

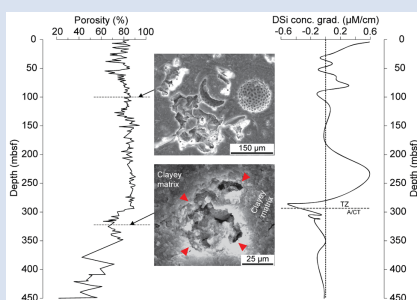
Pore water chemical constraints on petrophysical shifts following biosilica diagenesis

S. Varkouhi^{1*}, N.J. Tosca², J.A. Cartwright¹, Z. Guo³,
P. Kianoush⁴, A. Jurkowska⁵

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Abstract



Silica diagenesis commonly induces marked petrophysical changes to biosiliceous sediments across the sub-seafloor opal-A to opal-CT transition zone (TZ_{A/CT}). Integration of pore water thermodynamics with textural and mineralogical data reveals that sediments in the TZ_{A/CT} drilled at ODP Site 794 are sharply compacted *via* opal-A dissolution and matrix collapse. The porosity decline is linked to silica solubility through pore water saturation states with opal-A and opal-CT. A dissolved silicon (DSi) drop near the depth of the petrophysical shift implies that fluid expulsion removes the DSi produced by opal-A dissolution. Production of opal-CT from the pore fluid that is thermodynamically equilibrated with solubility of this silica mineral phase however slightly impacts the anomalous compaction at the TZ_{A/CT}.

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Introduction

Conventional perspectives on the compaction of deep water deposits deal with petrophysical properties (*e.g.*, porosity, density, and compressional velocity) as gently evolving traits with depth (Wrona *et al.*, 2017a). Nevertheless, siliceous hemipelagic sediments penetrated during the Deep Sea Drilling Project (DSDP) and Ocean Drilling Program (ODP) have undergone pronounced physical property variations (anomalous compaction) that typically accompany biogenic silica (opal-A) alteration toward opal-CT (α -cristobalite/ α -tridymite; Varkouhi *et al.*, 2020a). These petrophysical shifts occur through an early burial dissolution-redeposition process whose development is primarily regulated by temperature and time, with lesser controls from siliceous surface area, host lithofacies, and interstitial water chemistry (Williams *et al.*, 1985). Prominent opal-A content reductions ascribed to its dissolution, and the subsequent opal-CT precipitation compact the sediment framework and impose a porosity drop on the host lithology (Wrona *et al.*, 2017b). Distinct sediment density increases occur as porosity declines because pore volume decreases compress the solid matrix by densifying its constituents (Volpi *et al.*, 2003).

The drastic changes in petrophysics of opal-A deposits commonly generate a discrete reflector mainly identifiable on seismic sections when it cross cuts the neighbouring strata (Davies and Cartwright, 2002). This profound reflection marks the position of a transition zone, across which biosilica

appreciably dissolves and is replaced with opal-CT (TZ_{A/CT}; Varkouhi *et al.*, 2020a). The narrow (≤ 50 m thick) TZ_{A/CT} hosts both opal-A and opal-CT, whilst the latter phase dominates (Varkouhi *et al.*, 2021). Investigations into silica diagenesis and TZ_{A/CT} characterisation in sub-seabed sediments have been built upon 2D and 3D seismic stratigraphy, scientific drillings, and on-land surveys (Varkouhi *et al.*, 2022). Integration of these procedures with interstitial water chemistry has been applied to the current thermochemical state of biosilica diagenesis across the anomalous compaction section, *i.e.* active *versus* fossilised TZ_{A/CT} (Varkouhi *et al.*, 2020b). Following the original works on oxygen isotope relations among opaline polymorphs involved in silica diagenesis (Knauth and Epstein, 1976; Murata *et al.*, 1977; Pisciotto *et al.*, 1992), Yanchinila *et al.* (2020, 2021) and Ibarra *et al.* (2022) provided constraints on temperatures of biosilica transformation into opal-CT in deep water cherts using high resolution characterisation of oxygen-18 isotopic compositions. The alteration of opal-A to diagenetic opal (opal-CT) ceases in fossilised transitions, signifying that the TZ_{A/CT} is not at the phase predominance boundary between these silica polymorphs (Neagu *et al.*, 2010). Biosilica diagenesis progresses through an active TZ_{A/CT} instead. The view of an active silica diagenesis reaction front is based on the assumption that the TZ_{A/CT} has a regional sea bottom simulating geometry, which cross cuts the nearby stratigraphy, and is likely to be presently advancing upwards (Varkouhi *et al.*, 2022).

1. Department of Earth Sciences, University of Oxford, Oxford OX1 3AN, United Kingdom
2. Department of Earth Sciences, University of Cambridge, Cambridge CB2 3EQ, United Kingdom
3. School of Geographical Sciences, Hebei Normal University, Shijiazhuang 050024, China
4. Department of Petroleum and Mining Engineering, South Tehran Branch, Islamic Azad University, Tehran 1777613651, Iran
5. Faculty of Geology, Geophysics and Environmental Protection, AGH University of Krakow, Krakow 30-059, Poland

* Corresponding author (email: varkouhi1973@gmail.com)

Despite research on the TZ_{A/CT} structure and thermodynamics (e.g., Davies and Cartwright, 2002; Varkouhi et al., 2020b), the silica dissolution/precipitation kinetics driving petrophysical contrasts between porous opal-A deposits and more condensed opal-CT units is poorly understood. While Varkouhi et al. (2020b) describe diagenetic states of the TZ_{A/CT} using pore fluid chemical proxies, controls from pore water thermodynamics and kinetic modes of ion transport on petrophysics during silica diagenesis are inconclusive in their work. Elucidating the anomalous compaction drivers is crucial to understanding structural/thermal evolution in basin fills, and plays a role in petroleum systems as the TZ_{A/CT} contributes to drilling risks by often comprising hard cherts (Neagu et al., 2010).

This manuscript aims to link the sudden compaction of deep marine biosiliceous deposits that have undergone silica diagenesis to processes that transport dissolved phases in pore water through this reaction. To fulfil this objective, signatures of variations in interstitial water composition are combined with records of physical property changes and textural, mineralogical, and oxygen isotopic features of sediments subjected to biosilica diagenesis in Site 794 of the ODP Leg 127, the Japan Sea (Fig. S-1a–c). Interstitial water chemical constraints, including saturation states with silica polymorphs and dissolved silicon (DSi) and major ions concentration gradients indicate that opal-A transformation is active over the TZ_{A/CT} in this site and other borehole sites drilling the same transition zone (Varkouhi et al., 2020b). The pore fluid chemical variations can map the position of opal-A removal by dissolution and the resultant petrophysical shifts, pointing to where the diagenetic reaction remains active as diffusion is too slow to erase these chemical signals.

Materials and Methods

Essential data for this work come from the results of interstitial water analyses (Fig. S-2) and physical property measurements for Site 794 available through the International Ocean Discovery Program (IODP) database. Complementary mineralogical, textural, and chemical constraints were furnished with analyses of cored samples from this site. Optical petrography was conducted on smear slides with a Leica DM750 P microscope to describe the sediment biogenic and lithogenic components. Abundances of opal-CT were determined from X-ray diffraction (XRD) scans acquired using a PANalytical Empyrean diffractometer. The software Match! 3.16 identified the crystalline phases and determined their abundance in the sediments. The Reference Intensity Ratio (RIR) method, which scales diffraction data to the diffraction of reference material (corundum), was employed to quantify crystalline components in the samples with multiple phases. Distribution of frustules in the sediment, their dissolution and re-precipitation as opal-CT, and fabric evolution were characterised under a FEI Quanta 650 scanning electron microscope (SEM). Palaeo-temperatures of sediments with precipitated opal-CT were derived from δ¹⁸O values to trace opal-A inversion toward opal-CT through the anomalously compacted sediment interval. The operating procedures for these methods are detailed in the Supplementary Information.

Mineralogical and Textural Make-Up

Biosilica, mainly diatoms, is the sole major biogenic component, while clay-sized materials constitute the leading siliciclastic component of the sediment sequence in Site 794 (Fig. S-3a–e, b–h). The relative extent of opal-A assessed using visual

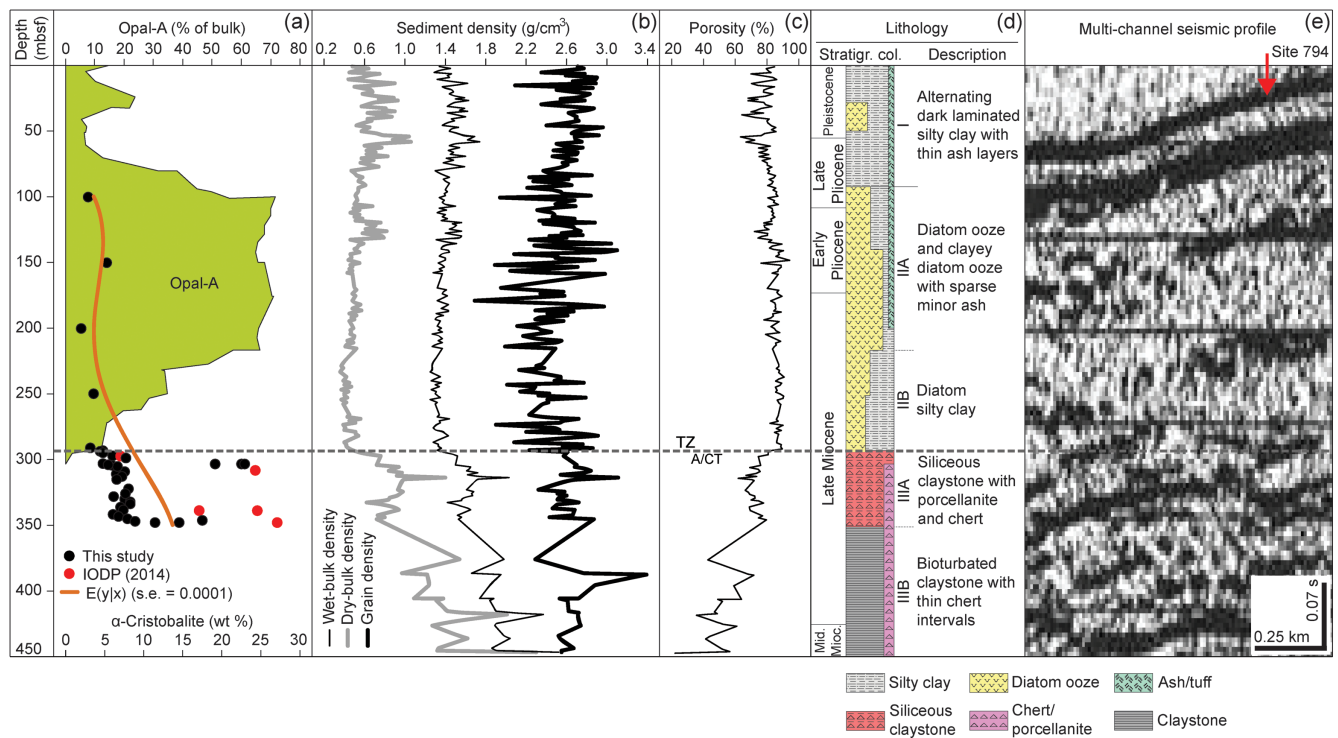


Figure 1 Integration of mineralogical and petrophysical measurements with seismic profiling, Site 794. (a) Reduced biosilica extents (green area) correlated with rising opal-CT contents (black and red circles), (b,c) anomalous density and porosity changes, and (d) onset of chert/porcellanite production across the TZ_{A/CT} at ~293.5 mbsf are tied to (e) exhibition of a high amplitude reflector ~0.37 s below the seafloor. The petrophysical profiles were modified from IODP (2014), and the biosilica distribution pattern with depth from Varkouhi et al. (2020a, 2020b). E(y|x)—non-linear regression, s.e.—standard error, Mid. Mioc.—middle Miocene, Stratigr. col.—stratigraphic column.

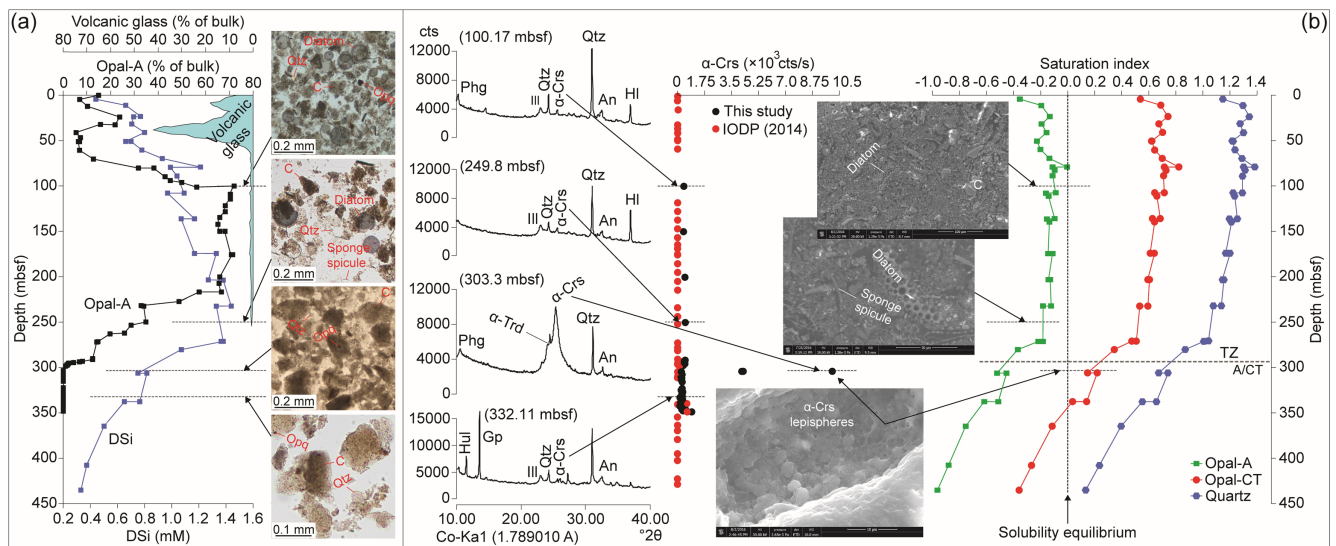


Figure 2 Pore water signatures of silica diagenesis at Site 794. (a) DSi profile closely corresponds to opal-A spatial distribution, and (b) pore water undersaturation with biosilica but supersaturation with diagenetic opal, and sediment textural evolution (detailed by cristobalite diffraction intensities and photomicrographs). Refer to Table S-1 for the sample identification. The saturation profiles were modified from Varkouhi *et al.* (2020b). The volcanic glass depth profile was based upon IODP (2014) smear slide data. α -Crs— α -cristobalite, α -Trd— α -tridymite, An—*anorthite*, C—*clay*, Gp—*gypsum*, HI—*halite*, Hul—*heulandite*, Ill—*illite*, Opq—*opaque*, Phg—*phengite*, Qtz—*quartz*.

estimation scheme of Marsaglia *et al.* (2013) records a peak of 72 % at ~100 mbsf for a diatom ooze, mixed with fine volcanic glass, sealing the $TZ_{A/CT}$ reflector of petrophysical changes (Figs. 1a–e, 2a and S-3a). The clayey fraction averaged at ~58 % in the opal-A sediment reaches 93 % within the opal-CT pile. The relative abundance of opal-CT was identified by peak intensities of planar spacing (d) (101) α -cristobalite at 26.18 – $25.14^\circ 2\theta$ and $d(110)$ α -tridymite grid at $23.98^\circ 2\theta$ in the XRD record of erratically aligned bulk samples (Fig. 2b). The XRD peaks of clayey and fine volcanic materials in multi-phase sediments overprint the diffraction signs of biosilica; making opal-A undetectable using this process. The intensity-depth profile of α -cristobalite as the opal-CT proxy (Damby *et al.*, 2014) displays the initial formation of this phase in the sequence over the $TZ_{A/CT}$ as a weak intensity diffraction set, while its intensity in the $TZ_{A/CT}$ largely varies between 0 and 10260 cts/s (Fig. 2b). The XRD detected normalised quantities of α -cristobalite vary between ~2 and ~27 wt. % and from 3.5 to ~31 wt. % for α -tridymite, with a mean value rise in the opal-CT-rich section to nearly three times that of the opal-A unit (Table S-1). The percentage of anorthite drops when shifting from the opal-A succession to the $TZ_{A/CT}$, but the mean quartz and phengite contents in the opal-CT pile remain close to that of the opal-A section. The siliceous textures in Site 794 exhibit a change in diatom content of the sediment with depth from significant to none (Fig. S-4a–h). The siliceous frustules are largely fragmented and dissolved even at shallow burial depths (Figs. 2a,b, S-4a), and their dissolution degree varies from moderately dissolved (for depths near the seabed) to low close to the base of opal-A interval (Fig. S-4b–e). The microfabric within the $TZ_{A/CT}$ and its underlying unit is featured by rather perfect biosilica dissolution and its replacement by growth of opal-CT lepispheres in a condensed clayey matrix with a reduced porosity (Fig. S-4f–h).

Ionic Transport Modes in Biogenic Silica Diagenesis

Biogenic silica diagenesis in Site 794 has crucially governed the pore water chemistry, particularly the gradient of major ions (Figs. 2a, 3 and S-2). Notable DSi and lithium releases to the

interstitial water above, and removal from the pore solution in the $TZ_{A/CT}$ imply that this transition behaves as a diffuse boundary that develops concentration gradients downwards below it and upwards into the ocean water (Varkouhi *et al.*, 2020b; Figs. 2a, S-2). Because the sediment diffusivity changes with burial (Fig. 3), the profiles linking ionic concentrations above and below the $TZ_{A/CT}$ are non-linear. The burial depth reliance of diffusivity is expressed by its connection to pore structures (Yong *et al.*, 2014), indicating the more reduced diffusion in lower porosity opal-CT sediments than in biosiliceous deposits (Fig. 3).

The exceedingly low permeability of biosiliceous deposits (5.35×10^{-20} to 1.97×10^{-14} m²; Holler, 1992) contributes to their greatly lowered sediment diffusivity with depth, especially in the $TZ_{A/CT}$ and deeper deposits at Site 794 (Fig. 3). Consequently, diffusive flows are negligible drivers for DSi removal from the pore water and bypassing of opal-CT documented across the $TZ_{A/CT}$ (Figs. 1a, 2a,b).

Conversely, a water content depletion coincident with the porosity shifts from biosilica oozes to opal-CT sediments (Fig. 3) supports the argument that DSi and other ions have not diffused in the sediment at rates comparable to those by advective ion transport modes. The opal-CT-rich sediment is thus more compacted than opal-A deposits because its transformation from biosilica involves pore volume reduction when pore water is expelled. The low permeable overburden sealing the $TZ_{A/CT}$ may impede the interstitial fluid expulsion because advection depends on higher permeability rather than diffusive fluxes (Kwon *et al.*, 2004). However, the pore fluid release when opal-A converts into opal-CT is influenced by over pressure, and the low permeability biosilica section above the $TZ_{A/CT}$ allows elevation and perpetuation of the over pressure that drives fluid outfluxes (Davies *et al.*, 2008).

Thermodynamic Indications of Active Silica Diagenesis

The ongoing opal-CT production is evident from highly undersaturated pore water state with biosilica while supersaturated with diagenetic silica polymorphs, including opal-CT and quartz along

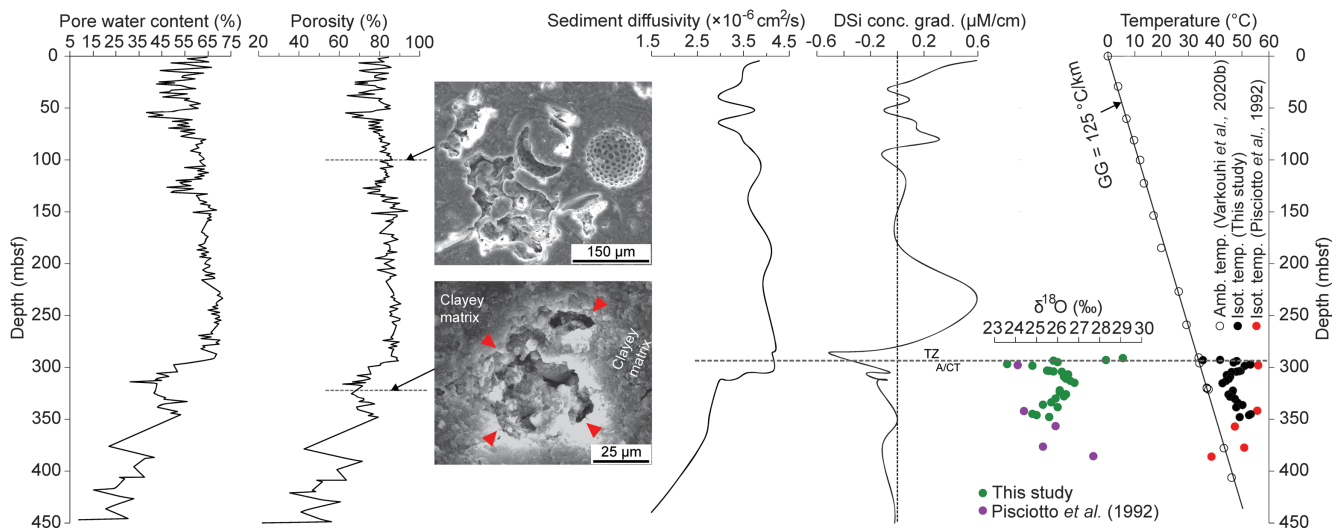


Figure 3 Pore volume drop due to sediment framework compactional collapse (red arrows) via pore water expulsion, documented by shifts in DSi gradient, oxygen isotopic composition, and temperatures of the $TZ_{A/CT}$ sediment; Site 794. The inverse DSi gradient indicates its removal, while highly reduced diffusivities occur, at the $TZ_{A/CT}$. The physical property profiles were modified from IODP (2014). The DSi gradient was estimated as concentration differences through each pore water sampling interval, and the sediment diffusivity as $\Phi^2 \times D$, where Φ is the porosity and D ($5.5 \times 10^{-6} \text{ cm}^2/\text{s}$; Dixit and Van Cappellen, 2003) the free solution silica diffusivity. The $\delta^{18}\text{O}$ values used to assess the isotopic temperature of opal-CT (see Table S-2) were computed by following Kita *et al.* (1985). Amb. temp.—ambient temperature, conc. grad.—concentration gradient, GG—geothermal gradient, Isot. temp.—isotopic temperature.

the sediment unit and the $TZ_{A/CT}$ in Site 794 (Figs. 1a, 2b). Consequently, biosilica dissolves throughout the sediment section, but synchronously the opal-CT supersaturated pore fluid in the biosiliceous unit reaches solubility equilibrium with this phase in the $TZ_{A/CT}$ (Fig. 2b). The solubility control is thus reached with opal-CT and yet this polymorph precipitates across the $TZ_{A/CT}$, where it has been largely produced since the late Miocene onset of the transition advancement (Varkouhi *et al.*, 2021).

This further argues for dominance of pore fluid advection with minor influences from diffusion across the interval of significant opal-CT production. The $TZ_{A/CT}$ coincides with the depth of DSi sink, where aqueous DSi leaves the pore water system through huge volumes of the interstitial water expelled, and thus a porosity decrease (Figs. 2a, 3). Co-occurrence of the DSi sink, the sharp physical property variations, and marked opal-CT production in Site 794 $TZ_{A/CT}$ (Figs. 1a–c, 2a,b) demonstrates that this DSi drop is essentially governed by active biosilica diagenesis. Under slow hemipelagic sedimentation (1–104 m/Myr; Varkouhi *et al.*, 2021), advection and diffusion potentially erase signatures of non-steady state DSi concentration drops unless diagenesis still continues or occurred recently (the 10–50 m/Myr accumulation rate in Site 794 falls within slow rate ranges; Shipboard Scientific Party, 1990). If solubility equilibrium with opal-CT and its production had ceased at Site 794, the late Miocene chemical signals of the last phase of silica diagenesis would have been erased in the present concentration profile under advective or even highly reduced diffusive rates.

The solubility equilibration at the boundary of diagenetic silica formation from dissolved opal-A conforms to temperature increases with depth as the assessment of saturation state involves temperature constraints. Hence, opal-CT equilibrates with the pore water as temperature increases with burial, highly suggesting that active opal-CT production in Site 794 links to the present day thermal gradient (Fig. 3).

Clustering of isotopic temperatures for the $TZ_{A/CT}$ sediment section and its underlying unit, extracted from the opal-CT $\delta^{18}\text{O}$ using the fractionation relation of Kita *et al.* (1985), near the present temperatures (Fig. 3, Table S-2)

supports the notion that biosilica diagenesis has thermodynamically equilibrated with the modern thermal gradient at Site 794 (Pisciotta *et al.*, 1992). Accordingly, the lack of signatures of sudden $\delta^{18}\text{O}$ changes with depth in the opal-CT-rich interval, *i.e.* a $\sim 5.5\text{‰}$ range of $\delta^{18}\text{O}$ variations maintained throughout the section (Fig. 3), implies that diagenetic opal has acquired these $\delta^{18}\text{O}$ records via equilibrium with its solubility in the pore water; an indication for the presently active state of the Site 794 $TZ_{A/CT}$.

Anomalous Compaction in Biosilica Deposits - A Synoptic Model

The chemical transition from biosilica to opal-CT concomitant with a porosity drop of $\sim 15\%$ (from ~ 80 to $\sim 65\%$; Fig. 1c) in the $TZ_{A/CT}$ at Site 794 is closely analogous to those formerly approximated for hemipelagic oozes from geographically diverse basins. For instance, biosilica diagenesis has led to 16% pore volume reduction in the early Miocene sediment of Norwegian Sea (Shipboard Scientific Party, 1987) and 15% in the Neogene claystones of New Jersey Atlantic continental slope (Shipboard Scientific Party, 1994). The porosity decline due to opal-A dissolution and consumption underlined here and in comparable works corresponds with other petrophysical shifts, *e.g.*, density and velocity increases and pore water release (Figs. 1b, 3). The association of these petrophysical contrasts with silica diagenetic transformation is described by a conceptual model shown in Figure 4.

Following our model, under oxygen depleted conditions, the pore water DSi extent has been continuously fed by dissolution of biosilica buried below the seabed. Despite low sedimentation rates in Site 794, elevated primary productivity has been able to sustain a continuous opal-A rain to the seabed, and hence is proportional to biosilica burial and DSi recycling (von Breyman *et al.*, 1992). The volcanoclastics (*e.g.*, volcanic glass) admixed with biosilica may impart DSi to the pore fluid in quantities near those of opal-A dissolution. The low volcanoclastic content of the biosilica sequence and its negligible distribution in the $TZ_{A/CT}$ however, display notably less DSi release to the

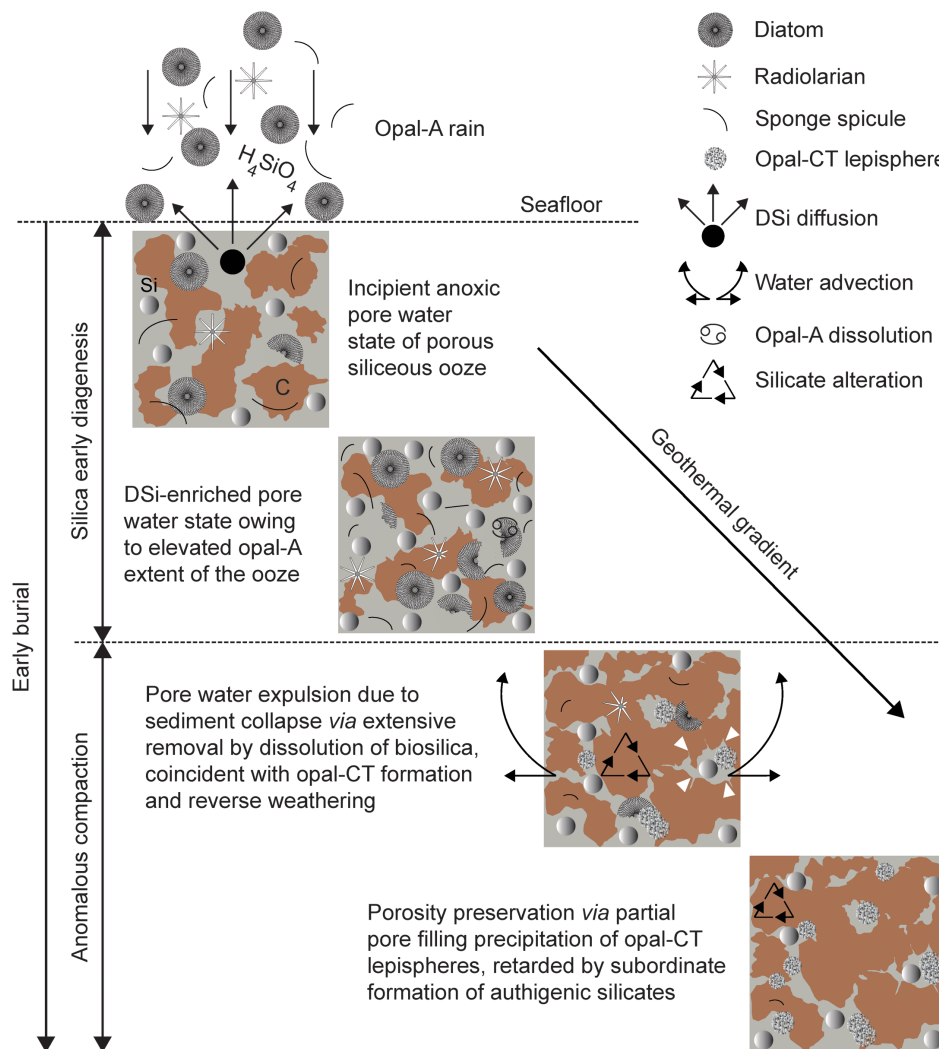


Figure 4 Compactional degradation of biosilica to opal-CT in hemipelagic deposits investigated. Silica diagenesis begins chemically near the seabed, but is not expressed as petrophysical changes until the reaction develops through opal-A dissolution and the removal of DSi by vertical/lateral pore water escape. Opal-CT production, retarded by reverse weathering, partially affects the porosity.

pore solution through volcanic glass alteration than biosilica dissolution (Fig. 2a; Varkouhi et al., 2020b).

The increasing DSi level with depth in the siliceous ooze implies the onset of biosilica early diagenesis (Fig. 2a), but is not expressed as pore fluid expulsion and porosity decreases until the reaction is complete via pore water DSi removal and subordinate opal-CT formation (Fig. 4). The anomalous compressional changes when opal-A undergoes appreciable dissolution happen abruptly (Wrona et al., 2017b). Disintegration and significant dissolution of biosilica destabilise the sediment structure and lead to the collapse of intergranular matrix (Fig. 3). Elevated pressures generated during opal-A dissolution and sediment compaction, and sustained by low permeability of clay-rich overburden, drive pore water escape from the compacted opal-CT sediment (Fig. 4).

Although opal-CT cements in the sediment fabric affect the porosity (Fig. S-4f-h), their impact on pore volume decrease is subordinate to precursor opal-A dissolution (Wrona et al., 2017b). This can be attributed to authigenesis of aluminosilicates, i.e. reverse weathering (Krissansen-Totton and Catling, 2020), which is associated with opal-CT production in deep marine deposits. These reactions compete with opal-A diagenesis for uptake of some ions, as evidenced by Si, Li, K, and Mg

removal from interstitial waters in Site 794 $TZ_{A/CT}$, but Ca and Sr additions through silicate alteration (Fig. S-2). Opal-CT precipitates in the $TZ_{A/CT}$, but its solubility equilibrium is slowly reached across this boundary (Fig. 2b) likely because of the secondary clay/zeolite formation. Reverse weathering restricts massive opal-CT cementation in the anomalous compaction interval, so that the diagenetic opal sediment retains appreciable porosities (~65 % in average; Figs. 1c, 3). The retardation in silica diagenesis by secondary silicate formation is barely assisted by the precipitation of minor opaque minerals, pyrite, and gypsum (Figs. S-3a-h, S-4d).

Synthesis

In line with recent research on geochemistry of transitions from biogenic to diagenetic opal, this study developed chemical criteria for thermodynamic state of silica diagenesis at pore fluid scales. This high resolution approach relates the anomalous petrophysical shifts across the $TZ_{A/CT}$ in Site 794 to pore fluid variations of DSi and major ions, oxygen-18 composition of opal-CT, and saturation states with biogenic and diagenetic silica. Accordingly, the transformation of opal-CT from biosilica has not been fossilised since the late Miocene initiation of the

advance of petrophysical TZ_{A/CT} at this site, as pore fluid advection (and diffusion) has not erased the signatures of active diagenesis. Given the implication of anomalous compaction dimensions for basin evolution future thermodynamic research may construct more inclusive pictures showing how silica diagenesis modifies the petrophysics of sediments in other basins of different thermal and geochemical histories.

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Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2440>.



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