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Key Points:

- REY flux from basalt weathering driven by SGD is a major natural source of seawater
- Coral Nd_N/Yb_N ratios with seasonal cycles are driven by the SGD and the adsorption-desorption processes of marine biogenic particles
- Coral Y/Ho ratio serves as a potential proxy for basalt weathering

Supporting Information:

Supporting Information may be found in the online version of this article.

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High-Resolution Coral Records of Rare Earth Elements and Yttrium in Seawater Driven by Submarine Groundwater Discharge in a Basalt Island: A Case Study in the Northern South China Sea

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Abstract Rare earth elements and yttrium (REY) are widely employed as tracers for oceanic geochemical processes, which require a thorough understanding of their sources, sinks, and drivers of variability in the marine environment. However, significant uncertainties exist in the marine REY cycle, the so-called "missing Nd flux," particularly regarding the contribution of submarine groundwater discharge (SGD) and basalt weathering in volcanic islands. Here, we present a 10-year record of monthly *Porites* coral REY parameters from Weizhou Island, a volcanic island built up underwater from basalt eruptions during the Quaternary, to investigate the sources and seasonal characteristics of surface seawater REY. Results demonstrate a robust seasonal cycle in the coral Y/Ho ratios, exhibiting a strong correlation with the rainfall-controlled SGD on monthly timescales and East Asian Summer Monsoon on interannual timescales, both of which are associated with basalt weathering. Combined with multiple climatic and environmental data, we find that coral Nd_N/Yb_N ratios may be mainly controlled by precipitation associated with SGD and the adsorption-desorption processes of marine biogenic particles, whereas coral REY/Ca ratios are influenced by the remobilization of sediment driven by winter monsoon. Our research suggests that the high coral Y/Ho ratios may be primarily influenced by basalt weathering during the wet season, when SGD from the island is the chief source of REY to the coastal waters. This study provides new insights into the sources and characteristics of marine REY in volcanic islands, highlighting the potential for SGD-driven REY fluxes from basaltic islands.

Plain Language Summary Rare earth elements and yttrium (REY) are able to record marine geochemical processes. To better understand this process, it is necessary to comprehend the sources and track the changes of REY in the marine environment. However, the contribution of submarine groundwater discharge (SGD) and basalt weathering of volcanic islands to marine REY is still unclear. Here, a *Porites* coral, reflecting the historical changes in the REY composition of dissolved seawater, was collected from a volcanic island in the South China Sea. Comprehensive geochemical analyses have been conducted to explore the temporal variations and sources of REY in surface seawater. We discovered that basalt weathering driven by SGD is the main cause of REY changes in seawater. Furthermore, seasonal fluctuations in biogenic particles, combined with the dissolution of particulate REY within sediment driven by winter monsoon, played a substantial role. Another key point is that the REY parameters (e.g., Y/Ho ratio) may hold promise as an excellent indicator for assessing basalt weathering intensity. This study deepens our understanding of the origin and behavior of REY in marine systems, and improves the comprehension of how basalt weathering influences REY recorded in corals.

1. Introduction

Rare earth elements and yttrium (REY) are regarded as excellent tracers for water-mass transport, current transport, and particle settling, for example, in marine geochemical processes (Behrens et al., 2020; Deng et al., 2017; Elderfield & Greaves, 1982; Murphy & Dymond, 1984), due to their subtle but systematic differences in chemical properties related to their unique electronic structure (Elderfield et al., 1988). The main sources of dissolved REY in the modern oceans are riverine flux, submarine groundwater discharge (SGD), atmospheric/eolian flux, and hydrothermal inputs (Chen et al., 2015; Jiang et al., 2018b). In addition, there is important evidence that a deep, benthic flux and boundary exchange also likely contribute to the marine REY cycle (Abbott et al., 2015; Pearce et al., 2013). Analyzing the budget and pattern of oceanic dissolved REY provides valuable

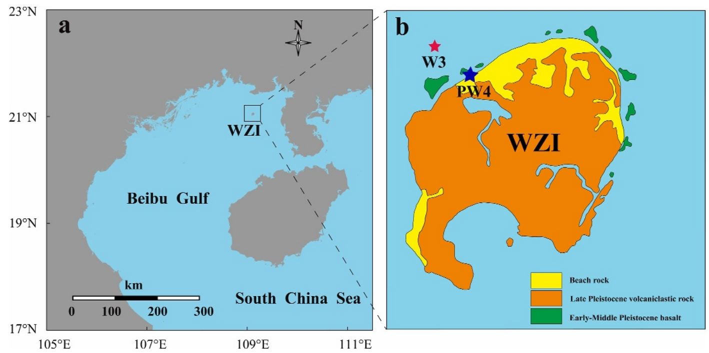

Figure 1. Location map of the study area. (a) Location map in WZI, and (b) Geological map in WZI (modified from Fan et al. (2006)) and sampling points of W3 coral and PW4 SGD (marked by stars).

insights into the geochemical signatures and climate evolution of the global or regional ocean (Adebayo et al., 2022; Garcia-Solsona & Jeandel, 2020; Garcia-Solsona et al., 2020; Pham et al., 2019). It is crucial to note that there is a difference between the combined river with atmospheric neodymium (Nd) fluxes and the flux required to balance the oceanic Nd budget, the so-called "missing Nd flux," which is a significant issue in marine REY research (Abbott et al., 2015; Arsouze et al., 2009; Lacan & Jeandel, 2005; Tachikawa et al., 2003). Recent research has shown that there are two promising candidates for "missing Nd flux," focusing on SGD (Chevis et al., 2015; Johannesson & Burdige, 2007; Johannesson et al., 2011) and the weathering of basaltic islands (Johannesson et al., 2017; Molina-Kescher et al., 2018). However, the geochemistry of REY in seawater remains relatively underexplored to date due to REY's low concentrations and complex chemical speciation (Schijf & Byrne, 2021). There is a lack of continuous high-resolution monitoring data to fully investigate the budget of REY in the oceans and their influencing factors.

Coral reefs, widespread in tropical and subtropical marine environments, provide an excellent high-resolution geochemical proxy due to their long growth histories, rapid annual growth rates, clear interannual boundaries, sensitivity to environmental changes, and suitable high-precision dating (Saha et al., 2016; Thompson, 2022; Yu et al., 2005). The concentrations of REY incorporated in coral skeletons are proportional to those in ambient seawater (Akagi et al., 2004; Sholkovitz & Shen, 1995), allowing for the historical changes in the dissolved REY composition of the surrounding seawater to be reliably represented by the changes in the REY composition along the growth bands in coral (Wyndham et al., 2004). Therefore, the records of REY provided by corals offer the potential to reveal past environmental and climatic parameters, including sea level change (Liu et al., 2006, 2011), human activities (Nguyen et al., 2013), continental weathering (Naqvi et al., 1996), SGD (Jiang et al., 2018b; Prouty et al., 2009), monsoon (Jiang et al., 2018a; Li et al., 2019), terrestrial runoff and associated coastal water quality (Saha et al., 2018). Despite numerous surveys of estuarine areas, much less is known about the coral REY variation in subterranean estuary areas as well as in basalt islands with SGD.

The Early-Middle Pleistocene flood basalts are the products of the largest volcanic eruption on Weizhou Island (WZI) (Zhang et al., 2020), which formed the volcanic terrain of the island (Fan et al., 2006). The natural conditions of WZI are conducive to the growth and reproduction of reef-building corals, which have developed since the mid-Holocene (Yu et al., 2019). Situated in a tropical zone with high precipitation and temperatures, the volcanic rocks in WZI primarily consist of basalt (Yu et al., 2021), which facilitates chemical weathering (Gislason et al., 2009). For this study, WZI was selected as the research area, where the high-resolution (monthly) coral records of REY were analyzed to investigate the impacts of climatic and environmental drivers (notably SGD and basalt weathering) on the seasonal variations of marine REY, aiming to enhance our understanding of marine REY budgets.

2. Materials and Methods

2.1. Study Area

WZI (20°00′–21°05′N, 109°04′–109°09′E), located in the northern South China Sea (SCS), is a volcanic island formed by subaqueous eruptions of Quaternary basaltic magma (Yue et al., 2024) (Figure 1). There are large areas of spheroidal weathered basalt in the north of the island (Fan et al., 2006). WZI is an independent hydrogeological unit surrounded by the sea and characterized by a tropical oceanic monsoon climate. The average annual temperature is 23.5°C, and the average annual precipitation reaches ∼1,423 mm, with nearly 85% occurring in the wet season (from May to October), which supports strong chemical weathering. Additionally, the island boasts abundant groundwater resources (Chen, 1999). The equal ^{224}Ra mass of the source and sink terms in the study area was used to estimate the SGD flux around the WZI to be 3.95×10^7 m^3 d^{-1} (26.57 cm d^{-1}) (Methods S1 and S2 in Supporting Information S1), which is comparable to those of Jeju Island (4.5×10^7 m^3 d^{-1}) (Kim & Kim, 2011).

2.2. Sample and Geochemical Analysis

A modern *Porites* coral labeled as W3 was collected at a water depth of 4 m in the northwest of WZI (21°4′7″N, 109°5′24″E) in October 2015 (Figure 1b). After washing with clean water, the coral was cut along the major growth axis using a rock saw to obtain a slab (approximately 24 cm long, 8 cm wide, and 1 cm thick). The annual growth bands of coral aragonite can be determined from the alternating light-dark density stripes of the X-ray image (Figure S1 in Supporting Information S1). Each high- and low-density band represents 1 year of growth (Knutson et al., 1972), while the outermost band represents the ongoing growth of coral skeletons in 2015. The slab was soaked in 10% H_2O_2 for 48 hr and washed 3 times in an ultrasonic bath with Milli-Q water to ensure the removal of any surface contamination that could potentially affect the accuracy of the trace elemental records. Subsequently, it was air-dried in an oven at 40°C for 48 hr.

Following pretreatment, preliminary grinding was conducted along the designated sample track of the slab, with the objective of removing approximately 1 mm of powder from the upper surface. Subsequently, based on the annual density stripes of the X-ray image, powdered calcium carbonate samples were taken at a fixed micro-sampling interval and continuously along the maximum growth axis to obtain 416 samples (32 years). The internal standard isotopes (^{103}Rh, ^{115}In, ^{187}Re) were used to prepare the 100 ml of 100 ppb internal standard stock solution. The national certified carbonate geochemical reference materials (GBW07129, GBW07133, GBW07135), as presented in Table S1 of the Supporting Information S1, were used as the external standards for the coral skeleton analysis (Jiang et al., 2022). Approximately 3 mg of the sample was weighed into an LDPE tube from the completely homogenized sample. Each tube was filled with 450 μl of 100 ppb internal standard stock solution, diluted approximately 3,000 times with 2% HNO_3, and then analyzed by inductively coupled plasma-mass spectrometry (ICP-MS) for geochemical analysis of REY. The signals of the REY isotopes (^{89}Y, ^{139}La, ^{140}Ce, ^{141}Pr, ^{143}Nd, ^{149}Sm, ^{153}Eu, ^{159}Tb, ^{160}Gd, ^{164}Dy, ^{165}Ho, ^{167}Er, ^{169}Tm, ^{174}Yb, and ^{175}Lu), other trace metals (^{43}Ca and ^{90}Zr), and the internal standard isotopes were simultaneously monitored and measured in the same aliquots. The signals were further corrected for any oxide, hydroxide, and isobaric interferences and calibrated against the certified elemental abundances in the standard to calculate the elemental concentrations in the coral samples (The levels of BaO and (REY)O were determined by analyzing pure elemental solutions, with appropriate corrections made for any potential interferences). All samples were measured 3 times with a relative standard deviation (RSD) of generally less than 5%. The details of the sample processing and analysis methods have been reported in our previous research (Jiang et al., 2022).

Previous studies have reported that the Ni/Ca and V/Ca ratios of the W3 coral were low and stable during the period 1994–2008, whereas these values fluctuated drastically and exhibited high levels during other periods of anthropogenic activity (Jiang et al., 2022; Wu et al., 2022). Thus, the coral REY time series was established for the decade of 1999–2008 in our study, which was the natural state relatively less affected by artificial disturbance in the WZI (Figure S2 in Supporting Information S1) and showed a more pronounced trend (Sun et al., 2022). The δ^{18}O values of W3 coral (selected from the same completely homogenized sample for the REY measurements) have been previously reported by Xu et al. (2018) and were applied in this study. The maximum δ^{18}O values corresponded to the lowest point of the sea surface temperature (SST) cycle and vice versa; then, each annual cycle variation of the same coral REY can be determined correspondingly (Tables S2 and S3 in Supporting Information S1). Subsequently, the REY data with the monthly resolution was obtained from linear interpolation. Because of the complex internal structure of the coral, the actual growth rate of the coral is not likely to be uniform. The interpolation method may result in slight deviations in the monthly resolution time series. Therefore, the data will be analyzed and discussed using a 5-month moving average, which effectively removes the disturbances in question.

2.3. SGD Sample Collection and Chemical Analysis

The groundwater sample (labeled as PW4, situated in close proximity to W3 coral) was collected in October 2022 in acid-cleaned high-density polyethylene (HDPE) bottles at the location shown in Figure 1b. In the field, salinity (~20‰) and pH (~6.66) were monitored using a portable multiparameter water quality analyzer until the parameters reached a stable state. Subsequently, the PW4 groundwater was filtered through a 0.45 μm filter membrane and collected in an acid-cleaned HDPE bottle. It was then acidified to a pH of less than 2 with ultra-pure HNO_3 and transported back to the laboratory under refrigeration. Since its return to the laboratory, the groundwater sample has been stored in the laboratory sample freezer at a temperature of approximately 0 degrees Celsius.

The PW4 groundwater sample was passed through Bio-Rad® Poly-Prep columns packed with ~2 mL of Bio-Rad AG® 50 W-X8 (100–200 mesh, hydrogen form) cation-exchange resin in order to separate the REY from the major salts. Two 3-mL acid rinses of 1.75 M ultra-pure HCl and 2 M ultra-pure HNO_3 were performed to elute Fe and Ba, respectively, from the columns. REY were then eluted from the column into a Teflon® beaker with 10 mL of 8 M ultra-pure HNO_3. Each sample was subsequently evaporated to dryness on a hot plate, and taken up in 10 mL of a 1% v/v ultra-pure HNO_3 solution for analysis (Johannesson et al., 2017). The sample was spiked with ^{115}In at 2 ppb for use as an internal standard and analyzed for the REY isotopes (^{89}Y, ^{139}La, ^{140}Ce, ^{141}Pr ^{146}Nd, ^{147}Sm, ^{153}Eu, ^{157}Gd, ^{159}Tb, ^{163}Dy, ^{165}Ho, ^{166}Er, ^{167}Er, ^{169}Tm, ^{172}Yb, and ^{175}Lu) by ICP-MS at Guangxi University. The analyzed data were assessed for accuracy and precision through quality assurance and quality control, which included reagent blanks, duplicate tests, and the National Research Council Canada Certified Reference Material (NASS-7). The analytical precision of REY analyses was consistently less than 10% RSD.

2.4. Climate and Environmental Data

Date on SST, rainfall, and wind speeds were collected from the WZI Weather Station. The East Asian Summer Monsoon (EASM) indices were obtained from the China Climate Bulletin (http://www.ncc-cma.net/channel/news/newsid/100060). Chlorophyll-*a* (Chl-*a*) concentration data were archived by the Environmental Research Division's Data Access Program (ERDDAP) of the National Oceanic and Atmospheric Administration (NOAA) (https://coastwatch.pfeg.noaa.gov/erddap/index.html). The sea surface salinity (SSS) data were obtained from the website http://www.ocean.iap.ac.cn/. The wet season of WZI is defined as the period between May and October, while the dry season is the interval between November and April.

2.5. REY Parameters

The REY parameters for W3 coral have been normalized using the Post Archean Australian Shales (PAAS) REY patterns (Pourmand et al., 2012). The ratio of $[Nd/Yb]_{PAAS}$ (Nd_N/Yb_N) is a common indicator to represent the fractionation of light rare earth elements (LREE, La-Eu) and heavy rare earth elements (HREE, Gd-Lu) (Henderson, 1984). Yttrium/holmium (Y/Ho) is commonly used in corals to indicate the degree of influence of continental sources, which are more enriched in Ho than in Y (Nguyen et al., 2013).

3. Results

3.1. Variation of the Coral REY

The total REE levels in W3 coral samples from 1999 to 2008 ranged from 136 to 775 ng/g, with an average of 202 ng/g (Figure 2). These values are substantially higher than those reported for sites in the SCS that have experienced minimal human disruption, such as Xiaodonghai Bay (average 106 ng/g) (Jiang et al., 2017, 2018b), Yongxing Island (average 42 ng/g) (Jiang et al., 2018a), and Longwan Bay (average 79 ng/g) (Liu et al., 2011), but lower than the areas that have been strongly impacted by human activities, such as Hong Kong (average 419 ng/g) (Liu et al., 2006, 2011) and Nha Trang Bay (average 340 ng/g) (Nguyen et al., 2013).

The monthly variation of REY parameters of W3 coral is shown in Figure 2. The coral had average REE/Ca and Y/Ca values of 149.03 ± 52.76 nmol/mol and 487.79 ± 41.57 nmol/mol, respectively. The coral REE/Ca showed a significant correlation ($r = 0.8$, $p < 0.001$, $n = 120$) to coral Y/Ca from 1999 to 2008. The average Y/Ho values for the coral are 152.53 ± 18.85. Notably, the Nd_N/Yb_N ratios (average 0.40, $n = 120$) exhibit a significant seasonal cyclicality, that is, high values in the wet season and low values in the dry season (Figure 2a). Additionally, a high value of REE/Ca (Y/Ca) occurs during the dry season (Figures 2c and 2d). The REY parameters in

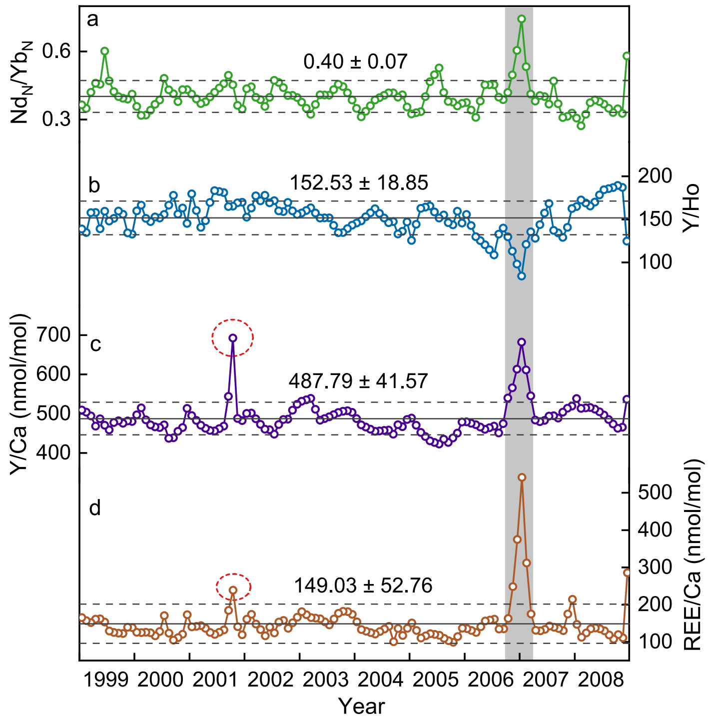

Figure 2. Monthly variation of REY parameters for W3 coral during 1999–2008. (a) Nd_N/Yb_N, (b) Y/Ho, (c) Y/Ca (nmol/mol), and (d) REE/Ca (nmol/mol). Solid lines represent the mean value, and dotted lines show the mean ± standard deviation. The red circles and gray background represent the anomalous peaks and valleys. The gray background corresponds to the middle and late construction period of the crude oil terminal project in the northwest of WZI (from October 2006 to March 2007).

the coral exhibit relative stability, except for anomalous peaks and valleys, which are primarily concentrated between September 2006 and March 2007. There was also an anomalous peak of REE/Ca (Y/Ca) in 2001, which may indicate an unconfirmed anthropogenic source (Jiang et al., 2022).

3.2. Normalized REY Patterns

The faithfulness and robustness of modern coral (e.g., W3 coral) marine REY proxies are empirical (Webb et al., 2009). The W3 coral samples have typical seawater REY patterns (Figure 3c), characterized by LREE depletions, HREE enrichments, negative Ce anomalies, and high Y/Ho ratios (∼152, $n = 120$). Additionally, the lack of a correlation between the REY and Zirconium (Zr) (Figure S3 in Supporting Information S1) in the coral samples suggests that the coral REY can accurately record the surface seawater conditions (Jiang et al., 2018b). The REY concentration in the W3 coral was higher than that in the WZI coral (Li et al., 2019), despite both corals being from WZI (Figures 3c and 3d). Deviating from the surface seawater in the SCS (Alibo & Nozaki, 2000), the REY patterns for the WZI soil (Cai et al., 2021) and WZI basalt (Cai et al., 2021) are characterized by enrichment in LREE, positive Eu anomalies, and high REY concentrations (Figures 3a, 3b, and 3f).

4. Discussion

4.1. Factors Affecting REY Variations in Surface Seawater

The variations of REY in the SCS surface waters could be caused by several factors, including river runoff, dust and precipitation from the atmosphere, erosion of surrounding peninsulas and islands, and sediment remobilization from the shelf (Alibo & Nozaki, 2000). The impact of riverine inputs on REY, however, is deemed negligible due to the W3 coral site being far from large rivers on the Asian continent (Li et al., 2019; Wu et al., 2022). Furthermore, the offshore seawater REY in the WZI is not affected by eolian dust, as evidenced by the remarkably low eolian dust indexes reported by Nozaki et al. (1998), Amakawa et al. (2000), and Li et al. (2019).

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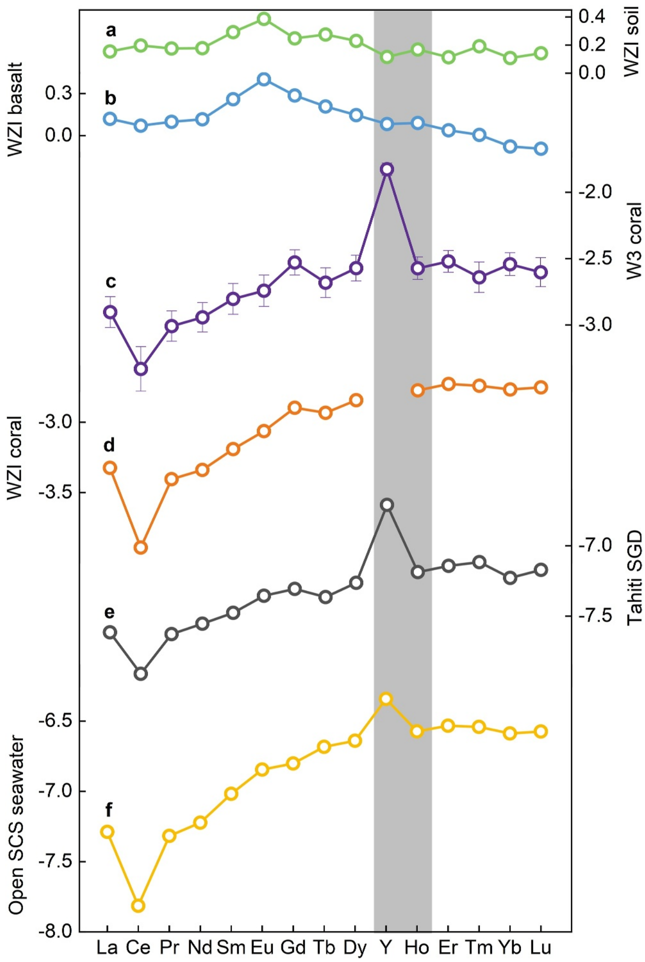

Figure 3. PAAS-normalized distribution patterns for REY. (a) WZI soil (Cai et al., 2021), (b) WZI basalt (Cai et al., 2021), (c) W3 coral (this study), (d) WZI coral (in the southwest of WZI) (Li et al., 2019), (e) Tahiti SGD (Molina-Kescher et al., 2018), and (f) open surface water of the SCS (Alibo & Nozaki, 2000). The ordinate is $\log_{10}$ (sample/PAAS) and the abscissa is REY. The gray background corresponds to the standardized Y and Ho.

Despite our previous research indicating that there were relatively small anthropogenic contributions of vanadium and nickel during the decade of 1999–2008 (Jiang et al., 2022; Wu et al., 2022), the REY parameters still demonstrate the impact of human activities due to their sensitivity. The REY parameters exhibited substantial anomalies between September 2006 and March 2007 (Figure 2). These anomalies are unlikely to be derived from natural sources and may be related to the construction of the crude oil terminal project in WZI during the middle and late stages. Coral V/Ca ratio is an effective indicator of oil pollution in coastal areas (Tanaka et al., 2013). It is worthy of note that an anomalous peak in the V/Ca ratio of W3 coral was also observed in late 2006 and early 2007, which may be correlated with the oil pollution events (Jiang et al., 2022). Furthermore, the observed depletion of Y/Ho ratios and the elevated Nd_N/Yb_N and REY/Ca in W3 coral during this period may be caused by the fact that when the dredged spoils were dumped into the seawater, the Ho-enriched and LREE-enriched terrigenous materials released an excess of Ho and LREE into the seawater (Nguyen et al., 2013). The REY/Ca ratios were significantly correlated with both the Nd_N/Yb_N ratios ($r = 0.99$, $p < 0.001$, $n = 6$) and the Y/Ho ratios ($r = -0.93$, $p < 0.01$, $n = 6$) (Figure S4 in Supporting Information S1) suggesting that total REY concentrations during this period exhibited a heightened sensitivity to increases in Ho and LREE, compared to HREE and Y. Consequently, the source of REY may be dominated by anthropogenic activity from September 2006 to March 2007. The study of natural drivers did not cover the changes in REY parameters during this period.

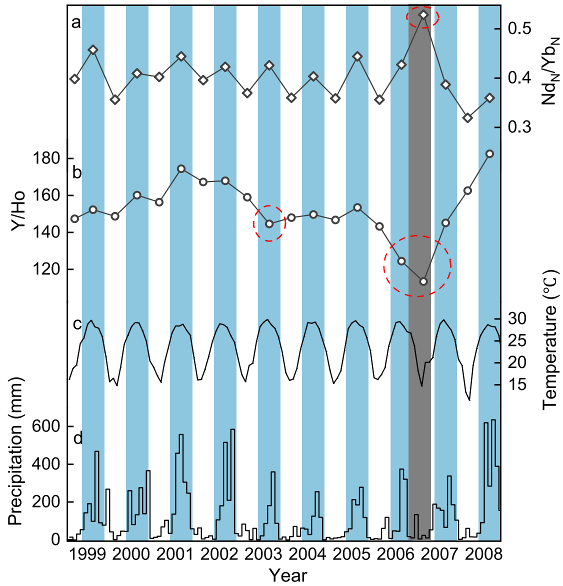

Figure 4. Seasonal variations of W3 coral REY parameters and environmental parameters during 1999–2008. (a) Nd_N/Yb_N, (b) Y/Ho, (c) temperature (°C), and (d) precipitation (mm). The white background represents the dry season of WZI (from November to April), and the blue background color represents the wet season of WZI (from May to October). The three red circles indicate a period characterized by inconsistent seasonal variation trends in REY parameters, and the gray background indicates a period with anthropogenic sources of REY.

In summary, to explore the natural drivers for the dissolved REY in WZI surface seawater, the following possibilities are studied: (a) Basalt chemical weathering driven by SGD, (b) remobilization of REY in the shelf sediments, and (c) biogeochemical cycle.

4.1.1. Basalt Chemical Weathering Driven by SGD

SGD is an important component of the global ocean hydrological cycle (Moore, 2010; Zhang et al., 2022) and is recognized as an essential flux of dissolved and particulate REY to coastal waters (Jiang et al., 2018b; Kim & Kim, 2011, 2014). The lithologic high permeability of marine islands is an important controlling factor for REY flux into the ocean (Kim & Kim, 2011). WZI exhibits high permeability due to its developed pore-fissure structure. This structure is composed of a series of basic volcanic rock types, including olivine basalt, basaltic tuff, and basaltic breccia (Chen, 1999). In contrast to the open surface seawater of the SCS (~0.023 μg/L), PW4 SGD (~0.087 μg/L) has been observed to exhibit elevated concentrations of REY. In general, terrestrial sources exhibit higher concentrations of REEs than seawater, especially LREEs. The coral Nd_N/Yb_N ratios exhibited a distinct seasonal variation pattern, characterized by elevated ratios during the wet season and reduced ratios during the dry season (Figure 4a). Thus, it is plausible to assert that SGD may contribute to REY fluxes of coastal areas in WZI.

Rock weathering converts primary minerals into secondary minerals and solutes, which are then transported to the ocean by SGD and surface runoff (Worthington et al., 2016). A similar effect occurs in volcanic rocks; thus, the weathering of volcanic islands is considered to be a significant source of REY in the seawater near volcanic islands (Johannesson et al., 2017; Molina-Kescher et al., 2018). Basalts are extremely susceptible to weathering (Louvat & Allègre, 1997), exhibiting a bulk weathering rate that is higher than that of many other silicate rocks (Dessert et al., 2001; Worthington et al., 2016). Previous studies have indicated that there are elevated REE contents in WZI basalt (ΣREE = 237 mg/kg) (Cai et al., 2021), with values approximately 160% of the upper continental crust (ΣREE = 146 mg/kg) (Taylor & McLennan, 1985). The chemical weathering of basalt may be an important source of REY in the surface seawater of WZI. In general, the rate of chemical weathering of groundwater on volcanic islands is always higher than that of surface water (Rad et al., 2007). Furthermore, young volcanic areas have generally higher hydraulic conductivity, facilitating the quick recharge of precipitation to

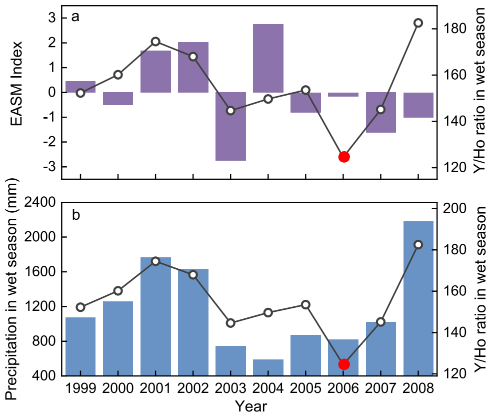

Figure 5. Comparison of the average W3 coral Y/Ho ratio of the wet season with the EASM index and precipitation of the wet season during 1999–2008. (a) Comparison of Y/Ho and the EASM index and (b) Comparison of Y/Ho and the precipitation. The red dot represents the coral Y/Ho value during the wet season of 2006, which was affected by the anthropogenic source of REY.

groundwater (Zang et al., 2023). In summary, the REY released by basalt weathering may be primarily discharged to the ocean via SGD rather than surface runoff due to the absence of rivers on WZI.

Weathering of basalt produces a solid mineral residue that is rich in Fe- and Mn- (hydro)oxides (Campodonico et al., 2019). The affinity of Y for Fe- and Mn- (hydro)oxides is substantially lower than that of holmium (Ho), despite their similar ionic radii (Bau, 1999; Bau et al., 1997). Thus, Y-Ho fractionation always occurs during basalt weathering (Thompson et al., 2013). The Y/Ho ratio of soil is lower than that of basalt on WZI (Figures 3a and 3b), confirming that Y is more mobile in weathering products from basalt compared to Ho. Notably, the majority of the erosion and migration of terrestrial materials in tropical islands predominantly occurs in the wet season (Nunes et al., 2023). The region's high precipitation during the wet season, combined with its high-permeability basalt aquifer, facilitates a rapid increase in groundwater fluxes, thereby promoting chemical weathering and water-rock interactions (Horta-Puga & Carriquiry, 2012; Kumar et al., 2019). Interestingly, the Y/Ho ratios of the W3 coral exhibit obvious seasonal cycles, with high values in the wet season and low values in the dry season, except for 2003 and 2006 (Figure 4b). The low Y/Ho ratios observed during the wet seasons of 2003 and 2006 may be related to the low EASM index (Figure 5a) and human activities, respectively. In particular, the PW4 SGD sample in WZI exhibited a high Y/Ho ratio (~123). Continuous rainwater leaching during the wet season provides a driving force for the downward migration of REY (especially Y), and groundwater with high Y/Ho ratios is transported to the surface seawater, where it is subsequently recorded by corals. Rainfall is the only natural source of replenishment for the abundant groundwater in WZI (Li & Qian, 2018). Therefore, precipitation data can be used to estimate SGD in WZI in the absence of groundwater data. The calculation of the coral residual $\delta^{18}O$ ($\Delta\delta^{18}O$) is achieved by subtracting the contribution of temperature from the coral $\delta^{18}O$ (Methods S3 in Supporting Information S1). Coral $\Delta\delta^{18}O$ can be used to indicate the seawater $\delta^{18}O$ isotope variations, which may correlate with SSS and precipitation (Corrège, 2006; Gagan et al., 1998). The 5-month moving average of $\Delta\delta^{18}O$ shows a significant positive correlation with SSS ($r = 0.39$, $p < 0.001$; Figure S5 in Supporting Information S1), whereas no significant correlation was observed with precipitation ($r = -0.22$, $p > 0.01$; Figure S5 in Supporting Information S1). It is interesting to note that the 5-month moving average variation of precipitation exhibited significant positive correlations with coral Nd_N/Yb_N ratios ($r = 0.34$, $p < 0.001$; Table S4 in Supporting Information S1) and Y/Ho ratios ($r = 0.45$, $p < 0.001$; Figure 6). Furthermore, there is also a notable correlation between coral Y/Ho ratios and precipitation during the wet season ($r = 0.37$, $p < 0.01$, $n = 60$; Figure S6 in Supporting Information S1), supporting that the Y/Ho may serve as an excellent proxy for basalt weathering in coastal seawater during the wet season, when SGD from the island represents the primary source of REY in the coastal waters.

Our findings suggest a substantial contribution of SGD from basaltic islands to the enrichment of REY in coastal seawater, which is supported by studies conducted in Kona, Hawaii (Johannesson et al., 2017), Jeju Island, Korea (Kim & Kim, 2011), and Tahiti Island (Molina-Kescher et al., 2018). It is noteworthy that our research, conducted in the WZI, may represent the inaugural report of this effect recorded by a coral.

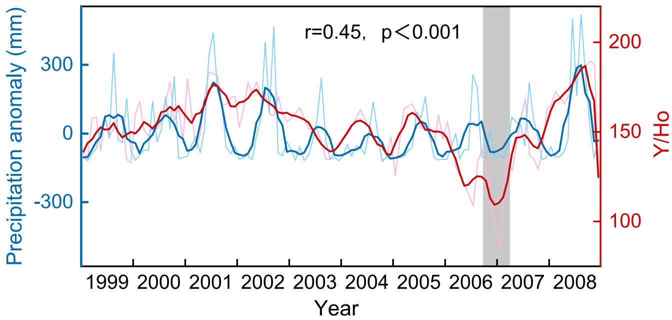

Figure 6. Comparison of W3 coral Y/Ho and precipitation anomaly during 1999–2008 (the average value of precipitation is 118.6 mm). The bold lines represent the 5-month moving average, and the fine lines represent the monthly variation. The gray background is the period with anthropogenic sources of REY.

4.1.2. Remobilization in the Bottom Sediments

Given that strong weathering and high SGD discharge would be expected to occur mostly during the summer seasons, however, this form seemed to contradict variations in the coral REE/Ca (Y/Ca) time series, which reach the peak value during the dry season (Figures 2c and 2d). In general, the majority of terrestrial dissolved and particulate REY in the near-coastal areas of WZI may be derived from SGD, due to the island's geographical positioning surrounded by the sea. Relevant to the high accumulation of REY in marine sediments, it is noteworthy that the resuspension of sediments and the upward diffusion of pore fluids may serve as important sources contributing to the presence of dissolved REY in surface seawater (Abbott et al., 2015; Crocket et al., 2018; Liao et al., 2022).

Strong winds have the potential to resuspend sediments, which may increase the concentration of dissolved elements in seawater. Approximately 50% of bottom sediments in shallow water areas can be resuspended when wind speeds exceed 4 m/s (Booth et al., 2000). The coastal seawater in WZI is very shallow, particularly at the W3 coral sampling site, with an average depth of approximately 5 m (Yu et al., 2019). The wind speed at WZI (average 4.2 m/s) exhibited a seasonal fluctuation under the impact of the strong East Asian Winter Monsoon (EAWM), displaying low wind speeds during the wet season and high wind speeds during the dry season (Figure 7 and Figure S7 in Supporting Information S1). During the dry season, wind speeds in excess of 4 m/s were a common occurrence in this region, creating favorable conditions for sediment resuspension. It is noteworthy that coral REY/Ca ratios also exhibited a similar trend, characterized by high values in the dry season (Figure 7). This pattern corresponds to the occurrence of winter monsoons and elevated average wind speeds observed during the same period. A 5-month moving average time series analysis showed a significant positive

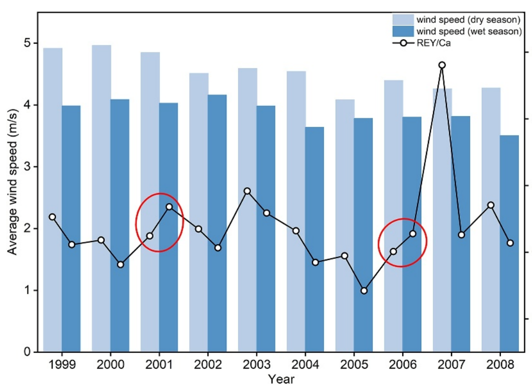

Figure 7. Seasonal variations of average wind speed and W3 coral REY/Ca ratios. The light blue and dark blue colors represent the dry season and the wet season of the average wind speed, respectively. The red circles indicate the year with inconsistent trends. The anomalous increase in REY/Ca values during the wet seasons of 2001 and 2006 interrupted the seasonal cycle of REY/Ca (an increase in the REY/Ca ratio was observed during the dry season), which should be related to artificial disturbances, as discussed above. This indicates that the increase in REY/Ca in surface seawater resulting from wind-induced sediment resuspension may be obscured by the effects of human-induced disturbances.

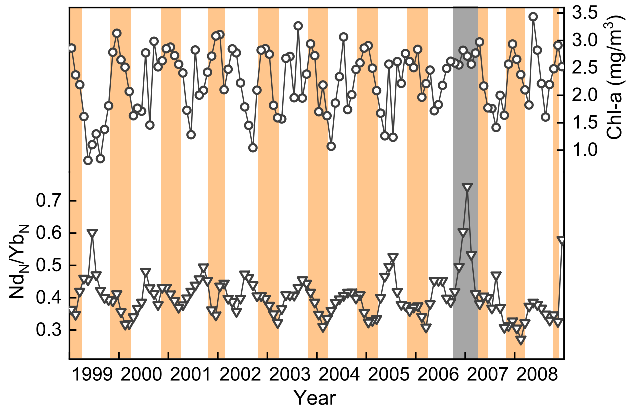

Figure 8. Comparison of W3 coral Nd_N/Yb_N and Chl-*a* during 1999–2008. The gray background is the period with anthropogenic sources of REY, and the orange background highlights that the high values of Chl-*a* correspond to low Nd_N/Yb_N ratios.

correlation between the average wind speed and coral REY/Ca in this area ($r = 0.34$, $p < 0.001$; Table S4 in Supporting Information S1). The occurrence of frequent hurricanes during the summer periods may also induce remobilization of bottom sediments. However, the absence of a correlation between coral REY/Ca ratios and the maximum wind speed ($p = 0.11$, $r > 0.05$; Table S4 in Supporting Information S1) precludes the possibility of an influence from the summer hurricane. In addition, we considered that the effects of summer hurricanes are not comparable to those of sustained winter winds in terms of their short-term impacts. Therefore, the concentrations of dissolved REY are substantially affected by the remobilization and resuspension of marine sediments in surface seawater caused by the strong winter monsoon. This conclusion is consistent with the findings reported by Li et al. (2019). Our study suggests that the signal of strong winter winds can be seen in the coral REY/Ca records on a longer timescale and with a monthly resolution.

4.1.3. Biogeochemical Cycle

Plankton plays an indispensable role in the biogeochemical cycle of trace elements in the ocean ecosystem (Martin & Knauer, 1973). For instance, rapid fluctuations in phytoplankton blooms and bio-particle variations can significantly impact the biogeochemical cycling of REY in shallow water environments (Hara et al., 2009). Seasonal variations in the WZI coral Nd_N/Yb_N ratios with peaks occurring during spring have been observed by Li et al. (2019), due to mineralization processes associated with biological particles.

Wind-induced sediment resuspension can also affect the light and nutrient limitation of phytoplankton, potentially increasing the nutrient supply for growth and enhancing phytoplankton production, as estimated by Chl-*a* (Schallenberg & Burns, 2004). A large number of biogenic particles are subsequently generated (Ma et al., 2014). The concentration of Chl-*a* in WZI generally exhibited a distinct seasonal pattern, with pronounced peaks during the dry season (Figure 8). Interestingly, the high Chl-*a* values observed in the dry season were inversely correlated with coral Nd_N/Yb_N ratios, and vice versa. The coral Nd_N/Yb_N ratios can still respond accurately to the changes in Chl-*a*, despite the potential presence of a time lag of 1–2 months in the time series due to the variable degree of annual growth rate of the coral (Gagan et al., 2012). The prevailing EAWM brings rich nutrients to the surface seawater in WZI during the dry season, promoting the proliferation of plankton. A substantial decrease in the Nd_N/Yb_N ratio was observed during the phytoplankton bloom (Figure 4a, shown with white background), due to biogenic particles preferentially absorbing LREE over HREE (Hara et al., 2009; Strady et al., 2015). Conversely, an increase in the Nd_N/Yb_N ratio was observed during the wet season, which may be caused by the release of LREE through the mineralization and desorption processes of biological particles during the waning winter monsoon. Interestingly, the 5-month moving average of coral Nd_N/Yb_N showed a negative correlation with Chl-*a* ($r = -0.29$, $p < 0.01$; Table S4 in Supporting Information S1). Therefore, the adsorption-desorption process of biological particles in the ocean may be an important factor influencing the REY pattern of seawater in WZI.

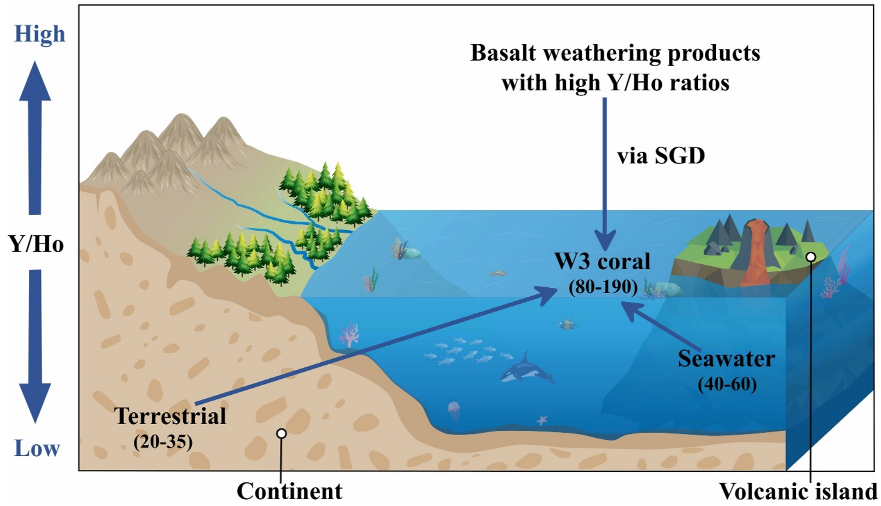

Figure 9. Schematic diagram of Y/Ho ratios in W3 coral and its end-members (terrestrial, seawater, and SGD), with ranges of Y/Ho values in parentheses.

4.2. Reliability of Coral Y/Ho Ratios as Basalt Weathering Proxies

Temperature, runoff, and precipitation are the most important parameters that control chemical weathering (Dessert et al., 2001; Shao et al., 2012). Indeed, the EASM controls the climatic regime of a region through changes in temperature and precipitation (Chen et al., 2017). The coral Y/Ho ratios are not only influenced by SGD associated with precipitation on monthly timescales but also affected by the EASM on interannual timescales (Figure 5). For instance, higher Y/Ho ratios were observed in 2001–2002 and 2008, which coincided with periods of elevated EASM index and heavy precipitation. Conversely, lower Y/Ho ratios were found in 2003 when the EASM index was weak and precipitation was low. In conclusion, the coral Y/Ho ratios exhibited a robust correlation with the EASM and rainfall-induced SGD, suggesting that the coral Y/Ho value is likely to be a potential novel and reliable proxy for chemical weathering intensity on basaltic islands.

The analysis showed noteworthy differences in the Y/Ho values of end-members between conventional continental sources and marine sources (Figure 9). However, the elevated Y/Ho values for W3 coral are too high to be explained by simple, binary mixing between seawater and terrestrial input. Instead, the high Y/Ho values for the coral indicate that there was an additional contribution derived from another flux. The W3 coral exhibits a REY pattern that is strikingly similar to that of Tahiti SGD, another Pacific volcanic island, particularly in terms of the elevated Y/Ho values (Figures 3c and 3e). In contrast to the open surface seawater of the SCS (Alibo & Nozaki, 2000), elevated Y/Ho ratios (~123) have also been observed in the PW4 SGD sample. The groundwater in the WZI was reported to be mainly basalt pore-fissure water (Ma et al., 2019). The coral average Y/Ho ratios during the wet season exhibit a significant positive correlation with precipitation, which controls SGD ($r = 0.95$, $p < 0.001$, $n = 9$; Figure 10). Accordingly, we suggest that the high Y/Ho ratio in W3 coral is most likely the result of the Y-Ho fractionation that occurred during basalt weathering. Ho is preferentially adsorbed onto secondary minerals during weathering, whereas Y is preferentially migrated away in solution, indicating that basalt weathering of the Y-Ho fractionation driven by the SGD contributes substantially to the coastal areas in WZI.

5. Conclusion

This study reconstructed the monthly variations of REY parameters in the surface seawater from 1999 to 2008, using the geochemical record of a coral from WZI. The Nd_N/Yb_N ratios exhibit a substantially seasonal cyclicity, which may be related to the precipitation associated with SGD and the adsorption-desorption processes of marine biogenic particles, with high values during the wet season and low values during the dry season. The combination of heavy rainfall and high temperature promotes basalt weathering and water-rock interaction during the wet season, which facilitates the release of REY and Y-Ho fractionation, resulting in high Y/Ho values in weathering solutions. Continued rainwater leaching provides a driving force for the downward migration of REY, and then SGD with high Y/Ho ratios is transported into the coastal seawater. Meanwhile, sediments resulting from the

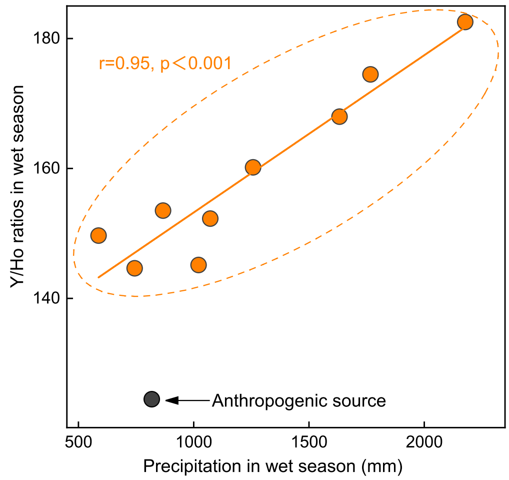

Figure 10. Comparison of the average Y/Ho ratios for W3 coral with the precipitation during the wet season during 1999–2008. The gray dot corresponds to the wet season of 2006, which was affected by the anthropogenic source of REY.

migration of basalt weathering products are deposited on the continental shelf around coastal areas of WZI. In the following dry season, the concentrations of REY in surface seawater are likely affected by the resuspension of marine sediments caused by winter winds, which is reflected in the coral REY/Ca ratios characterized by high ratios in the dry season.

The results suggest that REY fractionation in WZI is mainly controlled by basalt weathering associated with the EASM and SGD estimated by precipitation, as well as the biogeochemical cycle, whereas the total REY content is influenced by sediment resuspension driven by winter monsoon. REY flux from basalt weathering driven by SGD is the dominant natural source in the coastal areas of volcanic islands. This study presents for the first time that there is a substantial correlation between the coral Y/Ho ratio and basalt weathering, suggesting that coral Y/Ho could serve as a reliable proxy for basalt weathering intensity. Our study highlights the potential of this parameter in evaluating past and present basalt weathering. Furthermore, there is also the possibility that coral Nd_N/Yb_N could serve as a novel proxy for Chl-*a*. However, further high-precision data and longer time series are necessary for confirmation, and additional research is needed to strengthen the application of Y/Ho tracking for interannual EASM variability analysis.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

All data used in this study are accessible at Figshare (https://doi.org/10.6084/m9.figshare.28416318.v1) in Gu and Jiang (2025).

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