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Change of coral carbon isotopic response to anthropogenic Suess effect since around 2000s

Xi Liu [a,b,c], Wenfeng Deng [a,b,c,*], Hao Cui [a,b,c,d], Xuefei Chen [a,b,c], Guanqiang Cai [e], Ti Zeng [b,c,f], Gangjian Wei [a,b,c]

[a] State Key Laboratory of Isotope Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, 510640, China
[b] CAS Center for Excellence in Deep Earth Science, Guangzhou, 510640, China
[c] Southern Marine Science and Engineering Guangdong Laboratory, Guangzhou, 511458, China
[d] College of Earth and Planetary Sciences, University of Chinese Academy of Sciences, Beijing, 100049, China
[e] Guangzhou Marine Geological Survey, China Geological Survey, Guangzhou, Guangdong, 510760, China
[f] Key Laboratory of Marginal Sea Geology, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, 510640, China

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ABSTRACT

The stable carbon isotope composition ($\delta^{13}C$) in coral skeletons can be used to reconstruct the evolution of the dissolved inorganic carbon (DIC) in surface seawater, and its long-term declining trend during the past 200 years (~1800–2000) reflects the effect of anthropogenic Suess effect on carbonate chemistry in surface oceans. The global atmospheric CO_2 concentration still has been increasing since 2000, and the Suess effect is intensifying. Considering the coral's ability of resilience and acclimatization to external environmental stressors, the response of coral $\delta^{13}C$ to Suess effect may change and needs to be re-evaluated. In this study, ten long coral $\delta^{13}C$ time series synthesized from different oceans were used to re-evaluate the response of coral carbonate chemistry to Suess effect under the changing environments. These $\delta^{13}C$ time series showed a long-term declining trend since 1960s, but the declining rates slowed in eight time series since around 2000s. Considering that the declining rates of the DIC-$\delta^{13}C$ in surface seawater from the Hawaii Ocean Time-series Station and Bermuda Atlantic Time-series Station has not changed since 2000 compared with those during 1960–1999, the change in the coral $\delta^{13}C$ trends at eight of ten locations may indicate that the response of coral $\delta^{13}C$ to the anthropogenic Suess effect has changed since around 2000s. This change may have resulted from coral acclimatization to external environmental stressors. To adapt to acidifying oceans, coral may have the ability to regulate the source of DIC in extracellular calcifying fluid and/or the utilization way of DIC, therefore the response of coral $\delta^{13}C$ to anthropogenic Suess effect will change accordingly.

1. Introduction

The stable oxygen isotope ($\delta^{18}O$) in coral skeletons is usually used as a proxy for past sea surface temperature and salinity (e.g., Corrège, 2006). However, the interpretation of the stable carbon isotope ($\delta^{13}C$, routinely measured with $\delta^{18}O$) remains difficult. Complicated factors, such as solar radiation, cloud cover, rainfall, vital effects, kinetic isotope effect, terrestrial dissolved inorganic carbon (DIC) input and human activity, control the value of coral skeletal $\delta^{13}C$ on the seasonal time scale (e.g., Gagan et al., 1994; Grottoli and Wellington, 1999; Maier et al., 2003; McConnaughey, 1989; Moyer and Grottoli, 2011; Sun et al., 2008; Swart et al., 1996; Xu et al., 2018; Li et al., 2020). The $\delta^{13}C$ values of marine biogenic carbonates can be used to indicate the evolution of $\delta^{13}C$ in seawater DIC (Böhm et al., 1996; Graniero et al., 2017), and it is well accepted that the long-term declining trend in the coral $\delta^{13}C$ values over the past 200 years reflects the increase in the transfer of ^{12}C-enriched anthropogenic CO_2 from the atmosphere to the surface oceans (i.e., so-called Suess effect) (Al-Rousan and Felis, 2013; Dassié et al., 2013; Deng et al., 2017; Liu et al., 2014; Pereira et al., 2018; Swart et al., 2010).

Inorganic carbon used for the precipitation of coral skeletal aragonite originates from DIC in the extracellular calcifying fluid (ECF), which

* Corresponding author. State Key Laboratory of Isotope Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, 510640, China.
E-mail address: wfdeng@gig.ac.cn (W. Deng).

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is derived from the metabolic respiration of coral polyps and the external seawater (Gattuso et al., 1999). However, the relative contribution from these two sources remains uncertain (Al-Horani et al., 2003; Furla et al., 2000a; McConnaughey, 2003). Therefore, both internal biological and external environmental factors influencing the inorganic carbon input to the ECF can affect the $\delta^{13}C$ variations in coral skeletons. From this point of view, the response of coral skeletal $\delta^{13}C$ to oceanic Suess effect since the Industrial Revolution resulted from the predominant contribution of external environmental factor (i.e., the secular increase of anthropogenic CO_2) to the inorganic carbon pool in the ECF. Corals are animals, and they have the biological ability of resilience and acclimatization to external environmental stressors (Barshis et al., 2013; McCulloch et al., 2012; Putnam, 2012; van Oppen et al., 2015). Therefore, it is possible that the relative contribution of biological and environmental factors to inorganic carbon pool in ECF will change as oceans' uptake of anthropogenic CO_2 increases to some extent. If this possibility comes to fruition, the response of coral $\delta^{13}C$ to oceanic Suess effect will change accordingly.

Ten years ago, Swart et al. (2010) first demonstrated the effect of Suess effect on the secular decrease in coral skeletal $\delta^{13}C$ during the period ~1800–2000. The global atmospheric CO_2 concentration has increased from ~370 ppm to ~410 ppm during the past two decades (The keeling Curve, https://keelingcurve.ucsd.edu/), but there was a slowing of the growth rate of atmospheric CO_2 due to enhanced terrestrial carbon since 2002 (Keenan et al., 2016). The reconstruction on the history of anthropogenic CO_2 in the ocean over the Industrial period indicated that oceans' CO_2 uptake has declined by ~10% during 2000–2007 (Khatiwala et al., 2009). If the change in ocean and atmosphere CO_2 flux has actually occurred around 2000s, it should be imprinted in surface oceans and recorded by corals. Therefore, it is time to re-evaluate the response of coral carbonate chemistry to Suess effect since around 2000s. Here, the coral $\delta^{13}C$ time series covering the most recent decades from the different oceans were synthesized to re-evaluate the effect of oceanic Suess effect on coral skeletal $\delta^{13}C$ and to provide new insights for coral's response to global change stressors.

2. Materials and methods

2.1. Coral $\delta^{13}C$ data from the northern South China sea

In September 2015, modern coral cores with a diameter of 8 cm, referred to herein as XS15 and XS9, were extracted using an underwater pneumatic drill from two *Porites lutea* colonies with diameters of approximately 1.5–2.0 m, from a water depth of 2.5 m on the Qilianyu Reef in the Xisha Islands of the northern South China Sea (SCS) (Fig. 1). The coral cores were first cut lengthwise into slabs 1-cm thick and 7-8-cm wide. Then X-ray photographs were taken to reveal regular and well-defined annual density bands (Figure S1). Next, the coral slabs were ultrasonically cleaned in deionized water, rinsed three times, and dried at 40 °C prior to sampling. Guided by X-ray photographs, the annual resolution sub-samples of XS15 (1822–2015) and XS9 (1834–2015) were collected from annual bands from sampling grooves along the main growth axes, then ground into a fine powder (ca. 200 mesh) using an agate mortar and pestle set. Each high- and low-density band constitutes an annual couplet, generally representing 1 year of growth (Knutson et al., 1972). Coral skeletal $\delta^{13}C$ and $\delta^{18}O$ stable isotope analysis was conducted on a stable isotope ratio mass spectrometer coupled with a carbonate device MultiPrep™ that used 102% H_3PO_4 at 90 °C to extract CO_2 from the coral samples. Multiple measurements (n = 88) on a Chinese national standard GBW04405 yielded a standard deviation of 0.03‰ for $\delta^{13}C$ and 0.06‰ for $\delta^{18}O$. Duplicate measurements were made on approximately 15% of the samples. The details referred to the method of Deng et al. (2017).

2.2. Coral $\delta^{13}C$ data from published references

Aiming to obtain the most recent trend of the $\delta^{13}C$ in coral skeletons, almost all long coral $\delta^{13}C$ time series available from National Oceanic and Atmospheric Administration (NOAA) paleoclimatology archive and extending at least to 2007 from different locations (Fig. 1), including Belize in the Caribbean Sea (FR-02, 1912–2008 and BR-06, 1915–2008; Fowell et al., 2018), Kimberley in Western Australia (KIM16, 1919–2015; Chen et al., 2020), Brazil in the South Atlantic (13SS-1, 1961–2012; Pereira et al., 2018), Palau in the West Pacific (UC, 1960–2007 and NGB, 1963–2007; Osborne et al., 2013), and American Samoa in the South Pacific (Tau-1, 1521–2011; Linsley et al., 2019), were collected for study. Another $\delta^{13}C$ time series of coral Yongxing (1851–2007, Han et al., 2019) from the Xisha Islands of the northern SCS was also included. The original monthly resolution data by Osborne et al. (2013) and Pereira et al. (2018) were converted into the annual resolved time series by averaging all monthly data in each year. The detailed information of these coral $\delta^{13}C$ time series, including those newly generated from the northern SCS, is summarized in Table 1. To compare the data from different locations, all coral time series were normalized by centering the $\delta^{13}C$ values (subtracting the mean from each individual coral $\delta^{13}C$ value over the common time period

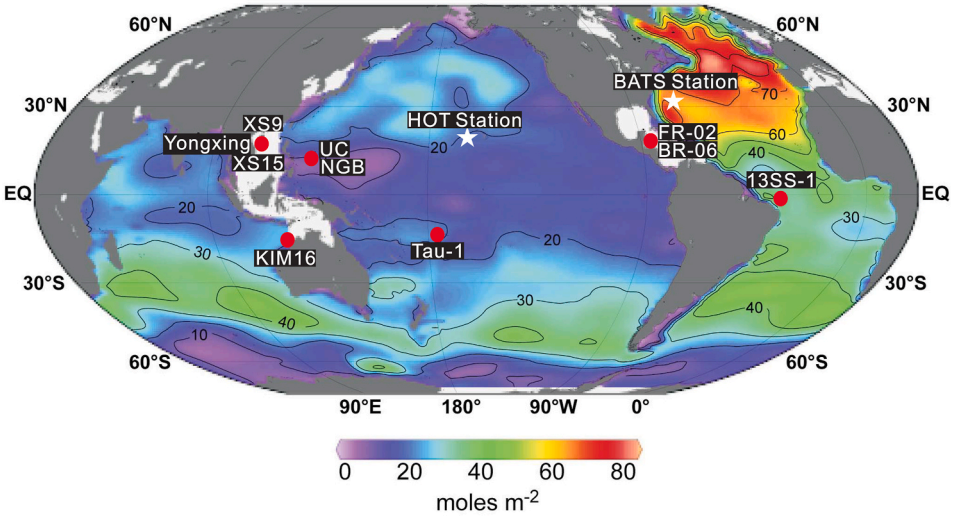

Fig. 1. Global map of the column inventory of anthropogenic CO_2 in 1994 (Sabine et al., 2004) and the locations of the coral $\delta^{13}C$ time series (red dots) and the HOT and BATS stations (white stars). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Locations, sample names and water depths of the shallow water corals, and the linear variation rates of the coral $\delta^{13}C$ time series.

Location	Sample Name	Latitude	Longitude	Water Depth (m)	Linear Variation Rate (‰·yr^{-1})				Reference
					1800–1959	1960 - Collection Year	1960–1999	2000 - Collection Year	
South China Sea	XS15	16.9438°N	112.3327°E	6	0.002	−0.028	−0.022	0.020	This study
	XS9	16.9720°N	112.2701°E	9	−0.007	−0.029	−0.032	−0.010	
	Yongxing	16.8400°N	112.3300°E	6	−0.002	−0.017	−0.020	0.072	Han et al. (2019)
American Samoa	Tau-1	14.2594°S	169.5004°W	7.5	−0.004	−0.031	−0.038	−0.008	Linsley et al. (2019)
Palau	UC	7.0045°N	134.0042°E	12	n.a.[a]	−0.014	−0.016	0.007	Osborne et al. (2013)
	NGB	7.0068°N	134.0073°E	3	n.a.	−0.029	−0.032	0.018	
Kimberley	KIM16	16.4771°S	123.0424°E	2	0.012	−0.024	−0.024	−0.007	Chen et al. (2020)
Brazil	13SS-1	3.8500°S	33.8167°W	7	n.a.	−0.021	−0.022	0.010	Pereira et al. (2018)
Belize	FR-02	16.1372°N	88.2529°W	3–5	−0.005	−0.009	−0.010	0.041	Fowell et al. (2018)
	BR-06	16.1405°N	88.2602°W	3–5	−0.008	−0.024	−0.019	0.001	

[a] n.a. = not available.

1963–2007 for all the time series). The long-term variation trends in all total coral $\delta^{13}C$ time series and those from 1960 to the collection years are shown in Figures S2 and S3, respectively. To quantitatively show the trend of change rates in coral $\delta^{13}C$ time series with time over the study period, a 20-year rolling window regression was performed on all coral $\delta^{13}C$ time series since 1960.

2.3. Oceanic and atmospheric CO_2 observational data

The time series data of the DIC-$\delta^{13}C$ (https://scrippsco2.ucsd.edu/data/seawater_carbon/ocean_time_series.html, Lueker et al., 1998) in surface seawater from the Hawaii Ocean Time-series (HOT) Station in the North Pacific (22°45′N, 158°00′W), and the Bermuda Atlantic Time-series (BATS) Station (31°50′N, 64°10′W) in the North Atlantic were employed for comparison with the coral $\delta^{13}C$ data. The two stations are long-term, ship-based ocean time series observation and research sites (Ducklow et al., 2009). The original observation data at the depths of 5 m and 10 m in HOT and BATS respectively were converted into annual resolved time series by averaging all the data in each year.

The time series of the global average annual CO_2 concentration in the atmosphere compiled by the Institute for Atmospheric and Climate Science (IAC) at Eidgenössische Technische Hochschule in Zürich, Switzerland (https://www.co2.earth/historical-co2-datasets), along with the compiled records of the annual mean $\delta^{13}C$ in the global atmospheric CO_2 for historical simulations in the Coupled Model Intercomparison Project 6 (https://www.geosci-model-dev.net/10/4405/2017, Graven et al., 2017), were also used for the comparative study with coral $\delta^{13}C$ data.

3. Results and discussion

3.1. Variation trends in coral $\delta^{13}C$ values since 1960s

The global oceanic carbon sink since 1960 was mainly attributed to rising atmospheric CO_2 (Séférian et al., 2014). For the corals dating back to pre-1960, the decline rates in their $\delta^{13}C$ values during the period pre-1960 were much lower than those during the period post-1960 (Table 1). For all coral $\delta^{13}C$ series, their variations after the 1960s are more concentrated than those during the period pre-1960 to match with the $\delta^{13}C$ variation of the atmospheric CO_2 (Figure S2), indicating the primary control of atmospheric CO_2 on the long-term variation trend of coral $\delta^{13}C$ since 1960. These characteristics mean that the change of $\delta^{13}C$ values in coral skeletons suffered from the intensifying ^{13}C Suess effect by more addition of ^{12}C-enriched anthropogenic CO_2 into the surface oceans and therefore can be used to discuss the issue of coral $\delta^{13}C$ response to anthropogenic CO_2. Generally, the decline rates of $\delta^{13}C$ values in different coral skeletons were very similar during the period post-1960, and their average decline rate −0.024‰·yr^{-1} (Table 1) was almost the same as that of the $\delta^{13}C$ values of atmospheric CO_2 (−0.025‰·yr^{-1}, Figure S2). Based on 37 coral $\delta^{13}C$ time series from different oceans, Swart et al. (2010) found that the decline rate in $\delta^{13}C$ values of coral skeletons from the Atlantic Ocean was essentially the same as that in the $\delta^{13}C$ values of atmospheric CO_2 and much higher than in those corals from the Pacific and Indian Oceans since 1960. The interoceanic difference was attributed to the different carbon cycle processes, i.e., CO_2 is being recharged into the oceans in the Atlantic while more deep water is being returned to the surface Pacific Ocean (Swart et al., 2010). This difference was not observed in this study, which may be caused by limited sample sizes used. It should be noted that there were large intercolony differences in the decline rates of $\delta^{13}C$ values for the corals from the SCS, Palau and Belize (Table 1). Assuming the $\delta^{13}C$ values of corals living in relatively shallow water are more vulnerable to atmospheric CO_2 than those living in relatively deep water, the difference between the two corals from Palau may attribute to the different water depths. However, the intercolony differences for the corals from the SCS and Belize should result from the vital effects and/or the water clarity since they lived in the same depth of water, because the increased water turbidity leads to the reduction in the amount of light reaching the coral and consequently affects the coral skeletal $\delta^{13}C$ values via decreasing the efficiency of endosymbiotic zooxanthellae photosynthesis (Yentsch et al., 2002; Shimamura et al., 2008).

All the time series of coral skeletal $\delta^{13}C$ data showed a long-term declining trend in recent decades, but the declining trend became slower after 2000 (Figure S3). To clearly display the difference between the declining trends before and after 2000, the variation rates during the periods from 1960 to 1999 and after 2000 were estimated (Fig. 2 and Table 1). There were increasing trends in the $\delta^{13}C$ time series of XS15 and Yongxing from the northern SCS, FR-02 and BR-06 from Belize, 13SS-1 from Brazil, and UC and NGB from Palau since 2000 (Fig. 2a and c and f-i, Table 1), and the declining trends in other coral $\delta^{13}C$ time series became slower from then on (Fig. 2b, and 2d-e, Table 1). In addition, eight of the ten results for the 20-year rolling window regression clearly revealed that the decline rates of coral $\delta^{13}C$ time series have decreased since around 2000s (Fig. 3a–f, and 3h-i). As for the other two time series (BR-06, Fig. 3g and NGB, Fig. 3j) in which the decline rates of coral $\delta^{13}C$ values didn't show a decrease but an increase, the decline rates in their corresponding duplicates (FR-02, Fig. 3f and UC, Fig. 3i) from respective sampling locations (Belize and Palau, respectively) decreased since around 2000s. This intercolony difference may due to the different water depths and vital effects as mentioned above. Considering that the coral $\delta^{13}C$ time series analyzed here were from the Pacific, Indian and Atlantic oceans, the change in the decline rates of coral $\delta^{13}C$ values since around 2000s possibly seems a very common phenomenon in different oceans.

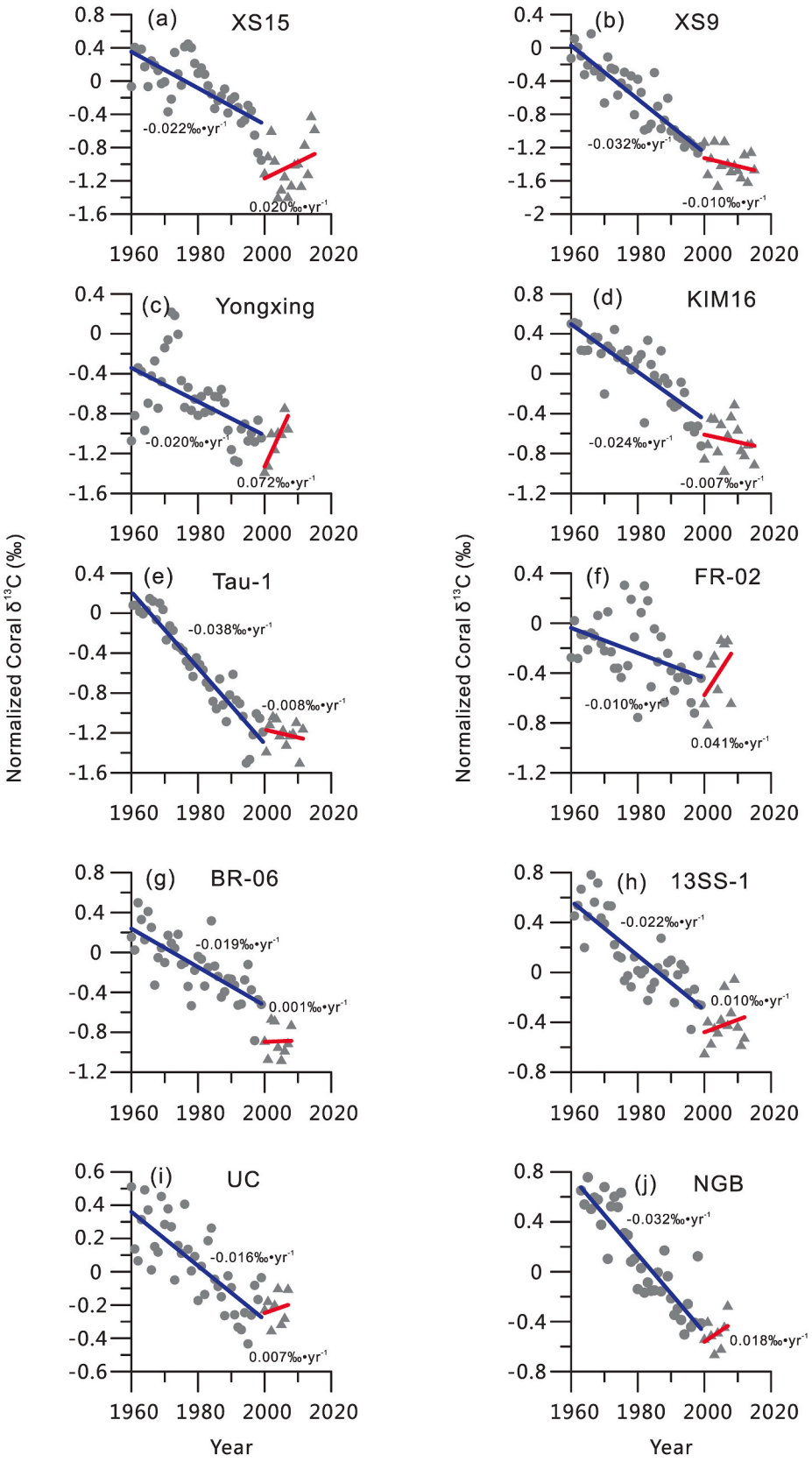

Fig. 2. The variations in the coral $\delta^{13}C$ time series during the periods from 1960 to 1999 and after 2000. The blue and red lines represent the linear fits of the variation trends during the periods from 1960 to 1999 and after 2000, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

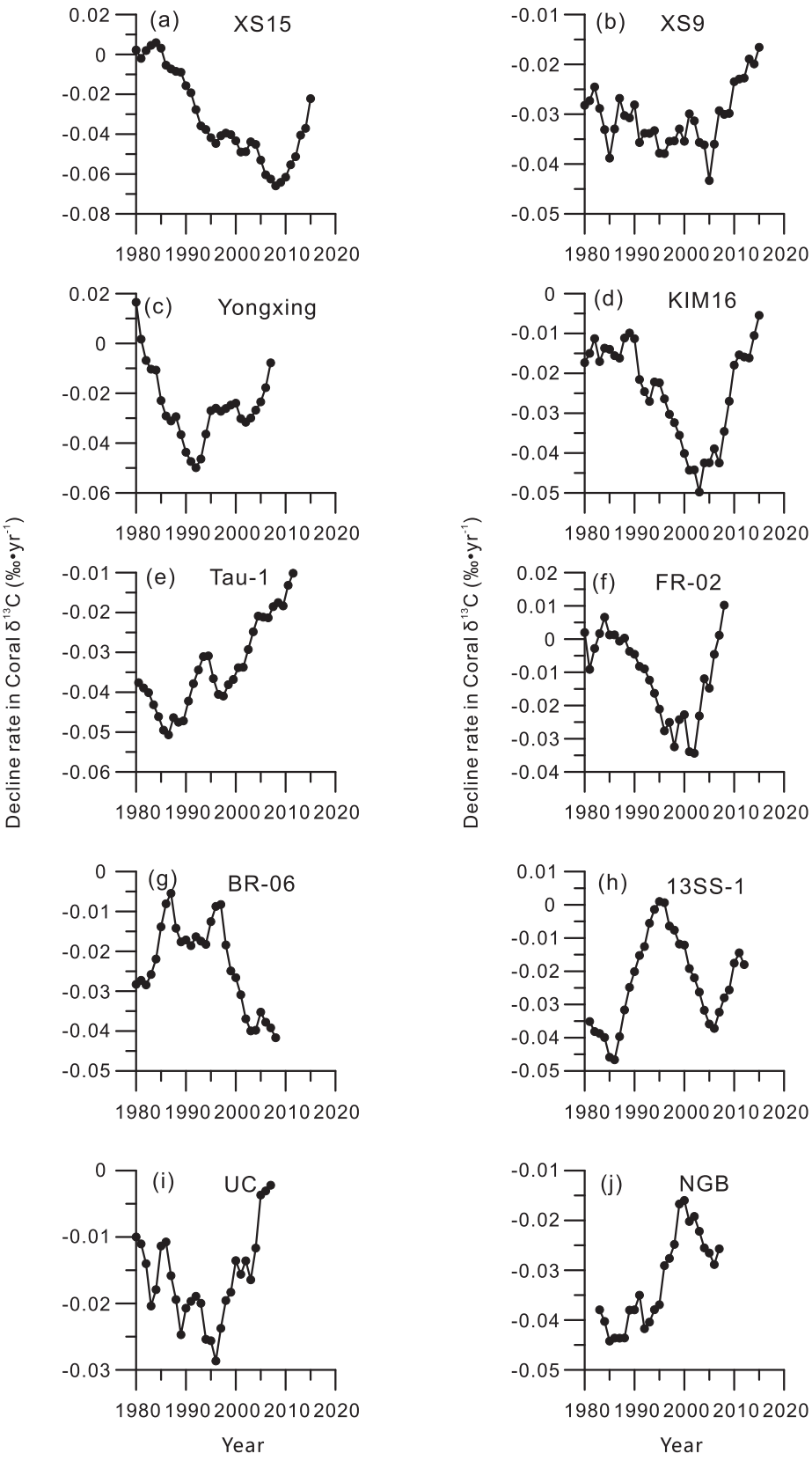

Fig. 3. The 20-year rolling window regression results showing the decline rates of all coral $\delta^{13}C$ time series since 1980.

3.2. Possible factors inducing the change of decline rate of coral $\delta^{13}C$

The kinetic effect associated with the coral growth rate may induce positive relationship between skeletal $\delta^{13}C$ and $\delta^{18}O$, and the faster coral growth leads to more negative $\delta^{13}C$ and $\delta^{18}O$ values and vice versa (McConnaughey et al., 1989). The potential of this effect has been considered carefully as follows. For the two Belize corals, their linear growth rates increased after around 2000 (Fowell et al., 2018). If the kinetic effect associated with growth rate plays a dominant role in regulating skeletal isotope compositions, their $\delta^{13}C$ should decrease rather than increase or keep stable. Therefore, the kinetic effect associated with growth rate should not be the main factor in changing the decline rate of coral $\delta^{13}C$ in Belize. The similar situation occurred for the corals form Palau and Western Australia, their $\delta^{18}O$ values decreased after around 2000 (Osborne et al., 2013; Chen et al., 2020), but their $\delta^{13}C$ values increased or slowed down the decrease since then (Fig. 2). The growth rate of the coral from Brazil has been decreasing since 1961 (Pereira et al., 2018). If the kinetic effect associated with growth rate played the leading role on this coral, the $\delta^{13}C$ should become more positive since then. However, the fact is that the coral $\delta^{13}C$ has been declining due to Suess effect since 1961 (Pereira et al., 2018). Therefore, the effect of the growth rate on the change of decline rate of coral $\delta^{13}C$ can be excluded. There is a weak positive relationship between $\delta^{13}C$ and linear growth rate ($r = 0.42$, $n = 11$, $p = 0.1$) during the period 2000–2010 for the coral from American Samoa (Linsley et al., 2019), which is not consistent to growth rate-related kinetic effect. The kinetic effect on the $\delta^{13}C$ of the coral Yongxing from the SCS has been ruled out by Han et al. (2019). The $\delta^{13}C$ and $\delta^{18}O$ values of the other two corals from the SCS are weakly negatively correlated ($r = -0.37$, $n = 16$, $p = 0.08$ for XS15 and $r = -0.32$, $n = 16$, $p = 0.11$ for XS9, respectively) during the period 2000–2015, indicating that the kinetic effect is not the primary factor responsible for the change in the decline rate of coral $\delta^{13}C$. Based on above analyses, the kinetic effect associated with the coral growth rate cannot explain the change in the decline rate of coral $\delta^{13}C$ since around 2000.

The increase of the solar irradiance can enhance the photosynthesis of endosymbiotic zooxanthellae and therefore lead to the increase of coral skeletal $\delta^{13}C$ (Swart et al., 1996). The annual-averaged total solar irradiance data (https://www.ncdc.noaa.gov/cdr/atmospheric/total-solar-irradiance) shows a significant decrease ($p < 0.001$) during the period 2000–2010 (Figure S4), which demonstrates that the change in the decline rate of coral $\delta^{13}C$ since around 2000 cannot be attributed to solar irradiance.

Ocean circulation also can affect coral skeletal $\delta^{13}C$ by changing seawater DIC (Druffel, 1997). For the SCS corals, the changes in their skeletal $\delta^{13}C$ cannot be accounted for by those in ocean circulation (Deng et al., 2017; Han et al., 2019). The long-term variation trends in skeletal $\delta^{13}C$ of the corals from Western Australia, Brazil and Belize were suggested to be subjected to Suess effect (Fowell et al., 2018; Pereira et al., 2018; Chen et al., 2020), and that of the coral from American Samoa was associated with growth rate (Linsley et al., 2019). However, the change in skeletal $\delta^{13}C$ of this American Samoa coral since 2000 is not consistent to the kinetic effect as mentioned above. Osborne et al. (2013) didn't discuss the $\delta^{13}C$ values in the two corals form Palau, so it is difficult to assess the effect of ocean circulation; but their long-term declining trend should also reflect the effect of Suess effect (Figure S1).

There are also other factors that can contribute to the change in coral skeletal $\delta^{13}C$, such as autotrophic photosynthesis (McConnaughey, 1989; Swart et al., 1996; McConnaughey et al., 1997), heterotrophic predation (Swart et al., 1996; Grottoli, 2002; Reynaud et al., 2002), and water turbidity (Yentsch et al., 2002; Shimamura et al., 2008). These factors were not regarded as the main contributions to the long-term variations of coral skeletal $\delta^{13}C$ values in all cited references (Osborne et al., 2013; Fowell et al., 2018; Pereira et al., 2018; Han et al., 2019; Linsley et al., 2019; Chen et al., 2020), therefore they were not discussed here. The sea surface primary productivity also can affect coral skeletal $\delta^{13}C$ in that enhanced primary productivity will increase the $\delta^{13}C$ of seawater DIC by preferential consumption of $^{12}CO_2$ in seawater during phytoplankton photosynthesis and vice versa (Park and Epstein, 1960). The skeletal $\delta^{13}C$ (with the ^{13}C Suess effect removed) of the coral Yongxing from the SCS has been suggested to link with the surface primary productivity on the decadal time scale (Han et al., 2019), and the primary productivity in the SCS increased during the period from 1998 to 2010 (Palacz et al., 2011). Thus, for the corals form the SCS, it is possible that the change in the decline rates of their $\delta^{13}C$ since around 2000 was associated with the increase of the surface primary productivity during the period from 1998 to 2010, which is exactly a manifestation of change in DIC source. The observation data are not available and were not discussed in cited references, and we therefore refrain from further interpretation of the effect of primary productivity on coral $\delta^{13}C$ from other sites.

3.3. The change of coral $\delta^{13}C$ response to anthropogenic Suess effect

As mentioned above, the long-term declining trends in the coral $\delta^{13}C$ values during the last 200 years reflect the increase in the transfer of ^{12}C-enriched anthropogenic CO_2 from the atmosphere to the surface oceans (Dassié et al., 2013; Deng et al., 2017; Swart et al., 2010). In addition, the increasing rate of atmospheric CO_2 concentration has increased from 1.357 ppm yr^{-1} during 1960–1999 to 2.031 ppm yr^{-1} from 2000 onward, and the decreasing rate of $\delta^{13}C$ value in global atmospheric CO_2 has remained almost constant at approximately $-0.025‰{\cdot}yr^{-1}$ during 1960–1999 and from 2000 onward (Figure S5). Theoretically, more ^{12}C-enriched anthropogenic CO_2 entering into the surface oceans will lead to a more rapid decline in DIC-$\delta^{13}C$ and coral $\delta^{13}C$ values and vice versa. Therefore, the change in the decline rate in coral $\delta^{13}C$ values since 2000 may mean that the decline rate of DIC-$\delta^{13}C$ values in the surface water in which corals grow has slowed down since around 2000. However, this inference cannot be supported by the observation data of the DIC-$\delta^{13}C$ in surface seawater from the HOT and BATS stations, which indicates a stable rate of declining DIC-$\delta^{13}C$ in surface seawater since 2000 compared with that during 1960–1999 ($-0.023‰{\cdot}yr^{-1}$ vs $-0.028‰{\cdot}yr^{-1}$ for HOT, and $-0.029‰{\cdot}yr^{-1}$ vs $-0.025‰{\cdot}yr^{-1}$ for BATS) (Figure S6). Therefore, the change of the decline rate in coral $\delta^{13}C$ values since 2000 should not be attributed to that of the DIC-$\delta^{13}C$ of surface seawater. Considering the large oceanographic differences between the western Pacific and the eastern-central Pacific (Deser et al., 2010; Chavez et al., 2011; McKinley et al., 2017), it should be noted that the DIC-$\delta^{13}C$ record at the HOT station in the central Pacific may not be appropriate for explaining the coral $\delta^{13}C$ records in the western Pacific from which most samples were used. However, the similar changes between the HOT and BATS stations seem to imply that the declining trends in DIC-$\delta^{13}C$ of surface seawater are consistent across global oceans. Therefore, the observation data at the HOT station was still used since the long-term observation on DIC-$\delta^{13}C$ of surface seawater in the western Pacific is not available.

The decrease of decline rate in coral $\delta^{13}C$ values may mean that the response in those corals with changes in the declining rate of skeletal $\delta^{13}C$ to the secular Suess effect has changed since around 2000s. The worldwide ocean CO_2-pH time series covering the past 40 years suggests that the anthropogenic Suess effect has led to the secular decreasing pH trend in surface oceans (Bates et al., 2014; Yao et al., 2016), and ocean acidification has shown an unambiguous impact on coral calcification (Guo et al., 2020). However, corals have the biological ability of resilience and acclimatization to climate change stressors (Coles et al., 2018; DeCarlo et al., 2018; Grottoli et al., 2018; McCulloch et al., 2012), and they can up-regulate the DIC and pH in the ECF to maintain higher aragonite saturation state and stable calcification rates (D'Olivo and McCulloch, 2017; McCulloch et al., 2017). Therefore, the change of coral $\delta^{13}C$ response to anthropogenic Suess effect is possible a manifestation of coral acclimatization to external environmental stressors. During the process of coral acclimatization and adaptation to acidifying

oceans, the change in source of DIC in the ECF and/or the DIC up-regulation may lead to the change of coral $\delta^{13}C$ response to anthropogenic Suess effect. As mentioned above, inorganic carbon from coral metabolic respiration and external seawater may both contribute to the carbon in the ECF used for calcification, and their contribution ratio varies from 1:1 to 9:1 (Goreau, 1977; Erez, 1978; Furla et al., 2000b). A suggested possible mechanism is that more seawater DIC is used to up-regulate the DIC in the ECF. Although the $\delta^{13}C$ of global surface seawater DIC has been declining, its value (~1.51‰, Quay et al., 2003) is still more positive than that of the inorganic carbon from coral metabolic respiration (~-17‰ to −9‰, Swart et al., 2005). Therefore, the change of the contribution ratio of these two DIC sources may lead to a change in the declining rate of skeletal $\delta^{13}C$. It should be considered that the respiration in hermatypic corals will strengthen under the stresses of high temperature (Coles and Jokiel, 1977; Castillo and Helmuth, 2005), which will produce more metabolic carbon depleted in ^{13}C and decrease the $\delta^{13}C$ in coral skeletons. Considering that the metabolic CO_2 needs time to transform into DIC species (CO_3^{2-} and HCO_3^-) for calcification, the seawater DIC with more positive $\delta^{13}C$ may be preferentially used to up-regulate the DIC in the ECF under warming and acidification. If this speculation holds true, the decline rate of coral skeletal $\delta^{13}C$ still can slow down even coral respiration increases under high temperature. However, the contribution ratio of the two DIC sources is difficult to constrain due to the complicated biological and environmental processes governing the DIC in the ECF, further study especially coral cultivation experiment is needed to explain the finding and clarify the underlying mechanisms.

It should be noted that there are interannual to interdecadal variabilities in almost all the coral $\delta^{13}C$ time series (supplementary Fig. S7). These variabilities in the coral $\delta^{13}C$ time series may inherit from those in the global ocean carbon sink (Landschützer et al., 2014, 2016), which may be associated with interannual to interdecadal climate phenomena such as the El Niño–Southern Oscillation and Pacific Decadal Oscillation. Therefore, it is also possible that the slowing declines from ~2000 in these coral $\delta^{13}C$ time series are only a temporary variation, and the final overall trend will be declining if the amount of anthropogenic CO_2 continues to increase. In this regard, continuous observations of $\delta^{13}C$ in coral skeletons and the seawater DIC time series are needed in the future.

4. Conclusions

The annual-resolution time series of coral $\delta^{13}C$ from the different oceans were used to study the response of coral carbon isotope to anthropogenic Suess effect. The results indicate that coral $\delta^{13}C$ time series showed a secular declining trend in recent decades, but the declining trend slowed or reversed after 2000. This change in coral $\delta^{13}C$ time series does not agree with those of DIC-$\delta^{13}C$ in surface seawater from the HOT and BATS stations, indicating that the response of coral $\delta^{13}C$ to the anthropogenic Suess effect has changed since around 2000s. The changing response is possible a manifestation of coral acclimatization to acidifying oceans, and suggests that using $\delta^{13}C$ in marine biogenic carbonates as a tracing tool for long-term evolution of $\delta^{13}C$ in seawater DIC may be subject to climate change and biological response.

CRediT authorship contribution statement

Xi Liu: Methodology, Formal analysis, Data Curation, Writing - Original draft preparation. **Wenfeng Deng:** Writing - Review & Editing, Supervision, Project administration, Funding acquisition. **Hao Cui:** Methodology, Investigation. **Xuefei Chen:** Data Curation, Writing-Original Draft. **Guanqiang Cai:** Investigation. **Ti Zeng:** Investigation, Resources. **Gangjian Wei:** Writing-Review & Editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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