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The role of supermountain belts and climatic controls on the genesis of copper deposits in the Kupferschiefer and the Central African Copperbelt

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Abstract

Sedimentary rock-hosted stratiform copper deposits are the world's second largest source of copper and the largest source of cobalt, with about 73% of the copper occurring in two basins: the Katangan Basin (Central African Copperbelt) and the Permian Basin (Kupferschiefer). Why these two sedimentary basins are so highly endowed in copper is puzzling because sedimentary rock-hosted stratiform copper deposits have formed since the Paleoproterozoic and they all share remarkably similar ore mineralogy, host-rock characteristics and basin settings. We suggest that this discrepancy is due to the development of these two basins close to the bases of ~8000-km-long supermountain belts. The supermountain belts were instrumental in raising oxygen levels in Earth's atmosphere, as well as providing a voluminous source of groundwater and a powerful and long-lived driver for the fluid-flow system. The elevated oxygen levels facilitated the diagenetic processes that converted copper-bearing labile minerals to amorphous iron-oxides and smectite and then in turn to hematite and illite. When oxidized brines flushed through the basin successions, the liberated copper was transported to units containing carbon-rich mudstone and the metals were deposited. For the Katangan Basin, development of the Transgondwanan supermountain belt along its margins between about 525 and 510 Ma explains the delay of several hundreds of millions of years between basin formation and mineralization in the Central African Copperbelt. In contrast, development of the Mid-Pangean supermountain belt formed penecontemporaneous with the Permian Basin explains the similarity in timing between basin formation and mineralization in the Kupferschiefer.

Keywords Sedimentary rock-hosted stratiform copper deposits \cdot Supermountain belts \cdot Red beds \cdot Kupferschiefer \cdot Central African Copperbelt

Introduction

Sedimentary rock-hosted stratiform copper deposits are the world's second largest source of copper, the largest source of cobalt and the fourth largest source of silver, based on an analysis by Singer (2017) of total production, reserves and

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² School of Geography, Earth and Atmospheric Sciences, The University of Melbourne, Parkville, Victoria 3010, Australia resources at the lowest available cut-off grade, and ranked among all deposit types on Earth. Singer (2017) also showed that although porphyry copper deposits contain about 5.75 times more copper than sedimentary rock-hosted stratiform copper deposits, the average ore grades of the latter are typically much higher. Another notable feature of sedimentary rock-hosted stratiform copper deposits is that despite occurring in numerous geological provinces around the world, they share remarkably similar ore mineralogy, host-rock characteristics and basin settings, and are only found in successions younger than about 2.06 Ga (Kirkham 1989; Knoll and Holland 1995; Hitzman et al. 2005; Perelló et al. 2017). These similarities are intriguing, because of all currently known reserves, resources and past production of copper in sedimentary rock-hosted stratiform copper deposits, about 73% occur in just two districts: the Katangan Basin in central Africa (Central African Copperbelt) and the Permian Basin



Fig. 1 a Contained tons of copper for sedimentary rock-hosted stratiform copper deposits through time, based on total production, reserves and resources at the lowest available cut-off grade (Singer 2017, D. Singer, pers. comm., 2022). b Simplified evolution of atmospheric oxygen through time and major episodes of supercontinent growth. After Wallace et al. (2017), Campbell and Allen (2008), and Campbell and Squire (2010). c (2007) Peak pressure versus age for metamorphic rocks (modified after Campbell and Squire (2010) and Brown (2007))

in central Europe (Kupferschiefer) (Fig. 1a, D. Singer, pers. comm., 2022).

In an attempt to understand why the Central African Copperbelt and the Kupferschiefer are such important districts for sedimentary rock-hosted stratiform copper deposits, most previous reviews have focussed on the architecture and evolution of the host basins during sedimentation and early diagenesis (Hitzman et al. 2005; Selley et al. 2005; McGowan et al. 2006; Kampunzu et al. 2009; Borg et al. 2012). However, mineralization in the Katangan Basin occurred over prolonged episodes and up to several hundreds of millions of years after sedimentation (Hitzman et al. 2005; Selley et al. 2005; Sillitoe et al. 2017a; Saintilan et al. 2018). Furthermore, the deposits of the Central African Copperbelt and Kupferschiefer formed near the boundaries between the Proterozoic and Phanerozoic eons and the Palaeozoic and Mesozoic eras, respectively, when Earth experienced several extreme shifts in global environmental conditions (Hay et al. 2006; Campbell and Squire 2010; Sperling et al. 2021) (Figs. 1 and 2). Most notable, in the context of this paper, were the major increases in atmospheric O_2 abundances that occurred near the Proterozoic/Phanerozoic and Palaeozoic/Mesozoic boundaries (Canfield 2005; Wallace et al. 2017; Zhu et al. 2022).

The major deposits of the Katangan and Permian basins also formed near the base of the two largest-known subaerial mountain ranges in Earth history, having formed during major continent-continent collisions associated with the amalgamation of the supercontinents of Gondwana and Pangea, respectively (Fig. 3; Torsvik and Cocks 2004; Squire et al. 2006; Cawood and Buchan 2007). Although tectonic controls and climatic influences have previously been considered by a number of workers (Selley et al. 2005; Hitzman et al. 2010; Saintilan et al. 2018; Sillitoe et al. 2017a), these authors principally focussed on the role of orogenesis that directly impacted the Katangan and Permian Basins. In addition, Hitzman et al. (2010) attributed mineralization in the two basins to major episodes of glaciation that preceded the assumed ages of mineralization. In our study, we investigate the role that major episodes of mountain building may have played during mineralization in the Central African Copperbelt and the Kupferschiefer. Our results suggest that the deposits are closely associated with two of the largestknown mountain subaerial ranges in Earth's history: the Transgondwanan supermountain belt (Squire et al. 2006) for the Katangan Basin and the Mid-Pangean supermountain belt (Matte 2001; Torsvik and Cocks 2004) for the Permian Basin (Fig. 3).

Age, architecture and palaeotectonic setting

To understand why the Katangan and Permian basins contain so many metal-rich sedimentary rock-hosted stratiform copper deposits, we begin by briefly reviewing the age and characteristics of the host successions, the age and relative timing of mineralization, the palaeotectonic setting of each region and the evidence for major shifts in climatic conditions that may have influenced metallogenesis. Several workers have provided comprehensive reviews of the age and characteristics of the sedimentary successions of the Katangan and Permian basins and the age of mineralization within each (Glennie 1989b; Hitzman et al. 2005, 2010; Selley et al. 2005; McGowan et al. 2006; Kampunzu et al.

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Fig. 2 Simplified stratigraphy of the Permian and Katangan Basins and a summary of robust radiometric ages of mineralization for associated sedimentary rock-hosted stratiform copper deposits, based on a review by Sillitoe et al. (2017a)



PALAEOTECTONIC SETTING OF THE KATANGAN BASIN (CENTRAL AFRICAN COPPERBELT)

Fig.3 Simplified palaeogeographic setting of the Katangan and Permian basins. \mathbf{a} - \mathbf{c} East – West Gondwana convergence created the ~8000-km-long Transgondwanan supermountain belt (Squire et al. 2006), of which the portion adjacent to the Katangan Basin did

2009; Alderton et al. 2016; Sillitoe et al. 2017a). We present only a brief overview of these data and interpretations for each of the basins. Instead, our discussion emphasizes the palaeotectonic and palaeogeographic setting of the regions surrounding the Katangan and Permian basins, plus several of the major shifts in climatic conditions that occurred broadly coincident with mineralization.

not form until about 525–510 Ma. Modified after Squire et al. (2006). **d–e** Convergence between Gondwana and Laurentia-Baltica produced the ~8000-km-long Mid-Pangean supermountain belt. Modified after Matte (2001) and Torsvik and Cocks (2004)

The Katangan Basin

Age and stratigraphic architecture

The sedimentary successions of the Katangan Basin are generally interpreted as intracratonic rift-fill sequences deposited between about 880 and 727 million years ago (Ma) (Fig. 2; Hitzman et al. 2005; Selley et al. 2005; Kampunzu et al. 2009; Rooney et al. 2015; Mambwe et al. 2020). The oldest of the rift-fill successions are the Lower Roan Group, which contains a basal unit of coarse, continentally derived sedimentary units that are commonly referred to as redbed units and an overlying unit of marine sequences. The Upper Roan Group contains abundant evaporitic units that are in turn overlain by the upward-grading successions of carbonate and siliciclastic units of the Mwashia sequence. Overlying these units is the Nguba Group, which grades upward from coarse basal successions of the Grand Conglomerate through dolomite and limestone into sandstone and is itself overlain by the mudstone, siltstone, glacial diamictites and carbonate units of the Kundelungu Supergroup. In the context of this paper, the Lower Roan Group is the most important unit in the region because most of the major orebodies occur in dark mudstone and siltstone layers located near the top of the red bedded units or within the basal portion of the overlying marine sequences.

Prior to about 2015, most geochronological data for the age of mineralization from the Central African Copperbelt recorded a spread of results with generally large uncertainties (Cahen et al. 1961, 1971; Darnley et al. 1961; Meneghel 1981; Richards et al. 1988a, 1988b; Torrealday et al. 2000; Muchez et al. 2015). These data were used to support interpretations that mineralization in the Central African Copperbelt was a multi-stage process spanning several hundred million years, commencing with syn-diagenetic copper mineralization penecontemporaneous with deposition of the host sedimentary successions and ending with an epigenetic event broadly coincident with orogenesis and peak metamorphism (Brown 1997; Hitzman et al. 2005, 2010). Hitzman et al. (2010) suggested that the peak mineralization event occurred at about 700 Ma, which was coincident with the end of the Sturtian glaciation period; they posited that the glacial event may have played a crucial role in the formation of the ore deposits by increasing the Mg/Ca ratios and sulfate contents of the oceans and evaporites which precipitated from the seawater. The younger ages for mineralisation were attributed by Hitzman et al. (2010) to resetting of the ages or minor remobilization of the metal.

However, Sillitoe et al. (2017a) presented a detailed review of the geochronological and observational data for the Central African Copperbelt in which they questioned the evidence for a major syn-diagenetic event. Their review highlighted concerns about the robustness of Re-Os dates for bulk copper±cobalt sulfide samples (Selley et al. 2005; Muchez et al. 2015) and the lack of textural evidence and widespread copper depletion in likely diagenetic sulfides at microscopic, sample or orebody scales. Instead, they presented 27 Re-Os ages for paragenetically constrained molybdenite from 9 deposits and prospects across the Central African Copperbelt that they considered robust (Fig. 2, Sillitoe et al. 2015; 2017a). These data indicate that copper mineralization involved several protracted events, each lasting 10–24 million years, which occurred mainly between about 525 and 490 Ma. Although debate continues about the possibility of an earlier syn-diagenetic Cu event (Hitzman and Broughton 2017; Muchez et al. 2017; Sillitoe et al. 2017b), recent work by Saintilan et al. (2018) provided evidence for copper-cobalt mineralization at about 609 Ma (Fig. 2).

Palaeotectonic and palaeogeographic settings

In the hundreds of millions of years between (a) the onset of deposition of the Roan Group at about 880 Ma (Hoffmann et al. 2004; Kampunzu et al. 2009; Rooney et al. 2015; Prave et al. 2016) and (b) the youngest-known age of mineralization in the Central African Copperbelt (473 ± 4 Ma, Saintilan et al. 2018), the most pronounced change in palaeotectonic and palaeogeographic setting of the Katangan Basin was between about 525 and 510 Ma (Squire et al. 2006). Prior to about 525 Ma, the Katangan Basin was predominantly influenced by rift-margin mountains located to the east and southeast (present-day coordinates). The presence of phengite- and kyanite-bearing mafic eclogites with garnet wholerock Sm-Nd isochron ages at 595 ± 10 and 638 ± 16 Ma suggest that oceanic units were present to the south (present-day coordinates) of the Katangan Basin until at least the latter stages of the Proterozoic (John et al. 2003). Although the high metamorphic grades of these oceanic units indicate burial to depths of about 90 km (John et al. 2003) and the possibility of large associated mountains, subduction was probably directed away from the Katangan Basin (Saintilan et al. 2018).

Between about 650 and 500 Ma, convergence between the continental landmasses of East and West Gondwana produced the 8000-km-long Transgondwanan supermountain belt (Figs. 3a-c, Squire et al. 2006). Growth of the giant mountain range occurred in three main stages (650-590 Ma, 580-550 Ma and 525-510 Ma), of which the third stage formed just to the east (present-day coordinates) of the Katangan Basin (Fig. 3c). Evidence supporting the existence of the supermountain belt at this time includes garnetbearing gneisses in Malawi that were buried to depths of about 60-70 km between about 530 and 500 Ma, then rapidly exhumed (Ring et al. 2002). In addition, eclogites and high-pressure granulites were generated in southern India at about 535 Ma (Santosh and Sajeev 2006; Santosh et al. 2010). This evidence for deep roots to the mountain range implies high elevations (Campbell and Squire 2010). Therefore, broadly coincident with peak metamorphism, deformation and a major mineralising event in the Katangan Basin, Himalayan-style mountains developed within a few hundred kilometres of its eastern margin. Importantly, the large volume of sedimentary debris generated by erosion of the

Transgondwanan supermountain belt was mainly deposited in the remanent ocean basin that occurred between East and West Gondwana before its final dispersion along the proto-Pacific margin (Squire et al. 2006). As a result, sedimentary evidence of a vast mountain range proximal to Katangan Basin is limited.

In addition to the Transgondwanan supermountain belt, two other episodes of mountain building occurred proximal to the Katangan Basin between about 580 and 500 Ma (Campbell and Squire 2010). To the west (present-day coordinates), the roughly 4000-km-long and 400-km-wide Adamastor Orogen formed between (a) the combined Amazon and Rio de la Plata Cratons of South America and (b) the Congo-Tanzanian and Kalahari Cratons of Africa (Cawood and Buchan 2007). Although the sparse exposures of these rocks indicate that continent-continent convergence occurred between about 590 and 550 Ma (Frimmel and Frank 1998; Pedrosa-Soares et al. 2001; Heilbron et al. 2004; Cawood and Buchan 2007), the relatively low metamorphic grades in the orogen and structural lineation fabrics are indicative of oblique convergence (see Goscombe et al. 2005; Goscombe and Gray 2008). Therefore, any mountain range along the western margin of the Katangan Basin was probably restricted in height compared to those of the Transgondwanan supermountain belt. The only other major mountain range in the region was probably associated with the roughly 2500-km-long and 750-km-wide Damara-Lufilian-Zambezi Orogen that formed during the collision between the Kalahari and Congo-Tanzania Cratons in southern Africa at about 530 Ma (John et al. 2004). It was during this collisional event that the successions hosting the deposits of the Central African Copperbelt reached peak metamorphic conditions and were strongly deformed. This means growth of the Trangondwanan supermountain belt was the dominant palaeotectonic and palaeogeographic event to influence the Katangan Basin between about 580 and 500 Ma.

Palaeoenvironmental conditions

Evolution of the Katangan Basin occurred broadly coincident with several pronounced shifts in palaeoenvironmental conditions. These included extreme glaciation events (Snowball Earth), during which Earth's surface was almost entirely frozen (Harland 1964; Kirschvink 1992). Snowball Earth events that coincided with evolution of the Katangan Basin were the 717–660 Ma Sturtian, the 639–635 Ma Marinoan and the 580 Ma Gaskiers glaciations (Hoffman et al. 1998; Hoffmann et al. 2004; Condon et al. 2005; Calver et al. 2013; Rooney et al. 2015; Prave et al. 2016). In addition, atmospheric and ocean oxygen levels rose sharply near the end of the Proterozoic eon (Holland 2002; Berner 2006), although the timing and magnitude of oxygenation is debated (Li et al.

2017; Wallace et al. 2017; Sperling et al. 2021; Qin et al. 2022).

Early interpretations of the rise of oxygen levels in Earth's atmosphere and oceans posited two prominent increases near the start and end of the Proterozoic eon (Holland 2002; Berner 2006). However, recent geochemical studies combined with an improved understanding of the timing of major ecological events associated with the rise of animals suggest that oxygenation near the Proterozoic-Phanerozoic boundary was probably protracted and irregular. For instance, rare-earth-element patterns in marine carbonates (Wallace et al. 2017) and Mo isotope values from black mudstone units (Qin et al. 2022) indicate that oceanic oxygen levels rose sharply from about 635 Ma, reaching moderate oxygen levels (compared to today) by the end of the Proterozoic eon. Ecological events supporting this pronounced rise in oxygenation include the transition from cyanobacteria to much larger and more complex eukaryotic algae in the ocean from about 635 Ma (Brocks et al. 2017), the emergence of architecturally complex organisms from about 575 Ma (Waggoner 2003; Canfield et al. 2007; Droser and Gehling 2015), and evidence of these organisms using energy- and oxygen-intensive processes to actively move across the seafloor from about 560 Ma (Droser et al. 2017).

Molybdenum isotopic data were initially used to suggest that oxygen levels continued to rise in the early Cambrian period, peaking at modern-day levels about 520 Ma (Wen et al. 2011; Chen et al. 2015). However, Qin et al. (2022) showed that in strongly euxinic conditions, intense scavenging of Mo can generate Mo-isotope values comparable to modern well-oxygenated oceans. This interpretation of lower oceanic oxygen levels during the early Cambrian period is consistent with sparse rare-earth-element data from marine carbonates (Wallace et al. 2017). But, Li et al. (2017) used Fe trace-element data to show that oxygen levels rose once again to moderate levels (compared to today) during Cambrian Stages 3 and 4 (about 521 and 509 Ma). The timing of these interpreted oscillations in early Cambrian oxygen levels also match closely the sudden extinction of Ediacaran biota about 541 Ma (Waggoner 2003; Droser et al. 2017), a prolonged interval of relatively sparse fossil data from small organisms from about 541 to 525 Ma (Landing 1994; Budd and Jackson 2016; Daley et al. 2018), and the Cambrian Explosion between about 525 and 505 Ma that included the appearance of animals with high oxygen demands, such as arthropods and echinoderms (Knoll and Carroll 1999).

A possible trigger for the punctuated rise in oxygen levels between about 635 and 509 Ma was a major episode of mountain building (Transgondwanan supermountain belt) associated with amalgamation of the supercontinent Gondwana (Squire et al. 2006; Zhu et al. 2022). According to Campbell and Squire (2010), the rise in oxygen production was triggered by an influx of nutrients into Earth's oceans

from the large and rapidly eroding mountain ranges. The algal blooms generated by this nutrient surge led in-turn to high levels of photosynthesis and thus oxygen production. Much of that oxygen was able to accumulate in Earth's oceans and atmosphere because the high depositional rates accompanying erosion of the vast mountain ranges (Squire et al. 2006) enabled large amounts of organic carbon to be stored among the voluminous sedimentary rocks being deposited at this time. Importantly, the timing of three main orogenic events associated with growth of the Transgondwanan supermountain belt (650–590 Ma, 580–550 Ma and 525–510 Ma) coincide closely with the punctuated rises in oceanic oxygen levels near the Proterozoic-Phanerozoic boundary (Li et al. 2017; Wallace et al. 2017; Qin et al. 2022).

The Permian Basin

Age and stratigraphic architecture

The stratigraphy of the Permian Basin is best understood in its southern parts around southwestern Poland and southern Germany where several major sedimentary rock-hosted stratiform copper deposits are located (Fig. 2a, Hitzman et al. 2005). There, the basement rocks, which include deformed Carboniferous coal deposits, are overlain by the Permian Rotliegendes and Zechstein successions. The Rotliegendes successions are divided into two units separated by an unconformity: the Lower and Upper Rotliegendes (Glennie 1998). The Lower Rotliegendes were deposited between about 300 and 290 Ma, vary laterally in thickness and are composed of continentally derived redbed units and bimodal volcanic rocks. The Upper Rotliegendes, which are > 1000-m-thick and dominated by redbed successions, may be distinguished from the underlying units by the abundance of fluvial-alluvial successions, consisting of sandstone and conglomerate units plus aeolian sandstone and sabkha sandstone, siltstone and mudstone facies. The unconformably overlying Zechstein were deposited between about 260 and 250 Ma and record a major transgression of seawater into the basin (Symons et al. 2011). The Zechstein consists of five depositional cycles that comprise halite, anhydrite, dolomite and mudstone. The Kupferschiefer, which is a thin (<2 m) bituminous unit of black mudstone that occurs at the base of the Zechstein, is the major host for the sedimentary rock-hosted stratiform copper deposits in the Permian Basin (Borg et al. 2012).

The age and timing of copper-sulfide mineralization in the Permian Basin is controversial. Possible evidence for the early commencement of mineralization include palaeomagnetic data from ore-related hematite alteration assemblages, obtained by Jowett et al. (1987) and recalculated by Symons et al. (2011) using a more recent apparent polar wander path to give an age of 254 ± 6 Ma. This early age is supported by Re-Os data of Alderton et al. (2016) on single types of ore-related sulfides from the Lubin Mine in Poland: 252 ± 1.7 Ma for bornite, 245.2 ± 1.6 Ma for chalcopyrite and 242.0 ± 1.6 Ma for pyrite. However, Sillitoe et al. (2017a) questioned the robustness of poorly documented data involving bulk samples and mixed assemblages for age determinations (Michalik 1997; Bechtel et al. 1999; Patzold et al. 2002; Pašava et al. 2007; Maliszewska and Kuberska 2009; Mikulski and Stein 2015; Alderton et al. 2016). Although Re-Os data do not preclude younger mineralization ages in the Kupferschiefer, additional well-documented age determinations are needed on single ore minerals from across the Permian Basin to properly constrain the length of the main mineralizing event.

Palaeotectonic and palaeogeographic settings

The Permian Basin was located near the base of the 8000-km-long Mid-Pangean supermountain belt (Fig. 3d-e). The Mid-Pangean supermountain belt developed when the continental landmasses of Gondwana and Laurentia-Baltica (plus several microplates) collided diachronously between about 480 and 250 Ma (Matte 2001; Torsvik and Cocks 2004). Climate models estimate that, at about 255 Ma, the section of the Mid-Pangean supermountain belt bounding the Permian Basin near southwestern Poland and southern Germany (present-day locations) had a mean altitude of at least 2000 m (Fluteau et al. 2001). Importantly, both the Rotliegendes and Zechstein were deposited in the continental interior of the foreland associated with the Variscan segment of the Mid-Pangean supermountain belt. Furthermore, the thick and extensive evaporite deposits of the Zechstein were deposited when seawater entered the expansive sub-sea level depression associated with, and located next to, the tall mountain range (Glennie 1989b; Warren 2010).

Palaeoenvironmental conditions

Oxygen levels in Earth's atmosphere and oceans were close to present-day values during development of the Permian Basin (Fig. 1b; Berner 2006; Wallace et al. 2017). As Campbell and Squire (2010) and Zhu et al. (2022) argued, elevated levels of oxygenation between about 350 and 250 Ma were strongly influenced by erosion and sedimentation associated with the diachronous growth of the Mid-Pangean supermountain belt. Broadly similar to the Proterozoic-Phanerozoic boundary, growth and erosion of the supermountains generated large algal blooms that in-turn drove high levels of photosynthesis and thus oxygen production. From about 400 Ma, large land plants and forests were important contributors to oxygen production (Wallace et al. 2017).

KATANGAN BASIN (CENTRAL AFRICAN COPPERBELT)

a ~877-635 Ma: Deposition of the Roan Group, 'ore shales' and overlying successions. Onset of early diagenesis



b ~635-525 Ma: Rapid increase in atmospheric oxygen levels, an acceleration of diagensis with extensive redbed formation and minor Cu mineralization



c ~525-510 Ma: Growth of nearby Transgondwanan supermountain belt triggers an influx of oxidized fluids and an acceleration of mineralization, which continues until about 473 Ma



◄Fig. 4 Schematic diagram showing the role played by the Transgondwanan supermountain belt in the genesis of the Katangan Basin and the Central African Copperbelt. a Deposition of the Roan Group, ore shale units and overlying successions commenced about 877 Ma. Low oxygen levels and modest volumes of groundwater prior to about 635 Ma led to a prolonged episode of early diagenesis and formation of smectite and amorphous iron oxides. b Late diagenesis commenced when oxygen levels increased sharply from about 635 Ma, leading to widespread formation of redbed units and minor Cu mineralization from about 509 Ma. c Growth of the Transgondwanan supermountain belt adjacent to the Katangan Basin from about 525 Ma led to a major influx of oxidized groundwater that remobilized the metals and deposited them in spatially restricted packages of reduced rocks, pre-existing sulfides or hydrocarbons. This major major influx of groundwater, and thus Cu mineralization, lasted until about 473 Ma

Palaeoclimatic conditions also played a crucial role in developing the sandy Rotliegendes successions and the Zechstein evaporite (Warren 2010). Because rising mountain ranges create large barriers to atmospheric circulation systems, they can modify jet streams and accentuate aridity in particular locations (Galy and France-Lanord 2001; Kendell et al. 2003; Lal et al. 2004; Warren 2010). For example, the rain-barrier effect was responsible for development of the Taklamakan desert, the second-largest sand desert in the world, following uplift of the Kunlun Mountains in northwestern China at about 5.3 Ma (Sun et al. 2008). By analogy, Warren (2010) argued that the sandy, hot, and arid deserts that generated the Rotliegendes were developed in arid to semi-arid conditions on the leeward side of the Variscan segment of the Mid-Pangean supermountain belt. Furthermore, the location of the Variscan component of the Mid-Pangean supermountain belt at latitudes between about 15 and 35° north of the equator provided the ideal hot and arid climatic window for evaporite formation (Kirkham 1989) once seawater filled the relatively shallow intra-cratonic sub-sea level foreland basin (Glennie 1989b).

Metal sources

The ability of basinal fluids to transport Cu, and other important metals, over a range of temperatures and pressures (Gregory et al. 2008; Hitzman et al. 2010; Champion et al. 2020) means the key metal sources for sedimentary rock-hosted stratiform copper deposits may include a mix of material up to a few hundreds of kilometers away. One potential metal source is the underlying volcanic and basement rocks (Koziy et al. 2009; Hitzman et al. 2010). Koziy et al. (2009) argued that the metals in the Zambian copper belt deposits were derived from basement rocks when oxidizing groundwater interacted with evaporites and as a result became saline and dense and were thus able to descend to depths of up to 12 km. There, the fluids were able to leach Cu from the volcanic basement before slowly heating up and thereby becoming less dense and able to ascend to higher levels where they formed Cu deposits in reducing environments. A variant of this hypothesis was proposed by Saintilan et al. (2018) for the Central African Copperbelt deposits, in which multi-stage hydrothermal processes remobilized metals from older deposits in the basement successions.

An alternate metal source is the precursor sedimentary successions of the redbed units that are associated with the sedimentary rock-hosted stratiform copper deposits (Rose 1976; Boyle et al. 1989; Brown 2009; Borg et al. 2012). These immature continental successions generally contain high abundances (>25%) of copper-bearing labile minerals, such as plagioclase, pyroxene and hornblende, that have copper contents of 120, 78 and 62 ppm, respectively (Walker 1989; Metcalfe et al. 1994). During the early stages of diagenesis of these units, we posit that oxidized groundwater interacted with the copper-bearing lithic fragments and mineral grains to create both alkaline, due to hydrolysis of the primary silicates, and reducing, due to oxidation of the ferrous iron in the silicates, conditions in environments in which a limited amount of water was available (Fig. 4). These reduced and alkaline conditions rendered the fluids incapable of leaching and transporting any copper liberated from the mafic minerals during their replacement by secondary minerals (Wood and Normand 2008). The iron in the mafic minerals formed stable amorphous or poorly crystallized iron oxides and oxyhydroxides while the plagioclase and other silicates in the sedimentary units were replaced by secondary smectite (Walker 1989).

Because the hydrolysis of plagioclase and other silicates involves the consumption of H⁺, conditions would have remained alkaline, or near-neutral, until all of these labile minerals were dissolved. Any metals, such as copper, that were released from the labile minerals would be adsorbed at moderate to high pH, either by the poorly crystallized Feoxides and/or by the smectite (Walker 1989; Zielinski et al. 1983). However, once the ferrous iron in the metastable ironbearing silicates had been oxidized, the incoming groundwater would remain oxidizing (Benjamin and Leckie 1981; Rose and Bianchi-Mosquera 1993). Importantly, elevated levels of atmospheric oxygen from about 635 Ma (Brocks et al. 2017; Wallace et al. 2017) enabled the process to operate faster and more efficiently.

With time, the sedimentary rocks underwent advanced diagenetic changes during which the amorphous Fe-oxides are replaced by hematite and the smectite by illite. A major factor in controlling the conversion of limonite and goethite to hematite is decrease in the pH of the fluids (Williams-Jones and Vasyukova 2018). This will occur once the primary minerals were hydrolysed. The formation of the hematite and illite results in dissolution of copper by chloride-rich

brines under near-neutral pH and moderately oxidizing conditions (Brown 2009). However, if little fluid is available, the copper remains immobile but will be leached if there is an increase in flow of oxidized fluids.

Metal transport

Copper in the redbed units was most likely leached and mobilised by chloride-rich brines under near-neutral pH and moderately oxidizing conditions (Brown 1971; 2005; 2009; Rose 1976; 1989). Groundwater water is ideal for this process because, although initially highly oxidizing and mildly acidic, it becomes moderately oxidizing because of the reddening process (Brown 2005). Also, when groundwaters intersect thick packages of evaporites, which are commonly associated with redbed units (Davidson 1965; Glennie 1989a; Kirkham 1989), dissolution causes the fluids to become saline and thus contain the appropriate chloride ligands to form soluble copper chloride complexes in lowtemperature fluids (Haynes 1986; Kirkham 1989; Jackson et al. 2003). Evidence supporting evaporite dissolution and brine advection in the Central African Copperbelt includes gypsum and anhydrite pseudomorphs, remnant sabkha facies, collapse breccias and stratigraphic gaps in the Roan Group (De Magnée and François 1988; Cailteux et al. 1994; Jackson et al. 2003), and propylitic alteration on the edges of evaporite megabreccias (Jackson et al. 2003; Kampunzu et al. 2009).

Because groundwater is strongly oxidizing (Brown 2009; Rose 1976, 1989), it would not transport the copper because the oxidizing conditions would prompt the formation of cupric chloride complexes, which have much lower solubilities than cuprous chloride complexes (Rose 1976). In addition, high O₂ levels at a pH>5 will result in the formation of atacamite ((Cu₂Cl(OH)₃). However, as suggested by Brown (2005), the redox state of groundwater entering an aquifer in a highland area will decrease if the fluids penetrate deep into pre-cursor redbed successions and interact with mafic minerals within those sedimentary rocks.

Supermountain belts and metallogenesis

We argue that the unusually large metal endowment of sedimentary rock-hosted stratiform copper deposits in the Central African Copperbelt and the Kupferschiefer was strongly influenced by supermountain belts that formed during amalgamation of the supercontinents of Gondwana and Pangea, respectively (Figs. 3, 4, 5). Our interpretation is consistent with the ages of copper-sulfide mineralization by Sillitoe et al. (2017a) and Saintilan et al. (2018) for the Central African Copperbelt and by Alderton et al. (2016) for the Kupferschiefer (Fig. 2). Also, our suggestion of gravitydriven fluid flow associated with major orogenic uplift is similar to models proposed for the genesis of Mississippi Valley Type (MVT) deposits (Bradley and Leach 2003; Garven 1985, 1995; Leach et al. 2010). However, we also argue that growth and erosion of the associated supermountain belts elevated the oxygen contents of not only Earth's atmosphere and oceans (Campbell and Squire 2010; Zhu et al. 2022), but also that of groundwater. Therefore, where the foothills of the supermountain belts developed proximal to the margins of the Katangan and Permian Basins, they provided the large and long-lived supplies of the oxidized groundwater that were necessary to generate the numerous large sedimentary rock-hosted stratiform copper deposits of the Central African Copperbelt and the Kupferschiefer.

The Central African Copperbelt

The metallogenic processes that formed the sedimentary rock-hosted stratiform copper deposits in the Katangan Basin were initially very slow. Prior to about 635 Ma, low levels of oxygen in the atmosphere and groundwater resulted in slow rates of conversion of labile minerals to iron oxides and smectite in the redbed units of the Lower Roan Group (Fig. 4a). In addition, the reaction of weakly oxidizing groundwater with labile minerals initially created conditions that were largely alkaline and reducing, such that most of the metals released by the reaction were incorporated at moderate to high pH by iron oxides or smectite. However, when oxygen levels started to rise from about 635 Ma (Brocks et al. 2017; Wallace et al. 2017), the rate of this diagenetic process also increased. With time, large amount of ferrous iron in iron-bearing silicates was converted into ferric iron in oxides and the incoming groundwater remained oxidizing. When the oxidizing and mildly acidic groundwater passed through the thick evaporite units and became saline (Davidson 1965; Glennie 1989a; Kirkham 1989), it was able to dissolve large quantities of copper, and other metals, from the surfaces of hematite and clay particles. We suggest that widespread formation of sedimentary rock-hosted stratiform copper deposits commenced in the Katangan Basin from about 635 Ma (Saintilan et al. 2018), although the initial rate of metallogenesis was probably restricted by the low volumes of groundwater (Fig. 4b).

When the Transgondwanan supermountain belt formed along the eastern margin of the Katangan Basin from about 525 Ma (Fig. 3c), the metallogenic processes accelerated, providing the Katanga Basin with a large source of groundwater and hydraulic head that drove the basinal fluid-flow system (Fig. 4c). The new groundwater system overwhelmed the pre-existing one, causing a marked increase in the volume and flow rate of highly saline, oxidized and mildly acidic fluids passing through **a** ~300-250 Ma: Deposition of the Rotliegendes, Kupferschiefer and Zechstein sequences. Rain-shadow effect over the Permian Basin inhibits groundwater influx and 'redbed' formation



b ~250 Ma: Mountains migrate into higher latitudes, triggering high rainfall in the Mid-Pangean supermountain belt. Influx of groundwater accelerates diagensis, redbed formation), and subsequent Cu mineralization



Fig. 5 Schematic diagram showing the role of the Mid-Pangean supermountain belt in the genesis of the Permian Basin and Kupferschiefer deposits. **a** Deposition of the Rotliegendes commenced about 300 Ma. Although oxygen levels were high, the Permian Basin was initially in the rain shadow of the Mid-Pangean supermountain belt, as suggested by formation of the evaporite units of the Zechstein. This limited groundwater supply and restricted redbed formation. **b** Migration of the Mid-Pangean supermountain belt led to a weakening of the rain shadow and an influx of oxidized groundwater into the Permian Basin the Lower Roan Group. Although orogenesis associated with the convergence of East and West Gondwana ceased about 510 Ma, the orogenic relief of the Transgondwanan supermountain belt remained for many tens of millions of years and continued to influence the hydrogeology and metallogenesis of the Katangan Basin until at least 473 Ma (Fig. 2; Sillitoe et al. 2015, 2017a). Furthermore, the size and strength of the fluid-flow system generated by the supermountain belt and its associated fracture network meant that some metals were likely leached by oxidized fluids that penetrated deep into the volcanic basement (Koziy et al. 2009; Saintilan et al. 2018). There, the oxidized fluids leached Cu from the volcanic basement before slowly heating up and thus becoming less dense and able to ascend to higher stratigraphic levels where they intersected reduced packages of rocks and formed the sedimentary rock-hosted stratiform copper deposits (Fig. 4c).

The Kupferschiefer

In marked contrast to the Katangan Basin, the diagenetic processes that led to redbed formation in the Permian Basin commenced very early in the history of basin evolution (Fig. 5a). The early onset of diagenesis was aided by the much higher oxygen levels in Earth's atmosphere and groundwater during basin formation. This was due to a rise in oxygen levels associated with the appearance of large vascular land plants in the late Devonian period and the flourishing of terrestrial flora in the Carboniferous period (Edwards et al. 2015; Wallace et al. 2017), plus increased carbon burial associated with growth of the Mid-Pangean supermountain belt (Campbell and Squire 2010; Zhu et al. 2022). However, because the Permian Basin was located in the rain shadow of the Mid-Pangean supermountain belt during deposition of the Zechstein evaporite sequence (Warren 2010), we posit that groundwater volumes were probably limited and thus restricted the rate of the diagenetic processes that formed the redbed units (Fig. 5a).

When the Mid-Pangean supermountain belt drifted north (Fig. 3d,e) and the rain shadow weakened, a large and long-lived supply of groundwater entered the Permian Basin (Fig. 5b). Also, when the oxidizing groundwater passed through the redbed units of the nascent Permian Basin, the copper-bearing labile minerals of the Lower Rotliegendes were converted to iron oxides and smectite. Subsequently, advanced diagenesis led to the conversion of amorphous iron oxides to hematite and of smectites by illite (Walker 1989). Therefore, widespread formation of sedimentary rock-hosted stratiform copper deposits commenced soon after 250 Ma (Fig. 5b) and probably continued for several tens of millions of years while the Mid-Pangean supermountain belt continued to drive the groundwater system of the Permian Basin.

Conclusions and implications for exploration

Four key factors are crucial for the genesis of sedimentary rock-hosted stratiform copper deposits (Hitzman et al. 2005, 2010; Selley et al. 2005; Large et al. 2019). First, a basinal fluid capable of transporting copper at low temperature, such as acidic and moderately oxidized groundwater. Second, a voluminous package of rocks that are rich in copper-bearing labile minerals (e.g., redbed successions). Third, a source of evaporite that generated high salinity levels in basinal fluids. And fourth, a mudstone rich in organic matter, or mobile hydrocarbons, that acted as a reductant to facilitate deposition of copper and other metals. However, to produce the large number of copperrich deposits that are found in the Katangan and Permian Basins, we posit a fifth key factor: a large intra-continental mountain range near the basin margin.

The supermountain belts that formed proximal to the Katangan and Permian Basins (the Transgondwanan and Mid-Pangean supermountain belts, respectively; Fig. 3) helped drive up oxygen levels and provided both a voluminous source of groundwater and a long-lived driver for the fluid-flow systems. For the Permian Basin, the Mid-Pangean supermountain belt formed penecontemporaneous with mineralization (Fig. 5). In contrast, the Transgondwanan supermountain belt formed several hundreds of millions of years after deposition of the host successions (Fig. 4).

These conclusions have important implications for the exploration of sedimentary rock-hosted stratiform copper deposits. In particular, the close temporal and spatial association between the formation of major sedimentary rock-hosted stratiform copper deposits and major mountain ranges means that sedimentary basins spatially associated with supercontinent-related collision zones are ideal targets for exploration. Although this association is similar to the one identified for MVT deposits (Garven 1985, 1995; Bradley and Leach 2003; Leach et al. 2010), mountain building must also coincide with an episode of increased oxygen levels in the atmosphere, as well as increased groundwater flow and the presence of evaporites. A useful proxy for major episodes of mountain building that coincide with sharp increases in oxygen production since the Great Oxidation Event about 2.4 Ga (Lyons et al. 2014) are the peaks of U-Pb detrital zircon ages in sedimentary material from 40 of the largest rivers in the world (Campbell and Allen 2008). These peaks in zircon ages coincide with the major growth phases of the supercontinents: about 1.95-1.6 Ga for Nuna/Columbia; about 1.25-0.95 Ga for Rodinia; 650-400 Ma for Gondwana; and 350-225 Ma for Pangea.

Evidence supporting our hypothesis is provided by the large Udokan deposit in Russia, which formed close to the Nuna/ Columbia supermountain belt about 1.9 Ga (Perelló et al. 2017) and White Pine in the USA, which formed close to the Rodinian, or Grenville, supermountain belt about 1.1 Ga (Mauk et al. 1992; Woodruff et al. 2020). A lower priority would be sedimentary basins that occur proximal to major mountain ranges during episodes of high oxygen levels, such as parts of the Alpine-Himalayan orogen since about 225 Ma (Le Fort 1975; Mahmoodi et al. 2018).

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