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Causes of Cretaceous Oceanic Anoxic Events (OAEs)

Chengjun Zhang

Henan Polytechnic University, Jiaozuo, 454000, China

ABSTRACT

Oceanic Anoxic Events (OAEs) during the Cretaceous—designated OAE1, OAE2, and OAE3, with OAE1 further subdivided into OAE1a, OAE1b, OAE1c, and OAE1d—are widely attributed to volcanic activity from the Kerguelen, Ontong Java, and Caribbean Large Igneous Provinces (LIPs), which triggered at least two global OAEs. These events are characteristically marked by widespread deposition of organic-rich black shales and positive carbon isotope excursions. By systematically reviewing the geochemical behavior of molybdenum (δ98Mo) and zinc (δ66Zn) isotopes, we find that δ98Mo primarily tracks the transition between locally euxinic and non-euxinic conditions, whereas δ66Zn serves to distinguish different responses of local marine systems—including primary productivity, continental weathering, and sediment burial/decomposition. Nevertheless, the fractionation mechanisms governing these metal stable isotopes in the oceanic realm remain incompletely understood, and existing records focus predominantly on OAE2. Future research must therefore adopt a more comprehensive and systematic approach across all OAEs.

KEYWORDS

Oceanic Anoxic Events (OAEs); Large Igneous Provinces; isotopes.

1. INTRODUCTION

Cretaceous Oceanic Anoxic Events, including OAE1a, OAE1b, OAE2, and OAE3, represent severe perturbations in the global carbon cycle and marine environments. These events are characterized by widespread deposition of organic-rich black shales and positive carbon-isotope excursions, and are generally attributed to massive volcanic activity from Large Igneous Provinces (LIPs) such as the Kerguelen, Ontong Java, and Caribbean plateaus. Metal stable isotopes, particularly molybdenum and zinc, have emerged as powerful proxies for reconstructing paleo-redox conditions, primary productivity, and continental weathering. However, the fractionation mechanisms of these isotopes in the ocean remain incompletely understood, and most existing records focus on OAE2. This paper systematically reviews the geochemical hallmarks of Cretaceous OAEs, synthesizes recent advances in Mo and Zn isotope applications, and compares records from different sections to unravel local versus global environmental changes. Future research directions are also proposed to achieve a more comprehensive understanding of OAE triggers and consequences.

2. LIPS

Large Igneous Provinces (LIPs) are predominantly mafic, comprising extensive extrusive and intrusive rocks. They formed over short durations (typically1–5 Ma) and cover areas exceeding 10^5 km². LIPs are implicated in extreme climatic and environmental changes, including continental rifting and plate drift, oceanic anoxic events (OAEs), and mass extinctions, and thus have received sustained research attention.

Three marine LIPs that formed during the Cretaceous—the Kerguelen, Ontong Java, and Caribbean LIPs—are causally linked to global OAEs. Figure 1.1 clearly illustrates the genetic relationship between these three LIPs and the OAEs.

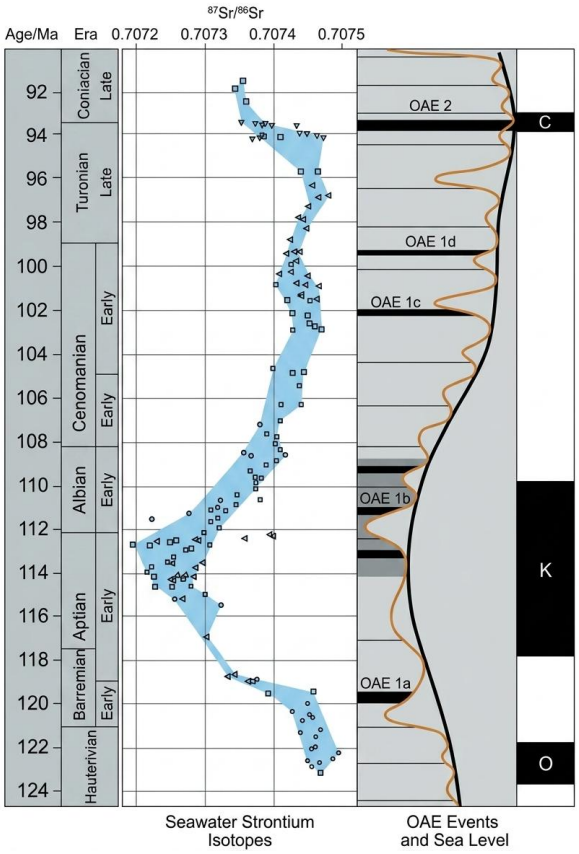

Fig 1 Formation of Cretaceous Large Igneous Provinces and their inferred evolutionary relationships (from [1])

Seawater strontium isotopes record the intensity of magmatic activity in the ocean. The mid-Cretaceous OAEs occurred repeatedly and roughly corresponded in genesis to LIP activity: OAE1a, which took place around 120 Ma, immediately followed the formation of the Ontong Java Plateau; OAE1b, about 110 Ma, occurred during the late stage of Kerguelen Plateau formation; and OAE2, about 93 Ma, was coeval with the emplacement of the Caribbean LIP (Fig. 1). These temporal coincidences suggest that LIPs were a major driver of Cretaceous OAEs.[18-20]

3. GEOCHEMICAL HALLMARKS OF OAES

3.1. Total organic carbon (TOC) characteristics

A hallmark of OAEs is the widespread deposition of black shales, which often have total organic carbon (TOC) contents of 10–30%, sometimes exceeding 30%. During the early stage of an OAE, elevated atmospheric greenhouse gas concentrations, intensified weathering, and nutrient input from Large Igneous Provinces (LIPs) caused a sharp increase in biological activity and biomass. As massive organism die-off occurred, the anoxic ocean prevented timely oxidative decomposition of the dead biomass, leading to substantial organic-matter burial on the seafloor. Concomitantly, before the OAE terminated, progressive nutrient depletion, together with seawater anoxia and acidification, reduced biomass and consequently decreased organic-carbon burial. As a result, TOC contents in sediments deposited during an OAE were markedly higher than those before or after the event. Typically, TOC contents rose rapidly during the early OAE, reached a peak, and then gradually declined.

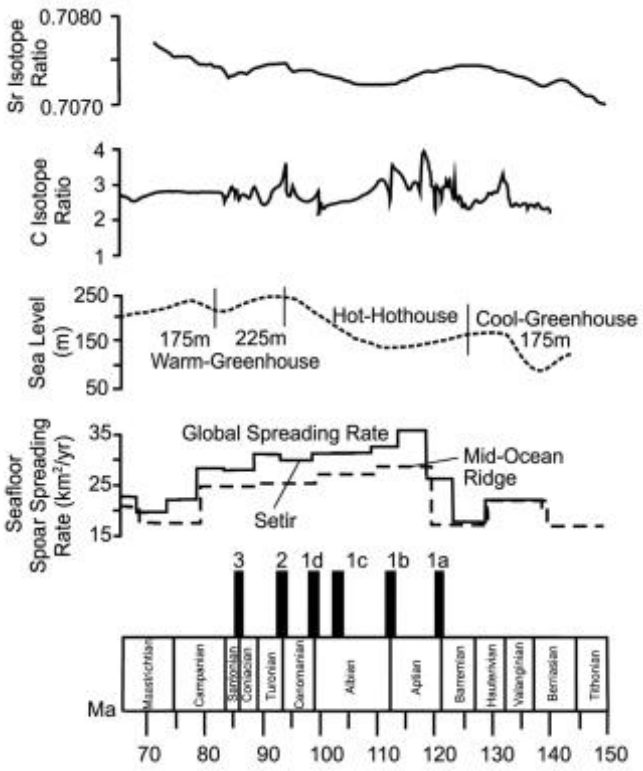

Fig 2 Mid-Cretaceous oceanic anoxic events and associated paleoceanographic/geochemical records (from [2])

3.2. Carbon isotope characteristics

OAEs exerted a profound influence on the global carbon cycle and left distinct carbon-isotope signatures that record paleoenvironmental changes. Across the global OAEs, sedimentary sections (including carbonates, sandstones, mudstones and shales) exhibit a common feature: a pronounced positive δ13C excursion. This positive shift can be explained as follows. In the early OAE, marine biomass increased dramatically, and phytoplankton preferentially took up ^{12}C during photosynthesis, fixing it into organic matter. Under oxic conditions, dead organic matter would be rapidly oxidized and ^{12}C would be released back into seawater, causing no net δ13C shift. However, during OAEs the ocean was anoxic, so large amounts of ^{12}C were buried with organic matter, thereby increasing the δ13C of seawater. Subsequently, although organisms continued to favor ^{12}C, the progressive depletion of ^{12}C in seawater forced them to take up more ^{13}C, causing a sustained positive δ13C shift in both sedimentary organic matter and seawater. Because the δ13C of marine carbonates is generally considered to reflect that of seawater, carbonates in the sediments also show a continuous positive δ13C excursion.

For OAE1a, OAE1b, and part of OAE2, the δ13C records exhibit a negative excursion immediately prior to the positive shift (Fig. 2). This negative excursion is currently interpreted as resulting from a massive input of ^{12}C to the ocean, either from LIP volcanism or from the dissociation of gas hydrates in the stratigraphic column before the onset of the OAE. In addition, some OAE2 records show a minor negative δ13C shift followed by a positive shift in the middle of the event, which is inferred to have been caused by transient re-oxygenation of the ocean during a brief cooling episode—the Plenus Cold Event (PCE). At the beginning of the PCE, oxidation of sedimentary organic matter released ^{12}C into seawater, driving a negative δ13C excursion. After the PCE ended, seawater δ13C resumed its positive trend.

4. METAL ISOTOPE SIGNATURES OF OAES

4.1. Strontium ($^{87}Sr/^{86}Sr$) isotope signatures

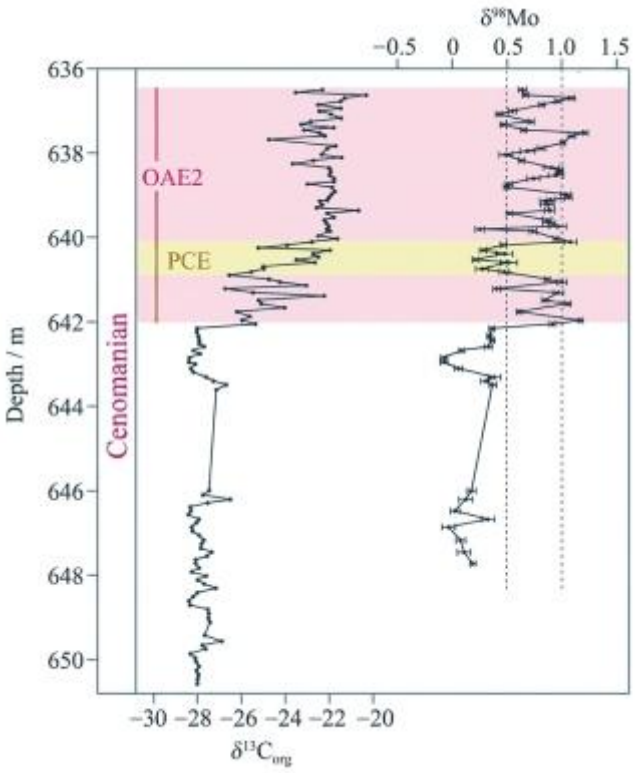

Fig 3 δ^{13}Corg and δ^{98}Mo records from the DSDP Site 367 section (from [7])

The isotope ratio $^{87}Sr/^{86}Sr$ in seawater—where ^{87}Sr is more enriched in the continental crust than in the mantle—is a widely used proxy for submarine hydrothermal activity and continental chemical weathering. A decrease in $^{87}Sr/^{86}Sr$ indicates enhanced submarine volcanic/hydrothermal activity, whereas an increase reflects intensified continental weathering. During most OAEs, a decline in $^{87}Sr/^{86}Sr$ precedes the event (Fig. 3), signaling a marked increase in submarine hydrothermal and volcanic activity. Subsequently, rising temperatures accelerate hydrological cycling and continental weathering, thereby increasing the flux of materials to the ocean.

4.2. Molybdenum ($\delta^{98}Mo$) isotope signatures

Molybdenum (Mo) is a redox-sensitive element whose speciation is primarily controlled by oxidation-reduction conditions. In rock-forming minerals, Mo mainly occurs as Mo^{4+}. Upon weathering, Mo^{4+} is oxidized and transported into water bodies as molybdate (MoO_4^{2-}), which has a wide pH–Eh stability range and therefore behaves as a relatively inert element in seawater [3,4]. Mo is considered the most abundant transition-metal element in seawater, with a concentration of ~105 nmol and a residence time of approximately 400 ka [5]. Seawater Mo is derived largely from continental weathering, with a minor contribution from low-temperature hydrothermal fluids. Once in seawater, Mo migration and enrichment are controlled by H_2S concentration, TOC, and Fe/Mn content. Under reducing conditions, Mo reacts with H_2S to form Mo–Fe–S aggregates on pyrite surfaces or precipitates as Mo-sulfide minerals. Under suboxic conditions, H_2S levels are substantially lower, and sedimentary Mo content decreases accordingly. In oxic environments, large amounts of Mo are adsorbed onto Fe-Mn oxides or hydroxides and subsequently incorporated into deep-sea sediments [6].

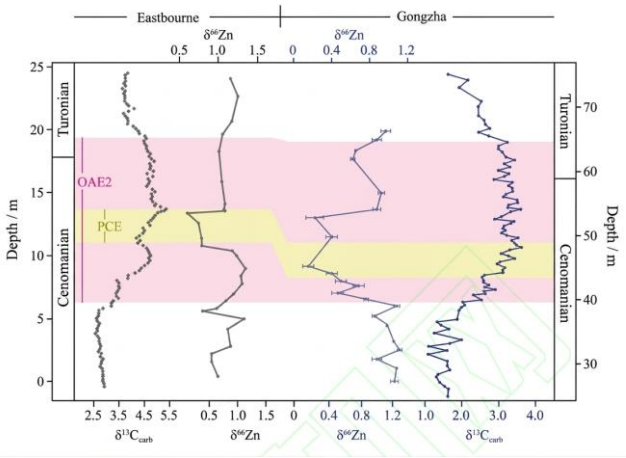

Fig 4 δ^{13}Ccarb and δ^{66}Zn records from the Eastbourne and Gongzha sections (from [12])

Previous studies have shown that Mo isotopes do not undergo significant fractionation in response to changes in temperature, pH, Eh, or biological processes; instead, redox variation is the primary control on Mo isotopic fractionation [8]. Under oxic conditions, MoO_4^{2-} is slowly removed from seawater by adsorption onto Fe-Mn oxides/hydroxides, during which light Mo isotopes are preferentially scavenged, leaving seawater enriched in heavy isotopes and thus raising δ^{98}Mo [8]. Under anoxic and fully sulfidic conditions ($H_2S > 11$ μmol/L), MoO_4^{2-} is quantitatively converted to thiomolybdate (MoS_4^{2-}) and rapidly removed via association with Fe-S particles or organic matter. Because essentially all Mo is transferred to the sediment, no Mo isotope fractionation occurs between sediment and seawater. Under anoxic but incompletely sulfidic conditions, partial conversion of MoO_4^{2-} to thiomolybdate and partial removal preferentially remove light isotopes, again increasing seawater δ^{98}Mo. Since no significant Mo isotope fractionation takes place during the removal of Mo from seawater into sediment under fully sulfidic conditions, reducing sediments formed in such environments can faithfully record the contemporaneous seawater δ^{98}Mo value, allowing reconstruction of the areal extent of sulfidic seafloor conditions.

Current studies using δ^{98}Mo records from sedimentary sections focus primarily on OAE2. Integrating the high-resolution δ^{98}Mo study of the DSDP Site 367 section by Dickson et al., the following pattern emerges (Fig. 4): δ^{98}Mo increased prior to OAE2, decreased then increased again during the Plenus Cold Event (PCE), and fluctuated within a defined range through the rest of OAE2. These observations can be interpreted as follows. Before OAE2, expansion of sulfidic marine conditions substantially reduced the fluxes of Mo removal via adsorption onto Fe-Mn oxides or thiomolybdate formation, thereby decreasing Mo isotope fractionation and leading to a positive δ^{98}Mo shift in the sediments. During the PCE, ocean re-oxygenation enhanced Mo isotope fractionation, causing a negative δ^{98}Mo excursion. After the PCE ended, sediment δ^{98}Mo again shifted positively.

In summary, variations in sedimentary δ^{98}Mo primarily reflect transitions between locally euxinic and non-euxinic conditions. Only sediments deposited under fully sulfidic conditions can potentially yield δ^{98}Mo values that represent the contemporaneous seawater δ^{98}Mo signature.

4.3. Zinc (δ^{66}Zn) isotope signatures

Zinc (Zn) is a transition-metal element with both lithophile and chalcophile affinities. It exists predominantly as Zn^{2+} in nature and participates widely in biological and geological processes. As an essential trace element, Zn serves as a cofactor for key enzymes involved in marine phytoplankton metabolism—including carbon fixation and phosphorus uptake/cycling—and under certain conditions can limit marine primary productivity [9]. The concentration of dissolved Zn in seawater ranges from 0.1 to 10.0 nmol/kg, with a minor fraction present as free Zn^{2+} and the majority complexed with Cl^-, CO_3^{2-}, humic acid, and fulvic acid. Zn exhibits a typical nutrient-type vertical

distribution: low concentrations in surface waters and higher concentrations in intermediate and deep waters [9].

Most studies indicate that phytoplankton preferentially take up light Zn isotopes, whereas diatom and bacterial surfaces preferentially adsorb heavy Zn isotopes, and organic matter preferentially complexes/adsorbs heavy Zn isotopes. These observations explain why in some regions surface seawater $\delta^{66}Zn$ is lower than that of deep seawater, while in other regions the opposite pattern occurs. Some research attributes the lighter $\delta^{66}Zn$ in modern surface waters of certain areas to anthropogenic influences.

Currently, most researchers reconstruct OAE-related paleoenvironmental changes using $\delta^{66}Zn$ from marine carbonates in OAE2 sections, assuming that carbonate $\delta^{66}Zn$ reflects contemporaneous seawater $\delta^{66}Zn$. In the Eastbourne section (UK), $\delta^{66}Zn$ during OAE2 exhibited two negative and two positive excursions (Fig. 4). This pattern can be interpreted as follows. Initially, LIP formation delivered large amounts of light Zn isotopes to the ocean via hydrothermal activity or seawater-basalt interaction, causing the first negative $\delta^{66}Zn$ shift in carbonates. Subsequently, greatly increased biomass, coupled with intensive biological activity that utilized abundant light Zn isotopes, together with the development of anoxic deep-ocean conditions that prevented rapid oxidative decomposition of dead biomass, led to the burial of large amounts of light Zn isotopes with organic-rich sediments, resulting in a positive $\delta^{66}Zn$ excursion. During the Plenus Cold Event (PCE), seafloor re-oxygenation released light Zn isotopes from organic-rich sediments, generating a second negative $\delta^{66}Zn$ shift. After the PCE ended, the deep ocean returned to anoxic conditions, and carbonate $\delta^{66}Zn$ again shifted positively until the termination of OAE2 [10].

The Tarfaya section (Morocco) shows $\delta^{66}Zn$ trends similar to those of the Eastbourne section, suggesting that the environmental changes during OAE2 at Tarfaya were likely similar to those at Eastbourne [11]. In contrast, the Gongzha section (Tibet) displays a different $\delta^{66}Zn$ pattern: during OAE2, $\delta^{66}Zn$ first shifted positively and then negatively (Fig. 4). The Gongzha section is interpreted to have remained under oxic conditions throughout, so Zn isotopes were little affected by burial of organic-rich sediments or by seafloor re-oxygenation during the PCE. In the early stage of OAE2, high atmospheric CO_2 partial pressure intensified continental basalt weathering, delivering light Zn isotopes to the ocean and causing a negative $\delta^{66}Zn$ shift in carbonates. In the late stage of OAE2, weathering weakened while sustained vigorous biological activity continued to utilize light Zn isotopes, leading to a positive $\delta^{66}Zn$ excursion.

Comparison of the $\delta^{66}Zn$ records from the Eastbourne, Tarfaya, and Gongzha sections demonstrates that although the pronounced changes in seawater $\delta^{66}Zn$ during OAE2 were a global phenomenon, regional variations in $\delta^{66}Zn$ trends exist depending on local redox conditions.

In summary, sedimentary $\delta^{66}Zn$ signatures are governed not only by global marine redox conditions but also by local oceanic responses to primary productivity, continental weathering, and sediment burial/decomposition processes.

5. CAUSES OF OCEANIC ANOXIC EVENTS (OAES)

During major oceanic anoxic events (OAEs), the widespread development of reducing conditions in deep-ocean bottom waters is typically accompanied by extensive black-shale deposition, rapid turnover of benthic and planktonic biota, and a positive carbon-isotope excursion in marine carbonates—indicating profound changes in the climate/ocean system. These changes may manifest as alterations in the ocean water-column structure (e.g., large-scale upwelling of bottom waters) and as an acceleration of hydrological cycling at the Earth's surface, enhanced physical-chemical weathering, and substantially increased nutrient input to the ocean, collectively raising marine productivity. Elevated productivity, in turn, likely contributed to initiating or sustaining bottom-water anoxia during OAEs. On balance, Cretaceous black-shale deposition represents an "anoxic,

high-productivity ocean." Extreme climatic conditions were the most direct drivers of OAEs, whereas tectonic activity may have been the ultimate determining factor.

OAEs at different times had distinct implications for Cretaceous climate evolution. For the Cretaceous as a whole, the primary driver of climate change—particularly the transition to greenhouse conditions—was intense tectonic activity. During the Valanginian (Early Cretaceous), basaltic eruptions of the Paraná Basin (Brazil) occurred, but their limited scale failed to trigger a greenhouse climate; instead, enhanced paleoproductivity and chemical weathering led to sequestration of large amounts of CO_2. The large-scale eruptions of the Ontong Java Plateau and other oceanic plateaus at ~120 Ma, marking OAE1a, signalled the onset of the Cretaceous greenhouse climate. Temperatures peaked near the Cenomanian/Turonian boundary (OAE2), reaching the highest global mean values from the Cretaceous to the present. Subsequently, from the late Turonian to the early Campanian, global temperatures remained relatively warm, with OAE3 occurring during this interval. From the early Campanian onward, global temperatures declined steadily, ending the greenhouse climate. This pattern underpins the three-phase mid- to late-Cretaceous greenhouse model of Huber et al. (2002): warm–hot–cool greenhouse. According to Erba (2002), OAE1a and OAE2 respectively mark the initiation, acme, and termination of the extreme greenhouse climate of the mid-Cretaceous, with intermittent cooler intervals.

Repeated OAEs during the mid-Cretaceous led to massive burial of organic carbon. For example, during the Cenomanian–Turonian OAE2 alone, approximately 1.6×10^{18} mol of organic carbon was buried within a short interval of 500 kyr. This large-scale organic-carbon burial profoundly impacted the ocean–atmosphere system. For OAE2, it caused a 40–80% drop in atmospheric CO_2 concentration to below 500 ppm, driving an "inverse greenhouse effect" that led to climate cooling and altered ocean-circulation patterns. Another direct consequence of massive organic-carbon burial was an increase in atmospheric oxygen concentration, enhancing the oxidation capacity of the ocean–atmosphere system. The widespread occurrence of red deep-sea deposits following Cretaceous OAEs may well reflect both of these effects.

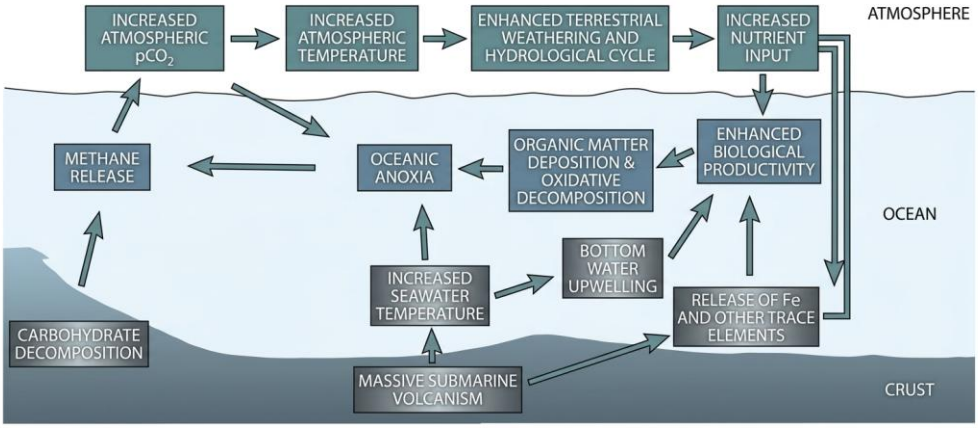

Fig 5 Genetic model for Cretaceous OAEs (from [2])

The causal mechanisms of Cretaceous OAEs are complex, involving interactions among Earth's internal and external spheres, and remain debated. Nevertheless, OAE genesis is generally considered to be closely linked to Large Igneous Province (LIP) activity (Fig. 5). Large-scale Cretaceous submarine magmatic eruptions altered ocean-basin volumes, triggered rises in seawater temperature and sea level, caused global transgression, modified land–sea configurations, enhanced absorption of solar radiation, and ultimately increased global atmospheric temperature [13]. In addition, massive volcanic eruptions released large amounts of heat and greenhouse gases from the Earth's interior, further raising atmospheric temperature [14]. Mid-Cretaceous atmospheric CO_2 levels were nine times those of the pre-industrial era and remained 1.5 times higher by the Late Cretaceous; Cretaceous global paleotemperatures were 3–8 °C higher than today [15–17]. Elevated CO_2 and seawater

temperature were preconditions for OAEs: together they lowered dissolved oxygen concentrations in seawater, a key factor promoting oceanic anoxia.

Concurrent with large-scale submarine volcanism, substantial nutrients were released into bottom waters. Warming of these bottom waters induced upwelling, triggering a widespread increase in marine productivity. Moreover, rising global temperatures enhanced continental chemical weathering and the hydrological cycle, intensifying the delivery of terrestrial nutrients to the ocean and further elevating marine productivity. The increased productivity, together with the sinking and decomposition of organic matter, consumed oxygen in the water column, exacerbating anoxic conditions. Seafloor anoxia inhibited oxidative decomposition of organic matter, thereby promoting its effective burial and preservation, ultimately leading to the global deposition of black shales during OAEs.

6. CONCLUSIONS

Cretaceous OAEs are representative extreme climatic events in Earth's history. Studying the processes, causes, and consequences of these major climatic perturbations is thus of great significance for understanding the operating mechanisms of the Earth's climate system and for addressing the modern challenge of "global warming.

Synthesis of metal-isotope data indicates that although the specific causes of individual OAEs may have differed, they were predominantly sourced from large-scale, intense submarine volcanic activity. Elevated paleo-seawater temperatures and atmospheric CO_2 concentrations were preconditions for Cretaceous OAEs. The increased relative abundance of strontium isotopes during this period also points to enhanced continental chemical weathering and hydrological cycling. The combination of high marine productivity and anoxic seafloor conditions facilitated the burial and preservation of large amounts of organic matter, and consequently the widespread global distribution of organic-rich black shales remains the hallmark of OAEs.

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