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## RESEARCH LETTER

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### Key Points:

- Biogenic silica diagenesis was examined for the first time in hadal trench sediments
- Availability of detrital materials may be a limiting factor for biogenic silica (bSiO<sub>2</sub>) diagenesis in bSiO<sub>2</sub>-rich sediments of the Mariana Trench
- ~40%–80% of dissolved silicic acid generated by bSiO<sub>2</sub> dissolution is fixed by authigenic silicate formation

### Supporting Information:

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

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## Active Silica Diagenesis in the Deepest Hadal Trench Sediments

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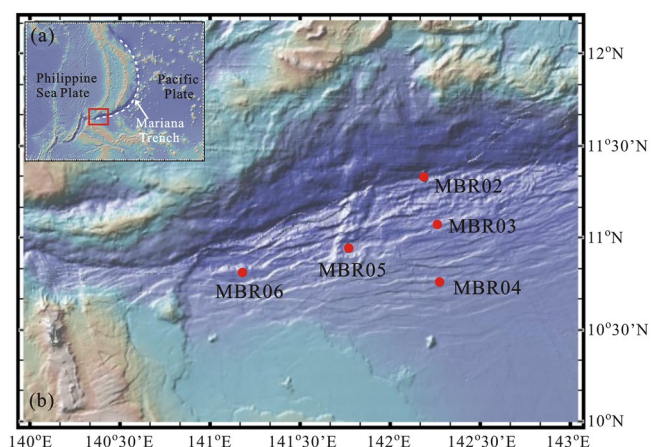
**Abstract** Porewater dissolved silicic acid (DSi) concentrations and stable Si isotope compositions ( $\delta^{30}\text{Si}$ ) together with biogenic silica (bSiO<sub>2</sub>) contents of sediments in five sediment cores collected from the southern Mariana Trench are presented. These data suggest the occurrence of bSiO<sub>2</sub> dissolution and concomitant authigenic clay formation in three bSiO<sub>2</sub>-bearing cores. A reaction-transport model constrained by the measured geochemical data was applied to quantify the rates of Si turnover. Model results predicted the greatest rates of both bSiO<sub>2</sub> dissolution and authigenic clay formation at the trench axis core that displayed low bSiO<sub>2</sub> contents and abundant detrital materials, suggesting that detrital materials may be a limiting factor for bSiO<sub>2</sub> diagenesis. Model results further predicted that ~40%–70% of DSi generated by bSiO<sub>2</sub> dissolution is consumed by authigenic clay formation. This is the first study that demonstrates active silica diagenesis in the hadal realm and has implications for understanding benthic Si cycling in deep-sea settings.

**Plain Language Summary** The marine Si cycle has received extensive attention because it is coupled with the marine carbon cycle, thereby playing a critical role in regulating global climate. While the importance of silica diagenesis in balancing the marine Si cycle has been recognized and studied mostly in estuarine and continental margin environments, very few data exist in deep-sea settings which comprise the majority of the seafloor area. In this study, we aim at understanding the Si cycling in the hadal trench sediments (>6,000 m in water depth) which represent the deepest and least explored sectors of Earth's surface. A reaction-transport model constrained by porewater dissolved silicic acid and stable Si isotope compositions as well as biogenic silica contents from the southern Mariana Trench predicted efficient conversion of biogenic silica to authigenic silicates, which may be controlled by the supply of detrital materials. Our findings provide significant insights into benthic Si and associated elemental cycles in extreme water depths.

## 1. Introduction

The global silicon (Si) cycle is of great interest as it is closely linked to the global carbon cycle and resulting climatic stability through the coupling of silicate weathering, reverse weathering, and the biological pump (Frings et al., 2016; Sutton et al., 2018; P. J. Tréguer et al., 2021; P. Tréguer & Pondaven, 2000). Silicifying organisms utilize dissolved silicic acid (DSi) in the photic zone of the ocean to build amorphous biogenic silica (bSiO<sub>2</sub>) frustules. After the death of the organisms, the frustules experience partial dissolution during sinking through the water column because seawater is highly undersaturated with respect to bSiO<sub>2</sub>. The rapid recycling of bSiO<sub>2</sub> to DSi in the water column results in approximately one third of the bSiO<sub>2</sub> produced in the photic zone reaching the seafloor (Ragueneau et al., 2000; P. J. Tréguer & Rocha, 2013). Further bSiO<sub>2</sub> dissolution can take place in the sediments, leading to <5% of newly synthesized bSiO<sub>2</sub> being permanently buried (P. J. Tréguer et al., 2021). Yet, bSiO<sub>2</sub> dissolution is often accompanied by the formation of authigenic aluminosilicates (i.e., reverse weathering), thereby potentially involving in atmospheric CO<sub>2</sub> levels over geological timescales (Dunlea et al., 2017; Michalopoulos & Aller, 1995; Rahman et al., 2016). Unraveling the fate of bSiO<sub>2</sub> (recycling vs. preservation) in the sediments represents an important component of the marine Si budget, thereby contributing to understanding the cycles of other pertinent elements, such as carbon (Dale et al., 2021). While the importance of silica diagenesis in balancing the marine Si cycle has been recognized and studied mostly in estuarine and continental margin environments, very few data exist in deep-sea settings which comprise the majority of the seafloor area.

Hadal trenches represent the deepest reaches of Earth's surface with water depth over 6,000 m. Growing numbers of in-situ O<sub>2</sub> uptake measurements have shown them to be dynamic hotspots of microbial activity, with generally



**Figure 1.** (a) Overview of the study area. (b) Enlargement of the red rectangular in (a) showing the location of the sampling sites.

enhanced but heterogeneous benthic carbon mineralization along the trench axes compared to the abyssal plain (Glud et al., 2013, 2021; Luo, Glud, et al., 2018; Wenzhöfer et al., 2016). These dynamic hotspots are thought to be formed as a result of downslope focusing of relatively labile organic materials triggered by unstable and complex depositional processes within hadal settings (Itou et al., 2000; Oguri et al., 2013; Turnewitsch et al., 2014). The recognition of these unique depositional environments could thus potentially lead to different perceptions on early diagenesis in hadal sediments from the general paradigm based on data in shallower water depths (Jørgensen et al., 2022).

In this study, our objective is to quantitatively examine Si cycling in the southern Mariana Trench sediments. To achieve this, porewater DSi concentrations and stable Si isotope compositions ( $\delta^{30}\text{Si}$ ) as well as bSiO<sub>2</sub> contents were measured in sediments at five sites with water depths ranging between ~6,000 m and ~10,900 m. Subsequently, the data were simulated using a reaction-transport model to quantify the rates of bSiO<sub>2</sub> dissolution and authigenic mineral precipitation. To our knowledge, this study is the first to explore silica diagenesis within hadal depths, and advances our understanding of benthic Si cycling in extreme deep-sea settings.

## 2. Materials and Methods

### 2.1. Sampling

The Mariana Trench was formed by the subduction of the Pacific Plate beneath the Philippine Sea Plate, which is famed for hosting the deepest point on the Earth's surface (Challenger Deep) in its southern sector. Sediments and porewater were collected during RV *Tansuoyihao TS15* cruise in 2019. The deepest short core (MBR02) was obtained by pushing a polycarbonate tube into the sediment block retrieved using benthic chambers mounted on an autonomous lander. The other two short cores (MBR05 and MBR06) were retrieved by a box corer. Two longer cores (MBR03 and MBR04) were collected by a gravity corer. Core locations are shown in Figure 1 and Table S1 in Supporting Information S1. Upon recovery, the cores were extruded and sliced into 1–2 cm intervals for porewater collection by inserting Rhizon samplers into the sediment cakes. Aliquots for shore-based DSi and  $\delta^{30}\text{Si}$  analyses were acidified with HNO<sub>3</sub> (suprapure) and stored in 8 ml Nalgene vials at 4°C.

### 2.2. Geochemical Analyses of Porewater and Solid Phases

Porewater DSi concentrations were determined by inductively coupled plasma optical emission spectrometry (ICP-OES, ThermoFisher Scientific iCAP 7000). Analytical precision constrained by repeat analysis of spiked IAPSO seawater standard was <3%. Porewater samples for  $\delta^{30}\text{Si}$  analysis were purified following the method of Georg et al. (2006). The pH of the samples was adjusted to about 2 with concentrated HNO<sub>3</sub>. One ml of the diluted samples with a concentration of ~140  $\mu\text{M}$  Si were loaded onto pre-cleaned cation-exchange resins (Biorad AG50 W-X8) and subsequently eluted with 2 ml MQ water.  $\delta^{30}\text{Si}$  measurements were done on the NeptunePlus HR MC-ICPMS at GEOMAR in medium-resolution mode in 'dry' plasma conditions, using an ESI Apex-HF sample introduction system. The instrumental mass bias was controlled by Mg doping. The blank to signal ratio was  $\leq 0.1\%$ . All samples were measured using the standard-sample bracketing method with the international Si standard NBS28 as the bracketing standard. The reference materials yielded  $\delta^{30}\text{Si}$  of  $-1.42 \pm 0.11\text{‰}$  (2SD;  $n = 24$ ) for IRMM018,  $+1.27 \pm 0.12\text{‰}$  (2SD;  $n = 68$ ) for Diatomite, and  $-10.63 \pm 0.09\text{‰}$  (2SD;  $n = 14$ ) for Big Batch, agreeing well with literature data (e.g., Savage et al., 2010). Additionally, an in-house porewater matrix standard was measured, yielding  $\delta^{30}\text{Si}$  of  $0.93 \pm 0.14\text{‰}$  (2SD;  $n = 13$ ), comparable within error to the  $\delta^{30}\text{Si}$  value measured at the NuPlasma MC-ICPMS at GEOMAR ( $+1.3 \pm 0.2\text{‰}$ , 2SD;  $n = 17$ ; Geilert, Grasse, Doering, et al., 2020; Geilert, Grasse, Wallmann, et al., 2020). The observed offset additionally agreed with the general observation that Si isotopes measured on the Neptune are lower compared to NuPlasma values, though it has not been confirmed yet, if this shift is due to potential instrument biases or the usage of Mg doping regarding isotope

measurements on the Neptune MC-ICPMS (Grasse et al., 2017). Fluid samples were measured at least three times and their  $\delta^{30}\text{Si}$  uncertainties ranged between 0.02 and 0.20‰ (2SD).

Porosity was determined from the weight loss before and after freeze-drying of the wet sediments. The volume fraction of porewater was calculated assuming a dry sediment density of  $2.5 \text{ g cm}^{-3}$  and a density of the porewater of  $1.023 \text{ g cm}^{-3}$  ( $P = 1 \text{ bar}$ ,  $T = 25^\circ\text{C}$ ,  $S = 35$ ).  $\text{bSiO}_2$  was extracted using the method described by Mortlock and Froelich (1989). The contents of  $\text{bSiO}_2$  were determined following the automatic NaOH leaching method of Müller and Schneider (1993) with a precision of 5%–10%. This classical operationally defined fraction has been shown to underestimate  $\text{bSiO}_2$  content as diatom frustules coated with authigenic metal oxides and authigenic aluminosilicate converted from  $\text{bSiO}_2$  dissolution are not extracted (Michalopoulos & Aller, 2004; Rahman et al., 2017). TOC contents were determined by high temperature combustion on a Vario Pyro Cube elemental analyzer after 10% HCl treatment. The analytical precision determined by duplicate measurements was better than 0.03%. The average sedimentation rate for each modeled core can only be estimated from  $^{14}\text{C}$  ages of bulk organic carbon as the cores were collected well below the carbonate compensation depth. The radiocarbon contents of sedimentary TOC ( $\Delta^{14}\text{C}_{\text{TOC}}$ ) were determined using AMS at Guangzhou Institute of Geochemistry, Chinese Academy of Sciences. AMS- $^{14}\text{C}$  ages were converted to calendar years before present (years B.P. relative to CE 1950) using the Calib.7.1 Program with the IntCal13 calibration curve and the global average reservoir age ( $\sim 400$  years; Reimer et al., 2013).

### 2.3. Reaction-Transport Modeling

A one-dimensional reaction-transport model modified from previous studies was applied to simulate Si turnover in the three  $\text{bSiO}_2$ -bearing cores (MBR02, MBR05, and MBR06) (Ehler et al., 2016; Geilert, Grasse, Doering, et al., 2020; Geilert, Grasse, Wallmann, et al., 2020). The diagenetic reactions considered in the model include  $\text{bSiO}_2$  dissolution and secondary authigenic aluminosilicate formation, which were mainly constrained by porewater DSi and  $\delta^{30}\text{Si}$  as well as  $\text{bSiO}_2$  content. Two partial differential equations were used to resolve the depth-concentration profiles of solid ( $\text{bSiO}_2$ ) and dissolved species ( $^{30}\text{Si}$  and Si), respectively (Berner, 1980; Boudreau, 1997):

$$(1 - \Phi) \frac{\partial C_s}{\partial t} = - \frac{\partial((1 - \Phi) \cdot v_s \cdot C_s)}{\partial x} + (1 - \Phi) \cdot \Sigma R \quad (1)$$

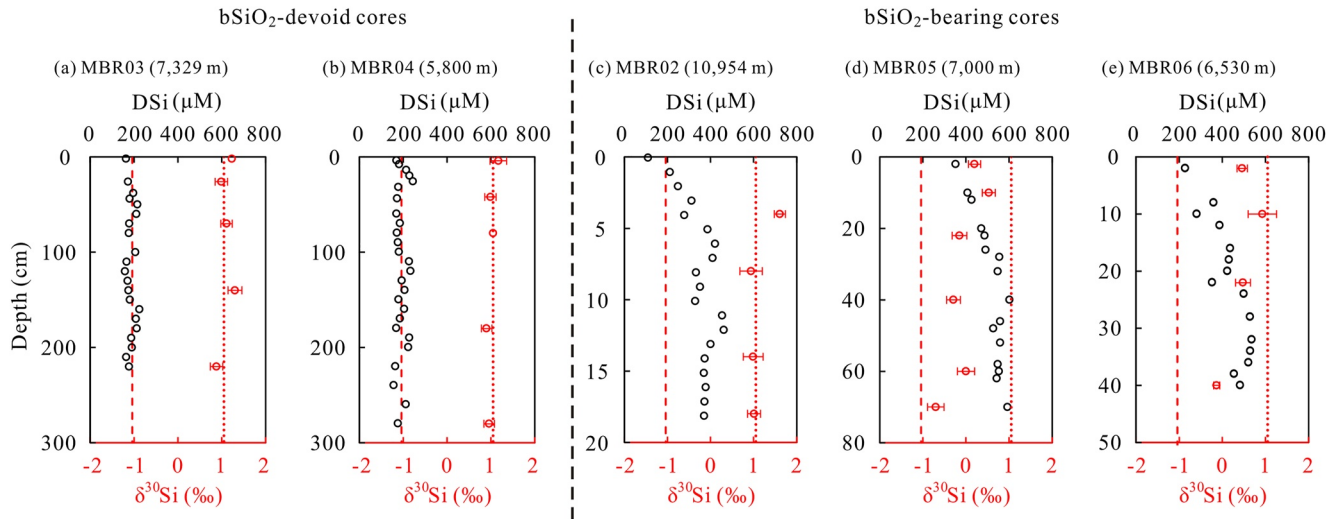
$$\Phi \frac{\partial C_a}{\partial t} = \frac{\partial(\Phi \cdot D_s \cdot \frac{\partial C_a}{\partial x})}{\partial x} - \frac{\partial(\Phi \cdot v_p \cdot C_a)}{\partial x} + \Phi \cdot \Sigma R \quad (2)$$

where  $x$  (cm) is depth in the sediments,  $t$  (yr) is time,  $\Phi$  (dimensionless) is porosity,  $C_s$  (dry wt. %) is the solid content,  $C_a$  (mmol  $\text{cm}^{-3}$ ) is the concentration of dissolved species,  $D_s$  ( $\text{cm}^2 \text{ yr}^{-1}$ ) is the molecular diffusion coefficient corrected for tortuosity,  $v_p$  ( $\text{cm yr}^{-1}$ ) is the burial velocity of porewater,  $v_s$  ( $\text{cm yr}^{-1}$ ) is the burial velocity of solids, and  $\Sigma R$  denotes the sum of the rates of Si diagenetic reactions considered in the model. A non-steady-state modeling approach was applied in order to obtain desirable simulation of observations, whereby  $\text{bSiO}_2$  rain rates were varied over time. The model parameters and kinetic rate expressions are listed in Tables S2–S5 in Supporting Information S1. A full description of the model can be found in the Supporting Information S1.

## 3. Results and Discussion

### 3.1. Evidence for $\text{bSiO}_2$ Dissolution and Authigenic Clay Formation

Two distinct types of lithology were observed from the retrieved cores. MBR03 and MBR04 were dominated by brown, homogenous, silty to clayed mud and were in absent of biogenic siliceous debris. In addition to the pelagic lithogenic sediments, light yellowish laminated diatom mats (LDMs) were found almost throughout the core MBR05 and in the interval of  $\sim 8$ – $20$  cm in core MBR06 with  $\text{bSiO}_2$  contents reaching up to  $\sim 60\%$  (Figure S1 in Supporting Information S1). The LDMs were predominantly composed of valve fragments of the giant diatom *Ethmodiscus rex* (*E. rex*) with extremely low abundance of other diatom species and radiolarians ( $< 1\%$ ). These giant diatoms, with valve length of  $> 50 \mu\text{m}$  (up to 2–5 mm), were also reported in the sediments of the Philippine Sea and the southern Mariana Trench (Figure S1 in Supporting Information S1) (Luo et al., 2017; Luo, Algeo, et al., 2018; Xiong et al., 2012). In contrast to MBR05 and MBR06, disseminated valve fragments of *E.*

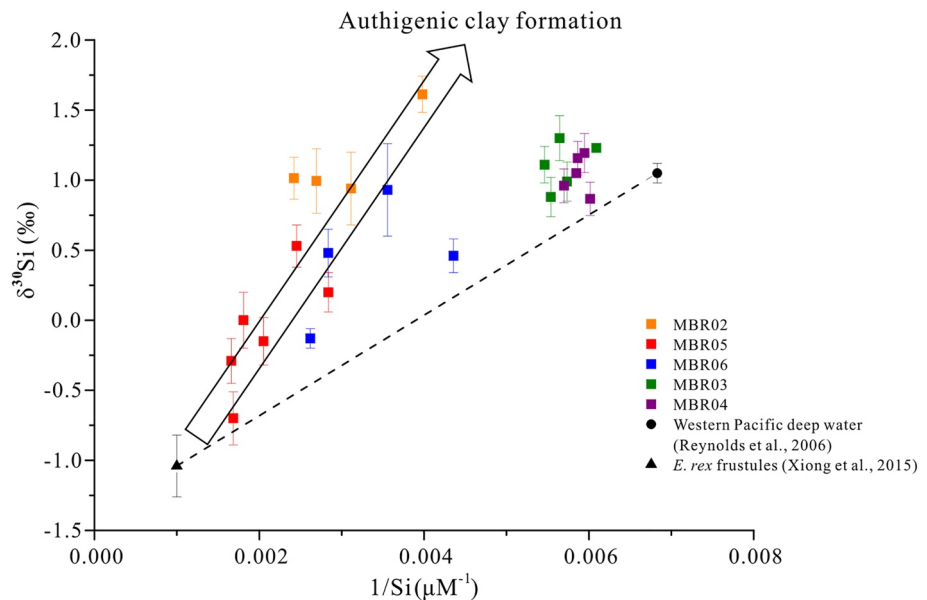


**Figure 2.** Downcore variations in porewater dissolved silicic acid (DSi) concentrations (black circles) and  $\delta^{30}\text{Si}$  values (red circles) for biogenic silica ( $\text{bSiO}_2$ )-devoid (a and b) and  $\text{bSiO}_2$ -bearing cores (c, d and e). Red dashed and dotted lines represent  $\delta^{30}\text{Si}$  values of *E. rex* frustules retrieved from the Philippine Sea basin ( $-1.04 \pm 0.22\text{‰}$ , Xiong et al., 2015) and the western Pacific deep water ( $1.05 \pm 0.07\text{‰}$ , Reynolds et al., 2006), respectively. Error bars for stable Si isotope data are shown except for those data points with error smaller than the symbol size. Note that the downcore trends of porewater DSi and  $\delta^{30}\text{Si}$  are distinct between  $\text{bSiO}_2$ -bearing (MBR02, MBR05, and MBR06) and  $\text{bSiO}_2$ -devoid (MBR03 and MBR04) cores. Also note the different depth scales.

*rex* instead of discrete LDM layers were observed in the trench axis core MBR02, with lower  $\text{bSiO}_2$  contents than in the discrete LDM layers of MBR05 and MBR06. The finding of significantly different sediment compositions in the southern Mariana Trench seems to agree with the assertion that heterogeneity of sediment textures and habitats features the hadal trench environments (Stewart & Jamieson, 2018).

In accordance with the contrast in the sediment  $\text{bSiO}_2$  contents, porewater DSi concentrations and  $\delta^{30}\text{Si}$  values showed marked differences in their downcore trends between  $\text{bSiO}_2$ -bearing (MBR02, MBR05, and MBR06) and  $\text{bSiO}_2$ -devoid (MBR03 and MBR04) cores (Figure 2). Porewater DSi concentrations in  $\text{bSiO}_2$ -bearing cores generally increased with depth, reaching quasi-asymptotic values of  $\sim 500\text{--}600\ \mu\text{M}$ . The DSi concentrations in these cores were much higher than those in  $\text{bSiO}_2$ -devoid cores where no obvious downcore trends were observed. Moreover, porewater  $\delta^{30}\text{Si}$  values in the  $\text{bSiO}_2$ -bearing cores exhibited noticeable deviation from that of the western Pacific deep water ( $1.05 \pm 0.07\text{‰}$ , Reynolds et al., 2006), whereas porewater  $\delta^{30}\text{Si}$  values in  $\text{bSiO}_2$ -devoid cores generally displayed seawater-like  $\delta^{30}\text{Si}$  value throughout the cores (Figure 2).

In  $\text{bSiO}_2$ -bearing cores MBR05 and MBR06, the general downcore increase in porewater DSi concentrations concomitant with lowered  $\delta^{30}\text{Si}$  values compared to the deep water  $\delta^{30}\text{Si}$  of western Pacific points to dissolution of *E. rex* frustules given that this dominant mat-forming giant species ( $\delta^{30}\text{Si} = -1.04 \pm 0.22\text{‰}$ ) is more depleted in  $^{30}\text{Si}$  than the small diatoms ( $\delta^{30}\text{Si} = 1.11 \pm 0.80\text{‰}$ ) (Sutton et al., 2018; Xiong et al., 2015). We surmise that the dissolution of *E. rex* is comparable to that of small diatoms when exposed to an undersaturated environment given their identical chemical composition and *E. rex*'s porous structure. Sensitivity tests of small diatom dissolution on porewater  $\delta^{30}\text{Si}$  are shown in Figure S2 in Supporting Information S1, which exemplifies the failure of simulating the observations if assuming small diatom dissolution. Similar  $^{30}\text{Si}$ -depleted porewater data have been reported for the Guaymas Basin, the Barents Sea, and the Antarctic region, and interpreted as indicators for dissolution of terrigenous clay and metal oxides apart from biogenic opal (Closset et al., 2022; Geilert, Grasse, Doering, et al., 2020; Ward et al., 2022). In the southern Mariana Trench sediments, however, dissolution of lithogenic silicates does not necessarily contribute to the low porewater  $\delta^{30}\text{Si}$  values because simple dissolution of *E. rex* frustules that represent the predominant diatom species can explain the observations. Indeed, porewater DSi and  $\delta^{30}\text{Si}$  in  $\text{bSiO}_2$ -devoid cores show no clear indication of lithogenic phases dissolution as they are mainly scattered close to the seawater endmember (Figure 3). Data from the three  $\text{bSiO}_2$ -bearing cores would be expected to plot along the mixing line between deep water and *E. rex* frustule if simple dissolution of  $\text{bSiO}_2$  was the only process taking place, which is contrary to observations. Rather, porewater  $\delta^{30}\text{Si}$  values tend to be shifted above the mixing line (Figure 3). The elevated  $\delta^{30}\text{Si}$  values can be explained by DSi precipitation in the form of



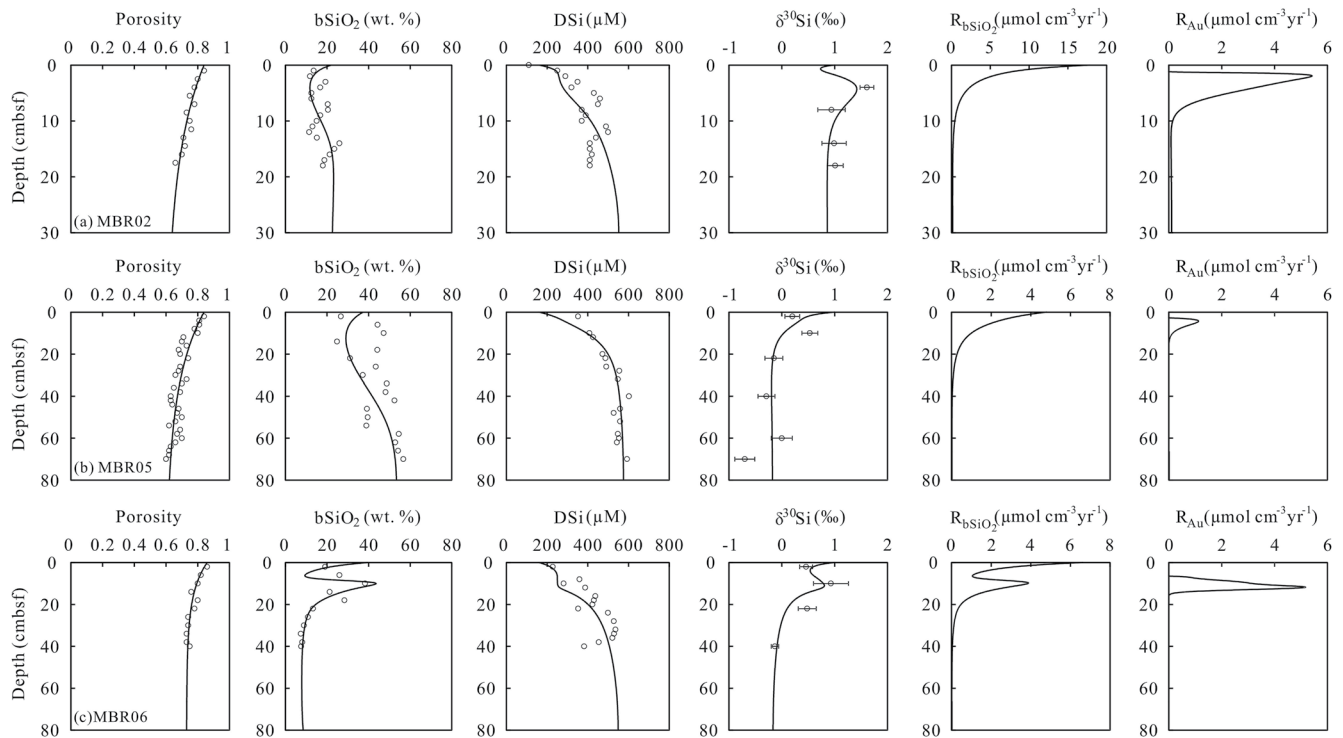
**Figure 3.** Crossplot of  $\delta^{30}\text{Si}$  versus  $1/\text{Si}$  in porewater. The dashed line represents the two-endmember mixing trend between the western Pacific deep water and *E. rex* frustules. The arrow indicates the potential influence of authigenic clay formation on porewater data. Error bars for stable silicon (Si) isotope data are shown except for those data points with errors smaller than the symbol size. Si concentration for the endmember of *E. rex* frustules was assumed to be the equilibrium concentration in respect to biogenic silica solubility ( $1000 \mu\text{M}$ ) under the ambient temperature and pressure of the Mariana Trench (Van Cappellen & Qiu, 1997).

authigenic clay minerals following  $\text{bSiO}_2$  dissolution, given that the lighter Si isotope is preferentially incorporated into the solid phase during low-temperature precipitation reactions (Ehlert et al., 2016; Geilert et al., 2014).

Formation of authigenic aluminosilicate minerals or reverse weathering driven by  $\text{bSiO}_2$  dissolution has been reported in a variety of marine settings, which represents a potentially important sink for elements (e.g., Mg, K) and alkalinity (Loucaides et al., 2010; Michalopoulos & Aller, 1995, 2004; Michalopoulos et al., 2000; Rahman et al., 2016). However, uptake of  $\text{K}^+$  and alkalinity is not observed in MBR02, MBR05, and MBR06 (Figure S3 in Supporting Information S1) where authigenic clay formation is inferred to take place based on porewater  $\delta^{30}\text{Si}$  data. Given that a variety of authigenic aluminosilicates could form in the sediments, porewater  $\text{K}^+$  would not necessarily show a downcore decrease trend if K-poor aluminosilicates (e.g., chlorite and smectite) are the major authigenic products. In addition, alkalinity, on the contrary, exhibits a downcore increasing trend in these three cores (Figure S3 in Supporting Information S1). These typical alkalinity profiles are generated by organic matter mineralization that represents the dominant early diagenetic process in marine sediments, despite the occurrence of reverse weathering (e.g., Wallmann et al., 2008). Therefore, we postulate that porewater  $\delta^{30}\text{Si}$  could be a more sensitive and straightforward indicator for authigenic clay formation than porewater major element compositions.

### 3.2. Trends in $\text{bSiO}_2$ Diagenesis

Our model predicts active  $\text{bSiO}_2$  cycling in the uppermost 10–20 cmbsf, with different rates of  $\text{bSiO}_2$  dissolution and resulting authigenic clay formation among the three  $\text{bSiO}_2$ -bearing cores (Figure 4 & Table 1). The highest depth-integrated rates of  $\text{bSiO}_2$  dissolution and authigenic clay formation were found for the trench axis core MBR02. High rates of authigenic clay formation were necessary to reproduce the positive excursion of porewater  $\delta^{30}\text{Si}$  at 4 cmbsf. Although MBR05 contained the most abundant *E. rex* frustules, with  $\text{bSiO}_2$  up to 60%, model results revealed significantly muted  $\text{bSiO}_2$  dissolution and authigenic clay formation (Figure 4). Apart from dissolved silicic acid from  $\text{bSiO}_2$  dissolution, authigenic clay formation is dependent on the availability of other cations (i.e., Al, K, Fe) in the porewater. In river delta settings where the input of reactive terrigenous materials is substantial, biogenic opal acts as the primary factor limiting the formation of authigenic aluminosilicates (Michalopoulos & Aller, 1995, 2004), whereas in opal-rich sediments, production of authigenic aluminosilicates is mainly regulated by Al and Fe released from reactive lithogenic phases (Ehlert et al., 2016; Loucaides



**Figure 4.** Measured (symbols) and simulated (curves) depth profiles of porosity, biogenic silica (bSiO<sub>2</sub>), dissolved silicic acid (DSi), and porewater δ<sup>30</sup>Si, as well as modeled rate profiles of bSiO<sub>2</sub> dissolution and authigenic clay precipitation in the bSiO<sub>2</sub>-bearing cores (MBR02, MBR05, and MBR06). Note the different scales for the rate of bSiO<sub>2</sub> dissolution for the three modeled cores.

et al., 2010; Van Cappellen & Qiu, 1997). Therefore, the relatively low Si turnover rates at MBR05 could be explained by limiting amounts of Al and Fe oxide-rich detrital phases, as indicated by lowest detrital fraction at this site compared to the other two bSiO<sub>2</sub>-bearing cores (Table S6 in Supporting Information S1). The negative correlation between asymptotic DSi values and the ratio of detrital fraction to bSiO<sub>2</sub> in a variety of marine environments highlights the potential important role of lithogenic materials in promoting production of secondary authigenic aluminosilicates (Dale et al., 2021; Dixit & Van Cappellen, 2003).

It is noteworthy that a peak in bSiO<sub>2</sub> content of ~40% occurs at 10 cmbsf of MBR06, which is accompanied by positive excursion of porewater δ<sup>30</sup>Si. Based on our model set-up, an elevated bSiO<sub>2</sub> content would increase the rate of bSiO<sub>2</sub> dissolution, whereby it would be expected that the porewater δ<sup>30</sup>Si values would shift to lower values. However, the observation of an opposite trend of δ<sup>30</sup>Si variation suggests that authigenic clay formation which preferentially takes up the light Si isotope has a greater impact on the porewater δ<sup>30</sup>Si than bSiO<sub>2</sub> dissolution

**Table 1**  
Compilation of Silicon (Si) Turnover in Different Marine Environments

Location	bSiO <sub>2</sub> dissolution (μmol Si cm <sup>-2</sup> yr <sup>-1</sup> )	Lithogenic silicate dissolution (μmol Si cm <sup>-2</sup> yr <sup>-1</sup> )	Authigenic silicate formation (μmol Si cm <sup>-2</sup> yr <sup>-1</sup> )	Fraction of DSi re-precipitated by authigenic silicate (%)	Dominant lithology
Guaymas Basin <sup>a</sup>	26.4	52.2	7.6	9.7	Siliceous silty clay (14%–22% of bSiO <sub>2</sub> )
Peruvian margin <sup>b</sup>	234	–	56	24	Diatomaceous mud (4%–12% of bSiO <sub>2</sub> )
Arctic Barents Sea <sup>c</sup>	5.5–61.7	97.1–490.9	4.7–193.8	3–37	Diatomaceous mud (0.2%–0.4% of bSiO <sub>2</sub> )
Mariana forearc <sup>d</sup>	–	773.2	776.9	99	Serpentinite mud
Mariana Trench <sup>e</sup>	6.38–28.9	–	2.46–19.1	39–66	Laminated diatom mats/diatomaceous mud (7%–55% of bSiO <sub>2</sub> )

<sup>a</sup>Geilert, Grasse, Doering, et al. (2020). <sup>b</sup>Ehlert et al. (2016). <sup>c</sup>Ward et al. (2022). <sup>d</sup>Geilert, Grasse, Wallmann, et al. (2020). <sup>e</sup>This study.

representing the source of low  $\delta^{30}\text{Si}$  value. In order to reproduce the observations, the kinetic constant for authigenic clay precipitation ( $k_p$ ) at MBR06 was elevated in the interval of 0–15 cm, with a maximum centered at  $\sim 8$  cm (Table S3 in Supporting Information S1), thereby leading to a substantial increase in the rate of authigenic clay precipitation corresponding to the increase in the rate of bSiO<sub>2</sub> dissolution at the same depth (Figure 4). In contrast, the kinetic constants for authigenic clay precipitation at MBR02 and MBR05 were set in synchronization with those for bSiO<sub>2</sub> dissolution exhibiting an exponential downcore decrease (Table S3 in Supporting Information S1). As the kinetics of authigenic clay formation in the sediments is poorly constrained, the rate laws assigned in the model can only be viewed as empirical and have no theoretical and experimental basis. Despite the occurrence of LDMs with high bSiO<sub>2</sub> contents at 8–20 cmbsf in core MBR06 that were similar to MBR05, model results yielded significantly intensified authigenic clay formation at MBR06. This can be attributed to the differences in the detrital content in both cores, which determines the amount of Al and Fe oxide-bearing phases required to form authigenic aluminosilicates.

Understanding the fate of biogenic opal (recycling vs. burial) in sediments has important implications for evaluating the global Si budget. Rapid recycling of biogenic opal in surface sediments supplies biologically available DSi to the overlying water, which feeds positively back to the growth of planktonic communities. Burial of altered and unaltered bSiO<sub>2</sub> represent permanent removal of Si from the ocean reservoir. It has been suggested that total bSiO<sub>2</sub> burial could have been substantially underestimated if authigenic clay minerals are unaccounted for; mounting evidence supports the ubiquitous occurrence of authigenic clay formation in sediments (Michalopoulos & Aller, 2004; Rahman et al., 2016, 2017). Our simulations indicate that 39%–66% of DSi released from bSiO<sub>2</sub> dissolution is sequestered as authigenic silicates (Table 1). The efficient conversion bSiO<sub>2</sub> to authigenic silicates emphasizes the significant contribution of deep-sea sedimentary processes in the oceanic Si sink, in agreement with the quantitative assessments of benthic Si cycling in other marine settings.

#### 4. Conclusions

Porewater DSi and  $\delta^{30}\text{Si}$  data from five sites in the southern Mariana Trench reflect mixed diagenetic activities of bSiO<sub>2</sub> dissolution and authigenic clay formation. Model simulations at three bSiO<sub>2</sub>-bearing cores reveal highest bSiO<sub>2</sub> turnover rates in the trench axis (core MBR02) characterized by the lowest bSiO<sub>2</sub> content and highest abundant detrital materials. These results imply that authigenic clay formation in the bSiO<sub>2</sub>-rich sediments of the Mariana Trench is likely governed by the supply of lithogenic materials. Around 40%–80% of DSi from bSiO<sub>2</sub> dissolution is re-precipitated to authigenic aluminosilicates. Our findings have important implications for understanding benthic Si and associated elemental cycles in extreme water depths.

#### Data Availability Statement

The geochemical data used in the study are available at Mendeley data repository via <https://data.mendeley.com/datasets/wdgpddhwcv/1>.

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