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Revisiting the greatness of Earth's great oxidation

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The accumulation of free oxygen in the atmosphere ≈2.45 billion years ago was one of the most transformative events in Earth's past. Since its identification, this interval termed the 'Great Oxidation Event' (GOE), has garnered a large amount of attention from a wide array of perspectives, with some suggesting it should define its own geologic era or period. Despite many new tools to interrogate the GOE, today the defendable range of possible atmospheric O_2 levels span orders of magnitude, begging the question, how great was Earth's Great Oxidation? The consequences of such disparate views on oxygenation levels are uncertainties regarding biospheric evolution, interpretations of the sedimentary record, and a limited ability to translate Earth's atmospheric history into insights for exoplanet research. In this review, we revisit the conditions immediately before, during, and after Earth's GOE to explore the key assumptions that underlie differing views on this critical interval of time. We then highlight new discoveries and outline extremely divergent but defendable interpretations of atmospheric oxygen trajectories across the GOE. Reducing such divergent scenarios should be a major target of research progress in the coming years.

The Great Oxidation Event (GOE) is the interval of time when oxygen first accumulated in Earth's atmosphere[1–3]. The first hints that such a transition took place in Earth's atmosphere can be traced to the mid-19th century, when Ebelemen (1845)[4] established that the burial of organic matter produced through oxygenic photosynthesis results in the buildup of O_2 in the atmosphere. However, Koene (1856)[5] was possibly the first to posit that through such a mechanism, the Earth likely began in an anoxic state and transitioned to an oxygenated one sometime in its deep history[6]. Almost a century later, MacGregor (1927)[7] firmly placed atmospheric oxygenation in the Paleoproterozoic with the observation of transported pyrite clasts in the Ndutjana Conglomerate of the Shamvaian Supergroup, followed by evidence for oxidizing conditions underlying the Lomagundi Formation of Zimbabwe. These early inferences tied the evolution of a metabolism to the ushering in of a substantial atmospheric transition. Subsequent geologic evidence has continued to build support for this hypothesis and established this event within the earliest Proterozoic. For example, the disappearance of redox-sensitive detrital minerals followed by the occurrence of red beds in Huronian successions led Roscoe (1973)[8], to coin this transition as the 'oxyatmoversion'. From these early studies, the concept of a 'Great Oxidation Event' has now been well established in the literature and colloquially used as an informal interval of Earth's past, beginning with an undefined boundary between 2.5 and 2.4 billion years ago (Ga)[9,10] and ending around 2 Ga[11].

Although the scarcity of geochemical records calibrated with geochronology continues to limit our ability to define the specific pacing of oxygenation across the earliest Proterozoic, modelling contributions have provided additional constraints. As oxygen accumulated past the threshold to establish an ozone layer, a suite of oxygen-consuming reactions would have slowed, further accelerating the accumulation of oxygen in the atmosphere[12]. Estimated timescales for this transition have ranged from 10^2 to 10^5 years[12–14]. The rapid introduction of large quantities of free oxygen into the surface environment had a profound impact. Continental environments witnessed a reshuffling of mineral weatherability and assemblages, which altered the chemistry of global river systems[15–17]. The biosphere was also forced to reorganize[18] but likely witnessed a sustained growth[19–22]. A key aspect of this growth in the biosphere was the expansion of aerobic metabolisms[23], and many organisms proliferating in shallow surface environments, which would have found a reprieve from harmful UV

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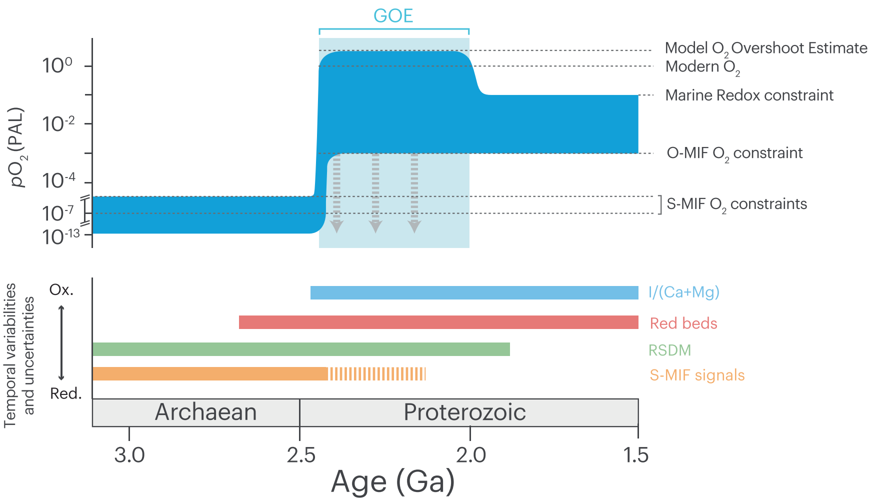

Fig. 1 | In the top panel, existing boundaries of atmospheric oxygen levels across Earth's Great Oxidation Event (GOE) are presented[28–34]**.** Here, specific boundaries are placed without previously interpreted trajectories or geologic hiatuses such as glaciations. In the lower panel, a subset of constraints utilized to derive the timing of the GOE is presented (RSDM = redox-sensitive detrital minerals).

radiation with the establishment of an ozone layer. At the same time, organisms would have had to contend with the deleterious consequences of free oxygen in their surroundings[23]. Finally, the introduction of O_2 to the atmosphere altered the broader atmospheric composition through destruction of other gas species such as nitrous oxide, sulfur dioxide, and methane[24], which potentially set the stage for early Paleoproterozoic glaciations[25–27].

While broad features of the GOE have largely found consensus within the Earth history community, divergent views have developed regarding the precise O_2 levels reached and the specific O_2 trajectory followed across the GOE. The firmest geochemical constraint to emerge since the recognition of the GOE derives from sedimentary sulfates and sulfides with a lack of mass independently fractionated sulfur isotope signatures that impose a lower limit on post-GOE pO_2 of ~10^{-7} to 10^{-5} present atmospheric level (PAL)[28–31]. Beyond this sulfur cycle-based oxygen threshold, modelling estimates exploring the implications of ozone-layer establishment and geochemical evidence through mass independently fractionated oxygen isotope signatures shift toward baseline pO_2 levels of 10^{-4} to 10^{-3} as a minimum[32]. However, no clear consensus has been reached on possible upper bounds across this interval above this ozone-based minimum, with some studies suggesting atmospheric O_2 may have climbed above modern levels[33–35]. Although model development is continuing to reveal potentially important dynamics across the GOE, at the core of this uncertainty is the challenge of constraining numerous factors that exert influence on the oxygen cycle, such as continental emergence, phosphorus fluxes, and organic carbon burial efficiency[33,36–41]. Together, this existing uncertainty on specific atmospheric O_2 levels across the GOE spans up to four orders of magnitude (Fig. 1).

Adding an additional layer of complexity, recent sulfur isotope evidence has now suggested that the GOE was not a singular rise of atmospheric O_2 but rather a protracted interval of fluctuating atmospheric and oceanic oxygenation[41–45], suggesting possible Great Deoxygenation Events across this interval of time. However, the magnitude, duration and global or local nature of such oscillations remain highly uncertain due to ambiguous stratigraphic correlations, limited age constraints, and the possibility of signal recycling[46,47]. Paleoproterozoic climate has found a similar degree of uncertainty, where it remains a challenge to ascertain the regional or global nature of Paleoproterozoic glaciations, as well as how to correlate some glacial units across different continents[10,48]. Moreover, what direction such glaciations may have pushed oxygen levels also remains debated. Although one would have hoped that the expansion of geochemical proxy records would offer key insights to resolve these issues raised above, such records have often added additional layers of uncertainty and have challenged the extent to which we can trust many interpretational frameworks that are applied to the preserved Paleoproterozoic rock record[49–51]. Together these observations limit our understanding of the GOE and add large uncertainties when placing the GOE in the context of planetary evolution through time in terms of the integrated size of the overall biosphere[21] and integrated fluxes of atmospheric and oceanic species such as O_2, SO_4, CH_4, and CO_2[52–54]. Plausible marine oxygen levels through the GOE also span ranges above and below the thresholds for eukaryotic or even metazoan aerobic metabolism[55,56], leaving it open for debate whether the later Paleoproterozoic appearance of eukaryotic microfossils reflects an immediate response to permissive environmental chemistry or an evolutionary delay[57,58].

Despite the large looming parameter space, new geochemical tools combined with community efforts to study the sedimentary record in its entirety[59–61] are fertile grounds to rule out competing O_2 trajectories across the GOE. For example, new efforts are allowing for diagenetic overprinting to not only be identified but to be quantified in ancient sequences[62–64]. The ability to geochemically identify terrestrial influences on marine records is another capability that is rapidly maturing[65,66]. Moreover, critical insights about the early evolution of specific metabolisms are being brought to the forefront through advances in molecular clocks[67–70], and advances in models designed to understand the rapid accumulation, maintenance, and possible decline of atmospheric oxygen are complementing geochemical efforts. These abilities, combined with improvements in mechanistically understanding the production, transport, burial, and lithification of proxy records[71–73] will likely lead to important advances in numerous areas that will aid in reducing the large uncertainties surrounding the GOE noted above. It is the aim of this review to emphasize defendable scenarios that may explain the existing GOE record, but that also depart from many existing syntheses of this interval. At the same time, we highlight new advances and project how they may come together in the coming years to ultimately answer the question: how great was Earth's Great Oxidation?

Oxygen before oxygenation?

Like the GOE, paleoenvironmental and ecological conditions of the Archean remain subjects of controversy. At the forefront of such debates are conflicting records and models of key properties such as marine phosphate concentrations, disagreement about when oxygenic photosynthesis emerged as a metabolism, and whether there are unambiguous hints of oxygenation left in the geologic record prior to the GOE[74–76]. Such ambiguity is found within both genomic and geochemical records. While uncertainties in the accumulation of mutations over time make molecular clock estimates highly model and calibration-dependent, anchoring such estimates with the geochemical record sacrifices the independence of genomic and geochemical histories. However, in the case of the emergence of oxygenic photosynthesis, most molecular clock estimates have placed the origins of Stem Cyanobacteria in the Archean prior to the GOE[68,69,76]. Despite this common practice, such estimates vary widely[77,78]. That said, a critical insight that has emerged from such efforts is that the ability to utilize oxygen in the environment through aerobic respiration pre-dates the GOE by hundreds of millions of years, and possibly even pre-dates the evolution of oxygenic photosynthesis[70,76,79]. Such an insight is important and forces consideration of how early oxygen may have been produced to promote this selective pressure and bring about this ordering of metabolisms. We expand on this below.

In the case of geochemical and geological calibrations, there have been two very different interpretations of such records. In the first case, studies have argued that the emergence of oxygenic photosynthesis occurred within the Paleoproterozoic, immediately prior to a purported Paleoproterozoic Snowball Earth at ≈2.4 Ga[22,27]. On the other hand, a growing number of geochemical studies argue for oxygen in numerous Archean environments[75] and infer that this oxygen was produced via oxygenic photosynthesis. Although originally reported Archean 'whiffs' of oxygen continue to be scrutinized[80–83], a growing number of studies have offered additional lines of evidence of oxidizing conditions well in advance of Earth's GOE[84–87]. For example, trace elements together with sulfur, molybdenum and iron isotope signatures from the Mozaan Group have been argued to be a result of aerobic iron and manganese cycling within Mesoarchean waters[88–90]. More recently, elevated iodine to calcium ratios, as well as cerium anomalies within shallow marine carbonates, suggest locally oxidized shallow water environments by ≈2.9 Ga[91,92]. In the case of cerium anomalies, the ability to date these signatures and not just their surrounding lithology has provided an important advancement in the confidence that such signals were not introduced long after initial deposition[92].

If one is convinced by the evidence presented for oxygenation well in advance of the GOE, it then presents an additional suite of questions. For example, if oxygenic photosynthesis evolved hundreds of millions of years before the GOE, what could have delayed global oxygenation? One hypothesis is that phosphorus concentrations were extremely low, placing a limit on global primary production at this time[93,94]. Moreover, if this metabolism did evolve very early, it has been argued that it may have maintained phosphorus at very low levels, which could have delayed the GOE[41]. However, estimates for phosphate availability in the Archean derived from sedimentary phosphorus records currently span six orders of magnitude, with some rising above modern levels[74,94]. Alternative explanations have invoked an inability of early oxygenic photosynthesizers to outcompete their anoxygenic neighbors[95,96], or a buffering effect imposed under an atmosphere with no ozone layer, which would have forced these organisms to inhabit deeper depths where they would have experienced light limitation relative to their progeny today[97–99]. A challenge to the ideas raised above is that there is fossil and geochemical evidence that early photosynthesizers occupied very shallow environments[91,100], suggesting that such locations were not so inhospitable to life[101]. If not biology, then abiotic explanations are required. For example, a stronger outgassing flux of reducing gases may have curtailed oxygenation of the atmosphere. Support for such a hypothesis is found in geochemical and modeling studies, which argue for a more reducing Archean mantle[102–104]. Moreover, other factors such as mineral evolution[105] and metal mineral sinks may have greatly inhibited the synthesis of critical proteins for early oxygenic photosynthesizers to thrive[106–112].

While many studies have explored a possible delay in the evolution of oxygenic photosynthesis and the GOE, few have wrestled with why aerobic metabolism may have evolved first, and whether the same selective pressure for such metabolic capability may have also left imprints on the Archean rock record[113]. So-called 'dark oxygen' production, that is, free oxygen produced independently of oxygenic photosynthesis, is increasingly being documented in a range of modern environments, Archean analog experiments, and in a varied suite of abiotic and microbial processes[114–120]. For example, silicate mineral abrasion, nitrous oxide, and chlorite dismutation, electrolysis, and radiolysis have all been documented to produce free oxygen in the absence of oxygenic photosynthesis. While the degree of importance of such mechanisms may vary widely by environment and through geologic time, it remains unknown whether the production of oxygen through these mechanisms may have reached requisite thresholds to preserve such signals within the sedimentary record. If such oxygen thresholds can be shown to be achievable through observations from modern environments, then geochemical anchors for molecular clocks used to calibrate the emergence of oxygenic photosynthesis will need to be revised, and the relationship between this metabolism and pre-GOE whiffs of oxygen reevaluated. Although answers to questions raised above do not have a bearing on 'how' and 'when' oxygen first accumulated in Earth's atmosphere to the point where clear indicators of atmospheric oxygenation were left in the geologic record, they do raise the question of 'why' it happened when it did.

Earth's first significant rise in atmospheric oxygen

Evidence of localized pre-GOE free oxygen notwithstanding, the beginning of the GOE has primarily been tied to the disappearance of mass-independent sulfur isotope signatures (S-MIF) within the sedimentary record[9,10,28]. The interaction of solar radiation with atmospheric sulfur species imparts a mass-independent discrimination of isotopes between photochemical products. Without O_2 in the atmosphere, these photochemical reaction products can settle on Earth's surface, followed by weathering, transport, and deposition within the sedimentary record. After O_2 is introduced to the atmosphere, the ability to preserve such signatures largely ceases[71,121], making the disappearance of S-MIF the so-called smoking gun for Earth's first large leap in atmospheric oxygen levels[28]. Although there has been some disagreement regarding a specific atmospheric O_2 threshold that leads to the disappearance of such signatures within the sedimentary record[29,30], it is worth noting that this is very much an ongoing area of research[122–124]. Accurately reproducing the pre-GOE atmospheric composition and then simulating the disappearance of S-MIF in the surface environment from this pre-GOE composition within a laboratory setting remains out of reach[125]. Moreover, sulfur photochemistry itself includes a complex network of reactions that remain challenging to create insightful experiments[126–129]. Despite these limitations, at present, estimates place the disappearance of S-MIF occurring when atmospheric O_2 levels rise beyond 10^{-7} to 10^{-5} present atmospheric levels[29,30].

An additional proxy that may offer insight into specific oxygen thresholds that were surpassed at the initiation of the GOE is the record of mass independently fractionated oxygen isotopes (O-MIF) within sulfate minerals (triple oxygen isotopes; $\Delta'^{17}O$). Given the bistability of Earth's atmosphere with respect to O_2 concentrations, oxygen levels are not predicted to remain near S-MIF thresholds but are estimated to either fall back to Archean levels or rise above those required to establish an ozone layer[12]. Although there remains a non-trivial age uncertainty of the Duitschland Formation, existin\g estimates place its deposition close to 2400 Ma[130]. Here, negative non-zero $\Delta'^{17}O$ values within sulfate suggest an active ozone cycle, placing a lower estimate for atmospheric O_2 levels of $\approx 10^{-3}$ PAL[32,131], which conforms to model predictions of O_2 levels rapidly climbing toward a new stable state across the GOE[12,13]. However, expanding on existing data sets to more closely align with the S-MIF record is dependent on new sulfate records being found and new model frameworks to interpret such records[132,133], which at present are sparse across this interval of time.

The disappearance of S-MIF and appearance of O-MIF alone is only one key ingredient for constraining the timing of the GOE, the other being high-precision geochronological constraints. At present, work targeting Paleoproterozoic basins in South Africa and Russia places Earth's first rise of atmospheric oxygen and the initiation of the GOE between ≈2501 and 2426 Ma[9,10]. This timing aligns with Macgregor's (1927)[7] estimate based on the disappearance of redox sensitive minerals, and more recent works are in agreement through further study on the disappearance of redox sensitive minerals, and the appearance of sulfate evaporites and terrestrial redbeds[131,134] as well as a suite of geochemical proxies[135]. Whether one can read such records as a progression of O_2 accumulation beyond certain thresholds remains an area of ongoing debate; it is virtually impossible to ever capture the very first or last appearance of any proxy signal in an imperfect and incomplete sedimentary record[136,137], which limits the temporal inferences that can be derived from such observations. In addition, there remains no clear consensus to date regarding how high O_2 in the atmosphere may have climbed during the beginning of the GOE.

How great was earth's great oxidation?

Assessing the significance of Earth's Great Oxidation can be broken down into many different components. Here we focus on the magnitude of oxygen introduced to the surface environment, which itself is the combination of proxy and model evidence for specific oxygen estimates, and the duration these estimates can be projected. As noted above, many geologic indicators of oxygen, such as the appearance of sulfate evaporites, the disappearance of redox-sensitive minerals from certain environments, and the appearance of red beds, only offer qualitative evidence of oxygenation[134,138,139]. Moreover, even when such evidence is found within the sedimentary record, and even when it can be radiometrically dated, there remains uncertainty regarding how much time has passed between its first occurrence on the ancient Earth and the deposition of the first occurrences available to study[136]. Proxies that have offered evidence for a rise of oxygen beyond a certain threshold are also limited in their extrapolation across Earth's GOE. Specifically, a rise of oxygen levels beyond S-MIF and O-MIF thresholds only places concentrations beyond a potentially unstable state[12,13,41]. However, deoxygenating the atmosphere becomes increasingly difficult to achieve as oxygen levels climb further from such thresholds. Therefore, it becomes important to constrain how high O_2 may have risen after initial oxygenation, how long such conditions may have persisted, and when clear hiatuses of extrapolation can be identified, such as possible global glaciation.

Oxygenation and glaciation. Plotting geochemical proxy data along a 4.6-billion-year time axis can offer insights into some broad features of the geochemical record, but it also biases one toward endmembers and leaves the impression that individual data points represent tens to hundreds of millions of years and scatter around a globally representative mean. If one agrees with some of the sentiments raised above, it motivates the question: how far can any individual geochemical indicator of oxygenation be temporally extrapolated beyond the time it took to deposit the sedimentary layer in which it was deposited? In the case of the syn-GOE interval between ≈2.5 and 2.0 billion years ago, there is extensive evidence of glaciations, and some have argued that at least one of them may have been a global, snowball Earth[25–27,140]. Importantly, arguments have been presented which not only link glacial initiation to a rise of O_2 but also articulate how the syn-glacial environment can evolve atmospheric composition far from pre-glacial levels[39]. If true, such a dramatic shift in climate places some model-based limits on extrapolating many oxygen estimates from proxies.

Snowball Earth, or panglacial climate states, would have had a profound impact on the global biosphere as well as fluxes of carbon and oxygen in the surface environment. The case for two Neoproterozoic Snowball Earth glaciations at ≈717 and ≈640 million years ago, which define the Cryogenian period, has been built upon similar sequences of globally distributed strata where evidence for grounded marine ice at sea level is extensively documented at low paleo-latitudes[141]. Above glacial units in such sequences are thick cap carbonate deposits which offer geologic evidence for predicted sinks of syn-glacial alkalinity and CO_2 build-up across long panglacial intervals[142]. Moreover, paleomagnetic evidence has placed many of these sequences within the tropics[143] and geochronological evidence has confirmed their synchroneity[144]. Additional study has documented a notable drop in average sedimentation rates during these glacial episodes and an absence of lithologies that require open waters and warm conditions[145]; such observations together with a suite of geochemical signatures that are challenging to explain in the absence of a panglacial state[146–148] sets up the Cryogenian as a benchmark for the presence or absence of global glaciations. In the case of Paleoproterozoic glaciations, it has remained difficult to achieve the same level of confidence in interpreting their global or regional extent.

Although there has been extensive documentation of glacial sequences between ≈2.45 and 2.2 Ga across many paleocontinents, few have been dated with the same degree of precision as their Neoproterozoic counterparts to establish clear correlations, and, furthermore, paleomagnetic evidence to place glacial diamictites in the tropics remains sparse. An exception to this is the Makganyene glaciation, which places ice in the tropics at ≈2.4 Ga[25,48]. In this case, evidence of low latitude glaciation together with possible correlative deposits in the Ramsey Lake Formation of the Superior Craton, the Campbell Lake Formation on the Wyoming Craton, the Polisarka Formation in the Kola-Karelia regions, the Meteorite Bore member on the Pilbara Craton, and the Padlei Formation of the Hearne Craton perhaps make this the strongest candidate for a Paleoproterozoic Snowball Earth[10]. Younger glacial units preserve more limited geographic expressions and suffer from poor paleomagnetic constraints. Even in the case of the putative older Panglacial, other key features found in Cryogenian sequences remain lacking, such as extensive cap carbonates[149], barite horizons[146,147] as well as other enigmatic sedimentary features[150].

In the context of the GOE, this uncertainty regarding the nature of Paleoproterozoic glaciations imposes limitations on oxygen estimates and reconstructions of productivity of the ancient biosphere. At present, it remains challenging to decipher whether there were two, three, or four glaciations in the Paleoproterozoic, and whether none, one, some, or all of them were global in extent. Moreover, even if they were all global in nature, it remains unclear what their precise durations were. Putting this current state of uncertainty together and extending inferences from the Cryogenian into the Paleoproterozoic, suggests a Paleoproterozoic panglacial climate state may represent between zero and many tens of millions of years[151]. In terms of oxygen fluxes to the atmosphere, the continuum of climate from greenhouse to regional glaciations to panglaciation does not extend to global primary production. Although a Snowball Earth climate state would drop primary production to extremely low levels, ice core evidence suggests that productivity increased during regional glaciations in the Pleistocene relative to interglacial levels[152]. Together, these factors make constraining the Paleoproterozoic climate a critical area to resolve in the assessment of the syn-GOE interval.

Predicted shifts in primary production imposed by a Snowball climate state also have important implications for oxygen dynamics of the syn-GOE atmosphere. Specifically, on one hand, there has been S-MIF evidence presented from a single drill core in South Africa, which argues for a return to an anoxic atmosphere across the deposition of the Rooihoogte formation, which some have interpreted as the aftermath of a third Paleoproterozoic glaciation at ≈2.35 Ga[42,130,153]. However, such evidence is counter to model predictions, which argue that global glaciations impose an oxidant–reductant flux imbalance to move the Earth system from a lower stable state of oxygenation to a higher one[39]. While models have been successful in reproducing atmosp

heric oxygen oscillations[13,41,43,44] which can potentially reconcile such arguments, such inferences can only speak to a limited suite of biogeochemical feedbacks that may or may not reflect the Paleoproterozoic Earth system. For example, such mechanisms have primarily explored oscillations of oxygen below 1% PAL and have not explored whether such mechanisms are plausible if O_2 levels approached those of the modern atmosphere. Indeed, an interesting preliminary inference from existing work may suggest that evidence of measurable atmospheric O_2 oscillations across the first half of the GOE discounts high oxygen 'overshoot' scenarios[13,41,43,44].

Geochemical records of the GOE. While glacial intervals and possible oscillations in atmospheric O_2 levels continue to drive much of the uncertainty across the first half of the syn-GOE interval, geochemical signatures are at the forefront of uncertainty for the remainder. Central to this debate is the interpretation of the Lomagundi-Jatuli positive carbon isotope excursion (LJE)[49–51]. Here, carbon isotope values of shallow marine carbonates are typically around 7‰ but have been documented to reach as high as 30‰[154–156]. Remarkably, values between ≈5-15‰ have been documented on every continent outside of Antarctica across overlapping age uncertainties. The canonical interpretation of these signatures has been that they represent an increase in the relative

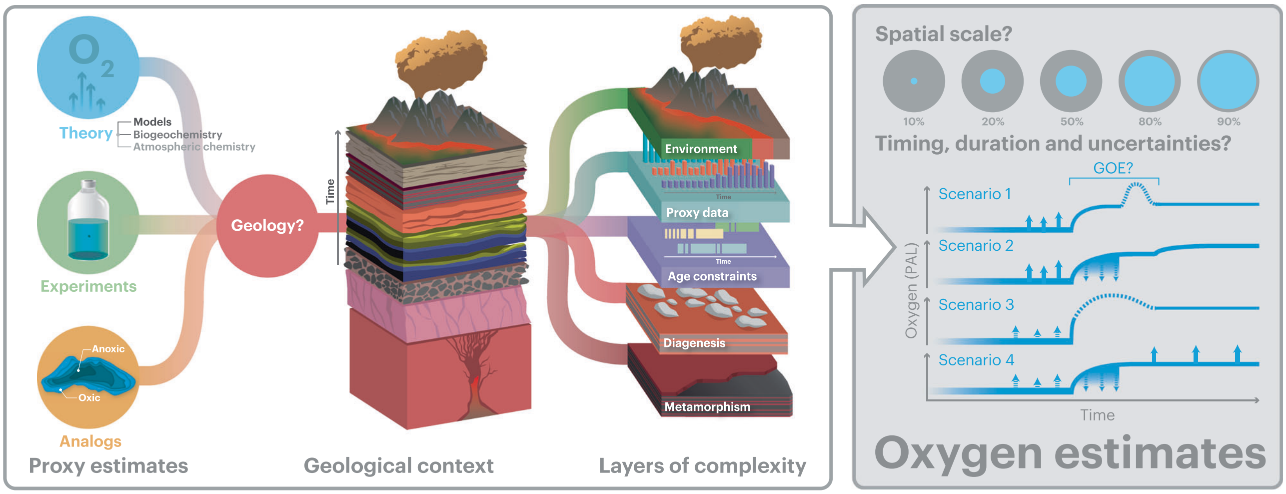

Fig. 2 | Challenges in building atmospheric oxygen proxies. The far-left panel depicts the genesis of atmospheric oxygen proxies from theory to experiment to modern analog applications. Next to this is a depiction of the geologic context and the complexity that this imposes on any proxy based on geochemical information extracted from the sedimentary record. The far-right panel indicates the challenges in translating this information into a specific atmospheric oxygen estimate and presenting it along with its inherent limitations.

proportion of carbon burial in the form of organic matter[157]. The implication of this interpretation is that the positive carbon isotope signatures can chemostratigraphically correlate Paleoproterozoic strata globally, and that extreme amounts of atmospheric O_2 may have been added to the atmosphere at this time as a result of high fractional organic carbon burial. Such an interpretation underlies the inference of a possible oxygen 'overshoot'[35]. Constraining exact atmospheric O_2 levels is challenging to determine since there would have been no buffering effect of terrestrial vegetation through fires, and there are no clear proxies to constrain specific syn-GOE oxygen concentrations beyond S-MIF and O-MIF thresholds. Without such constraints in place, model estimates have suggested oxygen may have reached up to 2.5 times modern[33,34]. Alternatively, such signatures have also been argued to be a possible consequence of an overexpression of preserved intertidal coastal environments where local carbon cycling could drive the extremely positive carbon isotope signatures[51,72,158,159]. While one would hope that the growing armada of redox proxies would clearly discriminate between these two very different interpretations, to date, this has not been the case.

Over the past two decades with the installation of numerous multi-collector inductively coupled plasma mass spectrometers (MC-ICPMS) as well as improvements in trace element analyses, a suite of proxies has emerged to constrain oxygen levels throughout Earth's past[45,160–166]. Such approaches can broadly be separated into four categories: proxies for specific O_2 thresholds (e.g., S-MIF, O-MIF), proxies for oxidative weathering fluxes (e.g., δ^{53}Cr, δ^{98}Mo), proxies for surface water oxygenation (e.g., I/Ca, Ce anomalies), and proxies for marine oxygenation (e.g., δ^{238}U, δ^{51}V, ε^{205}Tl, $\delta^{82/78}$Se, Fe-speciation). For many of these tools, following initial optimism, data sets have revealed a large amount of complexity, which has limited extrapolating such signatures in time and space. Such limitations are likely in part a combination of comparing disparate depositional settings and incomplete accounting of processes, which could drive specific signals. Moreover, as the ratio of the concentration of many of these elements between sediments and altering fluids is much less than carbon, in many cases, they are much more susceptible to diagenetic alteration[64], but quantifying this influence remains an enduring challenge.

Three examples of such proxies are triple oxygen isotopes in sulfate minerals as a proxy for biological oxygen production ($\Delta'^{17}O$), selenium isotopes in shales as a proxy for deep water oxygenation ($\delta^{82/87}$Se), and iodine to calcium ratios in shallow marine carbonates as a proxy for shallow water oxygenation (I/Ca). A challenge with each of these proxies is the degree to which they can be extrapolated to the global environment (Fig. 2). For example, $\Delta'^{17}O$ signatures in sulfate have been argued as a proxy for biosphere productivity. In this case, a large drop in $\Delta'^{17}O$ signatures across the end-GOE led to the interpretation of an extremely large drop in primary production between the syn-GOE and post-GOE intervals[54]. However, recent work has argued that such signals must be interpreted in the context of the sedimentary facies from which they were deposited, where it has been evidenced that terrestrial environments have the greatest capacity to preserve such signatures and marine environments the least[65]. Similarly, $\delta^{82/78}$Se values in shales have been leveraged as evidence for marine suboxia during the GOE, which could be interpreted as evidence against an oxygen overshoot or for evidence of fluctuating redox conditions across the syn-GOE interval[167]. Again, such results may also be highly localized and not representative of anything truly global[168]. As a final example, I/Ca signatures have been utilized as evidence for marine oxygen levels above 1 µM across the syn-GOE interval[135]. However, it is also possible that such signatures represent local conditions of unknown spatial extent.

Was there an end to Earth's Great Oxidation?

Although evidence for pre-GOE oxygenation has muddied the waters on a clear definition of what the beginning of the GOE represents with respect to the emergence of metabolism, and emerging records are suggesting a much more complex picture of oxygen levels during this interval, it is perhaps most challenging to define the GOE's end. Initially, Holland (2002)[11] suggested that the end of this informal interval can be tied to the disappearance of the extreme positive carbon isotope values that define the LJE. Additional geochemical arguments for an end-GOE transition, or for Earth's Exit from Oxygenation (OXIT), have been found through trace element concentrations, multiple sulfur isotopes of pyrites in shales, iron speciation, molybdenum isotopes, and $\Delta'^{17}O$ records noted above (Fig. 1.)[54,169–171]. However, the key criterion for each of these systems is that they record a clear shift in redox conditions within a similar environment and that such a shift was sustained beyond strata deposited immediately above those that define the syn-GOE interval. That is, geochemical signals should show a marked departure from both Neoarchean and post-GOE records. Moreover, such signals should be found in multiple geographically disparate locations to build confidence that they are indeed global in nature. However, to date, in most cases, such signatures do not clearly distinguish the syn-GOE interval relative to the following mid-Proterozoic[169,172], are typically based on a single location[170,173], or show clear evidence of large diagenetic influences[174,175]. Even with respect to carbon isotopes in shallow marine carbonates, finding a clear record of an exit from the LJE within a continuous stratigraphic sequence remains elusive[156].

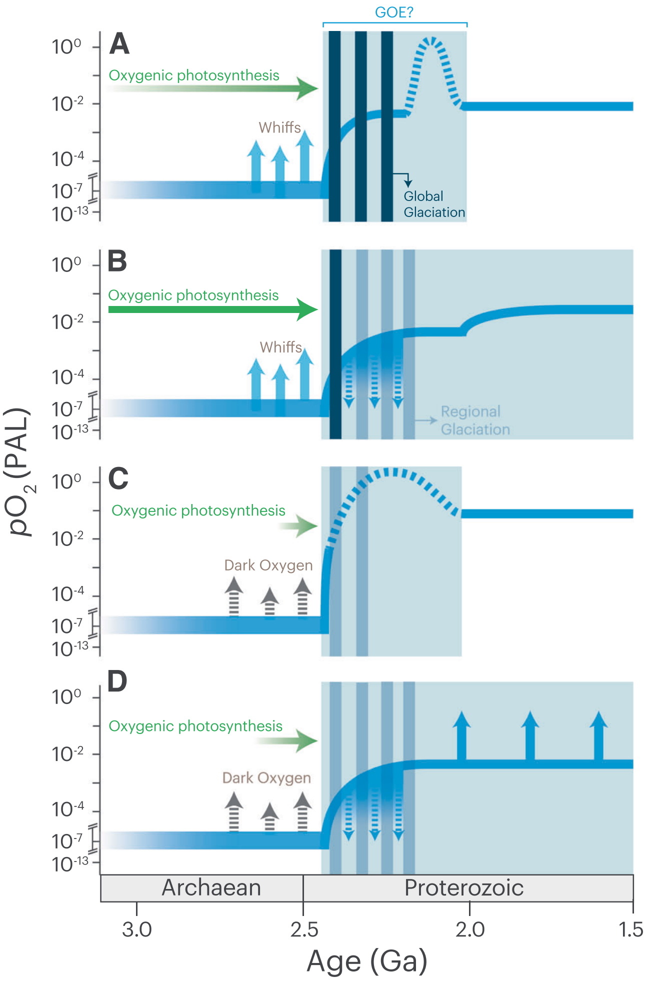

Fig. 3 | Defendable trajectories across Earth's Great Oxidation Event. In **A**–**D**, we present defendable trajectories of atmospheric oxygen together with possible Archean conditions leading up to the GOE. We note that different properties of each panel could be interchangeable and that these are a small subset of examples, which is not meant to be comprehensive.

Evidence against there being an end to Earth's GOE comes from multiple recent proxy studies that indicate oxic conditions after the end of the LJE. For example, large δ^{53}Cr signatures from the Mesoproterozoic have been interpreted to support a more protracted interval of oxygenation[53,176]. Such signals in the mid-Proterozoic do not only extend to the chromium system but also to I/Ca ratios, metal enrichments in shales, and molybdenum isotopes[176]. At present, such mid-Proterozoic signatures have typically been interpreted as transient oxygenation events. However, as noted above, the susceptibility to diagenetic alteration and challenges in deducing a possible sedimentary facies control of such signals may indicate that they are instead offering insight into mid-Proterozoic oxygen baselines and not excursions from it. Taking these inferences together with controversies surrounding the interpretation of the LJE and associated redox proxies, it remains unclear whether oxygen levels across the syn-GOE environment were truly much different than what followed.

One approach to explore possible carbon cycle and oxygen dynamics not only across the end-GOE but also throughout the GOE at large is through constraining the abundance of bioavailable phosphorus. Here, geochemical and model-based approaches have led to orders of magnitude uncertainties of this key property of ancient seawater[74,94,177,178,179]. At present, it appears that differing estimates are less a consequence of differing interpretations of specific data sets, but rather different data sets providing different estimates. For example, Fe-oxide, shale, and carbonate records have put forward very different reconstructions of ancient phosphorus, with Fe-oxide and shale records interpreted to record lower phosphorus levels relative to today[180,181], whereas carbonate-associated phosphate and apatite nano-particles suggest abundant phosphorus[94,178]. Some key next steps to better understand and possibly integrate such data sets will be to explore whether benthic records can be decoupled from shallow marine ones and how such records can be combined to offer a more holistic picture of ancient P-cycling. That is, whether some records, such as shales, are true passive records of phosphorus levels versus the possibility that if one measures lower phosphorus abundances, it could indicate more phosphorus in coeval seawater, sediment, or minerals such as phyllosilicates[182]. In the case of Fe-oxides, such records are challenging to interpret due to the suite of precursor minerals which would have initially formed from seawater, and how such minerals would have interacted with phosphorus during their initial precipitation all the way to burial, and mineral transformation during early and late-stage diagenesis is critical to constrain moving forward[106,107112]. Moreover, it is notable that today primary productivity is predominantly supplied with P through the recycling of organic-P in the water column. So far, the recycling efficiency is very poorly constrained for the early anoxic ocean with simple ecosystems[93,183]. While it is predicted that with increasing continental emergence, increased P would have been supplied to the ocean, it remains unclear how such inputs may have manifested as bioavailable P to the biosphere[17,177]. Continued progress on these areas, together with a focus on improving the mechanistic understanding of phosphorus cycling[184–187] across the array of possible conditions for the pre, syn, and post-GOE, may offer an important step forward in limiting possible oxygen trajectories (Fig. 2).

Moving forward

It has been almost one hundred years since arguably the most notable transformation of Earth's atmosphere was identified in the sedimentary record[7]. With all the advancements made over the past decades, it is worth revisiting the degree to which one can quantitatively or even qualitatively trace the specific trajectory atmospheric O_2 levels followed across the early Proterozoic. At this time, it appears there are numerous defendable paths that atmospheric O_2 may have taken in the earliest Proterozoic based on available data (e.g., Fig. 3). However, pooling proxies together to seek out a consensus view on such a trajectory can create the impression of certainty from many highly uncertain inferences, a phenomenon analogous to a mortgage-backed security. Although potentially insightful in some cases[1–3], such efforts may obscure sub-10s of millions of years of dynamics and environment-specific signals, which reduces the richness that the sedimentary record may offer. Below, we outline specific questions requiring further exploration in the coming years and decades.

The onset of the GOE. Was the GOE the product of the evolution of oxygenic photosynthesis? Its rapid expansion? Or was there some other set of circumstances which led to a dominance of oxygenic photosynthesis over other metabolisms? If oxygenic photosynthesis evolved 100 s of millions of years prior to the GOE, what is the significance of the syn-GOE build-up of atmospheric oxygen? Otherwise, is it possible that pre-GOE O_2 redox signatures in the sedimentary record are instead the product of dark oxygen production? Distilling the former set of questions into clear testable hypotheses will be important for making progress in this area and refining how oxygen fluxes may have operated on the Archean Earth.

The magnitude of the GOE. At this time, it remains challenging to quantify how high oxygen levels may have risen across discrete intervals within the GOE. Specifically, how high did oxygen rise above S-MIF and O-MIF thresholds after initial oxygenation[28–32]? At the center of this controversy is understanding what the LJE represents as either a reflection of global organic carbon burial or possibly a shift in local carbon cycling driven by local environment and metabolism[54]. Beyond specific thresholds is the degree to which oxygen fluctuated across the GOE. Does evidence of O_2 fluctuations infer that–at least for the first half of the

GOE–that atmospheric O_2 remained low, and even revisited Archean levels? How many Paleoproterozoic glacial intervals were there? Were any of them global panglacials? And how did they impact O_2 levels? Within these broad questions are specific geochemical and numerical lines of inquiry. For example, are there additional fluctuating S-MIF[42,46,47] signatures preserved in globally distributed units? And can remobilization of older S-MIF-bearing materials be definitively ruled out in the case of S-MIF reemergence[188]?

The end of the GOE. Did O_2 levels fall around 2.0 Ga? And was there a meaningful end to the GOE? At this time, with so much uncertainty regarding a specific O_2 trajectory across the GOE, it appears unclear whether one can confidently place a marker within the sedimentary record denoting when the GOE ended. Defining such a boundary will require a renewed focus on well-exposed and preserved Rhyacian and Orosirian, Paleoproterozoic records[189,190].

Reducing uncertainty across the GOE. In Fig. 3 we attempt to offer some examples of possible scenarios that depict very different trajectories of Earth's GOE, which are by no means exhaustive but demonstrate the current extent of uncertainty. In the coming years and decades, it should be a key goal to: (1) explicitly rule out competing scenarios such as those presented, (2) to decisively separate global, local, globally local, and diagenetic signatures within data sets, and (3) to revisit experimental underpinnings on which proxies are based. Community efforts are likely to be among the most promising paths forward to make meaningful progress on these questions[59–61]. No single formation will likely be able to provide definitive answers or present key global constraints on variables that may underlie oxygen accumulation across the GOE interval. In the absence of unambiguous direct proxies of atmospheric O_2, it is clear that renewed efforts will be required to combine indirect proxy evidence and refine existing uncertainties on such proxies. In addition, a major frontier in resolving competing GOE scenarios lies in shifting from static or threshold-based oxygen estimates toward fully dynamic Earth system modeling. Much of the existing modeling literature has focused on steady-state O_2 levels, critical thresholds (e.g., S-MIF collapse, ozone formation), or sensitivity analyses around prescribed parameter values. While these approaches have provided essential constraints, atmospheric O_2 is not simply a level to be estimated, but instead is the outcome of a nonlinear, feedback-rich system evolving over tens to hundreds of millions of years. Progress will require models that explicitly simulate how differing initial redox states, nutrient regimes, and reductant fluxes evolve through time to produce distinct O_2 trajectories. Rather than asking solely how high O_2 rose in the atmosphere, future work should seek to determine which trajectories are dynamically plausible given known feedback strengths and boundary conditions.

Equally important is the incorporation of evolving boundary conditions driven by deep Earth and tectonic processes. Secular changes in mantle redox state, volcanic and metamorphic reductant fluxes, continental growth, and crustal composition likely altered the global oxygen source–sink balance over the early Proterozoic. Expansion of continental area and shallow continental shelves may have increased the spatial footprint of primary productivity and organic carbon burial, while tectonic reorganization could have modified weathering regimes, nutrient delivery, and sedimentary burial efficiencies. Integrating these slow, tectonically driven boundary condition changes into dynamic oxygen models will be essential for evaluating whether oxygenation was triggered by internal feedback instabilities, gradual shifts in deep Earth forcing, or their interaction and combined effects. Together, these advances point toward a modeling framework focused less on reconstructing a single absolute atmospheric O_2 concentration and more on understanding the dynamical evolution of the oxygen cycle itself.

To conclude, the Paleoproterozoic occupies an interesting position within the geologic timescale. There is enough of a stratigraphic record to bring to light fascinating clues of how Earth's surface environment may have evolved from its early to middle history, but in some cases, not enough material (or related constraints) to move qualitative inferences into quantitative reconstructions. Although grass roots community-led efforts to pool resources and materials is reason for optimism, in some cases it is possible that intrinsic limits of an incomplete and altered sedimentary record may be identified. Constraining the stability structure, feedback strengths, and boundary condition evolution of the early Earth system may ultimately prove more decisive in resolving how 'great' the Great Oxidation was rather than refining any single proxy-derived oxygen estimate.

Data availability

Data sharing is not applicable to this article as no datasets were generated or analyzed for this study.

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Peter W. Crockford and Mojtaba Fakhraee conceptualized the manuscript. Ichiko Sugiyama and Lyle Nelson conceptualized figures with input from all co-authors. Ichiko Sugiyama produced figures for this manuscript. Peter W. Crockford, Ichiko Sugiyama, Michael Kipp, Sabs Wimmer, Jordon Hemingway, Jihua Hao, and Mojtaba Fakhraee contributed to writing and revising this manuscript.

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