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1 231Pa and 230Th in the ocean model of the Community Earth System Model

2 **(CESM1.3)**

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28 29 Abstract

Sediment ²³¹Pa/²³⁰Th activity ratio is emerging as an important proxy for deep ocean circulation in the past. In order to allow for a direct model-data comparison and to improve our understanding of sediment ²³¹Pa/²³⁰Th activity ratio, we implement ²³¹Pa and ²³⁰Th in the ocean component of the Community Earth System Model (CESM). In addition to the biotic ²³¹Pa and ²³⁰Th that is fully coupled with the active marine ecosystem module, another form of abiotic ²³¹Pa and ²³⁰Th have also been implemented with prescribed particle flux fields of the present climate. The comparison of the two forms of ²³¹Pa and ²³⁰Th helps to isolate the influence of the particle fluxes from that of circulation. Under present day climate forcing, our model is able to simulate water column ²³¹Pa and ²³⁰Th activity and sediment ²³¹Pa/²³⁰Th activity ratio in good agreement with available observations. For past climate, our model is able to simulate a comparable magnitude of the change of sediment ²³¹Pa/²³⁰Th activity ratio between the state with and without active AMOC in reconstruction. In addition, in hosing experiments, the biotic and abiotic sediment ²³¹Pa/²³⁰Th activity ratios behave similarly over large areas of low productivity, but can differ substantially in some regions of high productivity, indicating the importance of biological productivity in addition to physical circulation. Therefore, our model provides a potentially powerful tool to help our interpretation of sediment ²³¹Pa/²³⁰Th reconstructions and to improve our understanding of past ocean circulation and climate changes.

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1. Introduction

Sediment 231 Pa/ 230 Th activity ratio has been used as a proxy to reconstruct ocean circulation in the past (e.g. Yu et al. 1996; McManus et al. 2004; Gherardi et al. 2009). 231 Pa (32.5 ka half-life) and 230 Th (75.2 ka half-life) are produced at a constant rate approximately uniformly in the ocean by the α decay of 235 U and 234 U, respectively, with a production activity ratio of 0.093 (Henderson and Anderson, 2003). Water column 231 Pa and 230 Th are subject to particle scavenging and transport to sediments (Bacon and Anderson, 1982; Nozaki et al., 1987). Differential scavenging efficiency results in different ocean residence time: 231 Pa has a residence time of approximately 111 years and 230 Th has a residence time of approximately 26 years (Yu et al., 1996). Longer residence time of 231 Pa than 230 Th makes 231 Pa more subject to ocean transport and therefore in modern ocean about 45% of 231 Pa produced in the Atlantic is transported to the Southern Ocean (Yu et al., 1996), resulting a lower than 0.093 sediment 231 Pa/ 230 Th activity ratio in the North Atlantic and higher than 0.093 sediment 231 Pa/ 230 Th activity ratio in the Southern Ocean.

The application of the principle above to interpret sediment ²³¹Pa/²³⁰Th as the strength of overturning circulation, however, can be complicated by other factors, leading to uncertainties in using ²³¹Pa/²³⁰Th as a tracer for paleocirculation (Keigwin and Boyle, 2008; Lippold et al., 2009; Scholten et al., 2008). In addition to ocean transport, sediment ²³¹Pa/²³⁰Th is also influenced by particle flux and composition (Chase et al., 2002; Geibert and Usbeck, 2004; Scholten et al., 2008; Siddall et al., 2007; Walter et al., 1997). The region of a higher particle flux tends to have a higher ²³¹Pa/²³⁰Th (Kumar et al., 1993; Yong Lao et al., 1992), which is referred to as the "particle flux effect" (Siddall et al., 2005). High particle flux in the water column in a region will favor the removal of isotopes into the sediment, which leads to more isotopes transported into this region due to the down-gradient diffusive flux into this region and subsequently more removal of isotopes into the sediment. Since ²³¹Pa has a longer residence time, this effect is more prominent on ²³¹Pa than on ²³⁰Th and therefore sediment ²³¹Pa/²³⁰Th will be higher in high productivity regions. Also, opal is able to scavenge ²³¹Pa much more effectively than

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²³⁰Th, leading to higher ²³¹Pa/²³⁰Th in high opal flux regions such as the Southern Ocean (Chase et al., 2002). Moreover, sediment ²³¹Pa/²³⁰Th is suggested to record circulation change only within 1000 m above the sediment, instead of the whole water column, complicating the interpretation of sediment ²³¹Pa/²³⁰Th reconstructions (Thomas et al., 2006). For example, sediment ²³¹Pa/²³⁰Th approaching 0.093 during Heinrich Stadial event 1(HS1) from the subtropical North Atlantic is interpreted as the collapse of the Atlantic Meridional Overturning Circulation (AMOC) (McManus et al., 2004). If sediment ²³¹Pa/²³⁰Th only records deepest water mass, it is possible that during HS1, AMOC shoals, as opposed to fully collapse, yet an increase of deep water imported from the Southern Ocean featuring high ²³¹Pa/²³⁰Th can increase the sediment ²³¹Pa/²³⁰Th approaching the production ratio (0.093) (Thomas et al., 2006). All these suggest the importance of incorporating 231Pa and 230Th into climate models for a direct model-data comparison for a thorough understanding of sediment ²³¹Pa/²³⁰Th as well as past ocean circulation.

²³¹Pa and ²³⁰Th have been simulated in previous modeling studies (Dutay et al., 2009; Henderson et al., 1999; Marchal et al., 2000; Siddall et al., 2005). Here we follow the scheme of Siddall et al., (2005) to implement ²³¹Pa and ²³⁰Th into the Community Earth System Model (CESM). Siddall et al., (2005) uses prescribed particle fluxes. Our model ²³¹Pa and ²³⁰Th are coupled to a marine ecosystem model (biotic) and therefore can be used to study the impact of ecosystem change on ²³¹Pa and ²³⁰Th directly. To help to understand the influence of the particle flux, we have also implemented an abiotic version of ²³¹Pa and ²³⁰Th, for which the particle fluxes are prescribed. By comparing the abiotic ²³¹Pa and ²³⁰Th with the biotic ²³¹Pa and ²³⁰Th, we will be able to separate the effect of circulation change from particle field change. In addition, the abiotic ²³¹Pa and ²³⁰Th can be run without the marine ecosystem module, reducing computational cost by a factor of 3 in the ocean-alone model simulation and therefore making it a computationally efficient tracer for sensitivity studies.

This paper describes the details of ²³¹Pa and ²³⁰Th in CESM and serves as a reference for future studies using this tracer module. In section 2, we describe the

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model and the implementation of ²³¹Pa and ²³⁰Th. In sections 3, we describe the experimental design. We will finally compare simulated ²³¹Pa and ²³⁰Th fields with observations, show model sensitivities on the parameter and also sediment ²³¹Pa/²³⁰Th ratio response to freshwater forcing in Section 4.

2. Model Description

2.1 Physical Ocean Model

We implement ²³¹Pa and ²³⁰Th in the ocean model (Parallel Ocean Program version 2, POP2) (Danabasoglu et al., 2012) of CESM (Hurrell et al., 2013). CESM is a state-of-the-art coupled climate model and studies describing model components and analyzing results can be found in a special collection in Journal of Climate (http://journals.ametsoc.org/topic/ccsm4-cesm1). We run the ocean-alone model, which is coupled to data atmosphere, land, ice and river runoff under the normal year forcing of CORE-II data (Large and Yeager, 2008), using the low-resolution version of POP2 with a nominal 3° horizontal resolution and 60 vertical layers.

2.2 Biogeochemical component (BGC)

POP2 has incorporated a marine ecosystem module that simulates biological variables (Moore et al., 2013). The marine ecosystem module has been validated against present day observations extensively (Doney et al., 2009; Long et al., 2013; Moore and Braucher, 2008; Moore et al., 2002, 2004). The implementation of 231 Pa and 230 Th requires four particle fields: CaCO₃, opal, particulate organic carbon (POC) and dust. These particle fields can be obtained from the ecosystem driver from the ecosystem module (Jahn et al., 2015). The ecosystem module simulates the particle fluxes in reasonable agreement with the present day observations. The pattern and magnitude of the annual mean particle fluxes (CaCO₃, opal, POC) leaving the euphotic zone at 105m are similar to the satellite observations (Fig. 7.2.5 and 9.2.2 in Sarmiento and Gruber 2006) (Fig. 1 a~c): particle fluxes are higher in the high productivity regions such as high latitudes and equatorial Pacific; opal flux is high in the Southern Ocean. For ocean-alone experiments, atmospheric dust deposition to the surface ocean is prescribed from Luo et al. (2003) (Fig. 1d).

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2.3 ²³¹Pa and ²³⁰Th implementation

Two forms of 231 Pa and 230 Th are implemented in POP2: abiotic and biotic. Abiotic 231 Pa and 230 Th use particle fluxes prescribed as annual mean particle fluxes from the CESM marine ecosystem module under present day climate forcing (Fig.1). Biotic 231 Pa and 230 Th use particle fluxes computed simultaneously from the marine ecosystem module. Abiotic and biotic 231 Pa and 230 Th can be turned on at the case build time and the biotic 231 Pa and 230 Th requires the ecosystem module turned on at the same time.

The implementation of ²³¹Pa and ²³⁰Th is based on Siddall et al., (2005) (Eq.(3)). ²³¹Pa and ²³⁰Th are produced from the α decay of ²³⁵U and ²³⁴U uniformly everywhere at constant rate β^{i} ($\beta^{Pa} = 2.33*10^{-3}$ dpm m⁻³ yr⁻¹, $\beta^{Th} = 2.52*10^{-2}$ dpm m⁻³ yr⁻¹). ²³¹Pa and ²³⁰Th are subjective to radioactive decay with the decay constant of λ^i ($\lambda^{Pa} = 2.13*10^{-5}$ yr⁻¹, $\lambda^{Th} = 9.22*10^{-6}$ yr⁻¹). In addition to ocean transport, which includes advection, convection, and diffusion, another important process contributes to ²³¹Pa and ²³⁰Th activity is the reversible scavenging (term $w_S \frac{\partial A_p^l}{\partial a_s}$ in Eq. (3)) by sinking particles with a sinking velocity w_S , which describes the adsorption of isotopes onto sinking particles and desorption after the dissolution of particles (Detailed vertical differentiation scheme to calculate this term in the model is in the supplementary material). This process transports ²³¹Pa and ²³⁰Th downward and leads to a general increase of ²³¹Pa and ²³⁰Th activity with depth. We don't differentiate between particle sizes as in Dutay et al., (2009) and consider all particles as slowly sinking small particles with sinking velocity of $w_s = 1000 \text{ m yr}^{-1}$ (Arsouze et al., 2009; Dutay et al., 2009; Kriest, 2002). The reversible scavenging considers total isotope activity (A_t^i) as two categories: dissolved isotopes (A_d^i) and particulate isotopes (A_n^i) (superscript i=1 and 2 refers to ²³¹Pa and ^{230Th}, respectively) as in Eq. (1) and assumes these two phases are in equilibrium, which is a reasonable assumption in the open ocean (Bacon and Anderson, 1982; Henderson et al., 1999; Moore and Hunter, 1985; Roy-Barman et al., 1996). Any particulate isotopes at the ocean bottom layer are removed from the

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ocean as sediment. The ratio between the particulate isotope activity and the

dissolved isotope activity is set by a partition coefficient, K (Eq. (2)), where C_i is the

ratio of particle concentration to the density of seawater (1024.5 kg m^{-3}). Subscript j

refers to different particle types (CaCO₃, opal, POC and dust). The reversible

scavenging scheme applied here is the same as the neodymium implementation in

160 POP2 (Gu et al., 2017).

$$A_t^i = A_d^i + A_p^i (1)$$

$$K_j^i = \frac{A_{j,p}^i}{A_{j,d}^i C_j} \tag{2}$$

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Therefore, the conservation equation for ²³¹Pa and ²³⁰Th activity can be

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$$\frac{\partial A_t^i}{\partial t} = \beta^i - \lambda^i A_t^i - w_s \frac{\partial A_p^i}{\partial z} + Transport$$
(3)

where A_p^i can be calculated from Eq. (4) below by combining Eq. (1) and Eq. (2).

$$A_p^i = A_t^i \cdot (1 - \frac{1}{1 + K_{POC}^i \cdot R_{POC} + K_{CaCO_3}^i \cdot R_{CaCO_3} + K_{opal}^i \cdot R_{opal} + K_{dust}^i \cdot R_{dust}})$$
(4)

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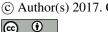
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3. Experiments

We run a control experiment (CTRL) and two experiments with different partition coefficients to show model sensitivity. We have both abiotic and biotic ²³¹Pa and ²³⁰Th in CTRL, but only show abiotic ²³¹Pa and ²³⁰Th in sensitivity experiments. Equilibrium partition coefficients for ²³¹Pa and ²³⁰Th vary among different particle types and the magnitude of the partition coefficients for different particle types remains uncertain (Chase and Robert F, 2004; Chase et al., 2002; Luo and Ku, 1999). Since the control experiment in Siddall et al., (2005) is able to simulate major features of ²³¹Pa and ²³⁰Th distributions, we use the partition coefficients from the control experiment in Siddall et al., (2005) in our CTRL (Table

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1). Two sensitivity experiments are performed with decreased (EXP_1) and increased (EXP_2) partition coefficients by a factor of 5 (Table 1).

All the experiments are ocean-alone experiments with the normal year forcing by CORE-II data (Large and Yeager, 2008). The ²³¹Pa and ²³⁰Th activities are initiated from 0 in CTRL and are integrated for 2,000 model years until equilibrium is reached. EXP_1 and EXP_2 are initiated from 1,400 model year of CTRL and are integrated for another 800 model years to reach equilibrium.

To test how sediment 231 Pa/ 230 Th ratio responds to the change of AMOC, we carried out a fresh water perturbation experiment (HOSING) with both abiotic and biotic 231 Pa and 230 Th. Starting from 2,000 model year of CTRL, a freshwater flux of 1 Sv is imposed over the North Atlantic region of 50° N \sim 70 $^{\circ}$ N and the experiment is integrated for 1400 model years until both abiotic and biotic sediment 231 Pa/ 230 Th ratio have reached quasi-equilibrium. The partition coefficients used in HOSING are the same as in CTRL.

4. Results

4.1 Control Experiment

Abiotic and biotic version of 231 Pa and 230 Th in CTRL show identical results. Abiotic and biotic dissolved and particulate 231 Pa and 230 Th in CTRL are highly correlated with each other (Fig. 2e-h) with correlations larger than 0.995 and regression coefficients are all near 1.0 (R²>0.995) (Fig. 2e-h). The correlation coefficient between abiotic and biotic sediment 231 Pa/ 230 Th activity ratios in CTRL is 0.99 (N=7879 points) and the regression coefficient between them is 0.9 (R²=0.98) (Fig. 4a). This is expected because the particle fields used in abiotic version are the climatology of the particle fields used in the biotic version for the present day. Therefore, under the same climate forcing, abiotic and biotic version of 231 Pa and 230 Th should be very similar. For the discussion of results in CTRL below, we only discuss the abiotic 231 Pa and 230 Th.

The residence time of both 231 Pa and 230 Th in CTRL are comparable with observations. Residence time in CTRL is 118 yr for 231 Pa and 33 yr for 230 Th (Table

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1), which are of the same magnitude as 111 yr for ²³¹Pa and 26 yr for ²³⁰Th in observation (Yu et al., 1996).

CTRL can simulate the general features of the water column ²³¹Pa and ²³⁰Th activities. Both dissolved and particulate activities for ²³¹Pa and ²³⁰Th increase with depth in CTRL except in the regions of deep water formation, as shown in the zonal mean Atlantic dissolved and particulate ²³¹Pa and ²³⁰Th activities (Fig. 2a-d). The dissolved and particulate ²³¹Pa and ²³⁰Th activities are also at the same order of magnitude as in observations (Colley et al., 1995; Luo et al., 2010; Mangini and Key, 1983; Moran et al., 1997, 2002; Rutgers van der Loeff and Berger, 1993; VOGLER et al., 1998; Walter et al., 1997). Our CTRL shows similar results as in Siddall et al., (2005) (their Fig. 2). The patterns of dissolved ²³¹Pa and ²³⁰Th activities are similar except in the Southern Ocean, where high opal flux effectively removes ²³¹Pa to sediment.

A more quantitative model-data comparison is shown in Fig. 3. The correlation between model and observations are significant at 0.01 confidence level for all dissolved and particulate ²³¹Pa and ²³⁰Th: [²³¹Pa]_d correlation is 0.65, [²³⁰Th]_d correlation is 0.73, [231Pa]_p correlation is 0.33 and [230Th]_p correlation is 0.62. The linear regression coefficient, an indication of model ability to simulate 231Pa and ²³⁰Th activity (Dutay et al., 2009), is near 1.0 for dissolved ²³¹Pa and ²³⁰Th (1.14 for [231Pa]_d and 1.04 for [230Th]_d), suggesting that CTRL can simulate the dissolved ²³¹Pa and ²³⁰Th in good agreement with observations. However, the simulation of the particulate activity is not as good as the dissolved activity. Particulate activity is overall somewhat smaller than observations in the surface ocean and larger than observation in the deep ocean for both dissolved ²³¹Pa and ²³⁰Th. The regression coefficient for particulate ²³¹Pa and ²³⁰Th is 0.14 for [²³¹Pa]_p and 0.42 for [²³⁰Th]_p (Fig. 3). This is also similar in previous modeling studies (Dutay et al., 2009; Siddall et al., 2005). One may think the performance of simulating [231Pa]_p and [230Th]_p can be improved by tuning model parameter: partition coefficient k. However, under current modeling scheme, changing partition coefficient k will have limited influenced on $[^{231}Pa]_p$ and $[^{230}Th]_p$, which will be discussed in section 4.2.

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The sediment ²³¹Pa/²³⁰Th activity ratios in CTRL is overall consistent with observations (Anderson et al., 1983, 1990, 1994; Bacon and Rosholt, 1982; François et al., 1993; Frank, 1996; Frank et al., 1994; Ku et al., 1972; Müller and Mangini, 1980: Schmitz et al., 1986: Scholten et al., 1995: Shimmield and Price, 1988: Shimmield et al., 1986; Walter et al., 1997; Yang et al., 1986; Yong Lao et al., 1992; Yong-Liang Yang et al., 1995; Yu et al., 1996). The North Atlantic shows low sediment ²³¹Pa/²³⁰Th activity ratio as in observations because ²³¹Pa is more subject to transport to the Southern Ocean by active ocean circulation than ²³⁰Th. The Southern Ocean maximum in the sediment ²³¹Pa/²³⁰Th activity ratio is also simulated in CTRL, which is caused by high opal fluxes in the Southern Ocean preferentially removes 231 Pa into sediment ($K_{opal}^{^{231}Pa} > K_{opal}^{^{230}Th}$), as well as upwelling in the Southern Ocean brings up deep water enriched with 231Pa, which is transported from the North Atlantic, to shallower depth and further contribute to the scavenging. CTRL can also produce higher sediment ²³¹Pa/²³⁰Th activity ratio in regions with high particle production (e.g. the Eastern equatorial Pacific, the North Pacific and the Indian Ocean) due to the "particle flux effect". However, this particle flux effect is less effective in the North Atlantic than in the North Pacific and the Indian Ocean due to active deep ocean circulation transporting ²³¹Pa southward in the Atlantic (Yu et al., 1996).

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4.2 Sensitivity on partition coefficient K

In this section, we show model sensitivity on partition coefficient by increasing and decreasing the partition coefficient, K, by a factor of 5, but keep the relative ratio for different particles the same (Table 1). Our model shows similar sensitivity to that in Siddall et al., (2005) as discussed below.

Increasing K will decrease water column dissolved 231 Pa and 230 Th activities but won't change particulate 231 Pa and 230 Th too much (Fig. 5). Larger K will lead to more 231 Pa and 230 Th attached to particles and further buried into sediment, which increases the sink for the 231 Pa and 230 Th budget. With the sources for 231 Pa and 230 Th staying the same, dissolved 231 Pa and 230 Th will be reduced. Increasing K will

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also reduce the vertical gradient of dissolved ²³¹Pa and ²³⁰Th as reversible scavenging act as the vertical transport and increase this vertical transport can decrease the vertical gradient. However, change in the particulate ²³¹Pa and ²³⁰Th is small. As stated in Siddall et al., (2005), if we neglect the transport term and the decay term in Eq. (3) and assume particulate phase activity at the surface as 0, when reach equilibrium, the activity of particulate phase will be as in Eq. (5). The particulate phase activity only depends on the production rate, the particle settling velocity and depth. The particulate phase activity will increase linearly with depth and any departure from this linear relationship with depth is due to ocean transport, which is suggested by observations (Bacon and Anderson, 1982; Roy-Barman et al., 1996). Therefore, changing K will have limited influence on particulate phase activity.

$$A_p^i(z) = \frac{\beta^i}{w_s} \cdot z \tag{5}$$

Increasing K will also reduce the spatial gradient in sediment 231 Pa/ 230 Th activity ratio and vice versa (Fig. 6). Larger K will decrease the 231 Pa and 230 Th residence time and most isotopes produced in the water column are removed into sediment locally (Table 1). Therefore, sediment 231 Pa/ 230 Th ratio becomes more homogeneous and approaching the production ration of 0.093 (Fig. 6b). The sediment 231 Pa/ 230 Th activity ratio in EXP_1 and EXP_2 departures from observations significantly, suggesting the partition coefficient in CTRL is of the right magnitude.

4.3. Sediment ²³¹Pa/²³⁰Th ratio in HOSING

With the AMOC collapsing, the ²³¹Pa/²³⁰Th ratio tends to increase over most of the North Atlantic, consistent with paleo proxy evidence there. In HOSING, after applying extra freshwater to the North Atlantic, AMOC strength quickly decreases to a minimum of 2 Sv at around year 300 (AMOC_off)(Fig. 7a). During the AMOC_off state, compared with CTRL of active AMOC (AMOC_on), both abiotic and biotic

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sediment ²³¹Pa/²³⁰Th ratio shows an overall increase in the North Atlantic and a decrease in the South Atlantic (Fig. 8b and d) because of the reduced southward transport of ²³¹Pa from the North Atlantic by AMOC. In most area of the Atlantic, the evolution of abiotic and biotic sediment ²³¹Pa/²³⁰Th activity ratio in HOSING are highly correlated (Fig. 9a). The change of sediment ²³¹Pa/²³⁰Th ratio from AMOC_on to AMOC_off are similar in abotic and biotic version (Fig.9b). The correlation between abiotic and biotic sediment ²³¹Pa/²³⁰Th ratio change is 0.72 (1455points) and the linear regression coefficient is $0.71 (R^2 = 0.52)$. This suggests that abiotic sediment ²³¹Pa/²³⁰Th activity ratio can capture the major feature of biotic ²³¹Pa/²³⁰Th activity ratio in our model and also circulation effect on sediment ²³¹Pa/²³⁰Th activity ratio is more dominant than the biological effect in HOSING. The pattern of abiotic (Fig.8a) sediment ²³¹Pa/²³⁰Th ratio in the Atlantic in AMOC_off state is similar to the opal distribution (Fig.1b) because, without active circulation, sediment ²³¹Pa/²³⁰Th ratio is more controlled by particle flux effect, which is similar to the case in the Pacific in CTRL. The overall increase of sediment ²³¹Pa/²³⁰Th ratio in the North Atlantic in response to AMOC collapse can be seen more clearly in the time evolution of the sediment ²³¹Pa/²³⁰Th ratio averaged from 20°N to 60°N in the North Atlantic in both the abiotic and biotic ²³¹Pa/²³⁰Th (Fig.7b). Quantitatively, the ²³¹Pa/²³⁰Th increases from 0.074 (0.074) in AMOC on to 0.098 (0.095) in AMOC off in the abiotic (biotic) version (Fig. 7b). Both abiotic and biotic version show average sediment ²³¹Pa/²³⁰Th ratio in the North Atlantic near the production ratio of 0.093. This increase of ²³¹Pa/²³⁰Th in both abiotic and biotic versions is also seen in the subtropical North Atlantic from the two sites near Bermuda Rise (Fig. 7e and f), which is, of comparable magnitude with the change from LGM to HS1 in reconstructions there (McManus et al., 2004). It is further noted that our abiotic sediment ²³¹Pa/²³⁰Th ratio in HOSING behaves similarly to that in Siddall et al., (2007).In spite of large scale patterns of sediment ²³¹Pa/²³⁰Th ratio response, the magnitude of the change between AMOC_on and AMOC_off varies with location in both abiotic and biotic version because of the distribution of particle flux (Fig.7 and 8). Take the abiotic version as an example, the maximum increase in sediment

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²³¹Pa/²³⁰Th ratio occurs near 40°N western Atlantic, where the opal production in our model is maximum (Fig. 1b). The sediment ²³¹Pa/²³⁰Th ratio in this region in AMOC_on is larger than production ratio of 0.093 because particle flux effect due to the opal maximum provides extra ²³¹Pa to this region, which overwhelms the active ocean circulation transporting ²³¹Pa southward outside this region. Therefore, sediment ²³¹Pa/²³⁰Th ratio in this region gets even larger (e.g. Fig. 7d). In AMOC_off, without active ocean circulation, the particle flux effect becomes more prominent because less ²³¹Pa is transported out of the North Atlantic.

The responses of abiotic and biotic sediment ²³¹Pa/²³⁰Th ratio to the collapse of AMOC show similar behavior over most ocean region of low productivity but can differ significantly in high productivity region because of the change of productivity. Productivity in North Atlantic is suggested to be halved during AMOC collapse because of increased stratification, which reduces nutrient supply from deep ocean (Schmittner, 2005). In the CESM, the productivity in mid-latitude North Atlantic is indeed greatly reduced after the freshwater forcing. For example, at year 100 in HOSING, opal production from 30°N-50°N in the Atlantic is reduced by 50%~90% of its original value in CTRL (not shown). Therefore, in the first 100 years in HOSING, most biotic sediment ²³¹Pa/²³⁰Th ratio show an initial decrease in the North Atlantic from the subtropics to the mid-latitude (Fig. 7 d, e, and f, red dash). In the subpolar region, the productivity is increased in the model, leading to an initial increase of biotic sediment ²³¹Pa/²³⁰Th ratio (Fig.7c). Furthermore, the detailed pattern of the difference between AMOC_off and AMOC_on in sediment 231Pa/230Th ratio is different. For example, the region (near 40°N west Atlantic), which has the maximum increase from AMOC_on to AMOC_off in abiotic sediment ²³¹Pa/²³⁰Th ratio discussed above, shows a decrease in biotic sediment ²³¹Pa/²³⁰Th ratio (Fig. 7d and Fig.8d) because there is no more opal maximum in this region in AMOC_off. A detailed discussion of the difference between abiotic and biotic sediment ²³¹Pa/²³⁰Th ratio in different regions is beyond the scope of this paper. Overall, our model is able to simulate the correct magnitude of the sediment ²³¹Pa/²³⁰Th ratio response to the change of AMOC, although the detailed difference between abiotic

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and biotic sediment 231 Pa/ 230 Th ratio response to fresh water forcing in different locations can be complicated.

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5. Summary

²³¹Pa and ²³⁰Th have been implemented into the ocean model of the CESM in both the biotic and abiotic forms. Our control experiment under present day climate forcing is able to simulate both ²³¹Pa and ²³⁰Th water column activity and sediment ²³¹Pa/²³⁰Th activity ratio consistent with observations by using the parameters that are suggested by Chase et al., (2002) and used in Siddall et al. (2005). Our sensitivity experiments with varying parameters suggest that these parameters are of the right magnitude. Furthermore, our model is able to simulate the overall sediment ²³¹Pa/²³⁰Th ratio change in the North Atlantic with a magnitude comparable to the reconstruction in response to the collapse of AMOC, although the detailed regional response can be complicated in different regions. Finally, the abiotic form is able to capture many major features of that of the biotic form over large ocean areas, although the two forms can also differ significantly in some regions, especially the region with large productivity. Therefore, with both abiotic and biotic ²³¹Pa and ²³⁰Th, our model can serve as a useful tool to improve our understanding of the processes of ²³¹Pa and ²³⁰Th and also interpretations of sediment ²³¹Pa/²³⁰Th reconstructions for past ocean circulation and climate changes.

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Code availability:

The Nd isotope source code of both abiotic Nd and biotic Nd for CESM1.3 is included as supplementary material here.

386 387

388 **Acknowledgement:**

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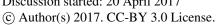
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	CTRL		EXP_1		EXP_2	
	²³¹ Pa	²³⁰ Th	²³¹ Pa	²³⁰ Th	²³¹ Pa	²³⁰ Th
K_{CaCO_3}	2.5*10 ⁵	1.0*107	5*104	2*106	1.25*106	5*10 ⁷
K_{opal}	1.67*106	5*10 ⁵	3.33*105	1*105	8.33*106	2.5*106
K_{POC}	1.0*107	1.0*107	2*10 ⁶	2*106	5*10 ⁷	5*10 ⁷
K_{dust}	0	0	0	0	0	0
T(yr)	118	33	501	143	27	9

Table 1. Partition coefficients for different particle types and residence time for 231 Pa and 230 Th in different experiments.

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681 Figures:

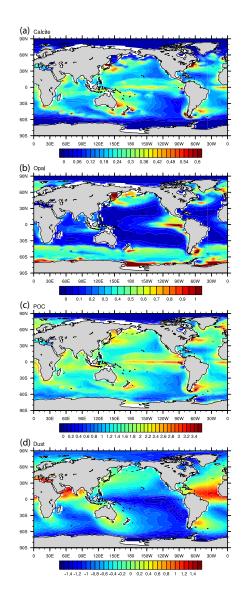


Figure 1. Annual mean particle fluxes in CESM. (a) CaCO₃ flux at 105m (mol m⁻² yr⁻¹). (b) Opal flux at 105m (mol m⁻² yr⁻¹). (c) POC flux at 105m (mol m⁻² yr⁻¹). (d) Log10 values of annual atmospheric dust deposition (g m⁻² yr⁻¹).

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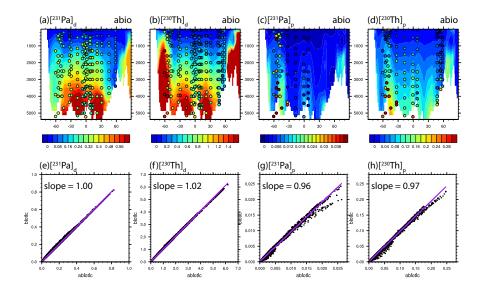
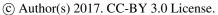


Figure 2. Atlantic zonal mean dissolved and particulate abiotic ²³¹Pa and ²³⁰Th in CTRL (unit: dpm/m³): (a) dissolved ²³¹Pa; (b) dissolved ²³⁰Th; (c) particulate ²³¹Pa; (d) particulate ²³⁰Th. Scatter plot of global dissolved and particulate ²³¹Pa and ²³⁰Th between abiotic and biotic in CTRL: (e) dissolved ²³¹Pa; (f) dissolved ²³⁰Th; (g) particulate ²³¹Pa; (h) particulate ²³⁰Th. Purple line is the least squared linear regression line and slope is the linear regression coefficient.

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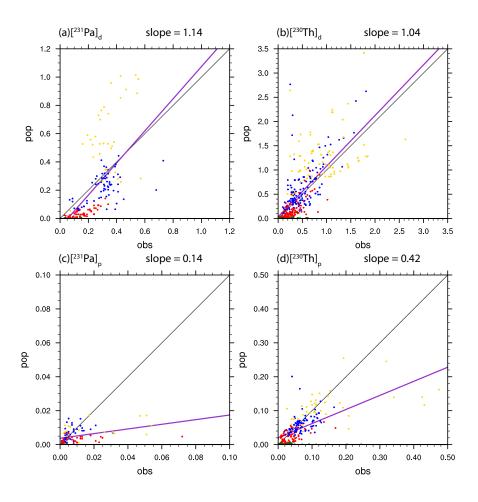


Figure 3. Scatter plot of global dissolved and particulate ²³¹Pa and ²³⁰Th between observation and CTRL (abiotic) (unit: dpm/m³). (a) dissolved ²³¹Pa; (b) particulate ²³¹Pa; (c) dissolved ²³⁰Th; (d) particulate ²³⁰Th. Observations in different depth range are indicated by different colors: green for 0-100m; red for 100m-1000m; blue for 1000m-3000m and yellow for deeper than 3000m. Purple line is the least squared linear regression line and slope is the linear regression coefficient.

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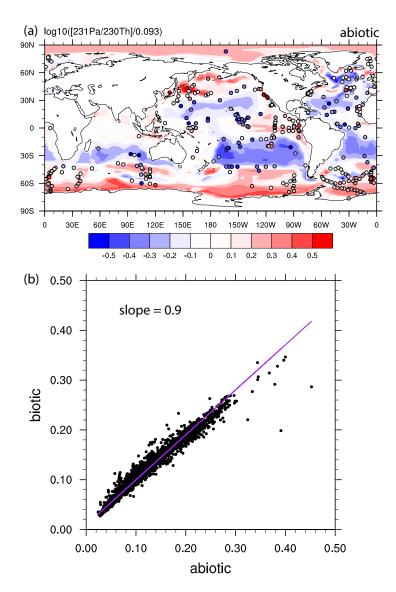


Figure 4. (a) Abiotic sediment 231 Pa/ 230 Th activity ratio in CTRL Observations are attached as filled cycles using the same color map. The 231 Pa/ 230 Th activity ratio is plotted relative to the production ratio of 0.093 on a \log_{10} scale. (b) Scatter plot of abiotic and biotic sediment 231 Pa/ 230 Th activity ratio in CTRL. Purple line is the least squared linear regression line and slope is the linear regression coefficient.

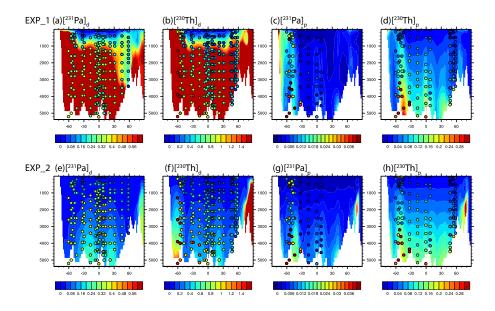
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Sediment 231 Pa/ 230 Th activity ratio is calculated using $[^{231}$ Pa]_p/ $[^{230}$ Th]_p is the bottom grid box in the model.

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Figure 5. Atlantic zonal mean dissolved and particulate ²³¹Pa and ²³⁰Th in EXP_1 and EXP_2 (unit: dpm/m³). EXP_1: (a) dissolved ²³¹Pa; (b) dissolved ²³⁰Th; (c) particulate ²³¹Pa; (d) particulate ²³⁰Th. EXP_2: (e) dissolved ²³¹Pa; (f) dissolved ²³⁰Th; (g) particulate ²³¹Pa; (h) particulate ²³⁰Th. Observations are attached as filled cycles using the same color map.

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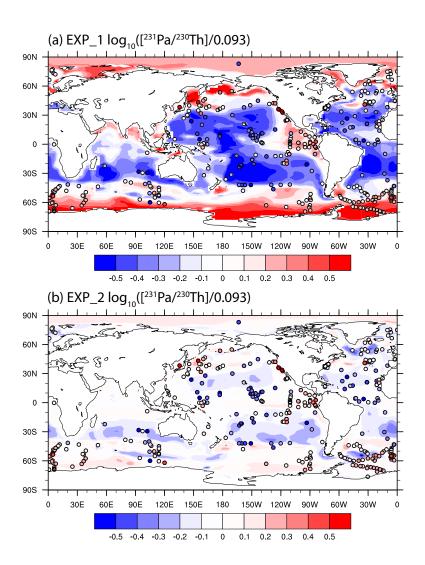


Figure 6. Sediment 231 Pa/ 230 Th activity ratio in EXP_1 (a) and EXP_2 (b). Observations are attached as filled cycles using the same color map. The 231 Pa/ 230 Th activity ratio is plotted relative to the production ratio of 0.093 on a log_{10} scale.

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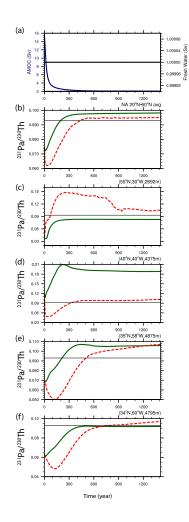


Figure 7. Time evolutions in HOSING. (a) Freshwater forcing (black) and AMOC strength (navy), which is defined as the maximum of the overturning streamfunction below 500m in the North Atlantic. (b) North Atlantic average sediment ²³¹Pa/²³⁰Th activity ratio from 20°N to 60°N: abiotic (green) and biotic (red). Production ratio of 0.093 is indicated by a solid black line (similar in c, d, e and f). (c) Sediment ²³¹Pa/²³⁰Th activity ratio at (55°N, 30°W). (d) Sediment ²³¹Pa/²³⁰Th activity ratio at (40°N, 40°W). (e) Sediment ²³¹Pa/²³⁰Th activity ratio at (34°N, 60°W). (e) and (f) are near Bermuda Rise. Locations of each site are shown as dots in Fig. 8b.

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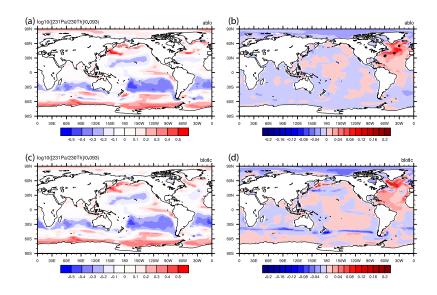
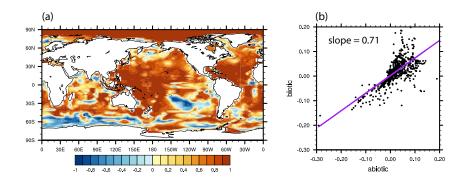


Figure 8. Sediment 231 Pa/ 230 Th activity ratio during AMOC off state and the difference between AMOC off and CTRL. (a) Abiotic $log_{10}([^{231}$ Pa/ 230 Th]/0.093) in AMOC_off. (b) Difference of abiotic sediment 231 Pa/ 230 Th activity ratio between AMOC_off and AMOC_on. (c) and (d) are similar to (a) and (b) for biotic sediment 231 Pa/ 230 Th activity ratio. Black dots in (b) shows the locations of sites in Fig. 7 from North to South.

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Figure 9. (a) Correlation of abiotic and biotic evolution of sediment 231 Pa/ 230 Th activity ratio in HOSING. (b) Scatter plot of abiotic and biotic sediment 231 Pa/ 230 Th activity ratio change from AMOC_on to AMOC_off in the Atlantic and the Southern Ocean (70°W-20°E). Purple line is the least squared linear regression line and slope is the linear regression coefficient.