

Emergence of the aerobic biosphere during the Archean-Proterozoic transition: Challenges of future research

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ABSTRACT

The earth system experienced a series of fundamental upheavals throughout the Archean-Paleoproterozoic transition (ca. 2500–2000 Ma). Most important were the establishment of an oxygen-rich atmosphere and the emergence of an aerobic biosphere. Fennoscandia provides a fairly complete record of the hallmark events of that transition: widespread igneous activity, its association with a possible upper-mantle oxidizing event, the global Huronian glaciation, a rise in atmospheric oxygen, the protracted and large-magnitude Lomagundi-Jatuli carbon isotope excursion, a substantial increase in the sea-water sulfate reservoir, changes in the sulfur and phosphorus cycles, a radical modification in recycling of organic matter, and the Shunga Event—the accumulation of unprecedented organic-matter-rich sediments and the oldest known inferred generation of significant petroleum. Current research efforts are focused on providing an accurate temporal framework for these events and linking them into a coherent story of earth system evolution.

INTRODUCTION

The Archean (3800 Ma–2500 Ma) earth system functioned under an oxygen-poor atmosphere, and although oxygen-rich habitats undoubtedly existed once cyanobacteria evolved and began producing oxygen (>2700 Ma; Summons et al., 1999; Brocks et al., 1999), these were restricted to microbial mats or perhaps ephemeral oxygen oases in the surface ocean or in lakes. During this time and the initial 500 m.y.

of the Proterozoic, Earth's lithosphere underwent the assembly of two supercontinents: one, Kenorland, comprising the Laurentian, Fennoscandian, and Siberian shields; the other, the amalgamated Zimbabwe, Kaapvaal, Pilbara, Sao Francisco, and Indian cratons (Aspler and Chiarenzelli, 1998; Williams et al., 1991). Subsequent breakup involved the emplacement of voluminous continental flood basalts, giant radiating dike swarms and layered gabbro-norite intrusions (e.g., Heaman, 1997; Vogel et al., 1998), and deposition of banded iron formations (Pickard, 2003). Profound climatic and biogeochemical events also were occurring, such as icehouse conditions (Young et al., 2001; Evans, 2003), the rise in atmospheric oxygen levels (Bekker et al., 2004), and the largest and longest duration positive excursion of carbon isotopic composition of sedimentary carbonates (Lomagundi-Jatuli Event), which predated deposition of anomalously organic carbon-rich sediments forming giant petroleum fields (Shunga Event; both events are reviewed in Melezhik et al., 1999a, 1999b). It was out of this period of geological upheaval that the aerobic earth system began to emerge, with biogeochemical cycles in the oceans and on land using the highly energetic aerobic pathway. Note though that some workers suggest that deep marine basins and perhaps the global deep ocean itself were euxinic (rich in H₂S) for much of the ensuing Proterozoic (Canfield, 1998; Anbar and Knoll, 2002; Poulton et al., 2004).

Here we present a synthesis of the hallmark features of the Archean-Paleoproterozoic transition in the Fennoscandian Shield (Fig. 1). It is representative of other shield areas and provides important insights into the biogeochemical evolution of the earth system.

Early Paleoproterozoic Environmental Upheavals: Insight from the Fennoscandian Shield

The Fennoscandian Shield comprises Archean, Paleoproterozoic, and Mesoproterozoic domains (Fig. 1). In particular, its eastern part is a composite of Late Archean granite-greenstone belts, Paleoproterozoic rifts, the ca. 1900 Ma Kola Orogen, and multiphase Svecofennian orogens. Paleoproterozoic sedimentary and volcanic successions reach a cumulative thickness of more than 20 km and contain a rich record of nearly 700 m.y. of earth history. Two of the best-explored and best-mapped areas are the Pechenga greenstone belt and the Omega Basin (Fig. 1); both contain exceptionally well-developed and preserved volcano-sedimentary successions (Fig. 2) that span >2505–1970 Ma and record intracratonic rifting, global glaciation (Fig. 3A–3D), and other profound alterations of Earth's surface environments. The research that has been conducted in the past on these rocks provides a series of fascinating vignettes of geologic events, but age control is poor, based largely on whole-rock Rb-Sr ages, and the linkages between these events are not well understood. The challenge for the future is to correlate Fennoscandian events with those recorded on other continents and to weave these observations into a coherent story of the emergence of the aerobic biosphere.

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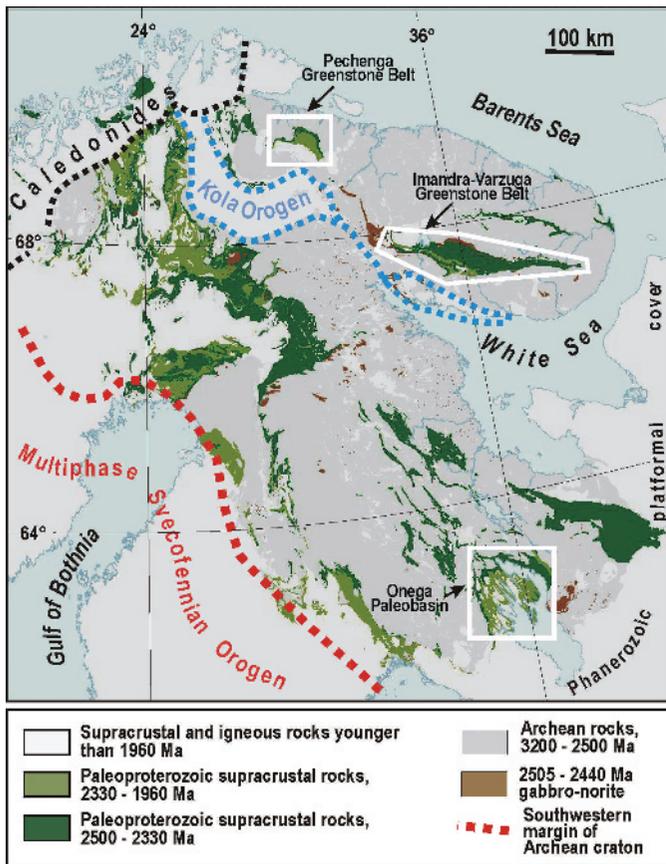


Figure 1. Simplified geological map of the Fennoscandian Shield.

Huronian Glaciation

The first significant environmental event known from the Archean-Paleoproterozoic transition is the seemingly rapid onset of global glaciations from otherwise climatically invariant conditions (Marmo and Ojakangas, 1984). The glaciation is known as the Huronian (after the eponymous Supergroup in Canada) and occurred at 2450–2220 Ma (Young et al., 2001). Some of the best age constraints come from South Africa where 2316 ± 7 Ma glacial diamictites (Hannah et al., 2004) rest unconformably on a variety of sedimentary rocks and banded iron formations (Bekker et al., 2001). Glaciogenic deposits of the Fennoscandian Shield are associated with the Sarioli Group and its equivalents (Marmo and Ojakangas, 1984) and consist of polymict conglomerates, sandstones, diamictites, and varve-like sedimentary rocks with dropstones (Fig. 3A and 3B).

The genesis of this global glaciation(s) remains poorly understood (Evans, 2003). Pavlov and Kasting (2002) linked the event to the collapse of an Archean methane-supported greenhouse effect driven by the rise of atmospheric oxygen (see below). Data from the Fennoscandian Shield and elsewhere suggest an alternative hypothesis involving a network of feedback loops as causative factors. At ca. 2500 Ma, mantle plume-driven continental uplifts emplaced voluminous continental flood-basalts (Heaman, 1997) in low latitudes (Mertanen et al., 1999). This global igneous event has otherwise been linked to mantle redox evolution and the rise of oxygen (Kump et al., 2001; see next section), but the weath-

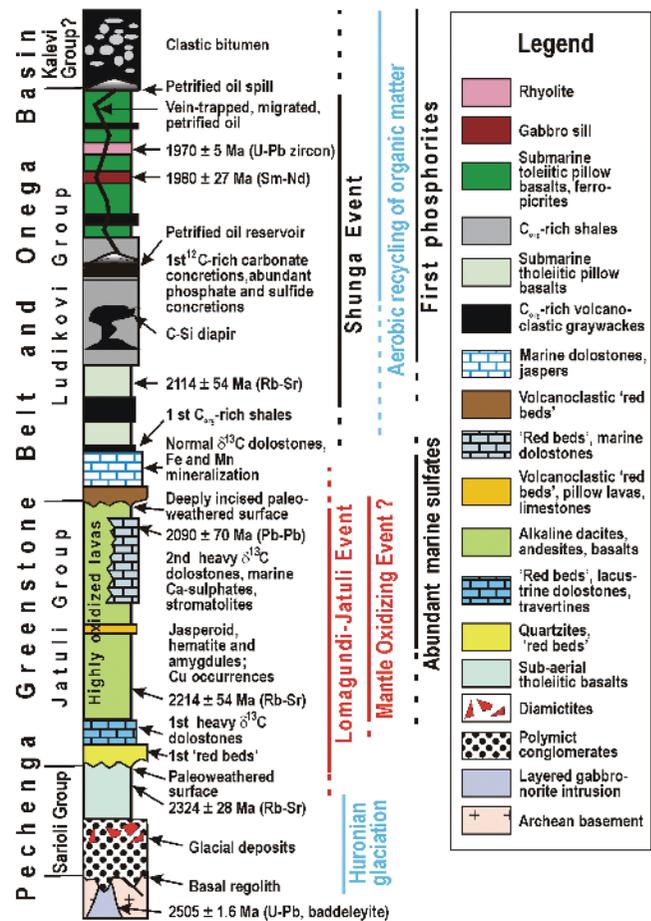


Figure 2. Composite (~10,000-m-thick) Paleoproterozoic section for the Pechenga Greenstone Belt–Omega Basin linked to major environmental events. Radiometric dates are from Hanski (1992), Amelin et al. (1995), and Balashov (1996).

ering of the basalts could have driven the climate system into glaciation without these other feedbacks. The basalt flows subsequently were dissected by rifting, producing major topographic features and drainage patterns in all likelihood similar to those of younger continental flood-basalt provinces (Cox, 1989). This would have enabled deep, subaerial weathering lasting over tens of m.y. of extensive fresh basalt-covered areas and consequent intense consumption of atmospheric CO₂ (Taylor and Lasaga, 1999; Dessert et al., 2001). Low ⁸⁷Sr/⁸⁶Sr ratios for this time interval (Bekker et al., 2003b) add credence to this scenario. Progressively diminishing volcanic CO₂ emissions and a constant burial ratio of inorganic/organic carbon would have reduced atmospheric CO₂ even further, all contributing to global cooling and the onset of icehouse conditions.

The Great Oxidation Event (GOE)

Interestingly, the igneous event in the earliest Paleoproterozoic may have been directly responsible for the rise in atmospheric oxygen content that occurred at this time. S isotope data showing the disappearance of mass independent signatures in sedimentary sulfides provide compelling evidence for a substantial increase in atmospheric oxygen

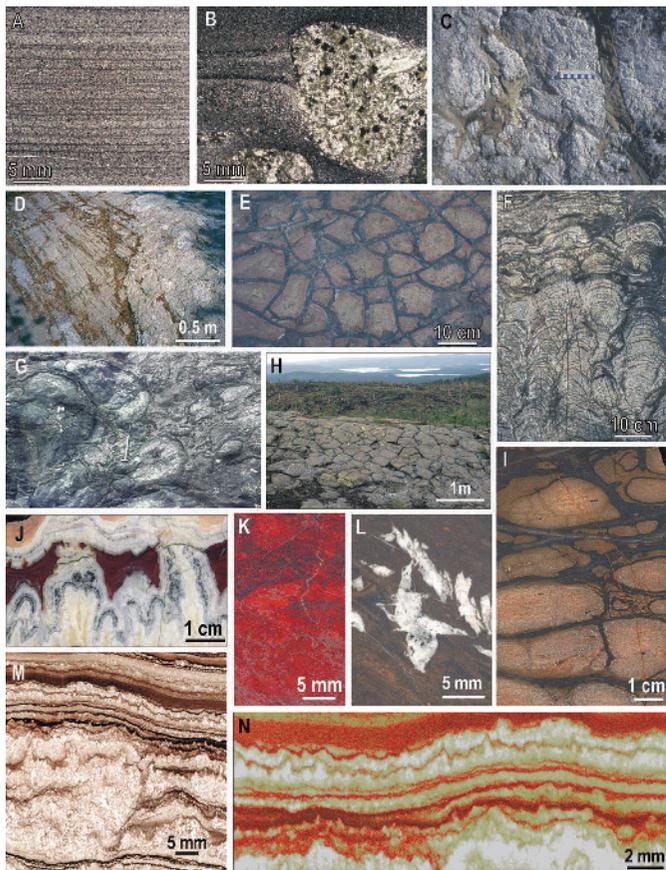


Figure 3. Rocks recording environmental events through ca. 2500–2060 Ma. Intracontinental rifting and glacial deposits from the Sarioli Group of the Pechenga and Imandra-Varzuga Greenstone Belts: (A) varves; (B) “dropstone”; (C) sand-filled cracks and fractures in Archean pegmatite recording incipient extension along the northern shoulder of the Pechenga paleorift; (D) erosional scouring at base of boulder conglomerates on Archean paragneiss (to left). Sedimentary features of Jatuli Group rocks from the Onega Basin: (E) desiccated red mudstone indicating an O_2 -rich atmosphere; (F) dolomitic columnar stromatolites. Volcanites of the Jatuli Group from the Pechenga Greenstone Belt: (G) slumped pillows in rift-basin lake; (H) columnar joints in alkaline basalts; (I) magmatically oxidized dacite. Chemical sedimentary rocks of the Jatuli Group from the Pechenga Greenstone Belt and the Onega Basin: (J) hot-spring travertine; (K) jasper; dolomite-pseudomorphed; (L) rosettes of gypsum with swallow-tail twin morphology; (M) sulfate nodule in former bedded evaporite; (N) bedded evaporite with enterolithic structure.

sometime after 2450 Ma but before 2320 Ma (Bekker et al., 2004) and about the time of the glaciation (Hannah et al., 2004). In Fennoscandia, this period was marked by intracrustal rifting and incursion of shallow-water epeiric seas with widespread deposition of terrestrial “red beds,” microbial carbonates, mature clastic sediments with Fe- and Mn-oxide mineralization (Melezhik, 1984), and voluminous alkaline to tholeiitic volcanites (Fig. 3E–3I) with native copper occurrences. Active hydrothermal systems associated with sub-aerial, rift-bound volcanism were responsible for spectacular travertine deposits and jaspers (Melezhik and Fallick, 2001; Fig. 3J and 3K).

Why and how the GOE occurred remains unknown, but its association with widespread igneous activity is suggestive of a causal connection. Anomalously high organic carbon

(C_{org}) productivity and burial driving high O_2 production, reflected in the Lomagundi-Jatuli $\delta^{13}C$ positive isotope excursion (2330–2060 Ma; see below), originally was considered to be the cause (Baker and Fallick, 1989; Karhu and Holland, 1996), but now is known to significantly postdate the initial accumulation of atmospheric O_2 , as indicated by the mass-independent sulfur isotope data. The lack of a global C isotope excursion between 2450–2330 Ma and the antiquity of oxygenic photosynthesizers (>2700 Ma; Brocks et al., 1999; Summons et al., 1999) suggest that the GOE may instead be related to a decline in the O_2 sink associated with the oxidation of reduced volcanic and metamorphic fluids rather than an increase in the O_2 source. The association of the GOE with widespread plume-related volcanism led Kump et al. (2001) to propose that mantle overturn associated with plume events at ca. 2700 and ca. 2500 Ma may have brought previously subducted, oxidized oceanic lithosphere (or perhaps banded iron formations) from the lower mantle back to the surface, increasing the average redox state of the upper mantle and the oxygen fugacity (f_{O_2}) of volcanic gases, and thus reducing the O_2 sink (Holland, 1978). The ca. 2214 Ma volcanic rocks (Fig. 3G–3I) in the Pechenga Greenstone Belt have high Fe^{3+}/Fe_{total} ratios (124 analyses average 0.37, of which 87% are >0.25; Predovsky et al., 1974), which are in sharp contrast to the majority of other volcanic units (ratios <0.25). While apparently too young to be the “smoking gun” for mantle redox evolution, these rocks highlight the heterogeneity of upper mantle redox state.

The lack of an apparent trend in upper mantle redox state (Delano, 2001; Canil, 2002; Li and Lee, 2004) argues against the mantle redox hypothesis, but the required increase in f_{O_2} is small (Holland, 2002) and may be within the scatter of the data (cf. Li and Lee, 2004). In response to these concerns, Catling et al. (2001) proposed that a change in the oxidation state of the continental crust created the necessary reduction in O_2 sink. Both of these hypotheses rely upon considerable Archean hydrogen loss to space to allow for the oxidation of the crust and/or mantle. Tian et al. (2005) proposed that hydrogen loss rates may have been much below the diffusion-limited rate and thus considerably lower than previously estimated, but this remains a controversial suggestion. Thus, the cause of the GOE remains unresolved and provides considerable motivation for future research on the Archean-Paleoproterozoic transition.

Abundant Marine Calcium Sulfates and Changes in Seawater Composition

Additional evidence for the GOE in Fennoscandia includes ca. 2100 Ma rocks (Fig. 2) containing relatively abundant syngenetic barite (Grinenko et al., 1989) and pseudomorphs after Ca-sulfates. The latter are abundant in the Onega Basin (Fig. 1) and are associated with magnesite and pseudomorphed halite crystals in a 500-m-thick succession covering more than 2000 km². They occur in varied facies: playa mudstones and fenestral stromatolitic sheets, sabkha and supratidal stromatolitic sheets, peritidal to intertidal biostromal and columnar stromatolites and variegated dolostones, and intertidal lenticular-bedded siltstone-mudstone couplets (Melezhik et al., 2000, 2005). Former Ca-sulfates occur mostly as dolomite and

silica pseudomorphs, commonly retaining relics of primary anhydrite (a variety of single and twinned crystal forms) and as diverse nodules, nodular masses with “chicken-wire” structure, and laminites with enterolithic structure (Fig. 3L–3N).

These evaporites postdate the events leading to irreversible oxidation of terrestrial environments that began prior to 2320 Ma, as evident by subaerial “red beds” (reviewed in Melezhik et al., 1999b). Combined, these features imply that the evaporites apparently precipitated syndepositionally from oxidized, evolved, and modified seawater (Melezhik et al., 2001), and their abundance in conjunction with those reported from elsewhere (Chandler, 1988; El Tabakh et al., 1999; Bekker and Eriksson, 2003) suggests that surface waters were oxidized and that sulfate had started to accumulate in the marine surface realm, a suggestion consistent with the sparse $\delta^{34}\text{S}$ data that exist for this interval (e.g., Strauss, 1993, 2004). Oceanic sulfate abundance remains unknown, but a sizeable sulfate reservoir as early as 2100 Ma would contrast with the view that, prior to the Mesoproterozoic, gypsum precipitation was inhibited by a small marine sulfate reservoir and higher marine carbonate saturation (Grotzinger, 1989; Kah et al., 2004). The irregular, cyclic, secular variations of geochemical parameters known for Phanerozoic terrestrial hydrosphere and atmosphere systems (Budyko et al., 1985; Veizer, 2005) may provide a means of reconciling these conflicting views.

Positive Carbon Isotope Excursion: The Lomagundi-Jatuli Paradox

The key discovery by Schidlowski et al. (1976) of unusually enriched ^{13}C Paleoproterozoic sedimentary carbonates in the Lomagundi province presented geologists with a puzzle. It took a decade for the first realization (Baker and Fallick, 1989) and a further decade for wide international acceptance (Karhu and Holland, 1996) of the Lomagundi-Jatuli Event as one of the major perturbations of the global carbon cycle in deep time, one associated with a series of fundamental changes of Earth’s surface (Fig. 4). In Fennoscandia, the excursion occurred after the glaciation and GOE, between 2330 and 2060 Ma (Karhu, 1993). Interestingly, it is primarily recorded in what we interpret as continental rift-basin lacustrine carbonates having little to no marine influences (Melezhik et al., 1997). The carbon isotopic composition of the carbonate ($\delta^{13}\text{C}_{\text{carb}}$) values display a broad range strongly correlated with environmental setting: playa and sabkha stromatolitic dolostones are most enriched, whereas those from intertidal settings exhibit lower $\delta^{13}\text{C}$ values (Melezhik et al., 2005). These facies-dependent trends suggest that the global $\delta^{13}\text{C}_{\text{carb}}$ excursion was smaller than previously thought, being amplified by up to 8‰ by local environmental factors such as rapid expansion of stromatolite communities in shallow-water evaporative and partly restricted environments, high bioproductivity and enhanced uptake of ^{12}C , and penecontemporaneous recycling of organic matter in cyanobacterial mats with the production and consequent loss of CO_2 (and CH_4 ?). Thus, extracting the global $\delta^{13}\text{C}$ excursion from the Fennoscandian Shield data has been controversial (Karhu, 1993; Melezhik and Fallick, 1996; Shields, 1997; Melezhik et al., 1999b).

Nevertheless, the Paleoproterozoic isotopic excursion is unique in terms of its duration (although age constraints are

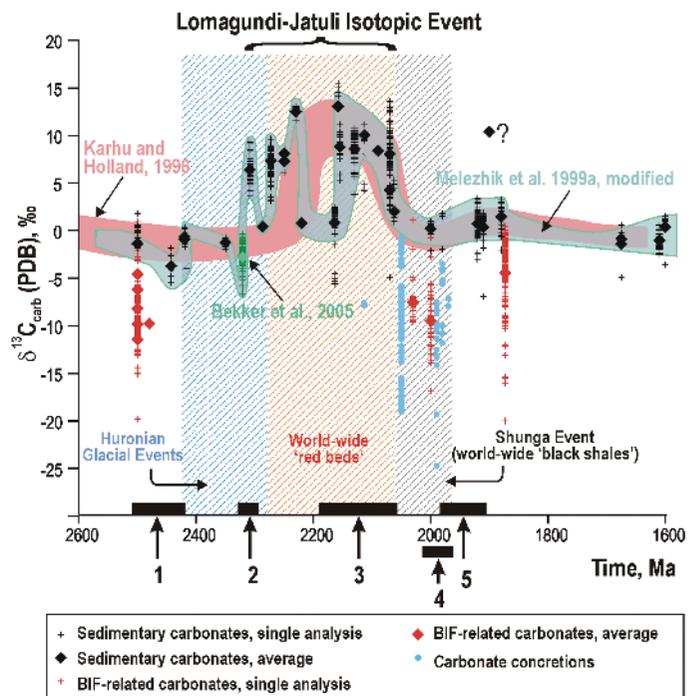


Figure 4. Paleoproterozoic $\delta^{13}\text{C}_{\text{carb}}$ trends of sedimentary and diagenetic carbonates and associated phenomena (modified from Melezhik et al., 1999b, including data from Bekker et al., 2005). BIF—banded iron formations; PDB—Pee Dee belemnite.

not robust, it appears to be >300 Ma) and ^{13}C enrichment. $\delta^{13}\text{C}$ values of purportedly synchronous deposits ranging from +8‰ to +28‰ (the latter interpreted as a global marine signal by Bekker et al., 2003a) have been reported from elsewhere around the world. Unlike the smooth $\delta^{13}\text{C}_{\text{carb}}$ trend generally depicted, the overall positive excursion is now argued to have two or three positive shifts separated by returns to 0‰ (Melezhik et al., 1999b, Fig. 4) that, unlike many younger isotopic events, do not go significantly below 0‰. Furthermore, there is no geological evidence for enhanced C_{org} accumulation prior to or synchronous with the excursion (although we cannot exclude preservational biases). Instead, it is the termination of the excursions that is followed by formation of a vast reservoir of variably ^{13}C -depleted organic material (–45‰ to –17‰ in the Onega Basin; see below) at a time when associated sedimentary carbonates are isotopically normal (Karhu, 1993; Melezhik et al., 1999a). Thus, determining the global timing of and regional influences on this event and identifying the mechanism(s) responsible for one of the most profound carbon isotopic excursions in earth history still represent major challenges for future research.

C_{org} -Rich Rocks and Earliest Significant Oil Deposits—The Shunga Event

Although Archean oil occurs elsewhere (e.g., Rasmussen, 2005), the most remarkable accumulation of organic matter and inferred generation of petroleum in the Paleoproterozoic

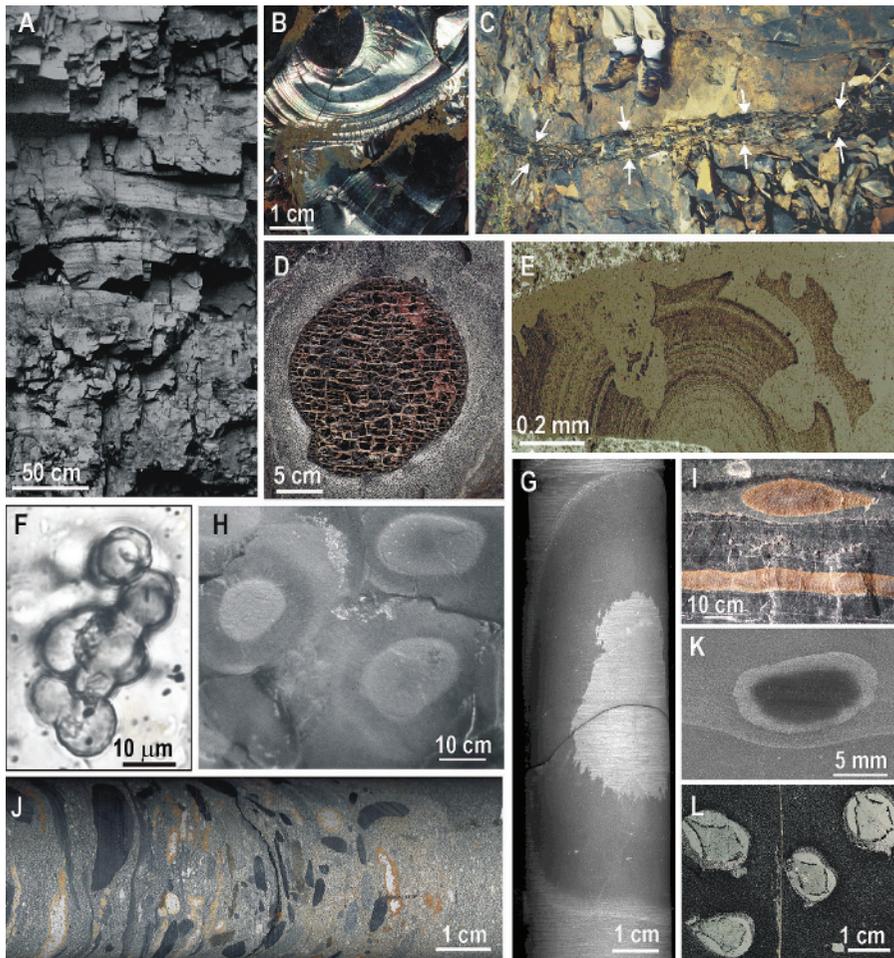


Figure 5. Rocks recording environmental events through 2060–1960 Ma. Petrified oil deposits of the Ludikov Group from the Onega Basin: (A) semi-lustrous shungite (originally oil shale; 45%–75% C); (B) lustrous layered shungite (originally interbedded-trapped oil; 98.4% C_{org}); (C) pyrobitumen-filled vein (petrified oil) in C_{org} -rich siltstone; (D) pyrobitumen clasts in lacustrine turbiditic graywacke; (E) organosiliceous rock with micron-scale concentric structure (reflected light). Microfossil from the Ludikov Group in the Pechenga Greenstone Belt: (F) coccoidal microfossil from nonstromatolitic chert interbedded with pillow basalts. Diagenetic concretions and phosphorites of the Ludikov Group from the Pechenga Greenstone Belt and the Onega Basin: (G) zoned, low- $\delta^{13}C$ calcitic diagenetic concretion in marine “black schist”; (H) calcite concretions in turbiditic graywacke; (I) sideritic diagenetic concretion in lacustrine turbiditic graywacke; (J) phosphorite clasts (black) in graywacke; (K) phosphorus-rich nodules in graywacke; (L) zoned sulphide concretions in “black schist.”

took place in the aftermath of the ca. 2330–2060 Ma Lomagundi-Jatuli Event (Melezhik et al., 1999a, 2004). 2000 Ma deposits rich in matured organic matter have been found in North America, Greenland, West Africa, and NW Russia (reviewed in Melezhik et al., 1999a) and represent the worldwide Shunga Event. In the Onega Basin, organic matter in unusually high C_{org} concentrations (up to 98%; average ~25%) occurs in a 1000-m-thick sedimentary-volcanic succession covering 9000 km². It is termed *shungite*, a black, noncrystalline, dense, glassy mineraloid with high semi-metallic luster and up to >98 wt% C (with traces

of N, O, S, and H). It accumulated in a non-euxinic, brackish lagoonal setting of a volcanically active continental rift (Melezhik et al., 1999a).

Shungite occurs either in situ and stratified, as migrated organosiliceous diapirs, or as redeposited clasts (Fig. 5A–5E; Melezhik et al., 2004). In situ shungite is metamorphosed oil shale ($C < 50\%$) containing autochthonous kerogen residue and allochthonous organic matter ($C = 50\%$ –75%) and bitumen (originally liquid hydrocarbons, $C > 80\%$). The type locality at Shunga (the most significant volume of trapped petroleum in the Onega

Basin) displays good examples of vertical and lateral migration of oil-defining cupola and diapiric bodies. These are organosiliceous rocks (35%–75% SiO_2 and 20%–55% C), considered to be originally gels or mud, with enigmatic concentric microstructures, abundant shrinkage cracks and cryptic fluidal textures, and brecciation caused by multiple fluidization processes. Lastly, clasts of lustrous shungite (<0.1–20 cm) eroded from subaerial oil spills occur in lacustrine volcanoclastic turbidites. In all occurrences, the organic matter suffered complex catagenetic and metamorphic alteration as evident by (Melezhik et al., 1999a): (1) four-modal distribution of C_{org} content (maxima at 5%, 30%, 65%, and 95%); (2) highly variable (–45 to –17‰) and bimodal distribution of $\delta^{13}C_{org}$ (maxima at –28 and –39‰); and (3) low H/C ratios (0.005–0.2). Abundant diagenetic carbonates associated with shungite rocks (Fig. 5H and 5I; $\delta^{13}C_{carb} = -5$ to –26‰) and the presence of diagenetic pyrite ($\delta^{34}S = -22$ to +31‰; Shatzky, 1990) reflect loss of organic matter via bacterial reduction of sulfate during diagenesis.

The integrated data suggest that the organic matter was most likely derived from planktonic microorganisms (it is noteworthy that peculiar microfossils [Fig. 5F] in non-stromatolitic cherts associated with extensive pillow lavas have been reported; Ivanova et al., 1988), but the cause of such unprecedented worldwide accumulation of organic matter at 2000 Ma remains unknown. If the fundamental features of the biologic carbon cycle were established by 3500 Ma (Schidlowski et al., 1975; Hayes et al., 1983; Grassineau et al., 2002), why did it take until 2000 Ma for the oldest known significant accumulation of organic carbon-rich sediments and petroleum deposits? Does this reflect an episode of enhanced biological productivity in a nutrient-enriched ocean that elsewhere supported widespread euxinia? Or was productivity more modest but preservation enhanced?

Fundamental Changes in the Early Diagenesis of Organic Matter

Compilation of global data shows that, with the exception of banded iron formations, $\delta^{13}C$ values of both primary and diagenetic carbonates of

pre-Paleoproterozoic rocks cluster near $0 \pm 3\%$ (Melezhik et al., 1999b). The end of the Lomagundi-Jatuli Event is marked by the first known appearance, and then worldwide development, of diagenetic carbonate concretions with negative $\delta^{13}\text{C}_{\text{carb}}$ values (Melezhik, 1992; Melezhik et al., 1999b). These are varied (Figs. 5G–5J) and abundant in the 2000 Ma sedimentary successions and are associated with other diagenetic products, such as phosphate nodules (Fig. 5K), all of which are seemingly absent from older rocks. Thus, this first appearance of isotopically light diagenetic carbonate concretions is an important hallmark in biospheric evolution linked to the emergence of “modern-style” recycling of organic matter. Perhaps prior to 2000 Ma fermentative methanogenesis rather than oxidation dominated early diagenetic organic matter remineralization (because of lack of O_2 and SO_4^{2-}). Escape of low $\delta^{13}\text{C}$ methane from the porewaters would have prevented the development of isotopically light porewaters. This hypothesis represents another subject for future research.

Sulfur isotope data ($\delta^{34}\text{S}$) display a narrow range around mantle values, 0‰ (e.g., Strauss, 2002). Bacterial sulfur metabolism has an early origin (ca. 3.47 Ga; Shen and Buick, 2004) and, although multiple lines of evidence of biogenicity are lacking, microbial sulfate reduction under either higher temperature and/or extremely low oceanic sulfate concentration leading to minimal isotopic fractionation has been suggested (e.g., Ohmoto and Felder, 1987; Ohmoto et al., 1993; Habicht et al., 2002). Recent data on mass-independently fractionated sulfur isotopes (Farquhar et al., 2000) suggest that photochemical dissociation of sulfur dioxide in an “oxygen-free” (O_2 partial pressure $<10^{-5}$) atmosphere might have dominated the global Archean sulfur cycle and enabled transfer of both oxidized and reduced sulfur compounds with distinct mass-independent isotope signatures to surface environments.

In contrast, sediments deposited during the early Paleoproterozoic contain sedimentary pyrite displaying a substantially larger isotopic fractionation ($\delta^{34}\text{S}_{\text{sulfate-sulfide}}$). The 2000 Ma successions differ markedly from older rocks by having a high abundance of various diagenetic generations of iron sulfides (Fig. 5L) with $\delta^{34}\text{S}$ values ranging between -22% and $+31\%$ (Melezhik et al., 1998; Shatzky, 1990). Even more negative values, as low as -30% , have been reported from the 2320 Ma Timeball Hill Formation, South Africa (Cameron, 1982; Bekker et al., 2004). Based on present understanding (e.g., Canfield, 2001), such values represent evidence for bacterial sulfate reduction and, given the absence of evidence for mass-independent sulfur isotopic fractionation in rocks younger than 2320 Ma, imply a profound change in the operational mode of the global sulfur cycle and concomitant increase in atmospheric oxygen concentration (Pavlov and Kasting, 2002). Subsequently, a rise in oceanic sulfate concentration enhanced the importance of bacterial sulfate reduction as a principal process of organic carbon and sulfur cycling in sedimentary environments. Hence, the global sulfur cycle changed into a mode quite similar to the modern world in which oxidative weathering, riverine delivery of sulfate to the ocean, and biological sulfur cycling dominate over volcanic/hydrothermal S cycling. However, a smaller than present-day isotopic fractionation associated with bacterial sulfate reduction and subsequent pyrite formation may be consistent with

an ocean low in sulfate through 2000 Ma, and probably until the Precambrian-Cambrian transition (e.g., Lyons et al., 2004). The driving forces of these changes remain to be established.

The oldest known phosphorites and phosphate concretions (Fig. 5J and 5K) are commonly associated with sulfides and organic matter in the 2000 Ma successions and reflect a critical change in the Precambrian phosphorous cycle. Although different processes have been proposed for concentrating dissolved phosphate in bottom sediments to levels required for authigenic precipitation, the most common pathway includes organic matter that carries phosphate concentrated by biological activity (Knudsen and Gunter, 2002). The association of phosphate concretions with organic matter in the C_{org} -rich 2000 Ma sedimentary rocks is consistent with such a mechanism. However, high abundance of organic matter is probably not the only factor controlling formation of Paleoproterozoic phosphate concretions. C_{org} -rich layers have also been found in sedimentary sequences older than 2000 Ma (Hayes et al., 1983), but no phosphate concretions have been reported. Thus, it is possible that 2000 Ma sediments record a major change in the diagenetic mineralization of organic matter, perhaps reflecting a substantial increase in the intensity of early-diagenetic sulfate reduction promoted by an overlying, sulfate-enriched ocean (Canfield and Raiswell, 1999) that in turn elevated the concentration of interstitial phosphate. However, determining the factors that controlled the formation of the oldest known phosphate concretions remains an unresolved problem.

QUESTIONS FOR FUTURE RESEARCH

The Archean-Paleoproterozoic transition was a major step in the eventual establishment of the modern earth system. A relative chronology of the transition has been determined, but fundamental questions remain unanswered. (1) Why did global glaciations only occur after the Archean? (Evidence for glaciation at 2900 Ma is restricted to South Africa; Young et al., 1998.) (2) Why did an oxygen-rich atmosphere only appear in the Proterozoic (around 2400 Ma)? Oxygen-rich habitats existed since at least 2700 Ma. (3) What drove the Lomagundi-Jatuli Event, the oldest known positive global carbon isotope excursion? (4) What caused radical change in biogeochemical recycling of organic matter at ca. 2060 Ma? (5) Why didn't large accumulations of organic matter similar to the Shunga Event occur in the Archean? (6) How, out of this suite of biogeochemical global-scale perturbations, did the modern earth system emerge? The recently launched International Geological Correlation Programme Project 509 “Palaeoproterozoic Supercontinents and Global Evolution” will be a focus for internationally coordinated research on this most interesting time period in earth history.

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