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Subduction modulated the long-term oxygenation of Earth's surface

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Abstract:

On Earth, atmospheric oxygen is inferred to have risen over three major intervals before reaching modern levels, with each interval having a profound impact on the evolution of the biosphere. However, the principal driver behind these stepwise increases remains elusive. Here, we compile metamorphic thermobaric ratios (*T/P*) through time and use them as a first-order, probabilistic proxy for the likelihood of 'cold' subduction (i.e., with T/P < 375°C GPa^{-1}) during secular cooling of Earth's mantle. Then, we couple this tectonic forcing to biogeochemical modelling to test whether more efficient cold subduction may have enhanced the net transfer of reduced organic carbon and pyrite to Earth's deep interior, thereby diminishing oxygen sinks and allowing surface oxygen levels to increase at geological timescales. Modelling results indicate that the progressive emergence of cold subduction could plausibly have contributed to the long-term oxygenation trajectory and associated secular trends in atmospheric carbon dioxide, seawater sulfate, sedimentary phosphorus, and marine redox conditions. Although the absolute magnitudes remain uncertain, the predicted trajectory of surface oxygenation is qualitatively consistent with the broad three-step pattern inferred from geochemical proxies. We propose that the progressive evolution of subduction may have been a key driver of long-term surface oxygenation, linking mantle cooling to the rise of conditions favorable for aerobic lifeforms.

Significance Statement:

How Earth acquired its oxygen-rich atmosphere remains a pivotal question in Earth science. By analyzing secular changes in the thermal state of subduction zones inferred from metamorphic thermobaric ratios (*T*/*P*), we evaluate whether the emergence of cold subduction could have progressively enhanced the atmospheric oxygen level via the net transfer of oxygen sinks (i.e., organic carbon and pyrite) into the mantle. Coupled biogeochemical modelling provides a quantitative framework linking mantle cooling, subduction redox fluxes, and long-term surface oxygenation. Our results offer a mechanistic hypothesis that helps place diverse geochemical records within a common Earth-system context, and suggest that secular evolution of subduction may have led to the development of conditions favorable for aerobic lifeforms.

Main Text:

Earth's atmosphere attained its present level of oxygen (O_2 = 21 vol.%) via three major steps (Fig. 1*A*) that ultimately led to a planet habitable to lifeforms of immense diversity and complexity, including humans (1, 2). The first major increase in atmospheric oxygen, the Great Oxygenation (or Oxidation) Event (GOE), occurred in the Paleoproterozoic Era (3, 4) between c. 2.43 and 2.0 billion years ago (Ga). The GOE includes the Lomagundi–Jatuli Event (c. 2.22–2.06 Ga), the largest positive carbonate carbon isotope ($\delta^{13}C_{carb}$) excursion in Earth history (5, 6) (Fig. 1*A, G*), during which the partial pressure of oxygen (pO_2) rose from <10^{-6} of the present atmospheric level (PAL) to 10^{-2} PAL or higher (6–8). The GOE enabled oxygenation of the surface ocean and increased mobilization (via oxidative weathering) and accumulation of redox-sensitive trace elements in marine sediments (8–10) (Fig. 1*B*), ultimately paving the way for the emergence of eukaryotic life (11) (Fig. 1*C*). Thereafter, pO_2 likely remained at levels between ~10^{-3} to 10^{-1} PAL for the next billion years or so (c. 1.8–0.8 Ga; commonly known as the 'Boring Billion') (Fig. 1*A*), during which deep water masses were mostly anoxic and ferruginous (12) (Fig. 1*B*).

The second proposed rise in atmospheric oxygen, commonly termed the Neoproterozoic Oxygenation Event (NOE)**,** occurred broadly from c. 0.8 to 0.52 Ga and was a potential trigger of the Ediacaran to Cambrian biotic explosion of metazoans, although its timing, tempo, and even coherence as a single 'event' remain debated (13–15) (Fig. 1*C*). The NOE was likely characterized by a high degree of instability in atmospheric and marine oxygen levels (Fig. 1*A*), as evidenced by extreme variability in the concentration of redox-sensitive elements in marine sediments (8–10) (Fig. 1*B*) and isotope mass balance modelling, which indicates that pO_2 likely fluctuated in the range of 0.1 to 0.6 PAL (16, 17). A third increase to present-day O_2 levels, termed the Paleozoic Oxygenation Event (POE), occurred during the mid to late Paleozoic (c. 0.45–0.25 Ga) (18) and is suggested to have driven an increase in the body size of organisms and the emergence of more diverse and specialized predators in the Devonian (19) (Fig. 1*C*).

Multiple explanations for the drivers of atmospheric oxygenation have been proposed. The evolution of oxygenic photosynthesis (20) and the subsequent succession of dominant primary producers from algae to vascular plants (21, 22) undoubtedly had a major impact on the production and accumulation of O_2 through time via burial of organic carbon (Fig. 1*C*). A transition from more mafic to more felsic continental crust at c. 2.7–2.5 Ga (23), an abrupt reduction in submarine volcanism after c. 2.45 Ga (24), and oxidation of the upper mantle (25) have all been proposed as potential links to the GOE via a decrease in sinks for O_2. Photochemistry (26), mineral–water interface reactions (27), hydrogen escape (28), and an increase in the length of a day associated with Earth's decreasing rotation rate (29) may also have contributed to the accumulation of atmospheric O_2. However, it is unclear how these processes could explain the long-term trajectory of Earth-surface oxygenation, including the potential step changes represented by the GOE, NOE and POE, such that the first-order driver of the long-term oxygenation of Earth's surface remains elusive.

Here, we use the record of thermobaric ratios (T/P) from metamorphic rocks (n = 876) formed over the past four billion years (30, 31) (Figs. 1*D* and *SI Appendix*, Fig. S1) to track the evolution of subduction styles on Earth and evaluate their potential impact on secular changes in deep carbon (and sulfur) cycling. When combined with biogeochemical modelling, this allows us to evaluate the effects of changes in subduction style as a driver of long-term oxygenation of the Earth's surface, and ultimately the development of the modern habitable world. Because the metamorphic record is incomplete and biased by preservation, we treat the T/P time series as a first-order probabilistic metric, and impose it as the sole time-varying forcing in a minimalist sensitivity experiment to generate falsifiable, first-order predictions.

Subduction style and the deep carbon cycle

Since the Archean, subduction of carbon has played a pivotal role in modulating Earth's climate and habitability, as well as the formation of deep (sub-lithospheric) diamonds (32) (Fig. 2*A*). On the modern Earth, subduction of carbonate and reduced species, i.e., organic carbon (OC) and pyrite, is highly variable and largely a function of geological happenstance (33). Carbon can be lost from the subducting slab by metamorphic decarbonation and partial melting, processes that are mainly controlled by pressure (P)–temperature (T) conditions and fluid availability and composition (33) (*SI Appendix*, Fig. S1). Although individual T/P estimates have associated uncertainties related to T (~50 ºC) and P (~0.1–0.2 GPa), the global distribution of T/P through time has been argued to capture first-order shifts in metamorphic thermal regimes (34). Therefore, we interpret secular changes statistically and use T/P

as a probabilistic proxy for the likelihood of subduction. Warmer subduction favors shallower devolatilization and melting of the descending slab top as well as oxidation and dissolution of OC in fluids, whereas cooler subduction allows deeper transport of carbonate and OC into the sub-arc mantle (35) (Fig. 2*A*).

Subduction zones act as net sinks of carbon on timescales of a few tens of millions of years (Myr), whereas at longer timescales (>50 Myr) subducted carbon is either recycled via volcanism and weathering or is sequestered into the mantle (35). Carbonate mostly breaks down during the subduction process, whereas OC (as graphite or diamond) and other reduced species can be transported through the mantle transition zone into the deep mantle (32), resulting in a net addition of O_2 at the Earth's surface. Subsequently, this deeply subducted OC may be entrained by mantle upwellings (including plumes) then released back to the surface through ocean-island volcanism (36). This cycling of OC from the surface to the deep mantle and back (the deep carbon cycle), which occurs on timescales associated with mantle mixing (37), has likely exerted a first-order control on long-term atmospheric O_2 buildup (see *SI Appendix* for further details).

Secular evolution of subduction style and surface oxygenation

The formation of the supercontinent Nuna/Columbia during the Paleoproterozoic is widely recognized as evidence that plate tectonics, driven mostly by subduction of oceanic lithosphere, has been Earth's tectonic mode at least since the late Archean (38) (Fig. 1*E*). However, subduction can also occur without being linked to plate tectonics, for example in sluggish lid or episodic tectonic modes that may have operated during the Archean due to higher mantle temperatures (39). Thus, subduction may have occurred prior to c. 2.5 Ga, but in a style that was episodic and short-lived (30, 40, 41). By contrast, since the Archean, subduction has probably been operating continuously as a necessary complement to seafloor spreading, allowing regional metamorphic terranes to be formed and preserved due to subduction-to-collision orogenesis at convergent plate margins (42).

To evaluate Earth-surface oxygenation impacts due to changes in subduction style linked to secular cooling, we constructed a time-series of metamorphic thermobaric ratios using *T*, *P* and age information from globally distributed localities with metamorphic rocks younger than the Hadean (<4.03 Ga; Fig. 1*D* and *SI Appendix*, Fig. S1; Materials and Methods). A direct proxy for the secular evolution of subduction style is provided by the temporal distribution of low *T/P* metamorphic rocks (those with *T*/*P* < 375°C GPa^{-1}), as represented by most eclogites and blueschists (30, 31). Because individual *T*/*P* estimates reflect complex *P*–*T*–*t* paths and can be influenced by exhumation rate and the balance between advective and conductive heat transfer, we interpret the *T*/*P* record statistically — we do not treat any single locality as uniquely diagnostic of tectonic mode. Since the Archean, the upper mantle has been cooling at a rate of ~100°C/Gyr (43). This cooling is consistent with an increasing proportion of localities characterized by low *T/P*, a decrease in average *T/P* ratios with time, the progressive development of bimodality in *T/P* ratios characteristic of convergent-margin metamorphism (see the upper panel of Fig. 1*D*), and the appearance of orogenic eclogites in the Paleoproterozoic and ultrahigh pressure metamorphic rocks and blueschists since the Cryogenian (44).

Low *T*/*P* values are observed during two major intervals (Fig. 1*D*): (1) in the Paleoproterozoic (c. 2.2–1.8 Ga), and (2) from the mid-Neoproterozoic to the present day (<0.8 Ga), broadly coincident with the purported GOE and the NOE–POE, respectively (Fig. 1*A*). The first period with low *T/P* values is consistent with the emergence of cold subduction associated with formation of Nuna/Columbia by plate convergence and collisional orogenesis (42) (Fig. 1*D*–*E*). Despite their scarcity, these localities are widely distributed and we argue to reflect an overall decrease in thermobaric ratios related to stable (continuous) rather than unstable (short-lived or episodic) subduction, which would have aided delivery of OC (and pyrite) to the deeper mantle on a global scale. However, the tectonic cycle that led to the formation of Nuna/Columbia and preservation of the earliest eclogites after c. 2.2 Ga may have begun some time earlier with progressive continental breakup. Based on the average lifespan of 186 Myr for passive margins initiated during the Neoarchean to Paleoproterozoic (>1.6 Ga) (45), we argue that this cycle likely began at c. 2.4 Ga. Notably, there is a small concomitant increase in the frequency of carbonatite and kimberlite magmatism during the mid-Paleoproterozoic (c. 2.1–1.7 Ga) (46) (Fig. 1*F*) that is offset by c. 100 Myr toward younger ages relative to the *T/P* metamorphic minimum (Fig. 1*D*), likely reflecting the time taken for slabs to sink through the mantle (47). In addition, eclogitic inclusions in Proterozoic diamonds record low $\delta^{13}C$ values between –10‰ and –5‰ (48, 49), consistent with subduction of isotopically-light surface carbon to the deep mantle (50) (Fig. 1*G*). Lastly, a peak in black shale and graphite deposition associated with Paleoproterozoic orogens (51) would have further contributed to OC burial and rising atmospheric oxygen levels, coincident with the Lomagundi–Jatuli Event (52).

Following the Boring Billion, a period likely associated with a slowdown of mantle convection and tectonic plate motion (45, 47), a second period characterized by low *T/P* values (c. 0.8 Ga to present) corresponds to the appearance of blueschists and ultrahigh pressure metamorphic rocks in the continental record, consistent with the emergence of modern-style plate tectonics (30, 35, 53) (Fig. 1*D*). Notably, after the initial drop in *T/P* associated with the NOE at c. 0.8 Ga, there was a further drop from c. 0.45 Ga to a nadir at c. 0.3–0.25 Ga, related to the POE. In both cases, a delay between the initiation of ocean-basin closure by subduction and the preservation of metamorphic rocks with low *T/P* during subsequent collision, is broadly consistent with the lifespan of the associated passive margins (averages of 174 and 137 Myr, respectively) for the Neoproterozoic and Cambrian-to-Carboniferous periods (45). Coeval with these changes were marked increases in carbonatite and kimberlite magmatism (46) (Fig. 1*F*), consistent with increasingly efficient delivery of carbon to the deep mantle.

Quantitative modelling

To test the hypothesis that the emergence and progressive evolution of cold subduction since the Archean contributed to the long-term oxygenation of Earth's surface, we integrated subduction style and mantle degassing into the COPSE biogeochemical model (54), which tracks the global cycles of C, S, O, N and P across a three-layered system comprising surface (atmosphere, ocean and biosphere), crust and mantle reservoirs (*SI Appendix*, Fig. S3). We introduced a key parameter, f_{cold}, in the model to estimate the development of cold subduction through time and its impact on the evolution of pO_2 (Figs. 2*B* and 3). This parameter represents the ratio between the number of low *T/P* localities and the total number of localities in the metamorphic dataset for a particular time interval normalized to the value for the last 100 Myr (0.63), taken as representative of modern-style plate tectonics (i.e., f_{cold} varies between 0, meaning no cold subduction, and a value of 1.2, corresponding to a maximum in cold subduction during the Carboniferous–Permian, see Fig. 3*A*). In this model, f_{cold} is treated as a first-order probabilistic metric rather than a deterministic mapping from *T/P* to a unique tectonic mode. At lower f_{cold}, inefficient subduction leads to greater recycling of subducted OC (and pyrite) through shallow volcanism and metamorphism, and to enhanced consumption of O_2 by reducing C–S–Fe fluids released from, for example, upwelling mantle plumes, maintaining low atmospheric pO_2. By contrast, at higher f_{cold}, efficient subduction enhances the transfer of OC (and pyrite) to the deep mantle, which in turn allows atmospheric pO_2 to rise (see Materials and Methods, *SI Appendix*, Table S1–S4 for further details). In the modelling, we hold all other COPSE model parameters at their pre-defined present-day steady-state values, imposing only the f_{cold} forcing derived from the metamorphic *T/P* record, to isolated the effect of a single tectonic forcing (i.e., cold subduction) within the simplified Earth system. As a result, we interpret the model outputs primarily in terms of broad secular trends rather than precise reconstructions of absolute paleo-pO_2 levels.

To evaluate the effect of varying f_{cold} on atmospheric pO_2, we ran our modified COPSE model backward in time for f_{cold} values varying from 10^{-6} to 1.2, with all other model parameters remaining at present steady-state values. For each value of f_{cold}, step-change experiments show that the modelled pO_2 relaxes toward a new, lower steady state value within c. 0.2–0.8 Gyr (Fig. 2*B*), basiclly consistent with the long response time of the mantle C–S reservoirs (55). Notably, there is a linear relationship between f_{cold} and the time-dependent response of new pO_2 steady states (Fig. 2*C*). For example, at f_{cold} ~0, buried OC (and pyrite) is totally recycled, resulting in pO_2 being maintained at very low levels <10^{-6} PAL, consistent with estimates for the Archean (8). Similarly, levels of pO_2 corresponding to the GOE (ca. 10^{-3}–10^{-1} PAL) occur when f_{cold} increases to values of 10^{-3}–10^{-1}. At f_{cold} values of 0.4–0.7, the predicted pO_2 levels correspond to the NOE–POE (16–18), before evolving to modern pO_2 levels at f_{cold} = 1 (Fig. 2*B*–*C*).

Next, we examined the potential of our model to simulate the oxygenation history of Earth's surface using a time-reversed evolution of f_{cold} (Fig. 3*A*) as defined by the metamorphic *T/P* data (Fig. 1*D*), again with all other parameters at present-day levels. Using Monte Carlo uncertainty propagation, we explicitly represent the uncertainty in the f_{cold} forcing, providing quantitative confidence bounds on trajectories and sensitivities (Fig. 3*B*–*F*) (see Materials and Methods for a full description of f_{cold} and Monte Carlo setup). Our model shows that, going backward from modern values, pO_2 decreases in two periods, including a stepwise decrease in pO_2 from the present day to c. 1.0 Ga, variable pO_2 during the 'Boring Billion', and then a 'great oxidation' of the atmosphere between c. 2.0 and 2.4 Ga, eventually predicting an Archean state with only trace levels of oxygen (<10^{-6} PAL) (Fig. 3*B*). Notably, the inset in Figure 3*B* shows a decrease in modelled O_2 from ~19 to ~12 mol.% at c. 0.4–0.6 Ga, supporting a distinct POE. Although by necessity qualitative, our modelled evolution of pO_2 through time shows broad agreement with other proxies over much of the past four billion years (Fig. 3*B*).

To test the robustness of our model output, we compared our predicted atmospheric partial pressures for carbon dioxide (pCO_2), concentrations of sulfate ($[SO_4^{2-}]_{sw}$) in seawater and sedimentary phosphorus abundance (Fig. 3*C*–*F*) with measured geochemical proxies and results from previous modelling studies. Moving back through time, modelled pCO_2 increases to ~100 PAL at c. 1.0 Ga and then progressively climbs to ~600 PAL by the late Archean (Fig. 3*C*), in broad agreement with previous estimates (56). Superimposed on this trajectory are two notable declines at c. 0.3–0.4 Ga and c. 2.1–2.4 Ga (Fig. 3*C*), which likely correspond to well-documented pCO_2 drawdowns during the Carboniferous and GOE, respectively (54,56). Predicted $[SO_4^{2-}]_{sw}$ decreases in a stepwise fashion to trace levels (<0.2 mM) (Fig. 3*D*), which broadly mimics reconstructions based on S-isotope and mass-balance modelling (57, 58). Modelled burial fluxes of marine organic (mopb) and authigenic (capb) phosphorus capture the broad secular records of phosphorus concentrations in marine shales and phosphorite occurrences (59), notably including two peaks during the Neoproterozoic-to-Paleozoic and Paleoproterozoic (Fig. 3*E*). The degree of ocean anoxia (ANOX) shows a monotonous increase from ~0.0025 (fully oxidized) to ~1 (fully anoxic) at ~0.4–0.6 Ga, but a pulsed decrease to 0.3 at 2.0–2.4 Ga (Fig. 3*F*), in line with documented trends for redox-sensitive elements in marine sediments (8–10) (Fig. 1*B*).

Notwithstanding the generally good fit of our calculations with existing proxies, with all other parameters held at present-day levels, our models show clear mismatches with existing estimates of O_2 levels at c. 0.5–0.7 Ga (approximating the NOE) (13, 16) and 1.4–1.7 Ga (1, 17) (Fig. 3*B*). They also fail to produce second-order fluctuations in surface environment, for example, the predicted $[SO_4^{2-}]_{sw}$ and capb values are higher than existing estimates during the interval from c. 0.8 to 1.8 Ga (the 'Boring Billion'). As the f_{cold} forcings vary only on tectonic timescales (≥10^8 years), these mismatches may reflect other short-term or local geological forcings (12, 58). Accordingly, we interpret the COPSE experiments only at tectonic timescales and do not aim to reproduce poorly-understood high-frequency changes within the 'Boring Billion' and/or the NOE (8–10, 12).

Discussion

Is the metamorphic record reliable?

Before interpreting secular changes in *T/P*, we acknowledge that metamorphic thermobaric ratios are an informative but imperfect constraint on past tectonic thermal regimes. Individual *T/P* estimates can be influenced by, for example, the broader *P–T* evolution of individual samples and their subsequent metamorphic overprinting, along with inevitable preservation and sampling biases (60). In particular, for low *T/P* eclogites and blueschists, prolonged residence at depth in the Precambrian increase the likelihood of overprinting (61). Thus, the absence of low *T/P* in older terranes cannot necessarily be interpreted solely as a lack of suitable protoliths or formation conditions. Consequently, our approach relies on the statistical behavior of a large, global compilation, treating low *T/P* occurrences as probabilistic indicators of the likelihood of cold subduction through time.

Previous numerical modelling has tested whether these rocks could have been produced in the past and, if so, whether they were lost to the mantle rather than being exhumed (39,40). Modelling results suggest that ultrahigh pressure eclogites were unlikely to have been produced and returned to crustal levels before the Neoproterozoic (39,40), consistent with isotopic evidence for an upper continental crustal component in the plume source only since the Precambrian (62). Secular change in protolith composition has been speculated as the cause for the absence of blueschists before the late Tonian (63). However, an alternative explanation is that suitable protoliths existed earlier, as the low-MgO compositions required for glaucophane-bearing mineral assemblages (≤11.2 wt % MgO) comprise more than half of the metabasalts analyzed from greenstone belts of post-Paleoarchean age (64). This suggests that the absence of earlier blueschists may have been primarily due to warmer subduction conditions rather than a lack of suitable protoliths (61,65). Furthermore, low-MgO metabasalts suitable for glaucophane-bearing assemblages are present in the preserved record since at least the Paleoarchean (62) and occur, for example, within block-in-matrix mélanges along an inferred arc–passive continental margin suture formed at ca. 2.5 Ga in the North China Craton (66, 67).

The survival of some Paleoarchean crustal terrains (e.g., the East Pilbara Terrane of the Pilbara Craton and the Barberton greenstone belt in the Kaapvaal Craton) (68) that escaped subsequent deep burial and erosion, and which do not preserve low *T/P* rocks, provides circumstantial weight to the interpretation that such rocks might not have been formed. It has also been suggested that eclogites might have been overlooked in the Archean record because they were overprinted by retrogressive mineral assemblages during exhumation (69,70). Notwithstanding, and regardless of age, retrogressed eclogite is distinctive because of the presence of a fine-grained vermicular intergrowth of diopside and sodic plagioclase replacing sodic pyroxene during exhumation (71). This microstructure is well preserved in Paleoproterozoic eclogites in the Belomorian Province in Russia (72) and the Snowbird tectonic zone in Canada (73), but has not been reported from Archean rocks. Thus, we argue that it is doubtful that the absence of orogenic eclogites in the Archean record is related simply to erosion or overprinting. Lastly, we note that the distribution of localities within the metamorphic dataset with *T/P* < 375ºC GPa^{-1} does not follow an exponential abundance distribution, as would be expected from a standard survivorship model (74).

The metamorphic dataset used in this study comprises 876 localities that cluster at times of supercontinent (<0.85 Ga, 0.85–1.35 Ga, and 1.35–2.25 Ga) or supercraton formation (>2.25 Ga). Of 124 localities older than 2.25 Ga, none record *T/P* < 375 °C/GPa, whereas 12 of 165 localities (7.3%) from 2.25 to 1.35 Ga, 1 of 72 localities (1.4%) from 1.35 to 0.85 Ga and 261 of 515 localities (40.7%) younger than 0.85 Ga do. Thus, there are two apparent step increases in the proportion of low *T/P* localities, the first during the early Paleoproterozoic and the second during the late Tonian. This contrasts with the steady decline in the surface area of continental crust on average per era or eon from 11.4% per 100 Myr for the Cenozoic to Mesozoic eras to 0.7% per 100 Myr for the Archean Eon (75). The two-step increase in the proportion of low *T/P* metamorphism since the Archean is consistent with a change from unstable (short-lived) to stable (continuous) subduction in the early Paleoproterozoic and to colder thermal gradients and deeper slab breakoff after the Tonian (76). Thus, while preservation bias is a concern, we contend that the statistical trends in our dataset record first-order changes in the evolution of mantle thermal structure and style and propensity of subduction.

The history of oxygenation of Earth's surface

Assuming that the record of metamorphism is broadly representative, we posit a series of linked events that enabled the rise of pO_2 in the early Paleoproterozoic. Although parts of some cratons had emerged above sea level during the Archean (77), their emergence globally was seemingly a late Archean phenomenon (78). The resulting increase in nutrient supply to the oceans could have promoted a dramatic expansion of the mass of marine cyanobacteria that led to the widespread occurrence of OC-rich black shales and graphite deposits within Paleoproterozoic orogens (51,52) (*SI Appendix*, Fig. S2*C*). An association of platform carbonates on passive margins with the breakup of the late Archean continental landmass (45), alongside evidence for increased subduction of sediment since the Archean (79), support a link between the growing importance of cold subduction (and rigid plate tectonics) and the rise of pO_2.

The decrease in the number of localities preserving low thermobaric ratios during the 'Boring Billion' (1.8–0.8 Ga) is argued to reflect a period of decreased mantle convection and plate tectonic activity with a diminished particulate weathering flux, thus starving trenches of lubricating sediment (47). In these circumstances, although it is likely that less OC was buried at convergent plate margins, occurrences of low *T/P* metamorphism associated with the Grenville Orogeny between ~1.1 and 0.9 Ga indicate local subduction-driven orogenesis, which may have prevented pO_2 dropping to particularly low levels (Fig. 3*B*). By contrast, the establishment of the modern-style plate tectonics involving global cold, deep subduction during the Gondwana–Pangea assembly (80) may have created a tectonic feedback that pushed Earth's surface environment toward persistently more oxygen-rich conditions that have been maintained to the present day, largely driven by organic matter (and pyrite) subduction around the margins of the Pacific Ocean (81). In this context, the decrease in $\delta^{13}C$ values of kimberlites and ultramafic lamprophyres since the Cambrian (Fig. 1*G*) can be interpreted to reflect an increase of isotopically-light carbon transported from the surface to the deep mantle during the formation of Pangea (49). Further support is provided by increases in the oxygen fugacity (ΔFMQ) (25) and whole rock $Fe^{3+}/\Sigma Fe$ (82) of mantle at c. 2.1–1.8 Ga and c. 1.1–0.8 Ga (*SI Appendix*, Fig. S2*A, B*), which overlapped with periods of lower thermobaric ratios (Fig. 1*D*). We note that the oxidation of the subarc mantle via subduction of surficial sediments and altered oceanic crust would have increased the oxygen fugacity of arc volcanic gases, decreasing their overall demand for O_2 (83,84), whereas our hypothesis concerns the net long-term export of reduced carbon and sulfur to the deep mantle, which removes reducing power from surface systems, processes that are complementary rather than contradictory.

Our results cannot incorporate the short-term effects of varied biological and tectonic processes related to Earth's climate or redox state. For example, the break-up of the Rodinia supercontinent would have increased the length of continental margins and the global area of nearshore environments and rifted basins (85), all of which serve as potential traps for carbon and sulfur burial. The invasion of plants with roots in the early Paleozoic undoubtedly enhanced continental weathering and erosion (22), thus facilitating sediment transport to oceanic trenches through turbidity currents and deep-sea fans (86). The expansion of pelagic calcifiers over the last 150 Myr are argued to have boosted deep seafloor carbonate deposition, subduction and degassing (87). These processes all operated on top of the baseline defined by the net flux of carbon (and sulfur) between Earth's interior and exterior, which we argue was controlled by the evolving efficiency of cold subduction on a cooling Earth. Despite the complexities, our study suggests that the progressive emergence of subduction exerted a first-order influence on the long-term oxygenation of Earth's surface, which was essential to the emergence of all complex life (1, 2).

Materials and Methods

Metamorphic dataset. In this study we use the 04-21-2025 update of the metamorphic T, P and age dataset based on ref. 30 (*SI Appendix*). This dataset comprises 876 localities from the late Eoarchean to the present. Here we provide age, T, P and T/P data for all localities in an online dataset (Dataset S1: Metamorphic thermobaric ratios throughout Earth's history, with the data shown as plots of T, P and T/P vs age in *SI Appendix*, Fig. S1). We note that peak P–T estimates carry method-dependent uncertainties and may be affected by disequilibrium and retrogression/closure effects. As a consequence, our analysis emphasizes the statistical distribution of T/P through time rather than concentrate on individual datum.

COPSE biogeochemical modelling. We calculated how the relative efficiency of cold subduction (T/P < 375°C GPa^{-1}) could have affected the long-term oxygenation of the Earth surface using the COPSE global biogeochemical model, which is solved in MATLAB using a variable-step variable-order implicit scheme for stiff systems of ordinary differential equations. A key modification relative to previous versions of the COPSE model is the introduction of a dynamic cold subduction flux to transfer C- and S-bearing sediments to Earth's interior, and compensatory mantle degassing fluxes of C and S to balance their subduction fluxes (*SI Appendix*, Fig. S3). Accordingly, we present these simulations as sensitivity experiments (proof-of-concept) rather than calibrated reconstructions of absolute paleo-levels. Here, we provide the key details of our modelling approach, while the full model description is provided in the *SI Appendix*, Table S1–S4.

Relative efficiency of cold subduction through time. Quantification of the transport efficiency of sediments from the Earth's surface to its interior lies at the heart of this study. Here, we extrapolate the relative efficiency of deep subduction of carbon and sulfur on the modern Earth to the past. We apply a calibrated parameter, 'f_{cold}', to define the relative efficiency of cold subduction in the past relative to the present, which represents the proportion of the total burial of carbon and sulfur that becomes deeply subducted. In all experiments, f_{cold} is the only imposed time-varying tectonic forcing; all other COPSE parameters are held at their published present-day steady-state values (except mantle degassing scaling, which follows an independent thermal-history constraint). This design intentionally minimizes tunability and helps reduce non-uniqueness. Variability in the f_{cold} parameter through time was calculated from the normalized ratio of the frequency of low *T/P* (<375°C/GPa) to total *T/P* localities in each time bin, as follows:

$$f_{cold} = \frac{\varphi_{low}}{\varphi_{total}} \cdot \frac{\varphi_{total_0}}{\varphi_{low_0}} \qquad [1]$$

where φ_{low} and φ_{total} are the numbers of low *T/P* to total *T/P* localities in a given time bin, respectively, and φ_{low_0} and φ_{total_0} are the numbers of low *T/P* and total *T/P* localities in the last 100 Ma. In this study we used a time bin of 100 Myr. As the highest proportion of low *T/P* localities occurs in the Carboniferous–Permian, this parameter reaches a maximum of ~1.2 during that period (Fig. 3*A*).

The value of f_{cold} is defined by probabilistic logic concerning the relative proportions of subduction zones representing different styles of subduction, potentially circumventing challenges when using *T*/*P* data to drive the COPSE model. Clearly, there is an inherent preservation bias in early Earth rocks, where plate tectonic activity progressively destroys evidence of its own past, potentially leading to a decrease in the presence and/or abundance of particular rock or mineral associations related to convergent margin settings as we look further back in time (30).

We assume that the subduction rate of C and S species are linearly dependent on the normalized crustal reservoir sizes and on f_{cold}, following the existing method of flux determination in models like

COPSE. The subduction equations for organic carbon (G), carbonates (C), pyrite (PYR) and gypsum (GYP) are:

$$G_{sub} = k_{ocsub} \cdot \frac{G_t}{G_0} \cdot f_{cold} \quad [2]$$

$$C_{sub} = k_{ccsub} \cdot \frac{C_t}{C_0} \cdot f_{cold} \quad [3]$$

$$PYR_{sub} = k_{pyrsub} \cdot \frac{PYR_t}{PYR_0} \cdot f_{cold} \quad [4]$$

$$GYP_{sub} = k_{gypsub} \cdot \frac{GYP_t}{GYP_0} \cdot f_{cold} \quad [5]$$

Parameters *k* with subscripts represent today's rates of organic carbon, carbonate, pyrite and gypsum subduction, and $\frac{G_t}{G_0}$, $\frac{C_t}{C_0}$, $\frac{PYR_t}{PYR_0}$ and $\frac{GYP_t}{GYP_0}$ are the normalized crustal reservoirs of organic carbon, carbonate, pyrite and gypsum, respectively.

Following the formulation of other processes in COPSE, we assume that subduction fluxes are balanced by the rates of mantle degassing to maintain a steady state at the present day. If the mantle is modelled as a convecting layer with heat-dependent viscosity, the production of hydrothermal fluids (i. e., C-, S-, Fe-bearing fluids and melts) is likely to have decreased with secular mantle cooling. Here, we scaled the release of these mantle-derived species to mantle carbon and sulfur degassing rates (MCDEG and MSDEG) (Fig. S2*D*), which are imposed as dimensionless forcings and mirror the history of heat flow from the Earth's interior. The mantle degassing equations for organic carbon and pyrite also include an oxygen-dependent feedback ($\frac{O_t}{O_0}$, normalized atmospheric O_2 level) that prevents oxidation if atmospheric oxygen concentration is extremely low:

$$C_{deg_m} = k_{ccdeg_m} \cdot MCDEG \quad [6]$$

$$G_{deg_m} = k_{ocdeg_m} \cdot MCDEG \cdot \frac{O_t}{O_0} \quad [7]$$

$$PYR_{deg_m} = k_{pyrdeg_m} \cdot MSDEG \cdot \frac{O_t}{O_0} \quad [8]$$

$$GYP_{deg_m} = k_{gypdeg_m} \cdot f_{cold} \quad [9]$$

where MCDEG follows the estimates of ref. 55 and references therein. For the case in which continents grew episodically from 10% to 80% of the current volume between 3.2 and 2.5 Ga, high-temperature heat flow is calculated to decrease from nine times the current level at 3.8 Ga, to 4.9 times at 3.2 Ga and 2.3 times at 2.5 Ga, thereafter declining exponentially. MSDEG is approximated relative to today with a polynomial fit of the history of heat flow from the Earth interior (89) as follows,:

$$MSDEG = 1 + 0.1217t + 0.0942t^2 \quad [10]$$

where *t* is time before present in Ga. Note that $\mathrm{GYP}_{\mathrm{deg}_\mathrm{m}}$ does not depend on MSDEG, and is instead linearly dependent on f_{cold}. This is because the mantle sulfate reservoir was extremely limited on the early Earth and $\mathrm{GYP}_{\mathrm{deg}_\mathrm{m}}$ was therefore likely controlled directly by the subducted flux of gypsum.

We modified the crustal degassing flux by including a dynamic 'rapid recycling' module to allow the buried carbon and sulfur that has not been subducted deeply to rapidly return to the atmosphere-ocean system. The crustal degassing equations were altered to remove the implicit equations present in the original COPSE model, to be replaced with a budget between burial, weathering and subduction rate at each time step. The new crustal degassing equations are as follows:

$$G_{deg_c} = mocb + locb - oxidw - G_{sub} \quad [11]$$

$$C_{deg_c} = mccb + sfw - carbw - C_{sub} \quad [12]$$

$$PYR_{deg_c} = mpsb - pyrw - PYR_{sub} \quad [13]$$

$$GYP_{deg_c} = mgsb - gypw - GYP_{sub} \quad [14]$$

where *sfw* is the seafloor weathering flux and *oxidw*, *carbw*, *pyrw* and *gypw* are the subaerial weathering rates already in COPSE. This modification modelled the scenario that an increase in the cold subduction flux would not necessarily enhance carbon and sulfur accumulation on continental shelves, whereas it would lead to a corresponding increase in carbon and sulfur sequestrations within the mantle reservoir.

Modern steady-state computations. Our modified COPSE model was run backwards in time to examine Earth-surface dynamics over the 4-billion-year interval from the present day to the beginning of the Eoarchean. We first ran this model to a modern-like steady state, with pO_2 = 1.0 PAL, pCO_2 = 1.0 PAL, $[SO_4^{2-}]_{sw}$ = 1.0 POL (present oceanic level), and f_{cold} = 1.0, maintaining it for 200 million years as the baseline, and we then imposed a dynamic decrease in f_{cold} yielded from the actual time-series record of T/P ratios lasting for 4.0 Gyr. All other dimensionless parameters (e.g., land area, river runoff, uplift, life evolution) were maintained at the present level during model runs but were perturbed in the sensitivity tests. All the reservoir sizes are in moles, and fluxes are in moles per Myr. Numerical integration used a stiff solver, and we verified stability over the explored forcing ranges: no singularities or unphysical states (e.g., negative reservoir sizes) occurred, and solutions evolve smoothly to new steady states under both step and time-varying forcings.

Monte-Carlo setup. To capture parametric uncertainty, we performed a Monte-Carlo analysis with 1,000 COPSE model runs, in which the key parameter f_{cold} was randomly sampled from an uncertainty range using a uniform distribution between minimum and maximum estimates (Fig. 3*B*–*F*). We chose a flat distribution because it better reflects the full range of uncertainty compared to a normal distribution. The uncertainty window for f_{cold} was scaled to reflect data availability: for the Phanerozoic, f_{cold} was varied linearly between 0.9 and 1.1 times the actual value, whereas for the Precambrian, it was varied between 0.5 and 2.0 times, reflecting larger uncertainty due to sparser data. To explore potential non-uniqueness in this nonlinear multiparameter system, we also tested the response to varying initial conditions. Six experiments were run with initial pO_2 values of 10^0, 10^{-1}, 10^{-2}, 10^{-3}, 10^{-4} and 10^{-5} PAL. All simulations converged to the same outcome of 10^{-6} PAL (i.e., the Archean level), indicating a single attractor in the model solution (Fig. S4).

Gaussian mixing model. The bimodal mixed-Gaussian distributions in Fig. 1*D* follow the method in ref. 44, and were calculated using the *fitgmdist* function and curve-fitting toolbox of MATLAB R2018a. The linear regressions and corresponding 95% confidence envelopes were calculated by Monte Carlo resampling (n = 10,000). For each regression, values of *T/P* and age were selected at random from each modelled Gaussian distribution, and the age ranges were used to bin the data for the mixed-Gaussian fitting. Global metamorphism at >2.2 Ga can be fitted by a unimodal Gaussian distribution (95% confidence interval, n = 124, p < 0.01); metamorphism at ≤2.2 Ga is non-Gaussian (95% confidence intervals; 2.2-1.8 Ga, n = 124, p < 0.0001; 1.8-0.8 Ga, n = 117, p < 0.001; 0.8-0.2 Ga, n = 336, p < 0.0001; 0.2-0 Ga, n = 175, p < 0.0001) but is well described by bimodal mixed-Gaussian distributions.

Data, Materials, and Software Availability. All study data are included in the article and/or supporting information (as *SI Appendix* and Dataset S1). Model code can be downloaded online at GitHub (https://github.com/bjwmills).

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Figures and Captions

Figure 1. Co-evolution of Earth-surface environments and deep-Earth processes over the past 4.0 Ga. (*A*) Earth's history of atmospheric oxygenation (1). (*B*) Marine redox evolution inferred from redox-sensitive element enrichments in marine sediments (8–10). (*C*) Timeline for biological evolution, with the number of species. (*D*) Metamorphic thermobaric ratios (*T/P*) (30) and histograms of age-binned *T/P* data with fits of bimodal Gaussian mixing models. (*E*) Supercontinent cycles indicated by zircon abundance and the emplacement of large igneous provinces (LIPs). (*F*) Age-frequency distribution of carbonatites and kimberlites (46). (*G*) Carbon isotopic compositions ($\delta^{13}C$) of marine carbonate (88) and kimberlite and ultramafic lamprophyres (49).

Figure 2. The carbon cycle in cold subduction zones and its impact on steady-state pO_2. (*A*) Illustration showing carbon transformations between Earth's exosphere (atmosphere, ocean and biosphere) and interior (lithosphere, mantle and core), regulated by subduction zones. F_{sub} and F_{deg} are fluxes of buried carbon-sulfur that being subducted and degassed, respectively. This omits the sulfur cycle, since pyrite and gypsum have similar circular pathways to organic carbon and carbonate, respectively. (*B*) Steady-state response of pO_2 in our modified COPSE model to variable cold subduction intensity (f_{cold}) through time. The modelled pO_2 drops gradually from the present steady state to a new steady state after hundreds of million years. (*C*) Linear response of pO_2 to the decrease in f_{cold} in our modified COPSE model.

Figure 3. Comparison of COPSE modelling results with the geological records, for a forced time-reversal in cold subduction intensity (f_{cold}). (*A*) Normalized f_{cold} derived from the metamorphic *T/P* dataset used in this study (for further details see *SI Appendix*). (*B*) Modelled relative atmosphere O_2 level (dark-blue lines and shading), with pO_2 estimated from geochemical proxies (light-blue lines and shading) (1) and models (17, 57). The inset shows the results on linear coordinate. (*C*) Modelled relative atmosphere CO_2 level (dark-pink lines and shading), with pCO_2 estimated from geochemical proxies (light-pink shading and bars) (56). (*D*) Modelled seawater sulfate concentration ($[SO_4^{2-}]_{sw}$; dark-purple lines and shading with reconstructions from geochemical models (17, 57) and sedimentary records (light-purple shading and bars) (58). (*E*) Modelled burial fluxes of marine organic (*mopb*; light-green lines and shading) and authigenic (*capb*; dark-green lines and shading) phosphorus (left Y axis) compared to the geological record of P contents in marine shales (right Y axis; light-green squares) and the occurrence of phosphogenic events (middle Y axis; dark-green bars) (59). (*F*) Modelled degree of ocean anoxia (thick-brown lines and shading), with marine redox records (I/Ca in carbonate, Mo and U in shales; brown symbols) (8–10). A value of 1 on the Y-axis reflects fully anoxia, while ~0.0025 reflects a fully oxidized global ocean.