



Oxygenation variations in the atmosphere and shallow seawaters of the Yangtze Platform during the Ediacaran Period: Clues from Cr-isotope and Ce-anomaly in carbonates

Wei Wei^a, Robert Frei^{b,c}, Geoffrey J. Gilleaudeau^{b,1}, Da Li^a, Guang-Yi Wei^a, Xi Chen^a, Hong-Fei Ling^{a,*}

^a State Key Laboratory for Mineral Deposits Research, School of Earth Sciences and Engineering, Nanjing University, Nanjing 210046, China

^b Department of Geosciences and Natural Resource Management, University of Copenhagen, Øster Voldgade 10, 1350 Copenhagen, Denmark

^c Nordic Center for Earth Evolution, NordCEE, Denmark

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ABSTRACT

The late Neoproterozoic Era witnessed a significant oxygenation in Earth's surface in line with the naissance and diversity of early animals; however, detailed variations of the oxygenation during this period remain controversial. Chromium-isotope in ancient carbonates has shown its ability to trace the oxygenation history of the atmosphere-hydrosphere system. In this study, we analyzed Cr-isotope and Ce-anomaly data of carbonates from the Ediacaran Doushantuo and Dengying formations in the Yangtze Gorges area, South China, in order to reconstruct oxygenation variations in the atmosphere and shallow seawaters of the Yangtze Platform during the Ediacaran Period.

After evaluating and excluding detrital and diagenetic effects, well-preserved $\delta^{53}\text{Cr}_{\text{auth}}$ signatures were obtained. Post-Marinoan carbonates record slightly positive $\delta^{53}\text{Cr}_{\text{auth}}$ values (0.18–0.43‰) and slightly negative Ce-anomaly, suggesting generally oxidizing conditions in the atmosphere (with $p\text{O}_2$ over 1% PAL) and in shallow seawaters of the Yangtze Platform during the post-Marinoan phase, which may have been related to the diversity of early Ediacaran organisms. Shuram/Wonoka correlative carbonates are characterized by the fluctuations of $\delta^{53}\text{Cr}_{\text{auth}}$ values (–0.25 to 0.97‰) and a significant negative $\delta^{13}\text{C}$ excursion, which may have resulted from partial oxidation of the large deep-water DOC reservoir under enhanced oxidative weathering conditions. This process released ^{13}C -depleted carbon into DIC pool and isotopically unfractionated Cr (III) into water columns, the latter of which immediately precipitated by adsorption onto the detrital materials. Meanwhile, an increase in local anoxia (and even euxinia) in the intrashelf basin of the Yangtze Platform driven by enhanced microbial sulfate reduction and the resultant H_2S , possibly limited regional biological evolution during this phase. Late Ediacaran carbonates record elevated $\delta^{53}\text{Cr}_{\text{auth}}$ values (0.40–0.96‰) together with the decreasing trend in Ce/Ce* values, indicating a more oxygenated environment in the atmosphere and shallow seawaters of the Yangtze Platform during the late Ediacaran phase, which may have been beneficial to the diversification of late Ediacaran animals.

1. Introduction

There is a consensus that Earth's atmosphere experienced two major pulses of oxygenation in the past. The first, the Great Oxidation Event (GOE; Holland, 2002) occurred in the Paleoproterozoic Era, though the exact timing, duration, and magnitude of this oxygenation remain debatable (e.g. Canfield, 1998; Holland, 2006; Konhauser et al., 2011; Lyons et al., 2014). After a supposedly prolonged period of relative stasis between ca. 1.8 and 0.8 Ga (e.g. Anbar and Knoll, 2002; Arnold

et al., 2004; Planavsky et al., 2014), a second atmospheric oxygenation took place in the Neoproterozoic Era, apparently temporally related to a couple of globally-scaled glaciations (the 'Snowball Earth' event; Hoffman et al., 1998, 2017; Hoffman and Schrag, 2002), and referred to as the Neoproterozoic Oxygenation Event (NOE; Shields-Zhou and Och, 2011). Atmospheric oxygen levels during the NOE are proposed to have nearly reached those of the modern atmosphere (e.g. Holland, 2006; Canfield et al., 2007; Sahoo et al., 2012; Lenton et al., 2014; Lyons et al., 2014), although studies also provide evidence for anoxic

* Corresponding author.

E-mail address: hfling@nju.edu.cn (H.-F. Ling).

¹ Current address: School of Earth and Space Exploration, Arizona State University, Tempe, AZ 85281, USA.

(ferruginous) and even euxinic conditions in many Ediacaran deep waters (e.g. Canfield et al., 2008; Li et al., 2010; Sperling et al., 2015; Sahoo et al., 2016). Recent molybdenum-isotope evidence suggests that at ~521 Ma, marine oxygen contents approached modern-like levels for the first time in Earth History (Chen et al., 2015).

It is noteworthy that major evolutionary advances broadly coincided with these two major oxygenation events, characterized by the transitions from prokaryotes to eukaryotes during the GOE, and from unicellular to complex multicellular organisms during the NOE (Och and Shields-Zhou, 2012; and references therein). From the Ediacaran to early Cambrian, bioturbation indices, diversities of animals, skeletal taxa, and eukaryotic phytoplankton, increased progressively leading up to the ‘Cambrian Explosion’, in line with stepwise oxygenation in the oceans to modern-like levels at ~521 Ma (Chen et al., 2015; and references therein). Therefore, it is believed that oxygen played an important role in ecosystem developments. However, detailed variations of the oxygenation in Earth’s atmosphere and oceans during the Ediacaran Period remain scarce, controversial, and enigmatic. This is probably due to the interpretative complexity resulting from many, relatively recent studies addressing redox fluctuations in the marine realm using a multitude of trace element and isotope geochemical tracers that respond to different threshold levels of oxygen (e.g. Anbar, 2004; Algeo and Maynard, 2008; Scott and Lyons, 2012; Lyons et al., 2014).

Chromium-isotope signature in ancient carbonates has shown its ability to trace the oxygenation history of the atmosphere-hydrosphere system (Frei et al., 2011; Gilleaudeau et al., 2016; Rodler et al., 2016a,b, 2017; Holmden et al., 2016; D’Arcy et al., 2017). The Yangtze Platform in South China is ideally suited for the study of redox fluctuations in Earth’s surface during the Ediacaran Period because of widely distributed and continuous Ediacaran successions. In this study, we analyzed Cr-isotope compositions ($\delta^{53}\text{Cr}_{\text{sample}} = [({}^{53}\text{Cr}/{}^{52}\text{Cr})_{\text{sample}} / ({}^{53}\text{Cr}/{}^{52}\text{Cr})_{\text{SRM979}} - 1] \times 1000$) of the stratigraphically coherent carbonates from the Ediacaran Doushantuo and Dengying formations in the Yangtze Gorges area, Hubei Province, South China. We discussed Cr-isotope data together with other seawater tracers, including C-isotope (Ling et al., 2013) and Ce-anomaly data from the same batch of samples. Our results provide new insights that help unravel oxygenation variations in the atmosphere and shallow seawaters of the Yangtze Platform during the Ediacaran Period.

2. Background: Cr-isotope and Ce-anomaly proxies

2.1. Cr-isotope

In Earth’s surface environment, chromium has two oxidation states, the soluble Cr (VI) anion (CrO_4^{2-} or HCrO_4^-) and the insoluble Cr (III)-compound. In soils and terrestrial rocks, it exists as Cr (III)-bearing minerals, commonly as chromite, exhibiting low solubility in natural environments (Fendorf, 1995; Oze et al., 2004). The $\delta^{53}\text{Cr}$ values of the Bulk Silicate Earth (BSE) reservoirs are within a narrow range of $-0.124 \pm 0.101\%$ (Schoenberg et al., 2008). During terrestrial oxidative weathering, insoluble Cr (III) can be oxidized to Cr (VI) via direct reaction with Mn-oxides which exist stably under high oxygen fugacities (e.g. Eary and Rai, 1987; Fendorf and Zasoski, 1992). This process results in the enrichment of isotopically heavier Cr in soluble Cr (VI) (e.g. Schauble et al., 2004; Zink et al., 2010; Wang et al., 2015), which is subsequently delivered to surface seawater via rivers (Oze et al., 2007). The operation of this release mechanism is supported by negative $\delta^{53}\text{Cr}$ values of modern weathering profiles (Frei et al., 2014; Paulukat et al., 2015; D’Arcy et al., 2016), and by positive $\delta^{53}\text{Cr}$ values of modern river waters (Frei et al., 2014; Paulukat et al., 2015; D’Arcy et al., 2016; Wu et al., 2017). Negatively fractionated Cr-isotope data have also been reported for paleosols (e.g. Crowe et al., 2013; Frei and Polat, 2013; Berger and Frei, 2014), although positive $\delta^{53}\text{Cr}$ values were recorded by ca. 1.85 Ga Flin Flon paleosols, which were explained

by either pedogenesis or subsequent hydrothermal/metamorphic alteration (Babechuk et al., 2017).

In modern seawater, the dominant Cr species is soluble Cr (VI) anion (CrO_4^{2-} or HCrO_4^-), although > 30% of dissolved Cr can occur as Cr (III) (e.g. Achterberg and van den Berg, 1997; Sirinawin et al., 2000), and up to 50% in some circumstances (Connelly et al., 2006), since the solubility of Cr (III) can be increased by complexation with dissolved organic matter (e.g. Yamazaki et al., 1980; Nakayama et al., 1981; Sander and Koschinsky, 2000, 2011). The oceanic residence time of Cr is estimated to be ~3000 years (McClain and Maher, 2016), not much longer than the mixing time of the ocean (~1600 yr; Broecker and Peng, 1982). This may partly explain the heterogeneity of Cr concentrations and isotope compositions of modern seawaters (e.g. Bonnard et al., 2013; Scheiderich et al., 2015; Paulukat et al., 2016). Moreover, there is a logarithmic negative relationship between Cr concentrations and isotope compositions of seawaters, which may have resulted from varying degrees of Cr (VI) reduction in surface waters and/or oxygen minimum zones (OMZ), and the subsequent back-release of scavenged Cr (III) into seawaters by oxidation or by complexation with organic matter, with associated isotopic fractionation (Scheiderich et al., 2015; Paulukat et al., 2016).

The major Cr sink in the global Cr cycle is authigenic burial in reducing sediments via reduction of soluble Cr (VI) to insoluble Cr (III) (e.g. Reinhard et al., 2013, 2014; Gueguen et al., 2016). In anoxic or even euxinic marine system, dissolved Cr (VI) anion can be efficiently reduced and precipitated by reaction with aqueous Fe (II), dissolved hydrogen sulfide (H_2S), or solid Fe-S phases (e.g. Reinhard et al., 2014; Gueguen et al., 2016; Bauer et al., 2018). Given that these reductions are fast and efficient, sediments deposited under a ferruginous or H_2S -containing water columns have the potential to mirror the Cr-isotope signal of seawater, which is ultimately linked to Cr release mechanisms on the continents (Reinhard et al., 2014; Gueguen et al., 2016; Bauer et al., 2018). This is supported by laboratory experiments designed to mimic natural aqueous systems (Døssing et al., 2011), and by the coincidence of $\delta^{53}\text{Cr}$ values of anoxic marine sediments with those of contemporary seawaters (Reinhard et al., 2014; Gueguen et al., 2016). However, in low-oxygen (but not entirely anoxic) and no H_2S environments ($\text{O}_2 < 5 \mu\text{mol kg}^{-1}$; Rue et al., 1997) such as those characterizing OMZ and mildly redox-stratified basins, Cr (VI) can be partially reduced to immobile Cr (III). This kind of reduction is accompanied by a significant isotope fractionation (e.g. Ellis et al., 2002; Sikora et al., 2008; Zink et al., 2010), in which isotopically light Cr (III) can be scavenged into sediments or transformed to soluble clathrates with organic ligands (e.g. Scheiderich et al., 2015; Gueguen et al., 2016; Paulukat et al., 2016).

Authigenic Cr in oxic siliciclastic sediments accounts for a negligible fraction of the Cr sink flux from the global oceans, while incorporation of Cr in biogenic and abiogenic carbonates constitutes another sink whose flux accounts for a relatively small fraction of global Cr cycle (Reinhard et al., 2013; Gueguen et al., 2016; Qin and Wang, 2017). Results obtained so far indicate that there is a significant isotopic disequilibrium between various biogenic carbonates (such as corals, green and red algae, foraminifera, molluscs) and coeval seawaters, indicating a distinctively negative isotope fractionation during Cr sequestration into the biogenic materials of carbonates (Pereira et al., 2016; Holmden et al., 2016; Wang et al., 2017). For abiogenic carbonates, laboratory experiments suggest that soluble Cr (VI) oxyanions coprecipitates with abiogenic carbonates without changing of its oxidation valence (Tang et al., 2007; Rodler et al., 2015), and isotopically heavier Cr preferentially enters the lattice of the carbonate structure with fractionation from 0.06 to 0.18‰ (Rodler et al., 2015). Fractionation associated with this process increases with increasing Cr concentration of the initial solution from which the non-skeletal carbonate precipitates, but can be expected to be negligible at low chromium concentrations typical of seawater (Rodler et al., 2015). Therefore, ancient carbonate can be advocated as a reliable archive for Cr-isotope compositions of

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