

Solar energy harvesting mechanisms of the frustules of *Nitzschia filiformis* diatoms: supplement

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1. Optical Confinement in Biosilica

The frustule was predominantly made of biosilica. Its refractive index was slightly higher than the surrounding water or inner cytoplasm. Due to this low index contrast and the thinness of the frustule, optical modes were only weakly confined to it. The number of modes and their confinement depended on the thickness, width, and perforation of the frustule as well as the wavelength of the mode. Regions that were perforated with holes had lower effective refractive index [1-3]. The E-field distributions of some of the lower order modes in the frustule are shown in Fig. S1.

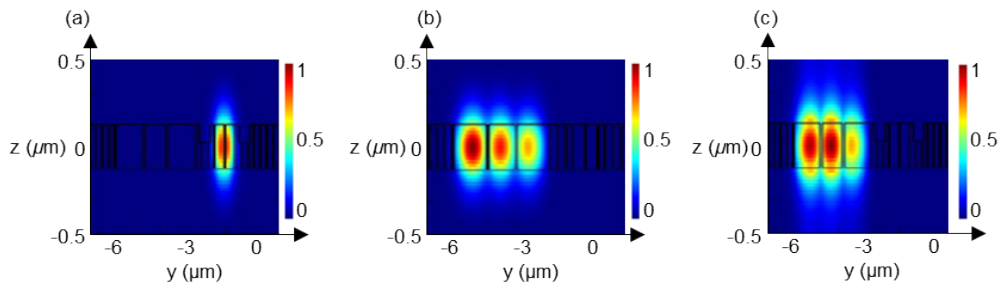


Fig. S1. Mode confinement in the frustule. (a) Fundamental mode inside the valve. (b) Lower order modes inside the girdle. (c) Higher order modes inside the girdle.

Due to weak confinement, the mode field expanded over a larger region such that it penetrated inside the cell. This high overlap facilitated evanescent coupling to the chloroplasts [4, 5] as well as directional coupling to adjacent frustules. Coupling to the chloroplasts was further supported by the marginally higher refractive index of the cytoplasm compared to water, which shifted the field distribution toward the inside of the cell. The location of the chloroplasts inside the cell was found to be variable. Outside the cell, directional coupling with neighboring frustules indicated a mechanism for light to be shared across diatoms in a colony. The weak mode confinement was also found to reduce birefringence resulting in a polarization independent optical response (characterized in Fig. S2(f)), which is beneficial for harvesting unpolarized sunlight.

2. Variations in the Photonic Band Structure of the Central Lattice

The central lattice occupied almost the entire exposed surface area of the valve and therefore, the majority of the frustule. This indicated the higher proportion of incident sunlight that it received in comparison to the other regions of the frustule. The lattice exhibited cylindrical holes from the outside. On the inside, they were covered by 10-nm-thick elliptical protrusions. The knobs were assumed to be artifacts from sputter coating the sample in preparation for scanning electron microscopy (SEM) imaging because otherwise, they would have fatally restricted chemical exchanges between the cell and its environment.

The arrangement of holes in the lattice formed a quasi-periodic photonic crystal (PhC). Different localized regions of the PhC exhibited different symmetries resembling square, triangular, and an intermediate oblique, depending on the angle subtended between the holes. Their corresponding photonic band structures are shown in Fig. S2(a). To quantify the effect of disorder in the PhC, its lattice parameters were varied to produce corresponding variations in the photonic band structure. Using the oblique symmetry as a basis, the dependence of the band structure on variations in angle (or symmetry), refractive index, thickness, spacing, hole diameter, and the polarization of light are shown in Fig. S2. To isolate the effect of a given parameter variation on the overall photonic band structure, only the selected parameter was varied while the others were held constant. The range of the spacing, hole diameter, and thickness parameter variations was two standard deviations from their mean value, that is, 2 σ as obtained from the SEM measurements.

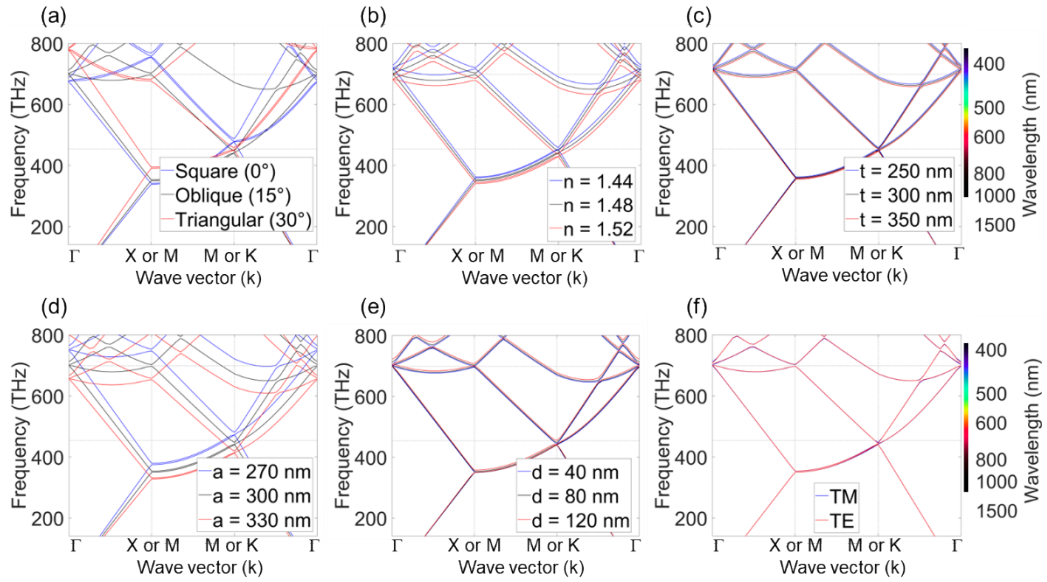


Fig. S2. Dependence of the photonic band diagram on the lattice parameters. Variations in (a) symmetry (corresponding to angle), (b) refractive index, n , (c) thickness, t , (d) spacing, a , (e) diameter, d , and (f) polarization.

For the valve, its guided modes were represented by the bands that were within the light cone of the photonic band structure. Their behavior depended on the localized parameters of the lattice. The strongest variations in the PhC response were due to the lattice symmetry, which could explain the high disorder in the angle between holes despite the longitudinal symmetry of the lattice. This also caused disorder in the spacing between holes. For example, an increase of 20 nm in the spacing proportionally lowered the position of the guided modes by 57 nm toward longer wavelengths. Varying the hole diameter resulted in a similar albeit weaker effect. Diameter variations of up to 100% (or 80 nm) still did not significantly shift the bands. The negligible effect of thickness [6] was attributed to the weak confinement of the modes. Varying the refractive index also had a negligible effect on the response, which justified approximating the biosilica material of the frustule as standard silica. Similarly, the negligible effect of polarization indicated the polarization independence of the optical response. This polarization insensitivity conferred an advantage on the frustule in the context of harvesting sunlight, which is unpolarized.

The central lattice exhibited long-range order despite having no translational symmetry due to the randomized variations of its parameters. This short-range disorder therefore modified the optical response of the PhC without eliminating it [7, 8]. As shown in Fig. S2, any variation of

the periodicity of the lattice induced a proportional change in its corresponding photonic band diagram. So, within a localized region where each unit cell contributed to the overall photonic band structure of the lattice, the disorder of the PhC proportionately shifted the bands into neighboring frequencies, ω , and wavevectors, k (or incident angles), corresponding to their (ω, k) position in the band diagram. This resulted in an effective ‘smearing’ of the photonic band structure. The effect was visualized in Fig. 2 of the main manuscript, whereby each hole was considered to be a unit cell based on the positions of its neighboring holes, and its corresponding band structure was calculated and plotted. The resulting ensemble average demonstrated an increased likelihood that a mode existed in the PhC to accommodate almost any (ω, k) combination of incident photons.

In this manner, the disorder facilitated random scattering in the lattice. Since the sharpness of peaks in the density of states (DOS) depended on long-range resonant scattering, these peaks got widened and flattened due to the perturbations of the lattice [9]. At each frequency, the DOS was calculated as the sum of all available wavevectors. Since this value depended on the resolution of the simulation, it was normalized within the wavelength range of interest. This broadened the spectrum over which a nonzero (or high) DOS was observed, and therefore increased the range of conditions for which modes were allowed. Additionally, the band gaps were collapsed due to the low refractive index difference, which caused significant overlap between neighboring modes. This effectively smoothed the overall mode field distribution at a given (ω, k) position. As a result, these hybrid modes better overlapped with incident plane waves and therefore, improved free-space coupling to the lattice. The combined DOS similarly increased retention.

The disordered PhC (DPhC) analysis showed that the structure was not only tolerant but in fact benefited from variations, which suggests that they might have evolved to enhance light absorption for a range of wavelengths and incident angles.

3. Resonances of the PhC

The bands in Fig. S2 indicated the modes of the PhC, that is, the allowed optical states for which the DOS was nonzero [10]. Conversely, empty (ω, k) positions in the diagram were indicative of regions in which there were no states for photons to occupy. For example, in the 550-820 nm wavelength range between the X-M symmetry points, the lowest order mode ran parallel to a higher order mode. Their proximity allowed an overlap of their corresponding DOS and field distribution, which averaged these quantities between both modes within that (ω, k) region. Such a combined mode field distribution offered a better overlap with incident light and therefore improved the coupling efficiency of the lattice. The modes also increased the overall DOS available for retention and waveguiding. Similarly, in the shorter wavelength range, the presence of multiple higher order radiation modes increased the coupling efficiency [11, 12]. However, this did not imply better retention in the frustule due to their position outside the light cone.

Positions in the band diagram where the slope of the band approached zero, implied a reduced group velocity [10]. Such regions typically occurred at band gaps or crossings near symmetry points. A band crossing was composed of at least two bands which crossed each other with a low, and possibly zero, slope. In contrast, a gap was composed of at least two bands near each other but not crossing. The width of the gaps in the central lattice were collapsed due to the low refractive index contrast and small hole diameter. For example, at 430 nm in Fig. S3(a), there was a convergence of multiple modes at the Γ point. The reduced slope and group velocity, combined with the presence of multiple bands, indicated a high DOS in those regions. This effect was amplified by the collapsed gap width, as seen at 900 nm. Here, the DOS was not reduced to zero between the bands, as it was for a typical band gap. On the contrary, these collapsed gaps allowed photon tunneling between the modes and therefore resulted in the DOS reaching a peak at the gap rather than on either side of it. An analogy in the spatial domain is a directional coupler, in which the separation between two waveguides can be small enough to

allow evanescent coupling between them, but when the separation is even smaller, then this results in a slot waveguide in which the intensity is actually higher inside the slot [13]. In this manner, the band gaps and crossings supported a continuously high DOS across the spectrum of photosynthetic active radiation (PAR), which contributed to the resonant behavior of the DPhC, as shown in Fig. S3.

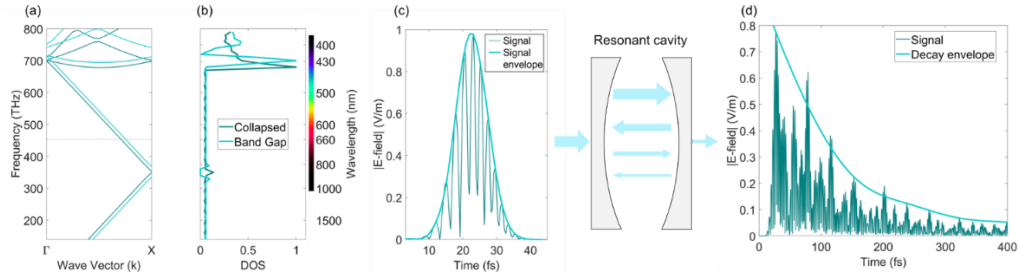


Fig. S3. Effect of resonances on the optical response. (a) Collapsed or open pseudogaps and band crossings with (b) their corresponding DOS. (c,d) Transient response of a resonance (represented by the resonant cavity) showing how (c) an optical pulse undergoes (d) decay due to the retention of light inside the cavity.

The resonances (collapsed pseudogap or band crossing) increased photon lifetimes [14] because photons remained confined or scattered in the frustule for longer durations, as depicted in Fig. S3. Hence, the retention of light by a collapsed pseudogap was more effective than if the gap was widened. Additionally, these resonances enlarged the localized DOS by reducing the group velocity and offering multiple modes for coupling, which increased the coupling efficiency. The increased light retention, light-matter interaction, and coupling efficiency to modes, all directly supported retention in the frustule which could assist the absorption rate of chloroplasts (from evanescent states [15]). In this manner, the resonances in the PhC contributed to the capture, redistribution, and retention of incident PAR.

4. Analysis of Scanning, Near-field Optical Microscopy (SNOM) Images

SNOM is a surface selective technique, which can be used to probe diatom frustules due to their transparency and thinness. Evanescent waves were coupled from the aperture to the sample in the near field at a fixed angle, which implied that the interaction for each pixel occurred only at the tip. During scanning, a constant separation between the tip and sample was maintained to ensure constant coupling despite the exponential decay of the wave with increasing separation distance [16]. Although SNOM is non-destructive, the tip itself damaged the sample in some cases.

In this transmission SNOM configuration, incident light was evanescently coupled into the sample by the apertured probe above it [16]. The amount of light coupled into the sample depended on the focal length of the lens and size of the aperture in the cantilever, both of which had a spectrally dependent optical response. This implied that long wavelengths (near infrared) were limited by the aperture size whereas short wavelengths (near violet) did not reach the aperture due to the correspondingly shorter focal length of the lens. Once coupled to the sample, the transmitted light was detected via an objective lens beneath it. The regions around the valve where light had coupled to, but diffused through, the cover slip provided a baseline intensity. Compared to this background, darker regions in the frustule indicated that light had not been transmitted to the collection optics. In this case, light had either not coupled into the sample due to poor contact (as seen inside the holes) or was redirected away from the point of contact (as seen in the diffraction pattern outside the valve edges). In contrast, the bright points indicated higher transmission through the sample, that is, where light was efficiently coupled to the valve without being diffused or redirected. A collection of these points formed bright lines which

were observed along the valve edges as well as laterally across the width of the central lattice. These continuous pathways indicated waveguiding.

The accuracy of the probed optical response was constrained by the measurement technique. For example, the angle at which the aperture was tapered, determined the fidelity of images obtained from surfaces with high curvature. Near the edges of the valve, the field was noticeably darker because the outer edge of the cantilever was physically restricted by its contact with the edge of the valve. As a result, the separation between the tip and the surface could not be maintained, which was confirmed by the blurriness of the AFM image. Similar dark spots were observed between the scaffolds of the raphe. Additionally, the convergence semi-angle of the bottom objective determined the aggregate of all k -vectors which were collected around the Γ symmetry point of the lattice. Hence, SNOM only probed the k -vectors around the Γ symmetry point for a single wavelength. However, note that the tip excited all modes in the lattice even if only the Γ symmetry point was preferred. Hence, light also coupled to modes that existed at other k -vectors. Due to scattering from imperfections in the lattice, residual light from these existing interacting modes would still reach the bottom objective. As a result, the sample was not dark despite the lack of modes at the Γ point for the SNOM wavelength, but the modes can be expected to appear brighter if probed at the correct angle. To do so, the angle of the detector would need to be adjustable, along with a sweep of the wavelengths within the range of PAR. The lack of mobility in our setup prevented the mapping optical bands in this manner.

Note that SNOM images did not produce a direct measurement of the localized DOS (LDOS) due to cross-coupling between the tip and sample [17]. However, given the experimental conditions and expected topography of the sample, the LDOS was inferred to have some correlation with bright pixels in the image. That is, the partial LDOS can be reconstructed from variations in the transmitted intensity resulting from the interactions between the aperture and the sample [18]. This is the sum of the partial DOS associated with the optical modes in each polarization direction, whereby its electric or magnetic part can be correlated with the near-field measurement [19]. So, the bright spots can be interpreted as the probability of detecting the electric field intensity of photons at that position. The bright points therefore indicated a higher LDOS at the point of contact in comparison to the surrounding regions. Since the LDOS indicated the availability of optical eigenmodes that allowed the existence of photons at a specific location, the bright points in the SNOM image were therefore assumed to map out the optical modes in the frustule. However, the total LDOS was not directly measured due to the vectoral nature of the optical field [19], which required a much larger convergence angles to obtain all k -vectors. Hence generalization was not straightforward. A complete LDOS measurement is only possible if all the modes are excited as well as detected, that is, illumination and detection by a wide solid angle. This was not possible using SNOM, hence, the image was generally interpreted as representing the k -vectors for the given laser wavelength, and possibly around the Γ symmetry point.

5. Photonic Circuit Model of the Frustule

The photosynthetic conversion efficiency of the diatom cell was assumed to match the absorption rate of its chloroplasts. The chloroplasts could receive incident PAR from the frustule via either direct transmission or evanescent coupling. Our results indicate that the frustule contributed to the PAR harvesting mechanisms of capture, redistribution, and retention. Each of these mechanisms was caused by a combination of the localized optical functionalities that were evoked by its architecture. We modeled this process as a photonic circuit, which is represented in Fig. S4(a). An analogous, simplified electrical circuit of this system is shown in Fig. S4(b) and its effect on photosynthesis is depicted in Fig. S4(c).

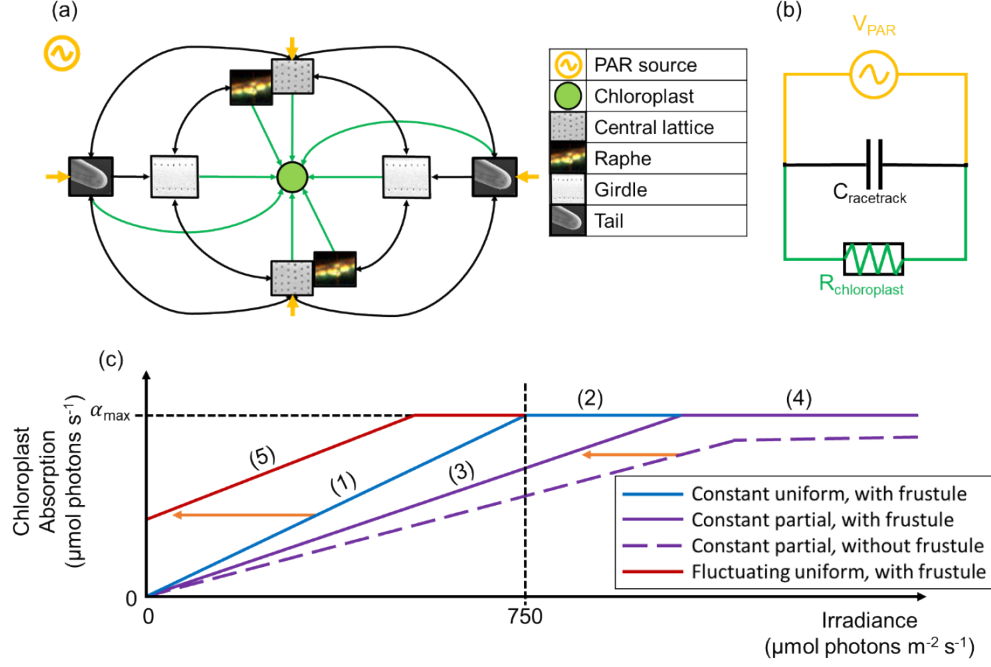


Fig. S4. Model of the optical response of the frustule. (a) Photonic circuit of the frustule showing (yellow arrows) incident PAR, (black arrows) light exchanged between different regions and (green arrows) absorption by the chloroplasts. (b) Electrical equivalent circuit with the chloroplasts, frustule, and PAR represented as a load resistor, capacitor, and variable power supply, respectively. (c) Contribution of the frustule to the absorption by the chloroplasts depending on the amount and distribution of irradiation on the cell. The slope represents the difference in the exposed surface area. The contribution of the frustule is depicted as orange vertical arrows in specific cases. The maximum absorption $\alpha_{\max}$ was reached under an irradiance of $750 \mu\text{mol photons m}^{-2} \text{s}^{-1}$.

The black, inner ring of Fig. S4(a) represents the cross-sectional racetrack formed by the central lattice, raphe, and girdle, which is shown in Fig. 7(a) of the main manuscript. It retained photons via circulation with an estimated Q factor of 271 to 822 at the wavelengths of 805 nm to 424.8 nm. For simplicity, this was assumed to be the main cause of retention in the frustule since the contributions from the raphe and DPhC merely amplified this effect. The remaining black arrows in Fig. S4(a) show how the racetrack received light via either butt-coupling from the tails or diffraction from the DPhCs in the central lattice and girdle. The radiation modes of the DPhC allowed light to couple into the racetrack from free-space with a simulated insertion loss (IL) of 4.56 dB at 450 nm [20]. Butt-coupling at the tail resulted in a simulated IL of 1.4 dB with free-space. Note that this is the approximate maximum value since the light source was aligned with the tail to obtain the lowest IL. For light being diffracted into lateral circulation by the central lattice and girdle, it incurred an approximately 7 dB loss [20]. Since these features also maintained lateral circulation by limiting longitudinal diffusion toward the tails, the loss from diffusion was neglected. The effective index of the racetrack was approximated as 1.38. Its optical loss per roundtrip was therefore calculated as,

$$Loss_{\text{racetrack}} = \frac{2\pi f T_r}{Q} \quad (\text{S1})$$

Here, f is the frequency, T_r is the transient time of the ring, and Q is its quality factor. This loss was calculated to be 2.6 dB per roundtrip.

The contribution of the capture, redistribution, and retention mechanisms depended on the initial state and boundary conditions of the system, which are represented in Fig. S4(c). We therefore present the following qualitative analysis of the contribution of the frustule to the absorption of chloroplasts using 5 cases which are based on the amount, location, and consistency of PAR that is incident on the diatom. Note that in all cases, the capture mechanism can be assumed to be always active and is therefore not discussed.

1. Under a constant, low, uniform irradiance (blue, solid, sloping line in Fig. S4(c)), any light in the frustule is immediately absorbed by the chloroplasts, which renders the retention mechanism redundant. Since the irradiance is uniform, the redistribution mechanism is also redundant. Hence, only the capture mechanism is useful to couple ambient light.
2. Under a constant, high, uniform irradiance (blue, solid, horizontal line in Fig. S4(c)), absorption of the chloroplasts is maximized, so excess photons are retained in the frustule. Although retention is activated, it is not used because these photons cannot be absorbed by the chloroplasts while they are already saturated. Similar to case 1, the uniformity of the irradiance renders the redistribution mechanism as redundant.
3. Under a constant, low, partial irradiance (purple, sloping lines in Fig. S4(c)), any captured light gets absorbed. A fraction might be redistributed for absorption by chloroplasts in the remaining regions of the cell, but this is only due to the passive optical behavior of the frustule rather than the presence of excess photons. Similar to case 1, the low irradiance renders the retention mechanism as redundant.
4. Under a constant, high, partial irradiance (purple, horizontal lines in Fig. S4(c)), the illuminated area of the cell absorbs photons at its maximum capacity until the chloroplasts are saturated. Excess photons can be made available to the remaining chloroplasts via the capture and redistribution mechanisms. This increases photosynthetic conversion in the cell. Similar to case 2, the retention mechanism is activated but not used.
5. Under a fluctuation from high to low, uniform irradiance (red, solid line in Fig. S4(c)), as represented by $t > 0$, the retention mechanism is most useful. Excess photons retained in the frustule are available to the chloroplasts so that photosynthesis can continue despite a drop in irradiance.

The redistribution mechanism was found to be useful when the frustule is partially irradiated. Since it effectively increases the exposed surface area under irradiation, it was represented by an increase in the slope of the corresponding lines in Fig. S4(c). Redistribution across the frustule allowed us to make an analogous assumption that the chloroplasts were distributed throughout the cell. The retention mechanism gets activated when the irradiation exceeds the maximum absorption of the chloroplasts and becomes useful when the irradiation drops below the maximum. Since it provides a smoothing effect during fluctuations in irradiance by releasing stored optical energy during periods of darkness, it was represented by a positive offset of the corresponding lines in Fig. S4(c). The adaptations of chloroplasts to available light were not accounted for in this model.

This retention mechanism was additionally modeled by a capacitor (the frustule) connected in parallel with a resistive load (the chloroplasts) to smoothen spikes or cuts in the power supply (incident irradiance). Similar to a resistor, the maximum absorption rate of chloroplasts, $\alpha_{\max}$, was assumed to be a constant 3.98×10^{12} photons/s using an irradiance of $750 \mu\text{mol photons m}^{-2} \text{s}^{-1}$ at 440 nm [21]. It was calculated as,

$$\alpha_{\max} = IA\epsilon \quad (\text{S2})$$

Here, I is the maximum irradiance, A is the surface area of the frustule, and ϵ is the absorbance coefficient of the chloroplasts approximated as 1 dB [20]. To find the contribution of the frustule to photosynthesis, we calculated the contribution of the racetrack to α . We assumed that all the

photons retained in the frustule were available for absorption by the chloroplasts at any time, that is, the retention rate, $Rate_{\text{retained}} = 1 - Rate_{\text{leak}}$, determined the maximum photons per second available for absorption. The leakage or decay rate, $Rate_{\text{leak}}$ was calculated using the total rate of photons/s that coupled into the racetrack due to the initial irradiance and harvesting mechanisms as well as $Loss_{\text{racetrack}}$.

For an irradiance of $550 \mu\text{mol photons m}^{-2} \text{ s}^{-1}$, the $Rate_{\text{retained}}$ that enabled $\alpha \rightarrow \alpha_{\text{max}}$ was calculated as 0.319×10^{12} photons/s. The total contribution to photosynthesis provided by the frustule was then estimated as $Rate_{\text{retained}}/\alpha_{\text{max}}$ because this represented the percentage increase in α due to the retention mechanisms of the frustule. It was found to be 9.83% at a wavelength of 450 nm. Note that this value represents an estimated percentage increase based on the specified conditions and was only calculated for a single wavelength. The accuracy of the calculation can be improved by conducting a full spectral analysis of the frustule within the wavelength range of PAR.

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