wildfire data from the same 158 geologic unit in the same location (e.g., section, region, or basin), they were considered as one 159 160 record (Lu et al., 2021). Depending on the type of evidence, wildfire occurrences in the same study 161 site can be identified in different studies and recorded as one occurrence. 162 The GPlates 2.2.0 model constructed by Scotece (2008) was used to estimate paleolatitudes 163 and paleolongitudes data for wildfire occurrence. To provide a visual representation of the spatial 164 distribution, the occurrence of wildfires was projected on a paleogeographic map of Gzhelian by 165 Scotese (2021) (Fig. 9).

# 4. Results

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### 4.1 Coal rock micro component and geochemical characterization

The microscopic component contents of the coal samples collected in the mineral-free state

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from No.9 coal of Yaogou Mine are shown in Table 1. Of all the samples, the vitrinite was dominant, with contents ranging from 64.5% to 84.7% (average = 73.5%). The inertinite content of the coal samples ranged from 14.1% to 30.8%, with an average content of 22.3%. The liptinite had the lowest content, ranging from 1.3% to 8.7%, with an average content of 4.2%. Nearly all inertinite preserved in coal was defined as an incomplete combustion product, comparable to fossil charcoal (Scott 2000, 2010; Scott and Glasspool 2006, 2007; Diessel, 2010). Because of this, subcomponent analyses were analyzed for the inertinite in the coal samples Table 1. In the 20 coal samples, the main component of the inertinite was semifusinite, with contents ranging from 7.4% to 17.2% with an average content of 11.5%. The content of inertodedetrinite was rather low, ranging from 1.8% to 13.4%, with an average content of 6.6%. The contents of fusinite, macrinite, and micrinite were low, with average contents of 1.2%, 2.3% and 0.7%. Among the 20 samples, the variation of inertinite/vitrinite ranged from 0.17 to 0.47, with a mean value of 0.31. The reflectance ( $R_{vo}$ ) of the vitrinite of the 20 coal samples ranged from 0.37% to 0.66. The TOC content of the 20 coal samples from the Taiyuan Formation ranged from 27.82% to 48.07%, with an average content of 39.49% (Table 2). The Hg content of the 20 samples varied greatly, ranging from 29.5 ppb to 393 ppb, with an average of 134.96 ppb (Table 2). Zhang et al. (2023a) analyzed the sulfur content, Al<sub>2</sub>O<sub>3</sub> content, and ash yield of 20 coal seams in the Taiyuan Formation in this study area (Table 2). The Spearman correlation analysis showed that the correlation between Hg and Al<sub>2</sub>O<sub>3</sub> in the 20 coal samples from Taiyuan Formation is highly significant, with positive correlation, the correlation coefficient is +0.529, and the significance is 0.016 (Fig. 6a). The correlation between Hg and sulfur is not significant with the correlation coefficient of -0.175 and significance of 0.46 (Fig. 6b). The correlation between Hg and TOC is significant, with a correlation coefficient of -0.659, and the significance is 0.002 (Fig. 6c). Since the





source of elemental aluminum in coal is mainly clay minerals, the correlation between elemental Al and ash yield was analyzed. The correlation between elemental Al and ash yield in the coal samples of this study was highly significant with a correlation coefficient of +0.776 and significance of 0. The correlation between elemental Hg and ash yield was significant and positive with the correlation coefficient of +0.481 and significance of 0.032 (Fig. 6d).

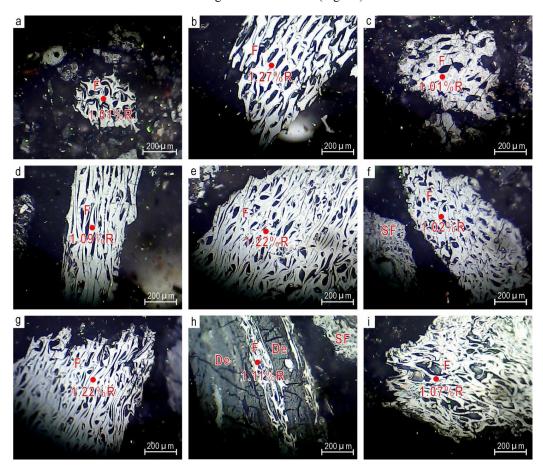


Fig. 2 Features of fossil charcoal in the Ordos Basin Taiyuan Formation observed under the oil immersion optical microscopy. F-fusinite; SF-semifusinite; Red circles show the test points, and numbers show the reflectance values of the points.





Table 1
 The coal micro component contents of 20 seams of No. 9 coal from Yaogou Mine in Ordos Basin

	Percentag	ercentage of the total organic Percentage of the total inertinite macerals acerals (vol.%)					TOM	Total minerals		
No.	Vitrinite	Inertinite	Liptinite	Fusinite	Semifusi nite	Macrinit e	Micrinit e	Inertode trinite	(vol.%)	(vol.%)
YG-1	79.6	14.6	5.8	1.1	9.3	0	0.2	4	53.9	46.1
YG-2	80.5	16	3.5	2.4	10.6	0.6	0.6	1.8	60.4	39.6
YG-3	68.3	27.4	4.3	0.9	14.1	1.5	2.6	8.3	81.2	18.8
YG-4	64.5	26.8	8.7	0.4	13.3	2.4	0.4	10.3	84.8	15.2
YG-5	84.7	11.9	3.4	0.4	7.4	1.5	0.2	2.4	84.9	15.1
YG-6	65.2	30.8	4	0	14.6	2.4	0.4	13.4	86.2	13.8
YG-7	72.2	23	4.8	3	10.7	3	0.8	5.5	87.1	12.9
YG-8	70.8	21.5	7.7	2.5	12.2	1.5	0.2	5.1	81.8	18.2
YG-9	78.4	19.1	2.5	0	9.8	2.5	0.4	6.4	80.1	19.9
YG-10	66.1	27.4	6.5	3.8	13.1	3.2	0.6	6.7	83.9	16.1
YG-11	75.2	23.5	1.3	0.6	12	3.4	0.2	7.3	79.6	20.4
YG-12	72.1	25.1	2.8	0.9	15.4	2.2	0.4	6.2	73.9	26.1
YG-13	65.8	30.2	4	0.8	17.2	4.1	1.8	6.3	86.2	13.8
YG-14	82	14.1	3.9	0	8.3	2.1	0	3.7	81.3	18.7
YG-15	80.2	18.4	1.4	0	11.2	0.8	0.6	5.8	77	23
YG-16	81.8	16.5	1.7	0	10.4	1.2	1.2	3.7	85.2	14.8
YG-17	78.1	18.4	3.5	2.7	9	1.7	0.5	4.5	59.5	40.5
YG-18	66.8	28.9	4.3	0.8	11.3	4.7	0	12.1	59.9	40.1
YG-19	73	24.9	2.1	1.1	8.9	2.9	0.4	11.6	71.5	28.5
YG-20	65	27.2	7.8	3.2	11	4.1	2	6.9	81.8	18.2





Table 2
 The chemical element data of 20 coal seams from the No. 9 coal of the Yaogou coal mine in the Ordos Basin. The
 Al<sub>2</sub>O<sub>3</sub>, TS, and TOC were from Zhang et al. (2023a).

Sample	Hg (ppb)	$Al_2O_3$ (%)	TS (%)	TOC (%)
YG-1	255	16.33	0.23	27.82
YG-2	145	11.94	0.39	29.92
YG-3	63.2	7.67	0.44	47.30
YG-4	118	9.46	0.77	44.14
YG-5	58.6	8.96	0.32	48.07
YG-6	70.4	12.75	0.41	42.04
YG-7	85.1	7.44	0.57	44.72
YG-8	35.4	7.42	0.52	48.04
YG-9	43.5	11.87	0.45	41.17
YG-10	269	10.5	0.59	41.51
YG-11	88.5	10.53	0.48	38.10
YG-12	83.9	10.74	0.5	35.33
YG-13	34.5	9.57	0.48	46.26
YG-14	156	12.77	0.35	40.37
YG-15	352	16.48	0.42	32.35
YG-16	237	14.23	0.32	36.72
YG-17	393	12.42	0.36	29.26
YG-18	152	17.88	0.29	32.05
YG-19	29.5	13.53	0.3	37.95
YG-20	29.6	7.57	0.44	46.68

# 4.2 Microscopic observations of fossil charcoal

Under the oil-immersion reflected-light microscope, a large amount of semifusinite was observed in the coal samples of the Taiyuan Formation. Some representative micrographs are provided in Fig. 2. The pore sizes of the semifusinite ranged from 40 - 120  $\mu m$ , with a small variation in pore size. The measured reflectance of the inertinite of 20 coal samples from the Taiyuan Formation ranged from 1.01% to 2.07%, with the average value of 1.66%.

The structural features of the fibre structure and the cell wall in the fossil charcoal fragments can be observed by SEM. The majorityt of the cellular structures were crushed or broken(Fig. 3a,c). As shown by the radial structure, the tubular cells were straight and 10-20 µm wide (Fig. 3a). There was one single row of circular or elliptical pitting with 0.5-2 µm width (Fig. 3b). And the cell walls



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# of the samples showed homogenization (Fig. 3d).

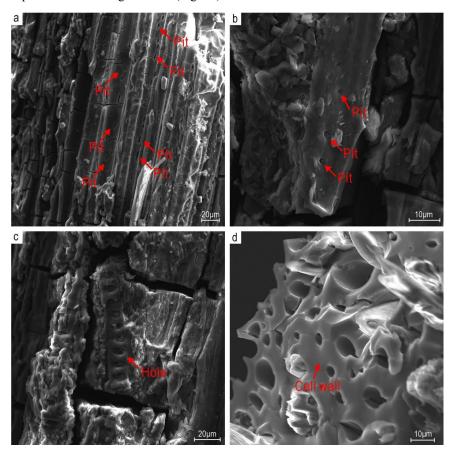


Fig. 3 Scanning electron microscope electron probe image of fossilized charcoal. (a-b) Red arrows point to uniseriate pits. (c) Red arrow points to larger holes in the cell wall that may represent pits, which were diagenetically enlarged during charring. (d) Red arrow points to homogenized cell wall.

# 4.3 The Gzhelian wildfire record in geologic context

Published wildfire records for the Gzhelian were compiled in Table S1. Based on the type of evidence these can be divided into five groups: 23 records of charcoal type I (79.4% of the total records), two records of charcoal type II (6.9%), one record of pyrogenic PAHs (3.4%), two records of charcoal type I and charcoal type II (6.9%), and one record of charcoal type II and pyrogenic PAHs (3.4%). The published Gzhelian wildfire data were observed in 19 geologic units (or





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formations), with two record from an unclear unit.

All of the 29 reported Gzhelian wildfires have occurred in the lower latitudes (30°N-30°S) (Fig.

230 9). According to Boucot et al.'s (2013) suggested paleoclimatic classification, the Gzhelian can be

divided into four different paleoclimatic zones: cool temperate, warm temperate, arid and tropical.

The Gzhelian wildfires all occurred in the tropical area (Fig. 9).

### 5. Discussion

### 5.1 Repeated wildfire in the Gzhelian of the Ordos Basin

The origin of the inertinite in coal has been controversial, and it has been suggested that the fusinite and semifusinite in the inertinite may be produced from fungal degradation of vegetation tissues in oxidizing environments or influenced by other microbial activities (Beeston, 1987; Teichmüller, 1989; Varma, 1996; ICCP, 2001). Some semifusinite in coal may also have been formed during diagenesis (Hudspith and Belcher, 2020). Nevertheless, research suggests that the fusinite and semifusinite in coal were equivalent to fossil charcoal (Scott and Glasspool, 2007; Hudspith et al., 2012), which was produced by the incomplete combustion of vegetation by wildfires (Scott, 2010; Liu et al., 2022). All coal samples from the Taiyuan Formation had low levels of both macrinite and micrinite (Table 1). Both fusinite and semifusinite preserved complete cellular structures (Fig. 2), suggesting that the fusinite and semifusinite in the study samples may be products of vegetation combustion. In addition, the low reflectance of the vitrinite in the coal samples, averaging 0.49%, indicated that the samples were poorly metamorphosed, and was not enough to support the claim that the semifusinite in the samples was due to diagenesis. Experiments had shown that the carbonization temperature for homogenization of vegetation cell walls needs to be at least higher than 250 - 300 °C (Scott and Jones, 1991; Osterkamp et al., 2017). Obvious homogenization of the cell walls of the https://doi.org/10.5194/cp-2024-42 Preprint. Discussion started: 20 June 2024 © Author(s) 2024. CC BY 4.0 License.





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fossil charcoal could be observed under the SEM (Fig. 3d), which more clearly suggested that the fossil charcoal in the samples was caused by wildfire burning. The inertinite content of the Taiyuan Formation coal samples showed a small range of variability (14.1% - 30.8%) and was dominated by semifusinite (7.4% - 17.2%), which indicates the presence of frequent small wildfires during this period. Under the SEM, it can be observed that the cellular structure of the fossil charcoal from the Taiyuan Formation is obviously broken, and some cellular cavities in the charcoal appeared to have different degrees of deformation (Fig. 3c), which may be due to the tectonic movement of the strata. Scott (2010) divided wildfires into three types based on their strength: surface fire, ground fire, and crown fire. Among these, surface fires whose fuels are mainly surface herbaceous, shrubs and various biological wastes, burn at lower temperatures, usually below 400 °C. Ground fires whose fuel is mainly the dry humus layer in the soil, which can burn thick layers of peat, burn at relatively high temperatures, up to 600 °C. It has been demonstrated that the reflectance of the inertinite is positively correlated with the combustion temperature, with higher inertinite reflectance indicating higher combustion temperatures (Jones et al., 1991; Guo and Bustin, 1998; Scott and Glasspool, 2007; McParland et al., 2009). The reflectance of the inertinite in the Taiyuan Formation samples varied in a small range (1.01%-2.07%), and it can be inferred that the Taiyuan Formation wildfire burned at temperatures ranging was about 300 °C to 430 °C, with an average burn temperature of 379 °C (after formula Scott and Glasspool, 2015). This suggests that mainly low-temperature fires occurred in the Ordos Basin during the Gzhelian (Fig. 4). Wildfire burning may have been fueled primarily by low-growing vegetation or biological waste, which would be consistent with the homogenization of fern cell walls observed under the SEM.



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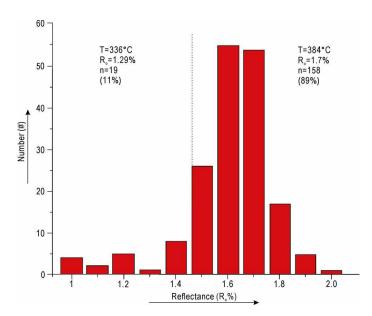


Fig. 4 The inertinite reflectance histograms indicate calculated combustion temperatures (Shukla et al., 2023).

Soils are the largest global reservoir of Hg and forest ecosystems are one of the key areas in

### 5.2 Sources of elemental mercury in coal

the Hg cycle (Wang et al., 2019). Peatland vegetation can retain atmospheric Hg in their bodies by absorbing it (Grigal, 2003), while humus and sulfides in peatlands have a high affinity for Hg, which makes them important sinks for Hg (Yudovich and Ketris, 2005; Woerndle et al., 2018). In the pre-anthropic era, the main sources of Hg in the surface environment were volcanic eruptions, intense continental weathering, and wildfires (Pavlish et al., 2003; Pyle and Mather, 2003; Selin, 2009; Shen et al., 2020).

In this study, Hg was negatively correlated with Total Organic Carbon (TOC) and Total Sulfur (TS) (Fig. 6), indicating that Hg is not predominantly present in organic matter and sulfides. Correlation of elemental concentration with ash yield may provide preliminary information on their affinity with organic and inorganic (Eskanazy et al., 2010; Kortenski and Sotirov, 2002; Dai et al., 2012a; Zhang et al., 2023a). Mercury has a relatively high correlation (r=0.481) with ash yield,





study (r=0.529), indicating that Hg was mainly enriched in clay minerals (Fig. 6). The Hg 288 289 concentrations in coal samples YG-1, YG-10, YG-15, YG-16, and YG-17 (237-352 ppb) were 290 generally higher than the average concentration of coals from around the world (100 ppb) and the 291 average concentration of Chinese coals (163 ppb) (Dai et al., 2012b). After normalizing for Al<sub>2</sub>O<sub>3</sub>, it 292 still showed anomalously high values in these five coal samples, indicating that these five Hg peaks are true Hg anomalies (Fig. 5) (Xie et al., 2022). 293 294 Xie et al. (2022) demonstrated that wildfire burning resulted in the release of most of the Hg 295 accumulated in vegetation and soils to the atmosphere, and that atmospheric Hg accumulated 296 quickly in peatlands, leading to Hg anomalies enrichment in peatlands. Comparing the Hg levels of 297 the samples in this study with the frequency of wildfires has not shown that the anomalously high 298 values of Hg are well correlated with the frequency of wildfires (Fig. 5). There were even 299 anomalously high values of Hg in samples YG-15, YG-16, and YG-17, but the frequency of wildfires was significantly reduced. This indicates that the anomalous enrichment of Hg in this 300 301 study was not caused by wildfire burning. 302 The mass of Hg in forest vegetation is about 0.1 mg/m<sup>2</sup>, however, the mass of Hg in peatlands is about 20 mg/m<sup>2</sup>, which is much greater than the amount in vegetation (Grigal, 2003; Turetsky et 303 al., 2006). The types of wildfire burning in Xie et al. (2022) were primarily ground and surface fires. 304 305 Ground fires were fueled primarily by a dry humus layer in the soil (Scott, 2010), which can burn a 306 thicker layer of peat, resulting in the release of Hg from the peatland. In this study, the type of wildfire burning was predominantly surface fires, with the primary fuels for surface fires were low 307 308 surface vegetation and biological litter (Scott, 2010), which coincides with the observation of ferns 309 as fuels in the SEM (Fig. 3). Ku et al. (2018) found that different wildfire intensities had different 310 effects on Hg volatilization from vegetation. Therefore, wildfire burning may not necessarily lead to

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311 Hg enrichment, and wildfire types may not have the same effect on Hg. Mercury enrichment is 312 favored when wildfires were primarily ground fires, where burning of the peat layer resulted in the 313 release of large quantities of Hg, which were absorbed by the peatland. 314 Excluding the anomalous enrichment of Hg in coal due to wildfire activities, the known causes 315 of the anomalous enrichment of Hg in coal are mainly the volcanic activities during the early 316 peatland accretionary phase that led to the increase of Hg input (Roos-Barraclough et al., 2002; 317 Yudovich and Ketris, 2005), and the invasion of low-temperature hydrothermal and magmatic fluids into coal-bearing strata (Sun et al., 2016; Zheng et al., 2018). Zhang et al. (2023a) identified 15 318 319 altered volcanic ashes in this study area, demonstrating the frequent volcanic activity during this depositional time. And it was confirmed that trace element enrichment in closed coal seams may be 320 321 due to the dipping of tonsteins by acid solutions. Correlation analysis of the Hg content of No. 9 322 coal with trace elements such as Ga, Zr, and Hf revealed a significant correlation (r=0.581-0.705). 323 Hg enrichment has been widely used as the indicator of volcanic sediment input (Shen et al., 2020), 324 so the anomalously high values of Hg in the study area may be attributed to the input of Hg 325 elements due to the frequent volcanic activities at that time, and the Hg in the tonsteins was leached 326 into the closing coals by leaching of the acid solution.





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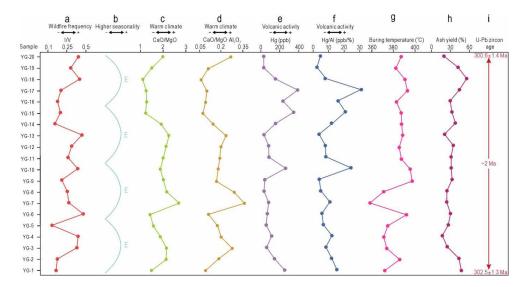


Fig. 5 Comprehensive analysis map of No.9 coal in Yaogou Mine. (a) Inertinite/ Vitrinite variations in 20 coal samples. (b) Long eccentricity orbital cycle variation. (c) CaO/MgO trends in 20 coal samples. (d) CaO/MgO • Al<sub>2</sub>O<sub>3</sub> trends in 20 coal samples. (e) Hg content trends in 20 coal samples. (f) Hg/Al trends in 20 coal samples. (g) Combustion temperature trends in 20 coal samples. (h) Ash yield trends in 20 coal samples. (i) Age of No. 9 coal, referred to Zhang et al. (2023a).

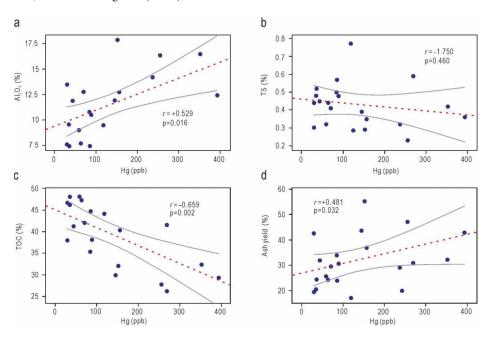
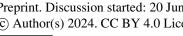


Fig. 6 (a) Correlation between Hg and Al<sub>2</sub>O<sub>3</sub>. (b) Correlation between Hg and TS. (c) Correlation between Hg and



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335 TOC. (d) Correlation between Hg and ash yield. Spearman's P and r values are shown. Red lines denote the

336 best-fit linear regression line. Gray curve lines denote the 95% confidence interval.

### 5.3 Gzhelian wildfires and climate change

### 5.3.1 Regional wildfire trends

Wildfire activity during geologic history has been influenced primarily by changes in atmospheric oxygen levels, climate and vegetation (Belcher and Hudspith, 2017; Baker, 2022). For example, Glasspool and Scott (2010) suggested that high levels of oxygen in the atmosphere influence increased wildfire activity on different time scales. Baker (2022) found that past periods of global climate change on Earth have been accompanied by increased wildfire activity. Vegetation type changes also influence wildfire occurrence, for example, the rise of angiosperms during the Cretaceous may have led to an increase in wildfire activity at that time (Lü et al., 2024). The level of oxygen in the atmosphere plays a key role in the occurrence of wildfires (Scott, 2000); when oxygen levels are below 15% it will not be possible to sustain vegetation burning, and when levels are above 23% humid vegetation will continue to burn. Based on model predictions, the oxygen level of 24.6% in the Gzhelian is higher than current oxygen levels and can sustain wildfire burning in humidter environments (Fig. 7).

CaO/MgO can be used as an indicator for paleoclimate reconstruction (Chen and Wan, 1999; Sun et al., 2010; Liu et al., 2013; Fu et al., 2018), and the ratio of CaO/MgO • Al<sub>2</sub>O<sub>3</sub> can more sensitively respond to the temperature changes at that time, with high values indicated for warm periods, and low values indicated for cold periods (Chen and Wan, 1999; Fu et al., 2018; Zhang et al., 2020b; Zhang et al., 2021). In this study, periods of high wildfire frequency (e.g., YG-10, YG-11, YG-12, YG-13) had higher values of CaO/MgO and CaO/MgO • Al<sub>2</sub>O<sub>3</sub>, indicated of a warm and humid climate (Fig. 5). Periods of lower wildfire frequency (e.g., YG-14, YG-15, YG-16, YG-17)

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had lower CaO/MgO and CaO/MgO • Al<sub>2</sub>O<sub>3</sub> values, indicated a cold and dry climate (Fig. 5). 358 359 Although wildfires were more easily caused when the climate was dry, cold climates with lower 360 temperatures were not favourable for vegetation development (Brovkin, 2002), and a dryer climate 361 meant less rainfall, both of which led to a decrease in fuels, hence a less frequent occurrence of wildfires. However, in warm and humid climatic periods, where seasonal variations were stronger, 362 sufficient rainfall and suitable temperatures promote the development of vegetation, leading to a 363 significant accumulation of fuels, hence more favourable for wildfires to occur (Litt et al., 2014; 364 365 Stockhecke et al., 2016; Swain, 2021). Furthermore, previous research has found that high frequencies of wildfire activity have occurred in moderately humid environments (Daniau et al, 366 2012; Marlon et al, 2013). This study predicts oxygen levels sufficient to sustain wildfires in humid 367 368 conditions for sustained burning, so sufficient fuel accumulation may have been a major factor in 369 the frequency of wildfires during this period. 370 Many scholars have examined charcoal abundance changes in Quaternary sediments to show 371 that orbital cycle forcing climate drives wildfire activity (Verardo and Ruddiman, 1996; Thevenon 372 et al., 2004; Zhou et al., 2007; Daniau et al., 2013, 2023; Kappenberg et al., 2019; Hao et al., 2020; 373 Shi et al., 2020; Zhang et al., 2023b). Zhou et al. (2023), based on a high-resolution charcoal fragment record from the Wushan Basin of the Tibetan Plateau during the Middle Miocene, found 374 375 that orbital period (short eccentricity, slope, and age difference) forced climate change to control 376 wildfire activity by influencing the amount of vegetative fuels and their combustibility. In this study, 377 Coal 9 has a floor age of  $300.6 \pm 1.4$  Ma, a roof age of  $302.5 \pm 1.3$  Ma, and a depositional age of ~1.9 Ma (Zhang et al., 2023a). We explored the effect of the orbital cycle on wildfires using long 378 379 eccentricity. As shown in Fig. 5(b-d), the variation of the long eccentricity had a good correlation with our climatic indicators (CaO/MgO, CaO/MgO · Al<sub>2</sub>O<sub>3</sub>). The maximum eccentricity is 380

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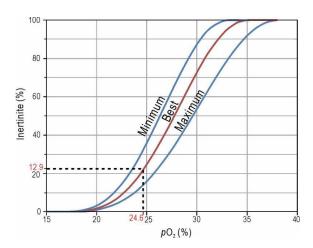
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382 with a colder and dryer period. When the orbital cycle eccentricity is at maximum, the sun was at 383 perihelion and the amount of insolation was relatively abundant, leading to a sufficient 384 accumulation of fuels, which in turn led to an increase in wildfire activity (Kappenberg et al., 2019; Hollaar et al., 2021; Qiu et al., 2023; Zhang et al., 2023b). This is consistent with the study by 385 Kappenberg et al. (2019) who found that orbital forcing leads to peak wildfire activity during warm 386 387 and humid periods. 388 Combined with the above research, the factors controlling wildfires in this study area are shown in Fig. 8. When the eccentricity was at a minimum, the Earth received insufficient insolation, 389 390 the climate was cold and dry, and seasonal variations were not evident (Hollaar et al., 2021; Huang 391 et al., 2024). The lack of suitable environments for survival at that time resulted the low amount of 392 vegetation. There may have been a low frequency of wildfires occurring at this stage, which produced less charcoal preserved in the peatland (Fig. 8a). When the eccentricity was at a maximum, 393 394 the Earth received sufficient insolation, the climate was warm and humid, and seasonal variations 395 were evident (Hollaar et al., 2021; Huang et al., 2024). Suitable environments lead to the proliferation of vegetation, which in turn accumulate large amounts of fuel. More rainfall was also 396 followed by more lightning, leading to increased chances of ignition. Thus there were more frequent 397 398 wildfires during the period, which produced more charcoal preserved in the peatlands (Fig. 8b). In 399 this study, intensive wildfires did not correlate to peaks in Hg concentrations, and the anomalous enrichment of Hg was most likely due to the frequent volcanic activity occurring at that time. When 400 401 the volcano erupts, the ash can carry large Hg transport (Coufalik et al., 2018). When volcanic ash 402 passes through peatlands, the Hg it carried naturally settles and the strong adsorption of Hg by 403 peatlands leads to anomalous enrichment of Hg at that time (Fig. 8a) (Yudovich and Ketris, 2005).

associated with a warmer and more humid period, while the minimum eccentricity is associated







405 Fig. 7 The atmospheric oxygen concentration during the Late Carboniferous Taiyuan Formation in the Ordos

Basin based on the prediction model proposed by Glasspool et al. (2015).

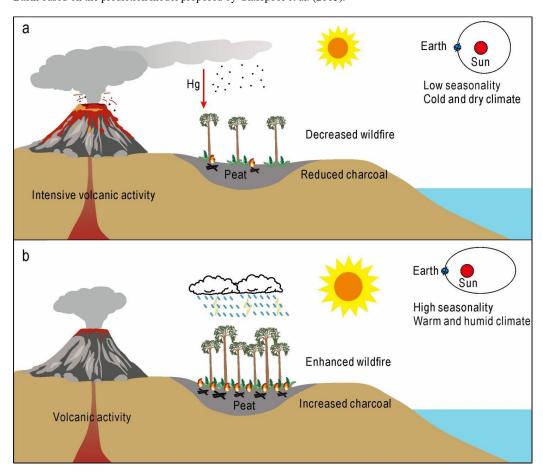


Fig. 8 Illustration of wildfires activity forced by the long eccentricity orbital cycle in the study area.

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# 5.3.2 The geographical distribution of published evidence in the Gzhelian

410 The distribution of wildfires on Earth is not random, and the spatial distribution of wildfires in 411 the modern world is affected by vegetation, climate, topography, and human activities (Krawchuk et 412 al., 2009). Studies have shown that frequent wildfires in modern environments mainly occur in tropical forests and savannas (Mouillot and Field, 2005; Giglio et al., 2006; Flannigan et al., 2009, 413 414 2013). In this study, based on the distribution of climate zones published by Boucot et al. (2013), it 415 was found that all of the wildfire records for the Gzhelian occurred in the tropical climate zone (Fig. 416 9). It was similar to the modern distribution of wildfires. In the tropics of the Gzhelian, different biomes are distributed, mainly divided into humid to semi-humid wetland groups and semi-humid 417 418 to semi-arid wetland groups, and the peatlands were under alternating humid and dry changing 419 environments (DiMichele et al., 2010; DiMichele, 2014). In warm and humid environments, 420 vegetation grows densely and generates large fuel accumulations. And in arid environments, fuel 421 burnability is enhanced, which in turn promotes more frequent wildfires (Denis et al., 2017; Baker, 422 2022). Hence the frequent occurrence of wildfires in the tropics with alternating seasons. 423 In view of the spatial distribution, the Gzhelian wildfire records were all located at low latitudes near the equator. In the Gzhelian, the southern part of the Gondwana was covered by a 424 large ice sheet, resulting in the lack of vegetation and the lack of fuel needed for wildfires to burn 425 426 (Scotese, 2021) (Fig. 9). Similarly in the northern part of the Gondwana and the northern part of the 427 Laurasia there were extensive arid climate zones spread across the mainland that lacked the fuels needed for wildfire burning, resulting in the absence of wildfire records (Fig. 9). However, it is 428 429 worth noting that widespread biases, such as taphonomy, research preferences, and sampling bias, 430 may result in global wildfire data for the Gzhelian not representing the true situation at that time 431 (Brown et al., 2012; Hamad et al., 2012; Lu et al., 2021; Lü et al., 2024). For example, this study





432 found no evidence of wildfires that have been reported in warm temperate regions (Fig. 9).

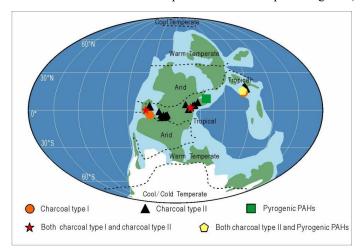


Fig. 9 Paleogeographic distribution of published wildfire occurrences during the Gzhelian. Plate reconstructions

from Scotese (2016) with paleoclimatic zones based on Boucot et al. (2013).

# 6. Conclusion

In this study, the detailed analysis of the inertinite in the No. 9 coal of the Yaogou coal mine in the Ordos Basin, China, demonstrated that frequent wildfires had occurred in the region during the Gzhelian of the Late Carboniferous and were mainly low-temperature fires. The frequency of wildfires during this period may be related to the forcing of the long eccentricity orbital cycle, where changes in the climate due to changes in the orbital cycle affect the fuel load. The review of the global wildfire record of the Gzhelian that wildfires were concentrated in low-latitude tropical climates, and the distribution of wildfires may have been limited by the climate and fuels available at that time. The anomalous enrichment of Hg in No. 9 coal was probably due to inputs of Hg from the frequent volcanic activity occurring at that time, and was not related to wildfire burning. The different types of wildfire burning may limit the release and enrichment of Hg.

# **Author Contribution**

All the authors have actively participated in the preparation of the manuscript. Wenxu Du was





responsible for data curation, methodology, and writing; Dawei Lv was responsible for funding
acquisition, project administration, and supervision; Zhihui Zhang was responsible for funding
acquisition and methodology; Munira Raji was responsible for review; Cuiyu Song was responsible
for conceptualization and methodology; Luojing Wang was responsible for visualization and
investigation; Zekuan Li was responsible for investigation; Kai Cao was responsible for software
and validation; Ruoxiang Yuan was responsible for visualization and investigation; Yuzhuang Sun
was responsible for review. All authors read and approved the final proof.

# **Competing Interests**

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The authors declare that they have no conflict of interest.

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464 comments on this paper.

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