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Variscan cycling of gold into a global coal reservoir

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ABSTRACT

The potential of coal and coal ash as a source of strategically and/or commercially valuable trace elements, including gold, is reflected in numerous analytical studies, reports to government, patents for new technology and the first commercial extraction facilities. It is shown here that there was a particular predisposition to the concentration of gold in the Upper Carboniferous (313–304 Ma) Carboniferous coals that dominate coal supplies in the northern hemisphere. Coal was deposited in foreland and other basins adjacent to the actively colliding Variscan-Alleghanian Orogen, while the orogenic belt was mineralized by gold ore from 410 to 310 Ma over 10,000 km from the Appalachians to China. High gold fertility in the orogenic belt, the additional concentration of gold along shear zones during transpression, and either rapid erosion (> 1 km/Myr) of the orogenic belt into adjacent basins or hydrothermal mineralization of the basins, made the coals a trap for the consequent flux of gold. This cycling of gold is evidenced by enriched coal and gold palaeoplacers in coal-bearing sequences in numerous basins along the orogen. Provenance data, especially detrital zircon ages of 350–300 Ma, help to identify successions that were sourced from gold-rich terrains, and thus can focus the search for anomalously enriched coal.

1. Introduction

In just a few years, awareness and interest in the resources of trace elements in coal has greatly increased (Tolhurst, 2015; Yao et al., 2015; Dai and Finkelman, 2018; U.S. Department of Energy, 2017). Measurements of whole coal and coal ash have shown that it can contain localized high contents of noble metals, rare earths, gallium, scandium, selenium and other elements (Franus et al., 2015; Dai et al., 2018; Chelgani and Hower, 2018). It has consequently been assessed as a potential source of a wide range of elements, from selenium (Bullock et al., 2018) to lithium (Qin et al., 2015). The abundance of coal, the availability of huge volumes of coal ash as a waste product from power stations (Abdullah et al., 2019), and the concentration of trace elements during the ashing process, make coal a potentially valuable reservoir of trace elements. Development of the technology to realise this potential, in Russia and China, was accompanied by claims that it was economically viable in those countries (Bratskaya et al., 2009; Taskin et al., 2016, 2017; Dai et al., 2018). This was rapidly followed by initiatives in western countries, including analytical surveys, report to U.S. Congress (U.S. Department of Energy, 2017) and the award of patents for extraction techniques. A major driver for this activity is the strategic need for trace elements in future technologies (Lusty and Gunn, 2014), and geopolitical controls on their availability. Geological studies have has focussed on analytical data rather than interpretation and prediction of deposits. This study documents the geological significance of gold concentrations in widespread Upper Carboniferous coal deposits to underpin exploration for trace element resources.

Occurrences of gold in coal sequences were first recorded by chance during routine analytical surveys, notably ppm-level gold recorded in coal ash in Wyoming (Stone, 1912), and as palaeoplacers (Klominsky et al., 1979). In the past four decades, data sets measured in support of understanding the impact of coal and its ash on the environment, and alternative ways of utilizing the ash, have highlighted more occurrences that suggest that coal may be an economically important reservoir of gold and other precious metals. However, the data is patchy, as many analytical studies of coal adopt a detection limit for gold which is much higher than the level that would make it valuable (see below). Evidence for gold in Carboniferous coal reviewed here is therefore assembled from a variety of types of data.

2. Geological context

The collision of Laurussia with Gondwana to form the supercontinent Pangea was marked by the Variscan-Alleghanian Orogen (Fig. 1) that extended 3000 km from the USA (Appalachian) to central Europe (Variscan, Central Pangean Mountains) (Simancas et al., 2005). Orogenesis at the margin of Laurussia with the closing Palaeothethys Ocean continued eastward through the Caucasus and Central Asia to

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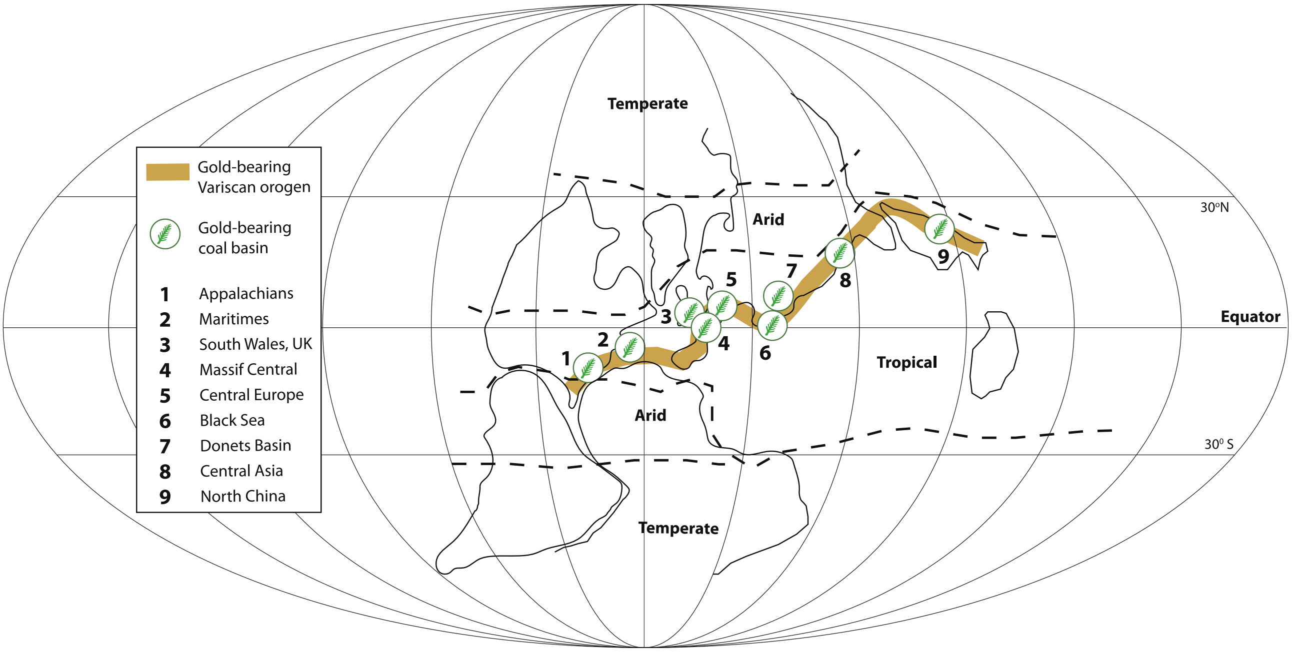

Fig. 1. Variscan orogenic belt, mineralized by gold deposits within zone of tropical climate during Upper Carboniferous (Boucot et al., 2013). Distribution of gold-bearing Pennsylvanian coal deposits shown adjacent to orogenic belt.

China (Fig. 1), nearly 10,000 km in total length. Progressive and rapid denudation of the orogenic belt fed detritus into adjacent foreland basins that developed as a flexural response to crustal thickening, parallel to the tectonic strike, and also into basins on the immediately adjacent cratons. New orogenic belts dominate the clastic budget by orders of magnitude compared to cratonic sources. Critically, the orogenic belt was oriented west-east to northwest-southeast in the vicinity of the equator (Fig. 1), in a tropical climate (adopting nomenclature for climatic belts of Boucot et al., 2013). A combination of wet climatic conditions and fluctuating sea level during the Upper Carboniferous gave rise to the deposition of coals in the foreland and intracratonic basins (Cleal et al., 2009). Coal was deposited in basins along the entire length of the orogen, and coal deposition continued at the eastern end (Central Asia, North China) through the following Permian period at latitudes up to 30° N while the equatorial region was becoming arid (Hilton and Cleal, 2007).

In the Euramerican region of Carboniferous basin formation, the Caledonian Orogeny and Variscan Orogeny associated with the closure of the Iapetus Ocean and the Rheic Ocean respectively were both extensively mineralized by gold (Goldfarb et al., 2001; De Boorder, 2012; Romer and Kroner, 2018), and the ores are consistently dated in the range 410–310 Ma in the Variscan Orogenic Belt (Romer and Kroner, 2018). Magmatism and gold mineralization continued progressively eastwards to China, where it peaked at about 280 Ma (De Boorder, 2012). Magmatism and metamorphism during Variscan crustal thickening introduced and concentrated gold (Romer and Kroner, 2018). The rich gold reservoir in the orogen became spatially concentrated by shear zones parallel to the belt, possibly reactivated from the geometry of thrust slices (Fig. 2). Shear zones are the primary pathways for gold-bearing fluids (Groves et al., 2018). The shear zones were active late in the history of the orogeny during the transition from compression to transpression driven by changing far-field behaviour (Groves et al., 2018). Gold deposits in the Variscides were particularly associated with this strike-slip behaviour (de Boorder, 2012). The result was an extensive chain of gold deposits through the Appalachians, Maritimes, North Africa, Iberia, Massif Central, Bohemian Massif, Caucasus, Central Asia and China. The concentration of gold to form discrete mineral deposits in this way enhanced the availability of particulate gold to the drainage systems during rapid exhumation and erosion of the orogenic belt. More generally, the reductive power of organic matter, which causes gold precipitation in black shales and graphitic schists (Hu et al., 2017) made the coal a chemical trap for the gold liberated from the orogen. The exceptional combination of gold and coal has resulted in a novel reservoir of gold in the coal deposits, at a scale of global importance.

3. Methods

Data were collated from published sources. Occurrences of gold in coal were registered as significant if they involved particulate gold or above 1 ppm gold. Quantitative exhumation data were recorded from massifs in the Variscan Orogen in Europe within the time frame 350 to 300 Ma. Zircon age data were collated for the time period 500 to 250 Ma, sampled from Mid-Upper Carboniferous sandstones (Table 1). Ages of gold deposits are reviewed by Romer and Kroner (2018), and sources for individual regions are recorded in Table 1.

4. Data

Gold in coal is anomalous if it occurs as discrete particles detectable

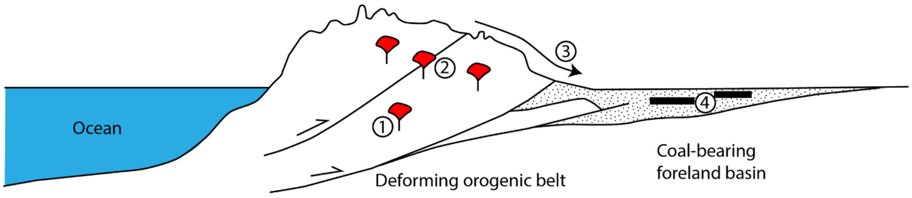

Fig. 2. Schematic cross-section through Variscan Orogen and related coal-bearing foreland (or intramontane rift) basin. Numbered stages indicate progressive stages in gold concentration: (1) gold introduction in magmas, (2) gold ore mineralization in shear zones, (3) exhumation to deliver gold to foreland basin, (4) gold concentration in coal by hydrodynamic and redox processes.

Table 1
Occurrences of gold in coal in Variscan orogen (regions shown in Fig. 1).

Region	Gold ore age in orogen (Ma)	Coal age (Ma)	Provenance	Gold in coal-bearing sequence	Type of gold in coal	References for gold in coal
1. Appalachian Basin	390–320 (Ayuso et al., 2005)	320–300	440–340 zircons (Thomas et al., 2017)	Up to 3.4 ppm gold in whole coal, Kentucky. Gold particles in coal.	Detrital, Hydrothermal	Goldhaber et al. (2007), Chelgani and Hower (2018)
2. Maritimes	410–380 (Morelli et al., 2005)	320–300	400–340 muscovite in Lower Carb. (Reynolds et al., 2010)	Up to 0.21 ppm gold in whole coal. Gold paleoplacers in Upper Carb.	Detrital	Zodrow (1987), Mills (1999)
3. South Wales	Local 410–390 Bohemian Massif 350–330 (Morávek & Pouba 1987)	320–300	370–355 white mica (Sherlock et al., 2000)	Up to 0.18 ppm gold in whole coal; 4.4 ppm in coal ash, Wales.	Hydrothermal	Gayer and Rickard (1994)
4. Massif Central	335–300 (Bouchot et al., 2005)	320–300	365–305 zircons (Pfeifer et al., 2018)	Gold paleoplacers in coal basins.	Detrital	Bouchot et al. (2005)
5. Central Europe (Poland, Czech Rep.)	350–330 (Zachariáš et al., 2014)	320–300	360–300 zircons (Mazur et al., 2008)	Up to 1.9 ppm gold in coal fly ash. Up to 0.14% gold in pyrite. Gold paleoplacers.	Detrital	Klominsky et al. (1979), Malec et al. (2012), Bielowicz & Misiak (2017), Franus et al. (2015)
6. Black Sea	360–260 then Alpine overprint (Mladenova et al., 2004)	320–300	350–330 zircons, Turkey (Okay et al., 2011)	Up to 1.1 ppm gold in whole coal, Bulgaria.	Detrital	Eskenazy (2009)
7. Donets Basin	300–260 (Sachsenhofer et al., 2012)	320–300	No data	Up to 1.4 ppm gold in whole coal.	Hydrothermal	Seredin (2007)
8. Central Asia	320–260 (Seltmann et al., 2011)	320–260	330–300 zircons (Huang et al., 2018)	Gold in coal sequence, Kazakhstan.	Hydrothermal	Levitan (2008)
9. North China	320–260 (Zhou et al., 2002)	320–260	320–300 zircons (Li et al., 2008)	Rare earths in Pennsylvanian coal, up to 3 ppm gold in Permian coal.	Hydrothermal	Nie et al. (2008)

by electron microscopy or other microscopy, and coal containing about 50 ppb noble metals (typically 50% as gold) is potentially an ore (Chelgani and Hower, 2018). This composition is an order of magnitude richer than the average gold concentration in coal (Dai et al., 2018). From west to east, Upper Carboniferous (middle-upper Pennsylvanian, Westphalian) coal-bearing basins show evidence for gold enrichment determined from independent studies along the length of the orogen (Fig. 1, Table 1):

Appalachian Basin coal contains detrital gold and up to 3.4 ppm gold in whole coal in eastern Kentucky (Hower et al., 2018; Chelgani and Hower, 2018), and gold-enriched pyrite in Alabama (Goldhaber et al., 2007). The data from Kentucky show a very clear positive correlation with ash, and inverse correlation with carbon, confirming residence in mineral form (Chelgani and Hower, 2018). Gold enrichment is associated with enrichment in rare earth elements (Hower et al., 2018).

Maritimes (Canada) coal has yielded up to 0.21% gold (Zodrow, 1987), and enrichments in other elements including rare earths (Birk and White, 1991). The distribution of metals indicates their residence in pyrite (Zodrow, 1987). There is also evidence of detrital gold in palaeoplacers of the Upper Carboniferous in Nova Scotia and New Brunswick (Mills, 1999), and also the Lower Carboniferous of Nova Scotia (Ryan et al., 1988; Jennex et al., 2000). High concentrations of other trace elements in coal ash have been attributed to a source in nearby orogenic rocks (Kaplan et al., 1985).

Massif Central (France) Pennsylvanian coal-bearing sediment includes gold-bearing palaeoplacers in at least two regions (Bouchot et al., 2005). Gold-bearing quartz-arsenopyrite pebbles occur at Bulidou, Alès Basin, and in the Argentat Basin. In both cases, the mineralogy of the pebbles provides a clear picture of the hydrothermal event in the Variscan basement before erosion (Charonnat et al., 1999; Nomade et al., 1999; Bouchot et al., 2005).

British coal-hosted Pennsylvanian anthracite gold occurs in the South Wales Coalfield (Gayer and Rickard, 1994), as particles accompanying a carbonate-quartz-clay-sulphide-selenide assemblage in cleats (joints). The gold exhibits a colloform morphology indicating growth into open space, and at least some post-depositional mobility. Coal ash in Wales has up to 4.4 ppm gold, although some ash yielded less than a detection limit of 0.005 ppm (Gayer and Rickard, 1994). Gold particles have also been recorded in siderite nodules within **Irish** Pennsylvanian coal, and are assumed to be derived from groundwaters draining off nearby gold-bearing basement rocks in the Leinster Massif (Bullock et al., 2019).

Central Europe records gold mineralization in the Pennsylvanian of Poland and the Czech Republic, adjacent to the Bohemian Massif. Gold occurs in palaeoplacers in several basins and mineralizes coalified plant material, in the Czech Republic (Klominsky et al., 1979; Malec et al., 2012). It is variably mixed with silver, mercury, platinoids and selenium. Traces of gold have been detected in pyrite in coal in Silesia, Poland (Bielowicz & Misiak, 2017), and the resultant fly ash has yielded about 1 ppm gold (Franus et al., 2015).

Black Sea region bituminous coal yields up to 1.1 ppm Au from the Dobrudzha Coalfield, Bulgaria (Eskenazy, 2009). Associated coaly shale yields a mean of 2.3 ppm Au. Limited data suggest that the Au is bound to organic matter in the coal (Eskenazy, 2009).

The Pennsylvanian coal in the **Donets Basin**, Ukraine and Russia, has yielded up to 1.4 ppm gold in whole sub-bituminous to anthracite coal (Seredin, 2007). The coal is also mineralized by mercury, which was mined with the coal extracted as a by-product, from Variscan structures (Kolker et al., 2009), and the gold is further associated with enrichments in arsenic and cadmium. The mineralization was associated with Permian magmatism and hydrothermal activity (Sachsenhofer et al., 2012).

In **Central Asia**, gold mineralization overlapped sedimentation in Pennsylvanian coal-bearing sandstone-mudrock successions in Kazakhstan (Levitan, 2008). The gold occurs in carbonaceous sediments, and cross-cutting quartz veins, and is particularly concentrated near thrust surfaces. It resides in pyrite and arsenopyrite in the quartz, at up to 150 ppm (Levitan, 2008).

Chinese coal of Pennsylvanian age contains anomalous concentrations of other rare elements, some to ore grade including gallium, attributed to derivation from the Variscan basement (Dai et al., 2018). Orogenic activity, and accompanying gold mineralization, continued into the Permian in Central Asia and China, and similarly did coal deposition. The record of enrichment of coal with gold is accordingly extended from the Carboniferous to the Permian in China (Nie et al., 2008). The coal-bearing Permian Longtan Formation contains up to 3 ppm Au (Nie et al., 2008).

Quantitative estimates for the Mid/Upper Carboniferous exhumation rate of the Variscan mountains across Europe from the Iberian Massif to the Bohemian Massif are consistently in the range millimetres to centimetres per annum (Brown and Dallmeyer, 1996; Schulmann et al., 2002; Capuzzo et al., 2003; Willner et al., 2002; Pfeifer et al., 2018), i.e. up to 10 km erosion in a million years. Beyond Europe, petrographic and provenance studies also indicate rapid exhumation at this time, in the Appalachians (Gehrels et al., 2011), the Anti-Atlas of Morocco (Sebti et al., 2009) and Central Asia/China (Han et al., 2011). The quantitative values reported from Europe are one to three orders of magnitude greater than in non-orogenic settings. Gold-bearing porphyry and epithermal gold deposits are emplaced within 3 km of the surface, and would be exposed and eroded within a million years at the estimated exhumation rates. Palaeoplacer gold, contributed by mechanical erosion, is therefore an expected product of orogenic evolution.

Provenance data help to link the availability of abundant gold in the orogeny and incorporation of gold in the coal. Dating of detrital grains (zircon, monazite, muscovite) along the entire orogen shows that the Carboniferous basins received a very young component whose age overlapped the time of gold mineralization. Quantitative zircon age data from the Appalachians, Massif Central, Poland, Black Sea and Central Asia (Fig. 3) all indicate that Mid-Upper Carboniferous sandstones were dominated by sources that were less than 100 million years age at the time of deposition (Mazur et al., 2008; Okay et al., 2011; Thomas et al., 2017; Huang et al.; 2018, Pfeifer et al., 2018). The zircons are dated 300 to 400 Ma, and especially 300 to 350 Ma (Fig. 3). Rocks of Mid-Upper Carboniferous age therefore included the newly exposed gold ores, dated at 410–310 Ma. In the Massif Central and Central Europe, where palaeoplacer gold is recorded in the Pennsylvanian, zircon data confirm the erosion of rocks simultaneous with local gold ore mineralization (410 to 310 Ma).

5. Discussion

The zircon data (Fig. 3) show that the provenance of Upper Carboniferous sandstones (modes 350–300 Ma) in coal-rich basins is the rapidly eroding Variscan orogen, a gold-fertile source terrane (Fig. 4). Gold deposits in the Variscan orogen may have been transferred to the adjacent foreland or intramontane basins by either erosion and deposition of detrital particles, or by mineralization of gold and post-depositional relocation by metal-rich fluids. In the Euramerican region (Appalachians to Black Sea in Fig. 4), the timing of mineralization (410–310 Ma) was up to 100 Myr prior to the timing of coal sedimentation in the adjacent basins (313–304 Ma), suggesting that detrital gold fragments were eroded from the orogen and deposited in the basins rather than being transported by post-depositional metal-rich fluids. Additionally, there is clear evidence for the supply of gold in the waning stages of orogenesis. For example, in South Wales, where discrete gold occurs in cleat-fills in the coal, mineralization of the coal was enhanced by fluid migration due to orogenic compression, as the Variscan thrust system propagated northwards into the coalfield even as Pennsylvanian sedimentation was continuing (Gayer et al., 1998). Clasts of coal containing cleats are found within the Pennsylvanian sequence, evidencing the overlap of mineralization and sedimentation. In the Asian region (Donets Basin, Central Asia and China) in particular, magmatism and hydrothermal activity continued during and after coal sedimentation, and there is more evidence of the post-depositional mineralization of coal (Fig. 4). This broad distinction between Euramerican and Asian regions must partly reflect that the Euramerican sequences have a longer, more detailed, history of study, in which the palaeoplacers are more likely to have been identified. The inferred hydrothermal activity is recorded along the whole orogen (Table 1, Fig. 4).

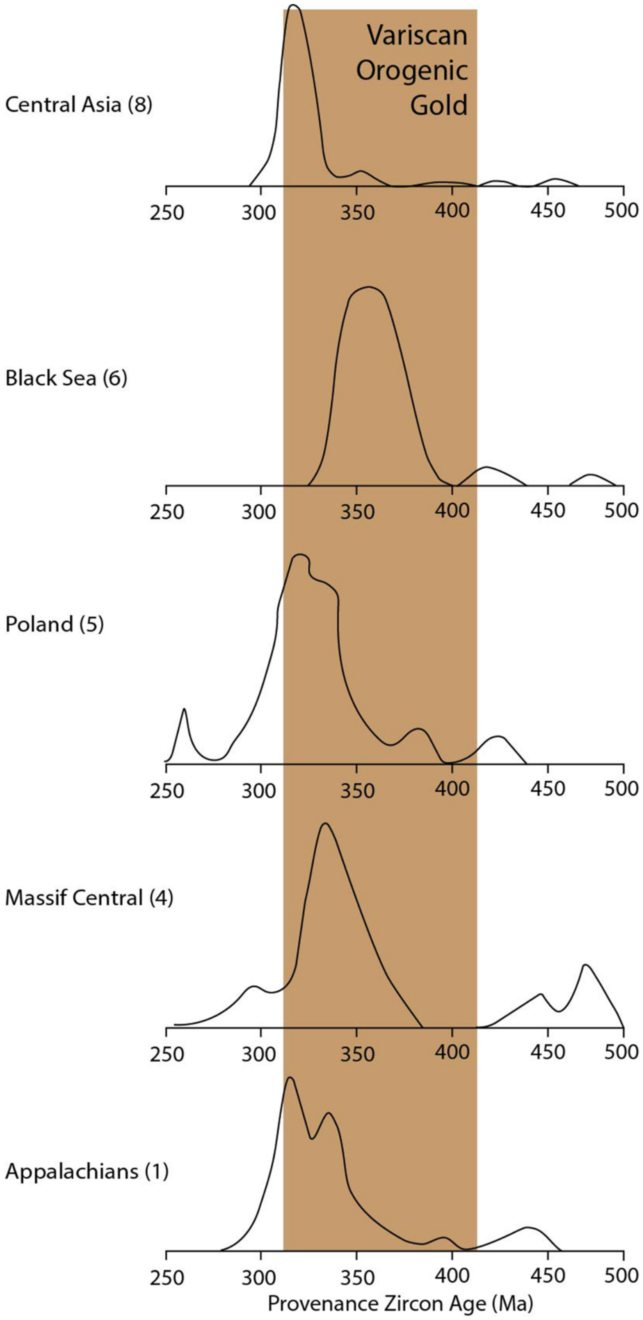

Fig. 3. Zircon age spectra for Mid-Upper Carboniferous sandstones in five regions along the length of the Variscan orogeny. The ages consistently peak at earlier stages of the Variscan Orogeny, i.e. the zircons were extremely young, and overlap the age of the gold-bearing Variscan basement, which was eroded. Data from Mazur et al. (2008), Okay et al. (2011), Thomas et al. (2017), Huang et al. (2018) and Pfeifer et al. (2018).

The zircon data imply that provenance data would refine the prediction of gold concentrations in coal and other Upper Carboniferous

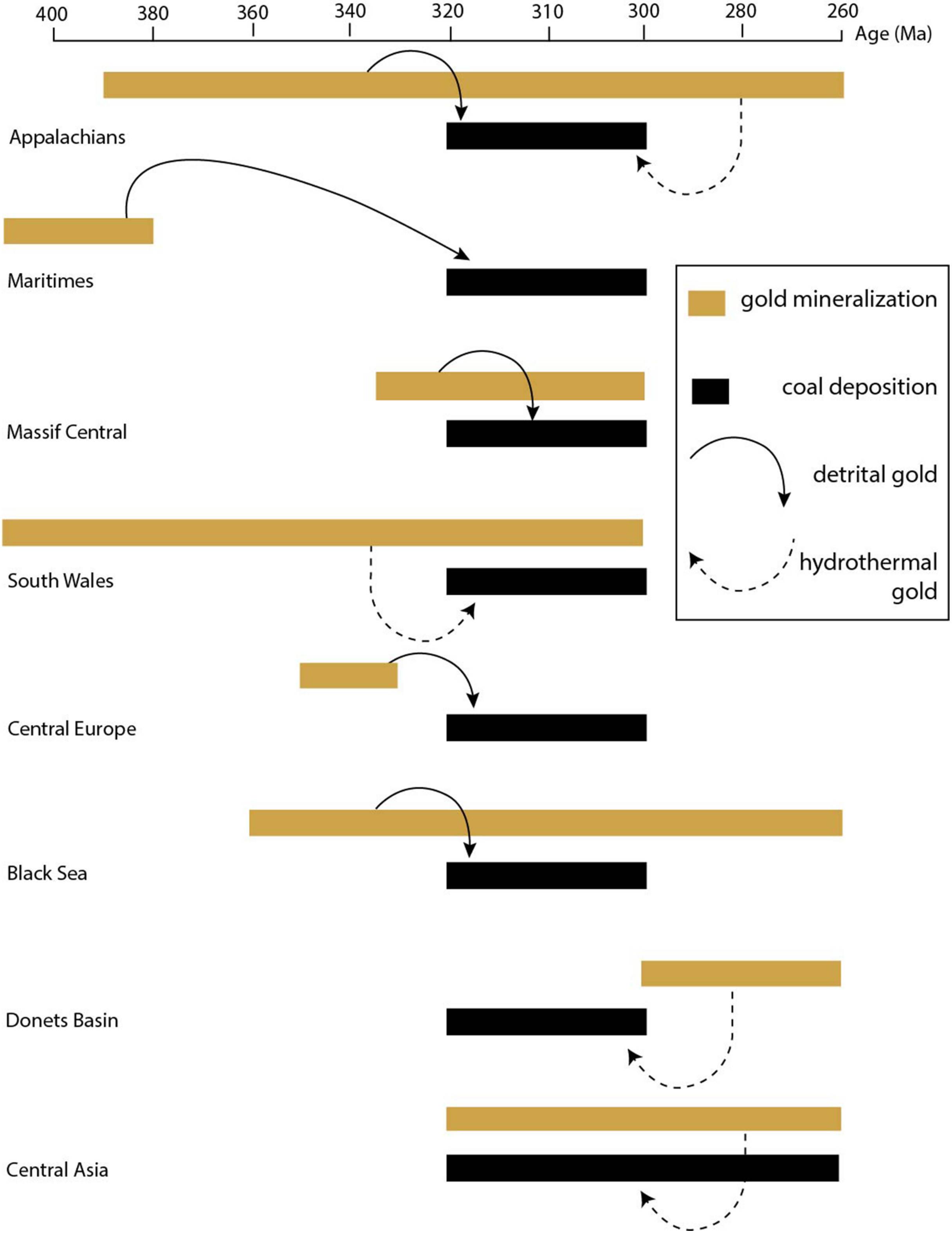

Fig. 4. Temporal relationships for gold mineralization and coal deposition along Variscan Orogen, and means of supply of gold to coal (detrital, hydrothermal). Coal age data from Cleal et al. (2009) and Nie et al. (2008). Gold ore age data from Ayuso et al. (2005), Morelli et al. (2005), Bouchot et al. (2005), Zachariáš et al. (2014), Morávek & Pouba (1987), Mladenova et al. (2004), Sachsenhofer et al. (2012), Seltmann et al. (2011), and Zhou et al. (2002). Table 1.

rocks. Zircon age data elsewhere has been used to detect the provenance of gold mineralization (Stephens et al., 2017; Kanouo et al., 2018), and suggest that it could be used to identify coal-bearing successions that have a gold-rich source and thus may be particularly gold-prospective.

Palaeoplacer gold requires a mineralized protolith, erosion of the protolith, a sedimentary system that includes the erosion products, a depositional environment that traps and concentrates the gold before it becomes dispersed, particularly through good sorting, and subsequent preservation of the deposit. The Variscan orogen met these requirements because its foreland basins were not only the immediate destination for particulate gold, but the mixed sequences containing coal and sandstone provided a combination of chemical and physical traps that caused the gold to be retained. Well-sorted sands are found in many Carboniferous coal-bearing basins (e,g. van Hinsbergen et al., 2015; Du et al., 2016). The relatively high sedimentation rates, and subsidence, in foreland basins (Allen et al., 1986) helped to retain material within the basins. But most important was the abundance of coal. The juxtaposition of gold and coal was ultimately a consequence of the palaeogeography of the Variscan orogen and palaeoclimate conditions in the low-latitude climate belt (Fig. 1) where plants flourished (Cleal et al., 2009; Boucot et al., 2013). The size of this climate belt varied through time, dependent on the disposition of the continents, but reached the 30° latitudes during the Upper Carboniferous (Boucot et al., 2013). Critically, the Variscan orogen *sensu lato* was positioned close to the equator for 10,000 km from North America to North China (Fig. 1). Coal-bearing basins were therefore sited along the length of the orogen, to provide the opportunity for a global-scale concentration of gold and a large-scale opportunity for trace element resource exploration.

There is limited data available to compare the occurrences from coals in the Variscan orogen with those from elsewhere. To some extent it reflects the fact that the Upper Carboniferous deposits have dominated coal resources in the developed world, and that much of them have been in the American and European portions of the Variscan orogen. There is negligible data that could be related to other, younger, orogens. Of the Upper Carboniferous deposits, those occurrences of gold that are recorded are proximal to the orogen, not distal, which suggests that the orogen is a critical control. A relevant data set, for rare earth element contents in Carboniferous coals in the USA, records higher contents in the Appalachian Basin compared to basins further west (Taggart et al., 2016), implying that ash composition is related in some way to geographic context, i.e. provenance is an important factor in coal composition. Although the Variscan basement was most important as a source to metal enrichments in immediately younger sediments, in regions where much younger coal was still able to access the basement erosion products it could take up the metal. In Turkey, coal-bearing sediments of the Thrace Basin were deposited on Variscan granitic basement, and the coal contains gold and a range of other metals attributed to the basement (Erarslan and Örgün, 2017). This emphasizes the exceptional nature of the Variscan basement as a source to sedimentary reservoirs for metals.

6. Concluding remarks

The relationship between Carboniferous coal and gold is distinctive. It relies on the combination of a fertile orogen, deposition in foreland and other basins related to the orogen, and the palaeogeography/palaeoclimate that saw extensive coal deposition in those basins. That combination did not occur again. Younger coal-rich periods (Jurassic, Palaeogene) were less related to foreland basins, and so less related to orogenic belts, partly reflecting the distribution of orogens in other climatic belts (Diessel, 1992; Thomas, 2012).

In addition to gold, coal has been considered as a possible source for a wide range of elements including selenium, gallium, scandium and rare earths. As governments, coal authorities, waste management companies and others evaluate the potential of coal and its combustion products for these resources (U.S. Department of Energy, 2017; Dai and Finkelman, 2018), it is clear that it is important to understand the paleoclimatic and tectonic context of coal deposition. The Pennsylvanian coals forming in the shadow of the Variscan Orogen were ideally placed to act as reservoirs for the trace element bounty released during orogenic exhumation. The coal that facilitated the industrial revolution in Europe and America may yet have more to offer.

Declaration of Competing Interest

The author 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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Appendix A. Supplementary data

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