

Supplementary Information

Structural and Spectral Adaptation of the Seagrass *Posidonia oceanica* Photosystem I to Seabed Light

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Supplementary Methods

Förster Resonance Energy Transfer (FRET) calculation

Excitation energy transfer within the PSI-LHCI-mega was modeled as Förster resonance energy transfer. FRET is a good approximation for transfers between Chl *b* and Chl *a* pairs, since the coupling strength is much smaller than the energy separation between the Q_Y energy levels of the molecules ($\sim 450 \text{ cm}^{-1}$)¹, but it can also be assumed for Chl *a*-Chl *a* pairs, since Chl *a*-Chl *a* pairs with distances less than 1 nm (where multipole expansion usually breaks up) contribute by less than 1% of the total pairs in PSI complexes². Thus, pools of Chl *a* and Chl *b* were considered isoenergetic (except for low-energy Chl forms), in calculating structure-based Förster efficiencies. General site energies based on the chemical nature of the pigment, obtained from³, were only considered to determine EET direction between pigments of different chemical nature. Since the specific site energies for each chromophore have been experimentally determined only in a few cases⁴ and, thus, are not available for a complete PSI-LHCI supercomplex⁵, we devised a proxy for site energies based on the assumption that the site energy of a pigment is lower the greater the strength of interaction with other pigments^{6,7}. Site energies allow rescaling the interactions between different pairs, giving a more realistic directionality to the energy flow.

FRET transfer rates between Chls were computed using a point-dipole approximation (PDA). The Chlorophyll point dipoles were placed at the geometric centers of the four nitrogen atoms of the Chlorine ring⁸. The Chlorophyll Q_Y and Q_X transition dipole moments were taken parallel to the nitrogen ND-NB and NC-NA axis, respectively, as in^{1,9}. The Q_Y transition dipole moments were used for couplings between Chls (Chl *a*-Chl *a*, Chl *a*-Chl *b*, and Chl *b*-Chl *b*). The FRET transfer rate k_{ij} (s⁻¹) between two chlorophylls *i* and *j* is computed as:

$$k_{ij} = \left(\frac{R_{0ij}}{R_{ij}} \right)^6 \frac{1}{\tau_i}$$

where R_{0ij} is the Forster distance between Chlorophylls *i* and *j*, R_{ij} is the distance between the point-dipoles and τ_i is the fluorescence lifetime of chlorophyll *i* in the absence of chlorophyll *j*. Chl *a* and *b* fluorescence lifetime values were set at 5.6 ns and 3.1 ns¹⁰, respectively. The Forster distance R_{0ij} (nm) is computed as:

$$R_{0ij}^6 = \frac{9 \ln(10)}{128\pi^5 N_A} \frac{k_{ij}^2 Q_i}{n^4} J_{ij}$$

where N_A is Avogadro's number, k_{ij} is the orientation factor between chlorophylls *i* and *j*:

$$k_{ij} = (\hat{\mu}_i \cdot \hat{\mu}_j) - 3 \cdot (\hat{\mu}_i \cdot \hat{r}_{ij})(\hat{\mu}_j \cdot \hat{r}_{ij})$$

Q_i is the fluorescence quantum yield of Chl i (0.25 and 0.11 for Chl a and Chl b , respectively¹¹, n is the refractive index of the medium (equal to 1.55¹), and J_{ij} is the spectral overlap between the emission of Chl i and absorption of Chl j :

$$J_{ij} = \int f_i(\lambda) \epsilon_j(\lambda) \lambda^4 d\lambda$$

where f_i is the normalized emission spectrum of pigment i and ϵ_j is the extinction spectrum of pigment j . Chl a and Chl b emission and extinction spectra taken from PhotochemCAD (<https://www.photochemcad.com>).

The site energy of a pigment i , E_i (cm⁻¹), is therefore calculated as:

$$E_i = \frac{\sum_j V_{ij}}{N - 1}$$

Where V_{ij} is the excitonic coupling between the two pigments i and j , and $N-1$ is the total number of pigments interacting with pigment i (in the case of a fully connected network). In the PDA, couplings between Chls and Cars were computed with the Q_X transition dipole moments for Chls¹². The carotenoid point dipole was placed on the C15 atom, and the transition dipole moments (transition S0-S2) were considered oriented parallel to the central part of the polyene chain (atoms C11-C33), as modeled in previous studies⁹. The excitonic couplings V_{ij} (in cm⁻¹) in the PDA were calculated using the following formula:

$$V_{ij} = \frac{f_1^2 |\mu_i| |\mu_j|}{\epsilon_r} \cdot 5.04 \frac{k_{ij}}{R_{ij}^3}$$

Where f_1 is the local field correction factor, $|\mu|$ the module of the transition dipole moment, μ is the normalized transition dipole moment vector, ϵ_r the relative dielectric constant (here equal to 2.4^{1,9}), k_{ij} is the orientation factor between pigment i and j dipoles, and R_{ij} the module of the distance between the point dipoles. Dipole moment values were taken as 4 D, 3.4 D, and 4.5 D for Chl a , Chl b , and Cars, respectively^{1,9}. In the case of $\epsilon_r = 2.4$, $(f_1^2 \mu^2)/\epsilon_r$ is calculated to be 17.6 D² for Chl a ¹. Excitonic couplings have been computed for all the pigment pairs in the PSI-LHCI-mega, giving rise to a fully connected network with N nodes (pigments). From this perspective, E_i can be seen as the average coupling of pigment i with other pigments. In *network theory, $E_i = S_i^{\text{TOT}}/N-1$ where S_i^{TOT} is the total strength of the node i . The value $1/E_i$ can be interpreted as the energy level of a pigment.

Assigning site energies to pigments allows re-scaling FRET rates between different pairs. According to this model, while both directions of energy transfer are allowed between two pigments, the transfer rate changes according to the detailed balance equation, giving the rescaled FRET transfer rates k_{ij}^E :

$$k_{ij}^E = k_{ij} e^{-1.439 \frac{E_j - E_i}{T}}$$

with the temperature T set at 298K.

As the site energies already consider the interaction and effect of the carotenoids, by this approach, we can implicitly include their contribution to the rescaling, even if the transfer rates are computed only for Chl pairs.

Two-dimensional Electronic Spectroscopy (2DES)

2DES uses a sequence of three delayed broadband femtosecond pulses that interact with a sample. This results in the emission of a third-order nonlinear signal, which depends on the delays between the pulses. Specifically, the first and the second pulses excite the sample and the delay between them is referred to as ‘coherence time’ and labeled t_1 . The third pulse acts as the probe, and the delay between the second and third pulse is called ‘waiting time’ and labeled t_2 . A single 2DES map at fixed t_2 delay is obtained by recording the transient absorption (TA) spectrum of the system with a spectrometer as a function of the delay t_1 . This yields the TA map $\Delta I(t_1, t_2, \omega_3)$, where ω_3 is the detection frequency. Fourier transforming with respect to the delay t_1 yields the map $\Delta I(\omega_1, t_2, \omega_3)$, where ω_1 is the excitation frequency. Repeating this procedure for different t_2 delays yields a collection of time-resolved 2D maps as a function of the excitation and detection frequencies.

In this work, we used the partially collinear pump-probe¹³, in which the first and the second pulses propagate along the same direction, and all the pulses are spectrally degenerate. These pulses were generated by a non-collinear optical parametric amplifier (NOPA) pumped by a Ti: sapphire laser with a wavelength of 800 nm, a repetition rate of 1 kHz, and a pulse duration of 100 fs¹⁴. The NOPA output is a broadband visible pulse with a spectrum spanning from 580 nm to 720 nm. This pulse is temporally compressed down to 15 fs with a pair of chirped mirrors. We performed the 2DES experiment by placing the sample in a 200- μ m-pathlength cuvette and flowing it through a peristaltic pump whose reservoir is placed in ice to keep the temperature below 10°C. For both samples, we used a fluence of 44 μ J/cm², to avoid singlet-singlet annihilation. Supplementary Figures 17 and 18 show the 2DES maps of the PSI-LHCI and PSI-LHCI-mega at different t_2 delays, respectively.

2DES Global analysis

The 2DES data of the *P. oceanica* PSI-LHCI and PSI-LHCI-mega are modeled as a sum of four exponential functions, as described by the following expression¹⁵:

$$\Delta I(\lambda_1, t_2, \lambda_3) = \sum_{i=1}^4 A_i(\lambda_1, \lambda_3) e^{-\frac{t_2}{\tau_i}}$$

The amplitude matrices A_i are called 2D Decay Associated Spectra (2D-DAS) and represent the amplitude of the i -exponential function, which is characterized by the time constant τ_i . The choice of four exponential functions is based on the minimum number of exponentials that

satisfactorily reproduce the experimental data. Supplementary Figure 19 shows the 2D-DAS for PSI-LHCI and PSI-LHCI-mega. Here, amplitude sign, and the corresponding time constant, reveal the signal decay (positive amplitudes) and signal rise (negative amplitudes). For this reason, 2D-DAS enabled us to isolate and temporally characterize the distinct ultrafast energy-transfer pathways in the system. The results of the global analysis of PSI-LHCI isolated from *S. oleracea*, as well as the discussion, can be found elsewhere¹⁶.

Mean decay lifetime (τ_m) from 2DES

From the global analysis, we modeled the entire 2DES data set obtained for each sample with a sum of exponential decays. In this condition, the mean decay lifetime τ_m can be expressed with the following formula:

$$\tau_m(\lambda_{exc}, \lambda_{det}) = \frac{\sum_i A_i(\lambda_{exc}, \lambda_{det}) \tau_i^2}{\sum_i A_i(\lambda_{exc}, \lambda_{det}) \tau_i}$$

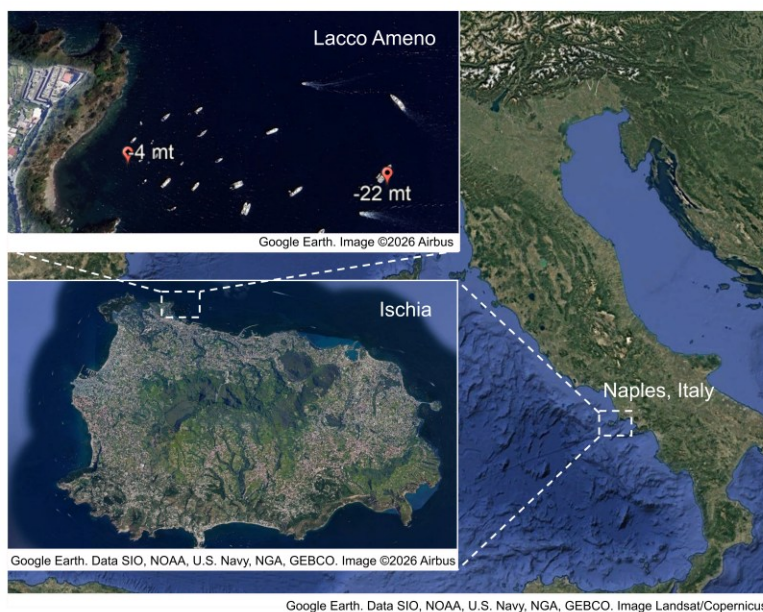
Where A_i are the 2D-DAS obtained with the global analysis and τ_i are the corresponding time constants. The result is a 2D matrix representing the first moment of the normalized decay distribution as a function of the excitation and detection wavelengths. This value can highlight potential differences in decay rates depending on the excitation or detection site, revealing which energy equilibration mechanisms among the chromophores are faster or slower, depending on the excited chlorophylls. Although this parameter does not correspond to the trapping time, it obviously depends on it. Such analysis had already been applied to 2DES data, highlighting the energy transfer limitations imposed by the low-energy Chl forms in the supercomplex PSI-LHCI isolated from land plants¹⁶. Supplementary Figure 20 shows the τ_m maps obtained from the PSI-LHCI and PSI-LHCI-mega of *P. oceanica*. We focused on the 670-700 nm spectral window, which corresponds to the spectral signature of the bulk Chls and P700. Such maps are calculated neglecting the non-decaying component of the global fit due to the long-lived signal of the P700. The data are cut up to 700 nm since no signal above such detection wavelength is observed, causing artifacts in the calculation of the mean decay.

Supplementary Figures

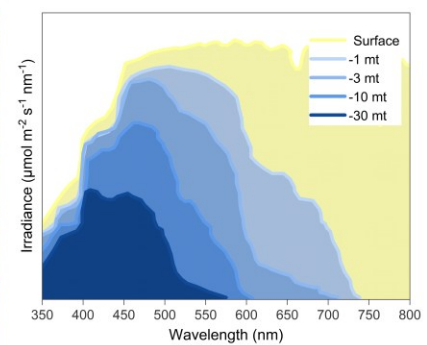
a



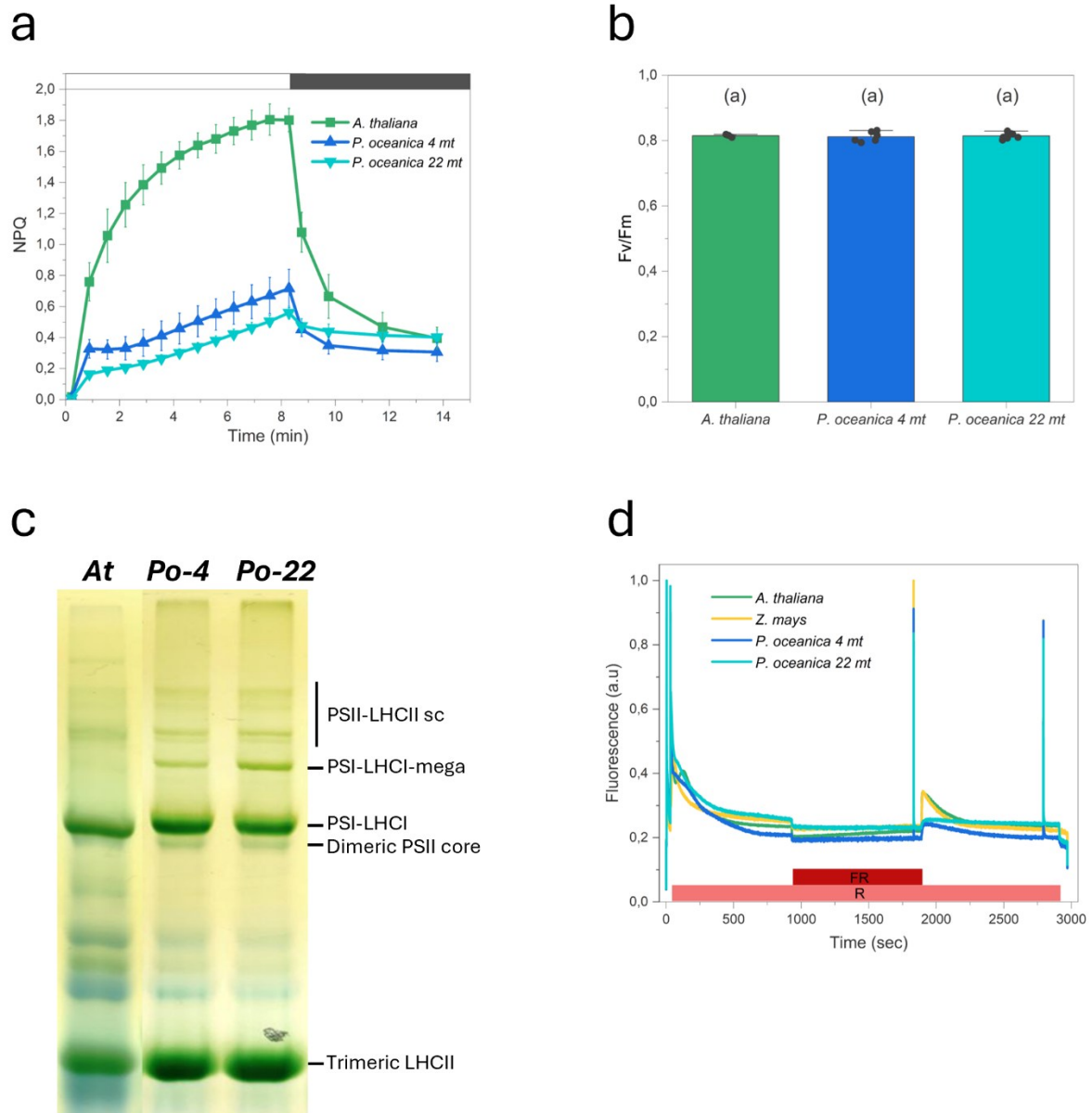
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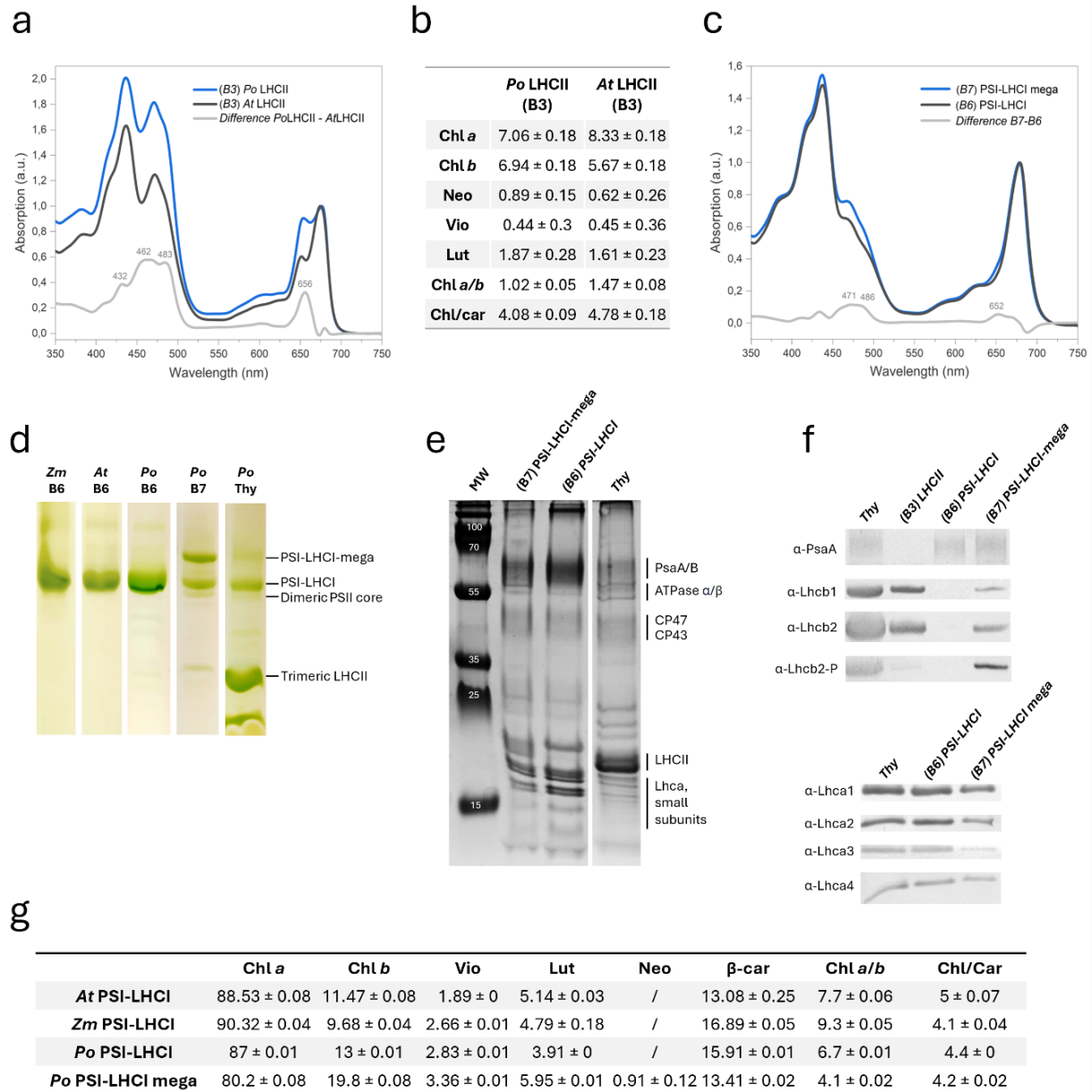


Supplementary Fig. 1. *P. oceanica* meadows, sampling sites, and irradiance spectrum underwater. (a) Representative photograph of a *P. oceanica* meadow observed during SCUBA diving in the Mediterranean Sea. (b) Sampling sites of *P. oceanica* plants used in this study, located at two depths (-4 mt and -22 mt) in the Mediterranean Sea near Lacco Ameno, Ischia Island, Naples, Italy. Sites are indicated by red markers: -4 mt (40°45'26"N, 13°53'09"E); -22 mt (40°45'25"N, 13°53'25"E). Base maps generated using Google Earth Pro (Google LLC; <https://www.google.it/intl/it/earth/index.html>). Map data and imagery attribution: Map data ©2026 Google; Data SIO, NOAA, U.S. Navy, NGA, GEBCO; Image Landsat / Copernicus; Image ©2026 Airbus. (c) Attenuation of solar irradiance with depth in the water column. Data are extracted from ¹⁷.



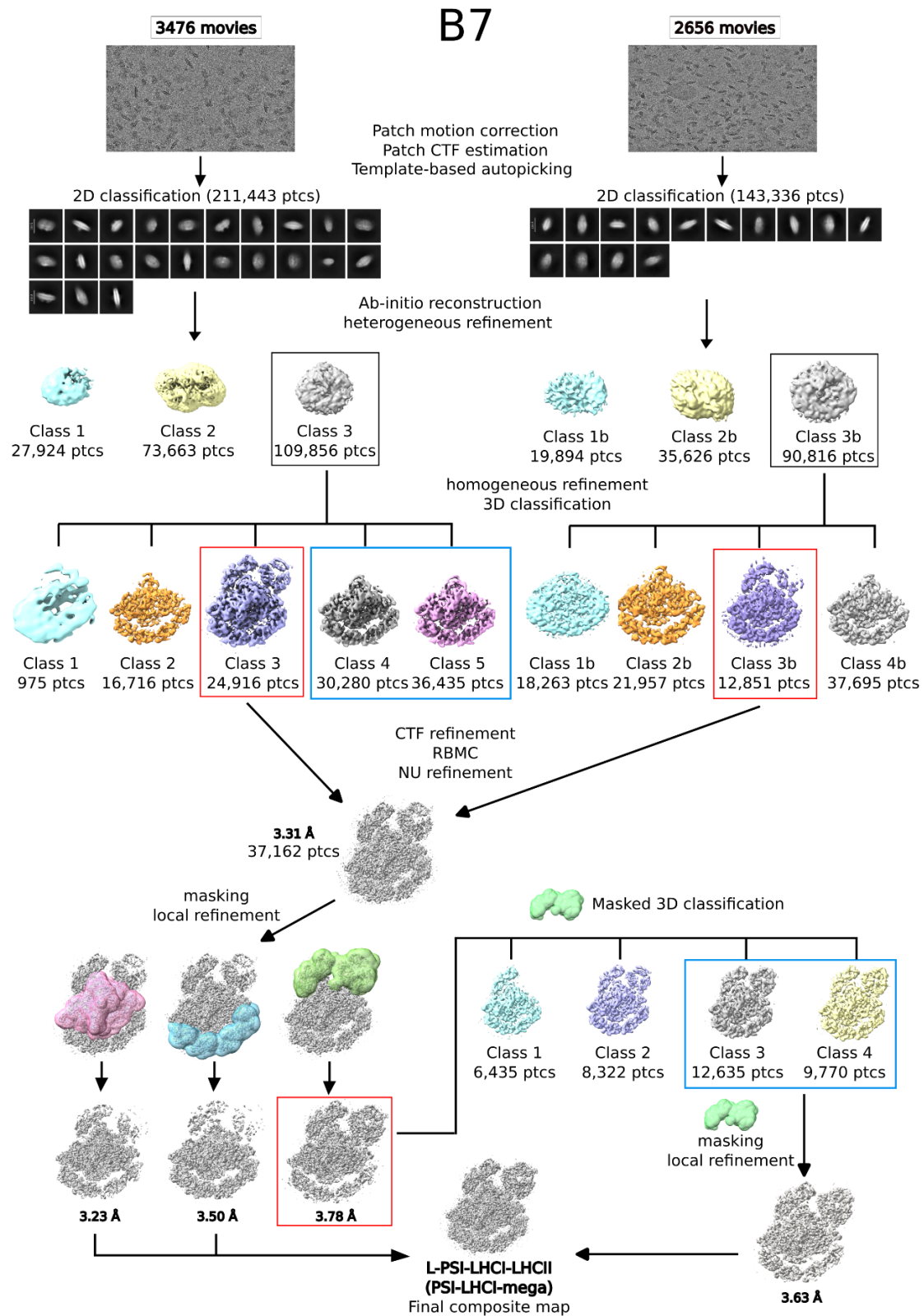
Supplementary Fig. 2. Physiological and biochemical characterization of *P. oceanica* and land plants. (a) NPQ induction and relaxation kinetics measured under actinic light ($\sim 1150 \mu\text{mol photons m}^{-2} \text{s}^{-1}$) followed by dark recovery in *P. oceanica* at 4 and 22 m depth and in *A. thaliana* plants (n=5, 5, and 3 biological replicates, respectively). (b) Maximum quantum yield of PSII (Fv/Fm) of *P. oceanica* at 4 and 22 m depth and *A. thaliana* plants (n=6, 6, and 3 biological replicates, respectively). (c) Large pore Blue Native-PAGE (4-12.5% acrylamide) of thylakoid membranes to assess the organization of Chl-binding proteins. For each lane, 35 μg of thylakoid were solubilized with 1% β -DDM (w/v) at a final concentration. *P. oceanica* thylakoids from plants collected at 4 and 22 mt depth were analyzed, with *P. oceanica* 22 mt samples showing increased abundance of the PSI-LHCI-mega compared to those from 4 mt. The estimated average PSI-LHCI-mega/PSI-LHCI ratio in 22 mt thylakoids is 0.29 ± 0.08 (n=3). Densitometric analysis was performed using ImageJ. (d) Dual-PAM fluorescence

measurements of the state 1-2 transition on *P. oceanica* 4 and 22 mt depth, *A. thaliana* and *Z. mays* plants. Quenching kinetics of PSII fluorescence were extracted from these recordings to calculate qT (Figure 1d). State 1 was established by applying far-red (FR) light in addition to a basal red (R) light, whereas state 2 was triggered by FR removal.

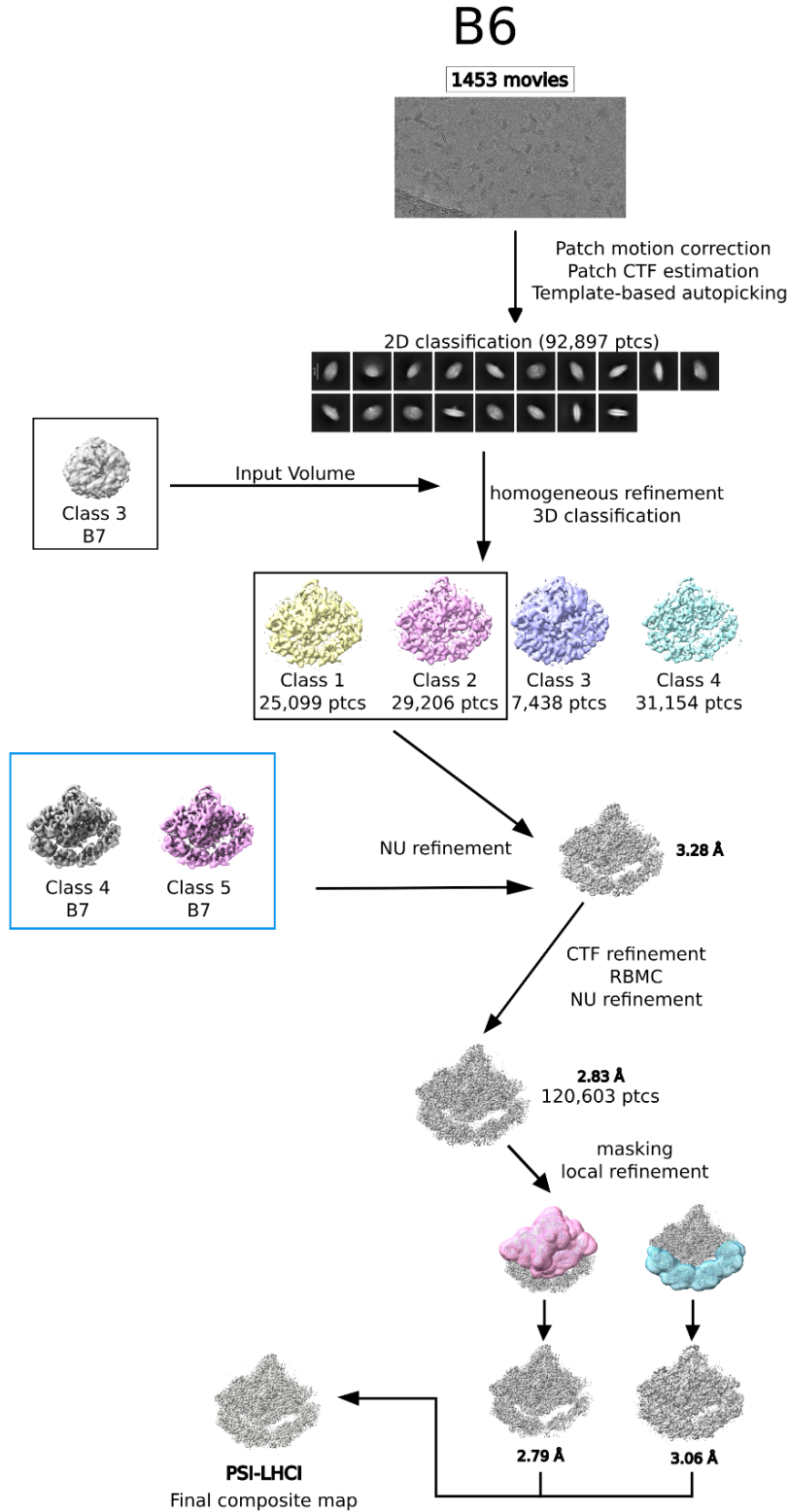


Supplementary Fig. 3. Spectroscopic and biochemical characterization of photosynthetic complexes. Species abbreviations: *Po*, *P. oceanica*; *At*, *A. thaliana*; *Zm*, *Z. mays*. Bands (B3, B6, B7) correspond to those indicated in Figure 1e. **(a)** Room-temperature absorption spectra of *Po* and *At* trimeric LHCII (B3). *Po* LHCII displayed enhanced absorption at 460-483 nm

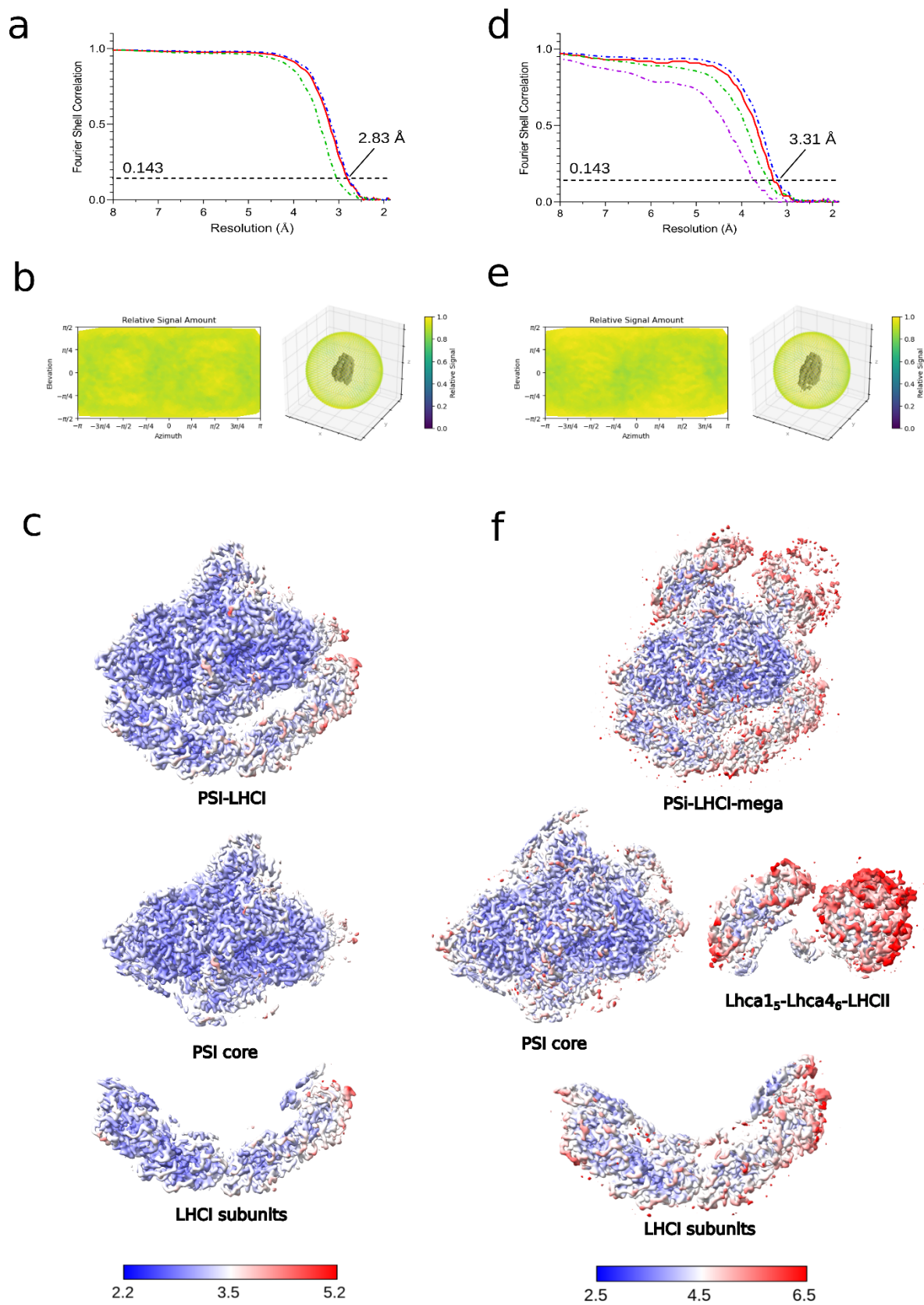
and 650-660 nm, consistent with increased Chl *b* levels compared to *At* LHCII. **(b)** Pigment composition of trimeric LHCII (B3) from *Po* and *At*, determined by spectral fitting of acetone extract absorption spectra (n=6). **(c)** Room-temperature absorption spectra of *Po* PSI-LHCI-mega (B7) and PSI-LHCI supercomplex (B6). The mega-complex exhibited increased absorption at 460-486 nm and 652 nm (highlighted in the difference spectra), indicative of higher Chl *b* content derived from trimeric LHCII and Lhca1₅-4₆ dimer. Spectra were normalized to the Q_y maximum. **(d)** Non-denaturing Deriphat-PAGE (4-12% acrylamide) of *Po* thylakoid (thy) membranes (22 mt depth) and bands from Figure 1e to assess Chl-binding protein composition. Thylakoids were solubilized with a final 1% β -DDM (w/v). **(e)** Coomassie-staining SDS-PAGE of thylakoid membranes (22 mt depth), *Po* PSI-LHCI (B6), and PSI-LHCI-mega (B7). The mega-complex contained an additional band corresponding to LHCII. **(f)** Immunoblot analysis of protein subunits from *Po* thylakoids (thy) membranes (22 mt depth), trimeric LHCII (B3), PSI-LHCI (B6), and PSI-LHCI-mega (B7). Detection of phosphorylated (P) Lhcb2 was performed using the α -Lhcb2-P antibody. **(g)** HPLC pigment quantification of *Po* PSI-LHCI-mega (B7) and *At*, *Zm*, *Po* PSI-LHCI (B6) complexes, (n=3). Chl *a*, Chlorophyll *a*; Chl *b*, Chlorophyll *b*; Vio, violaxanthin; Lut, lutein; Neo, neoxanthin; β -car, β -carotene.



Supplementary Fig. 4. Cryo-EM data processing workflow of *P. oceanica* PSI-LHCI-mega complex. B7 corresponds to the sucrose gradient band shown in Figure 1e (see Methods for details). Abbreviations: CTF: Contrast Transfer Function; ptes: particles; RBMC: Reference Based Motion Correction; Non-Uniform (NU) refinement.

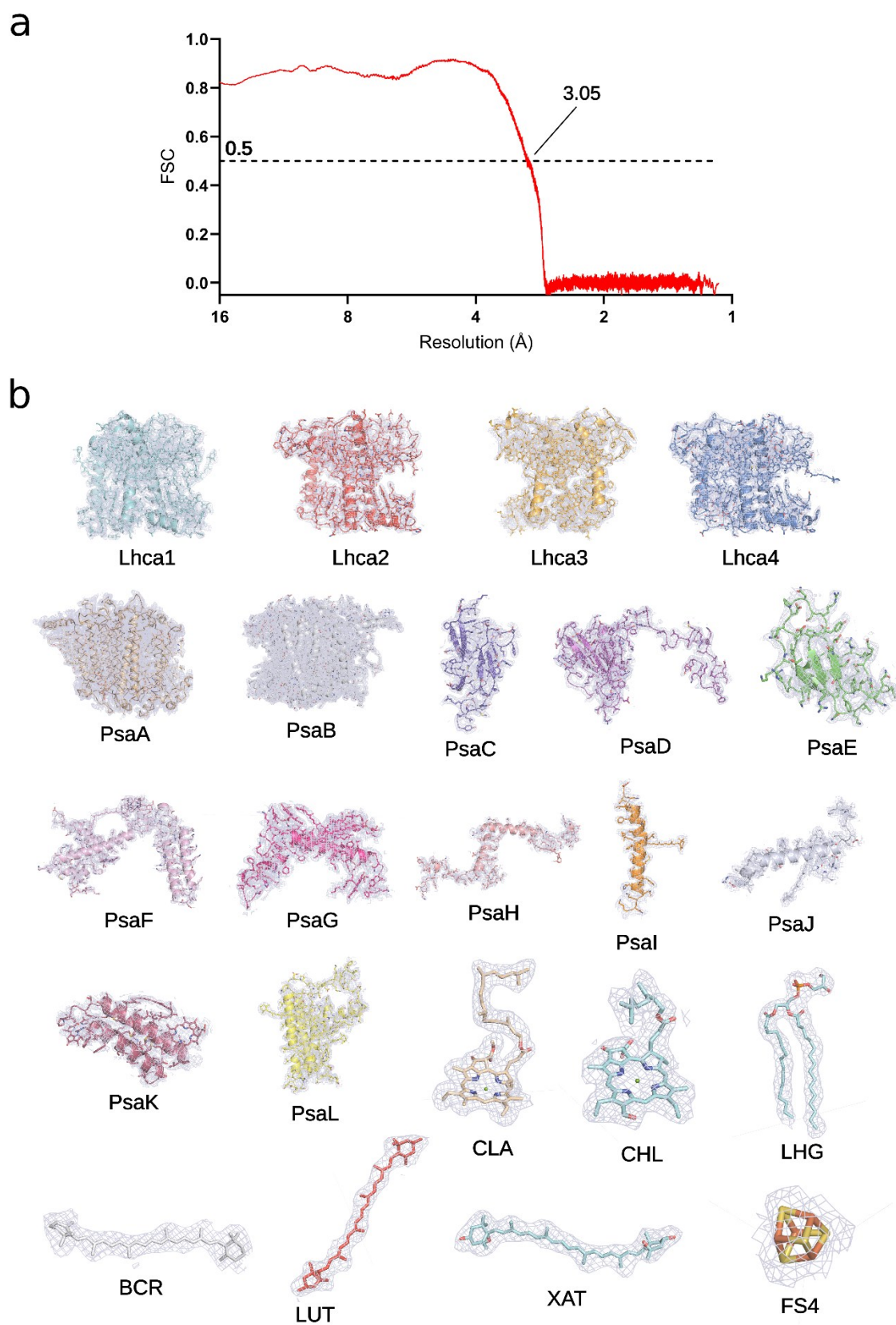


Supplementary Fig. 5. Cryo-EM data processing workflow of *P. oceanica* PSI-LHCI complex. B6 corresponds to the sucrose gradient band shown in Figure 1e. Abbreviations: CTF: Contrast Transfer Function; ptcs: particles; RBMC: Reference Based Motion Correction; Non-Uniform (NU) refinement.



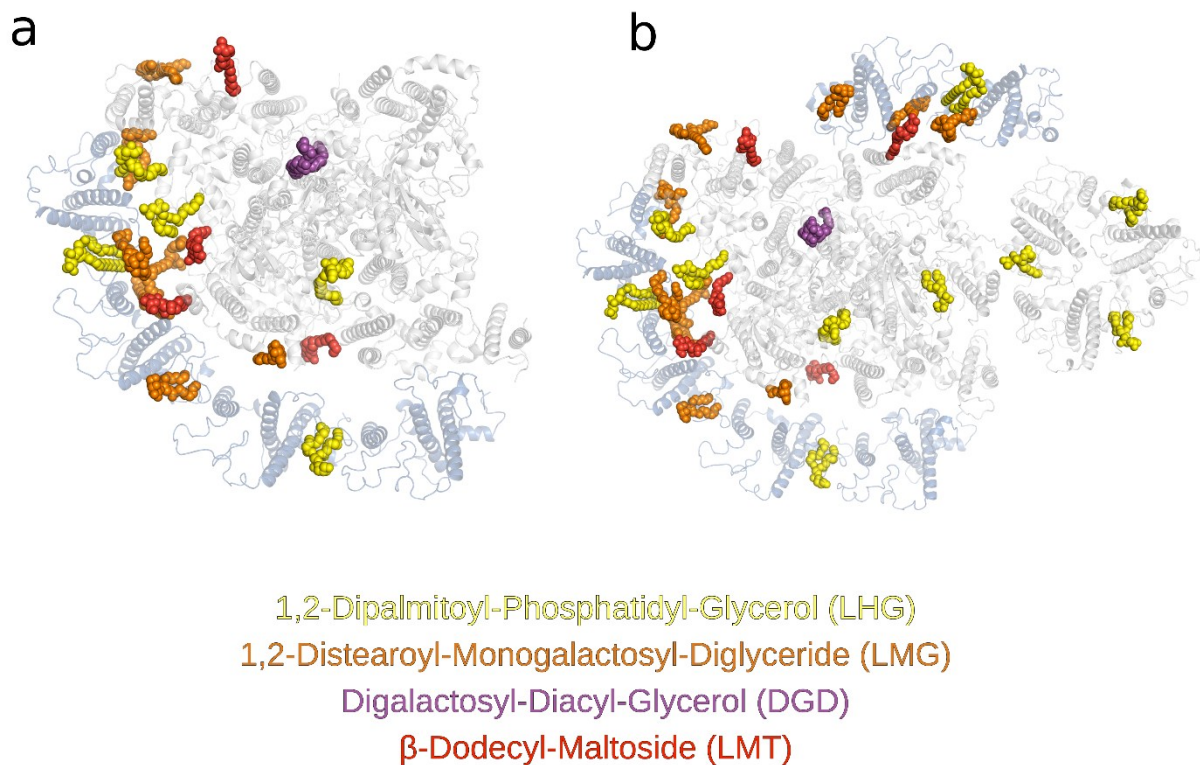
Supplementary Fig. 6. Cryo-EM structure determination of *P. oceanica* PSI-LHCI and PSI-LHCI mega-complexes. The Gold Standard Fourier Shell Correlation (GSFSC) curves

of the PSI-LHCI (continuous red line, consensus map; dashed blue line, focused map PSI core; dashed green line, focused map LHCI subunits) (**a**) and PSI-LHCI-mega (continuous red line, consensus map; dashed blue line, focused map PSI core; dashed green line, focused map LHCI subunits; dashed magenta line, Lhca1₅-Lhca4₆-LHCII) (**d**) consensus maps with a resolution-cutoff threshold at 0.143. Relative Signal Amount vs. Viewing Direction plots for PSI-LHCI (**b**) and PSI-LHCI-mega (**e**). Local resolution estimation in the Cryo-EM maps of *P. oceanica* PSI-LHCI (**c**) and PSI-LHCI-mega (**f**) supercomplexes.

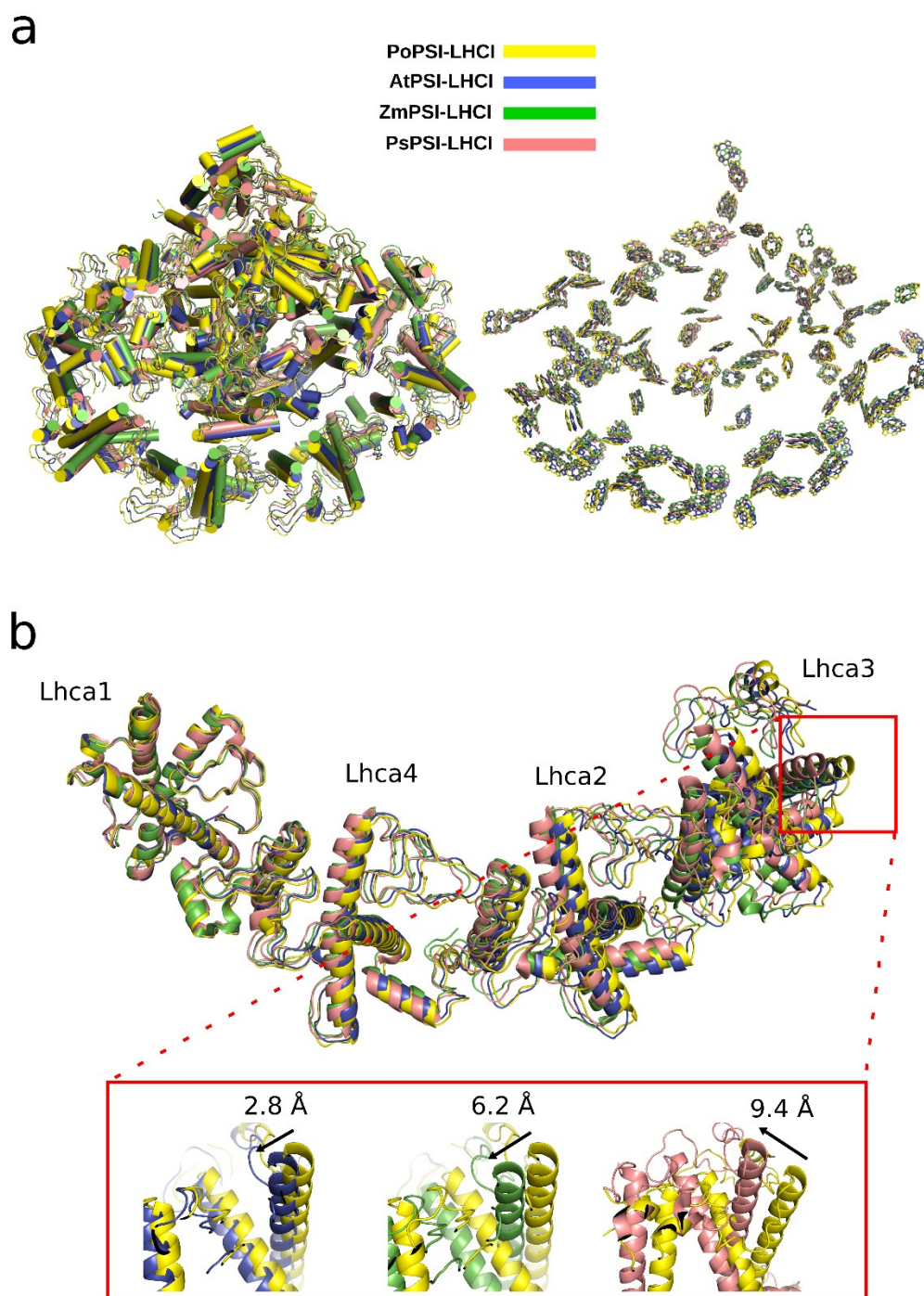


Supplementary Fig. 7. Cryo-EM density maps of protein chains and representative cofactors of *P. oceanica* PSI-LHCI. (a) The Fourier shell correlation (FSC) curve between

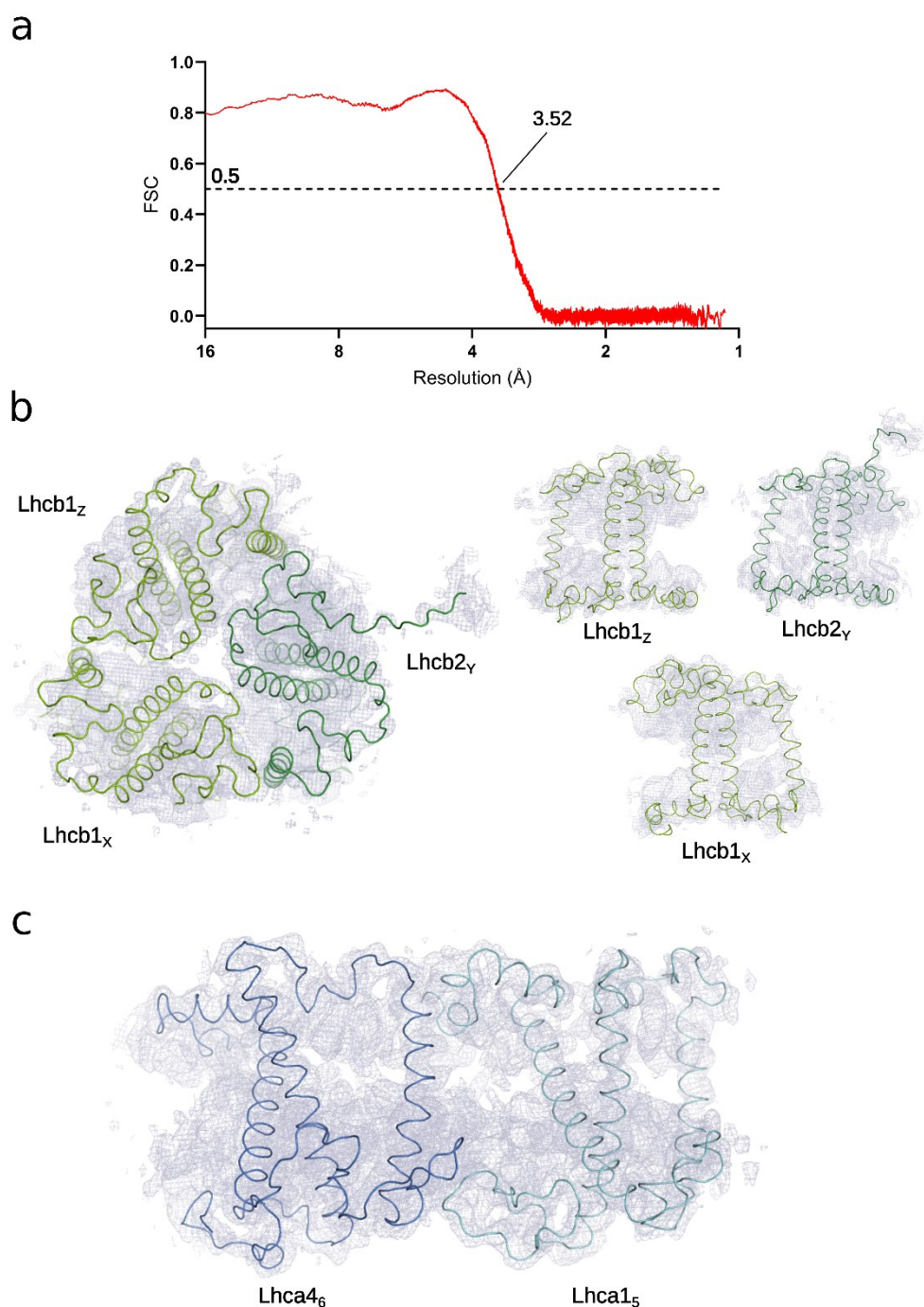
the model and the composite Cryo-EM density map of PSI-LHCI. **(b)** LHCI and PSI core subunits (shown in cartoon) and representative ligands (stick models) surrounded by the Cryo-EM density maps. CLA, Chlorophyll *a*; CHL, Chlorophyll *b*; BCR, β -carotene; LUT, Lutein; XAT, Violaxanthin; LHG, 1,2-dipalmitoyl-phosphatidyl-glycerole; FS4, Fe₂S₂ iron-sulfur cluster.



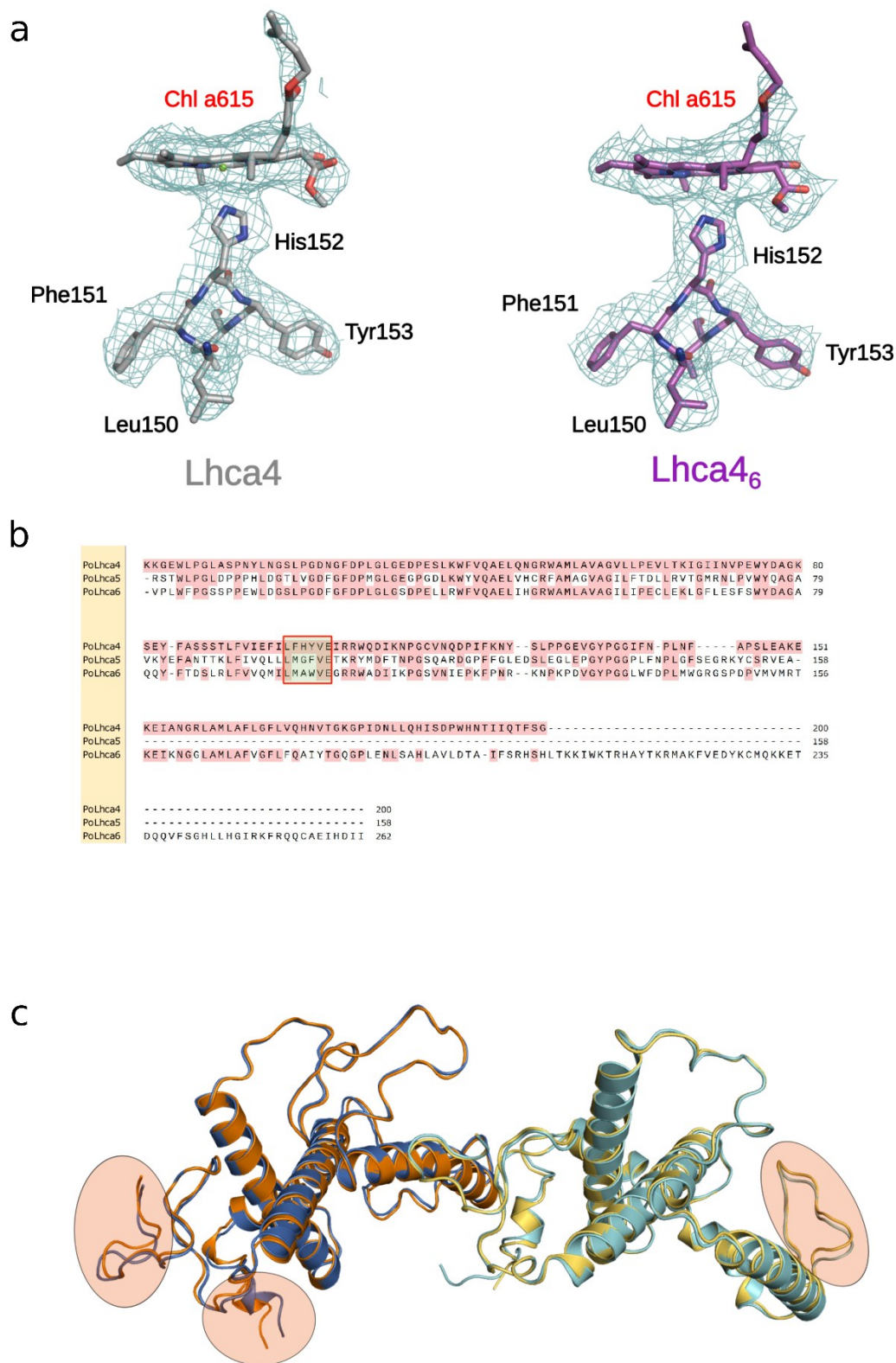
Supplementary Fig. 8. Lipid and detergent molecules found in the structures of *P. oceanica* PSI-LHCI supercomplexes. The molecules identified in the structures of PSI-LHCI **(a)** and PSI-LHCI-mega **(b)** are depicted as sphere models in different colors.



Supplementary Fig. 9. Structural comparison of *P. oceanica* PSI-LHCI with other higher land plant PSI-LHCI. Species abbreviations: *Po*, *P. oceanica*; *At*, *A. thaliana*; *Zm*, *Z. mays*. **(a)** Global Ca superimposition of *Po* PSI-LHCI (yellow, PDB 9S1L) with that of *At* (blue, PDB 9GBI), *Zm* (green, PDB 5ZJI), and *Ps* (salmon, PDB 7DKZ). The location of the Chl rings is shown on the right panel. **(b)** The LHCI belt of the same complexes aligned on the Ca position of Lhca1. The shift in the position of the third transmembrane helix relative to *Po* PSI-LHCI in the three structures is shown in the red box.

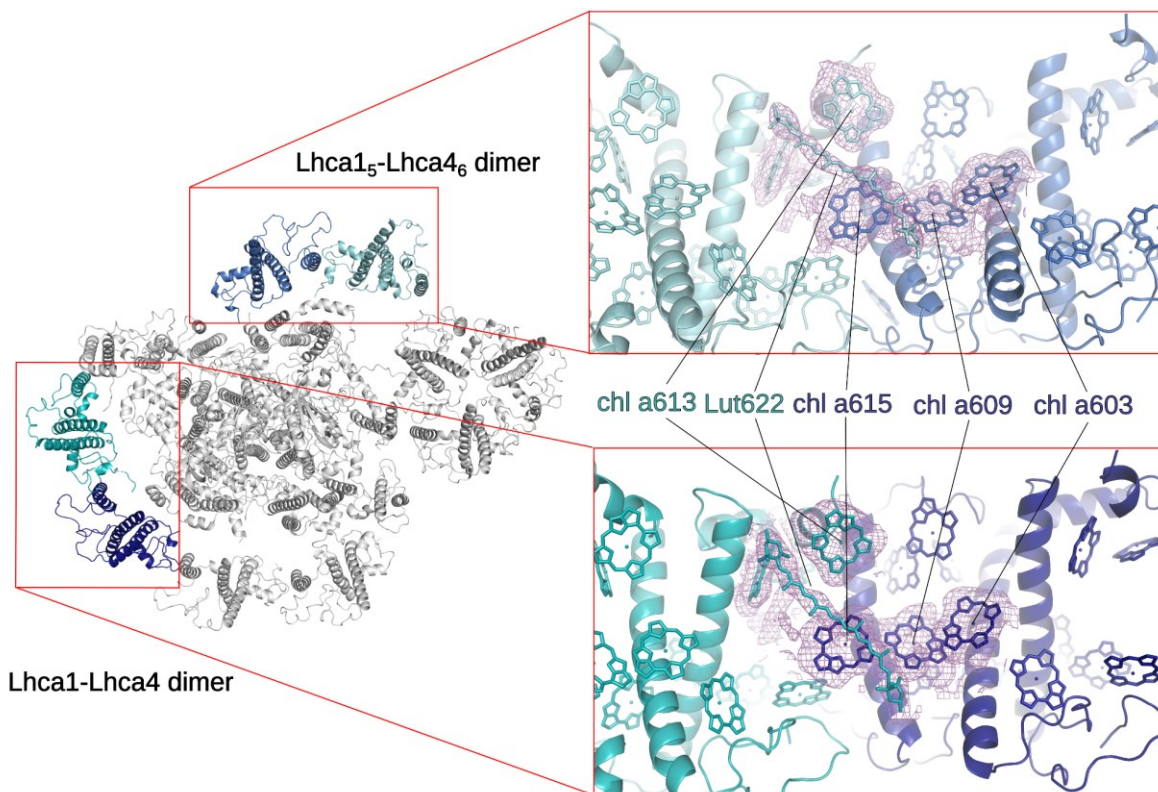


Supplementary Fig. 10. Density maps of the additional LHC subunits in *P. oceanica* PSI-LHCI-mega complex. (a) The Fourier shell correlation (FSC) curve between the model and the composite Cryo-EM density map of PSI-LHCI-mega. (b) Cryo-EM density (contour level 3.5σ) of the LHCII trimer (left) and of the single Lhcb subunits (right). (c) Cryo-EM density (contour level 3.5σ) of the Lhca1₅-Lhca4₆ dimer. The subscripts indicate the name of the chain in the PDB file.

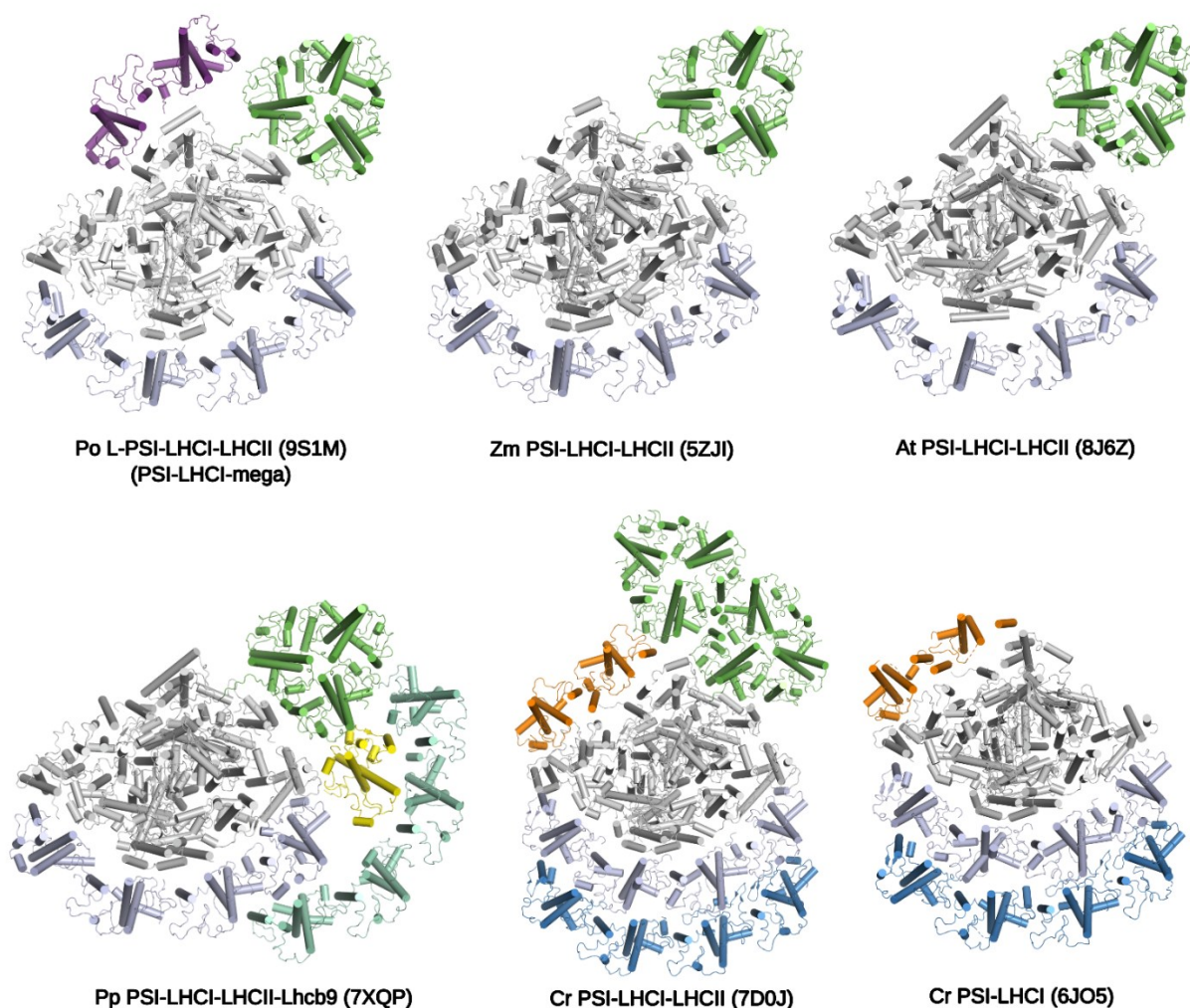


Supplementary Fig. 11. Comparison of the two Lhca1-Lhca4 dimers of PSI-LHCI-mega.
(a) Cryo-EM density maps (3.5σ contour level) surrounding the Leu150-Tyr153 and Chl a615

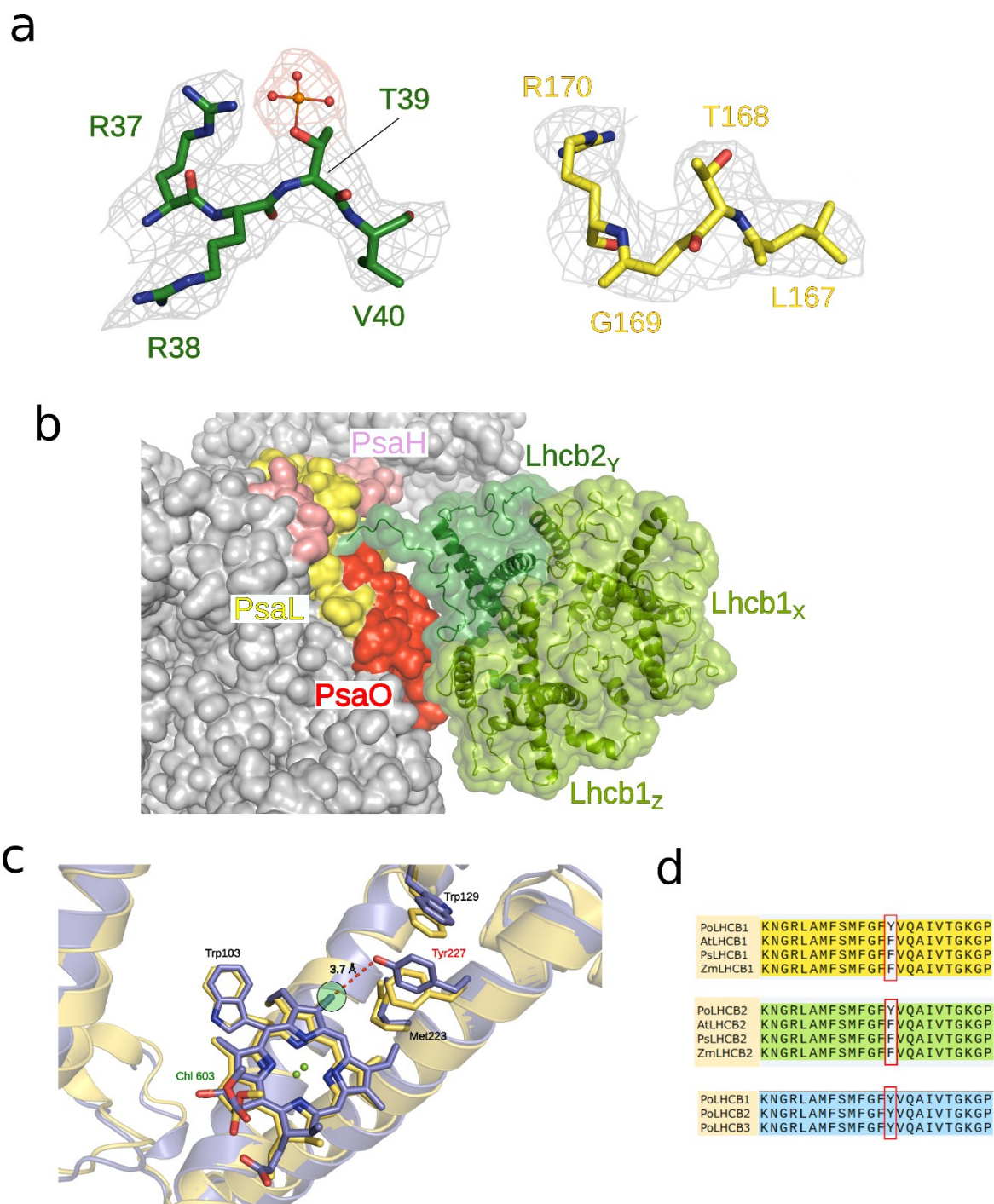
residues in Lhca4 and Lhca4₁. (b) Sequence alignment of Lhca4 with the other two Lhca isoforms (Lhca5 and Lhca6) identified in the *P. oceanica* genome. The red box indicates the stretch of residues shown above. The different residues and the absence of the axial Chl ligand at these positions exclude the presence of alternative isoforms in the additional Lhca dimer of PSI-LHCI-mega. (c) Structural alignment of the two Lhca1-Lhca4 dimers (C α RMSD = 0.57 Å). The greater structural diversity (highlighted by the ovals) between the two dimers can be observed in the regions where they establish contacts with different subunits within the supercomplex.



Supplementary Fig. 12. Comparison of chromophores in the two Lhca1-Lhca4 dimers of *P. oceanica* PSI-LHCI-mega complex. The chromophores located near the Lhca1-Lhca4 interface (Chl a613₁, a603₄, a609₄, a615₄, and the Lut622₁) are shown surrounded by their Cryo-EM density maps (3.5 σ contour level) in the Lhca1-Lhca4 and Lhca1₅-Lhca4₆ dimers.

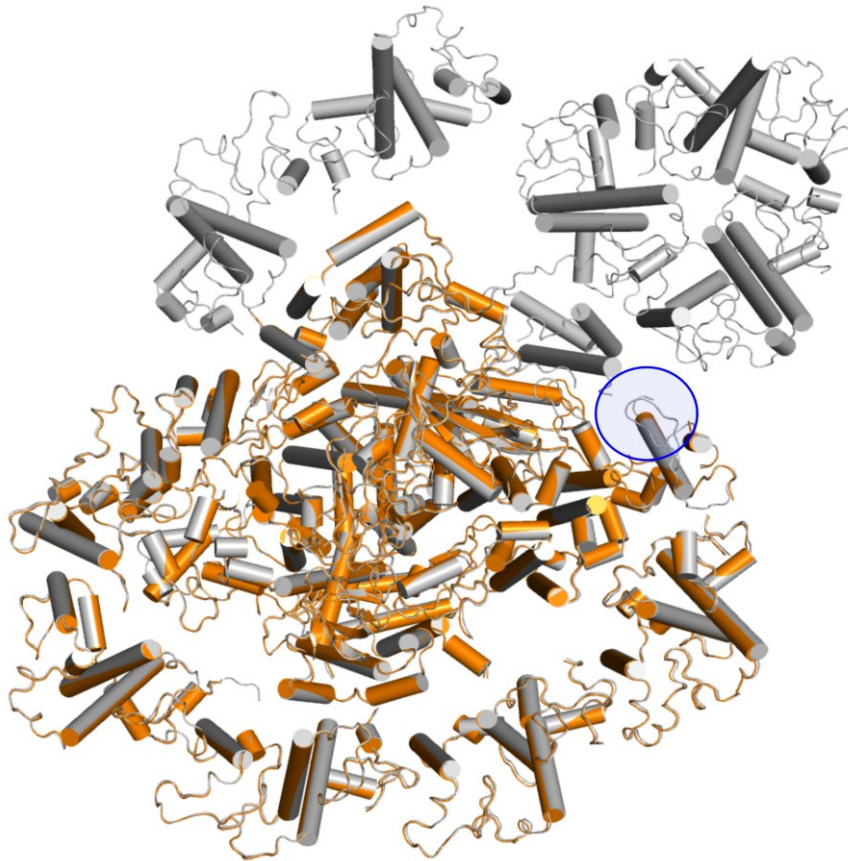


Supplementary Fig. 13. Structural models of PSI-LHCI-LHCII supercomplexes from different species. Cartoon models of different PSI-LHCI-LHCII supercomplexes. The PSI core subunits are shown in gray, the first LHCI belt in lavender, the second LHCI belt in blue or light blue, and the LHCII trimer in green. The additional LHCI dimer is depicted in violet for *P. oceanica* and in orange for *C. reinhardtii*. The Lhcb9 subunit of *P. patens* is shown in yellow. PDB codes of the different structures are given in parentheses. Species abbreviations: *Po*, *P. oceanica*; *Zm*, *Z. mays*; *At*, *A. thaliana*; *Pp*, *P. patens*; *Cr*, *C. reinhardtii*.

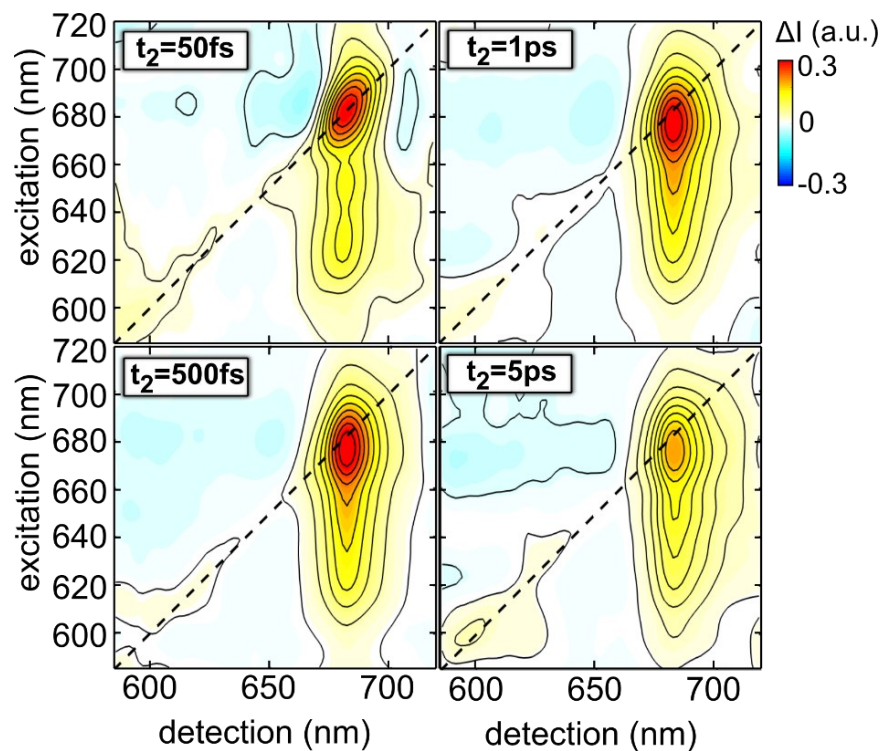


Supplementary Fig. 14. Structure and interaction of the LHCII trimer with the PSI core in *P. oceanica* PSI-LHCI mega-complex. (a) The phosphorylated RRpTV motif of Lhca2 γ (green) surrounded by the Cryo-EM density map contoured at the 3.5 σ level. The extra density of the phosphate group (ball-and-stick model) is shown in red. The density map (with the same local resolution) of a non-phosphorylated Thr (T168 of chain L, in yellow) is shown for comparison. (b) Surface model of the core subunits PsaH (pink), PsaL (yellow), and PsaO (red) in contact with the LHCII trimer (light green, Lhcb1; dark green Lhcb2). The cartoon model of LHCII is visible in transparency. All the other subunits are shown in gray. (c)

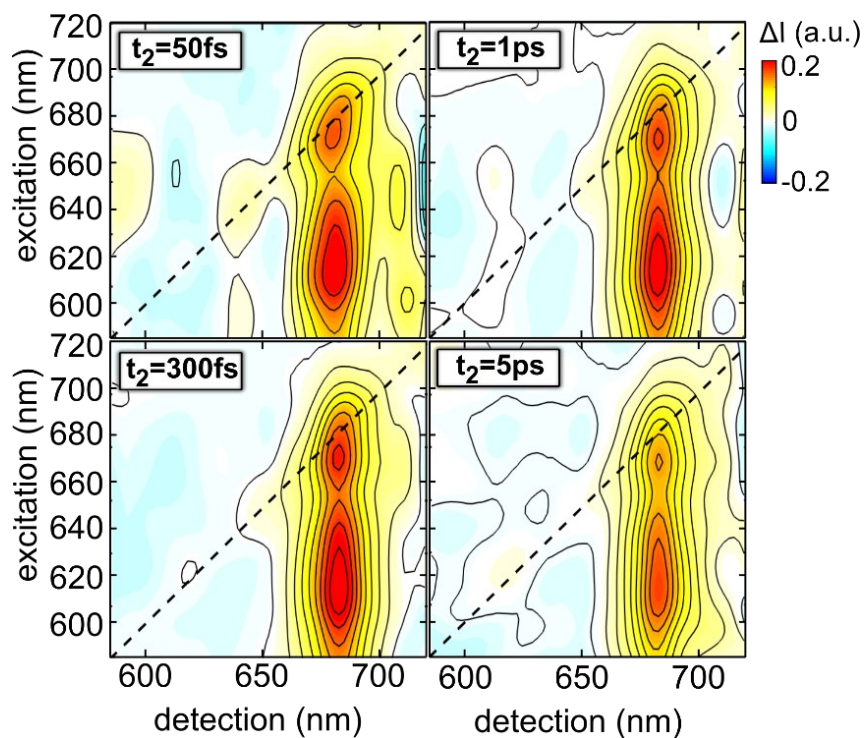
Superposition of Chl 603 binding site in Lhcb1 of *P. oceanica* (lavender) and *Z. mays* (gold).
(d) The amino acid sequence alignments of *P. oceanica* Lhcb1-2 with the land plant orthologs (*At*: *Arabidopsis thaliana*; *Ps*: *Pisum sativum*; *Zm*: *Zea mays*) and the Lhcb1-3 isoforms of *P. oceanica*.



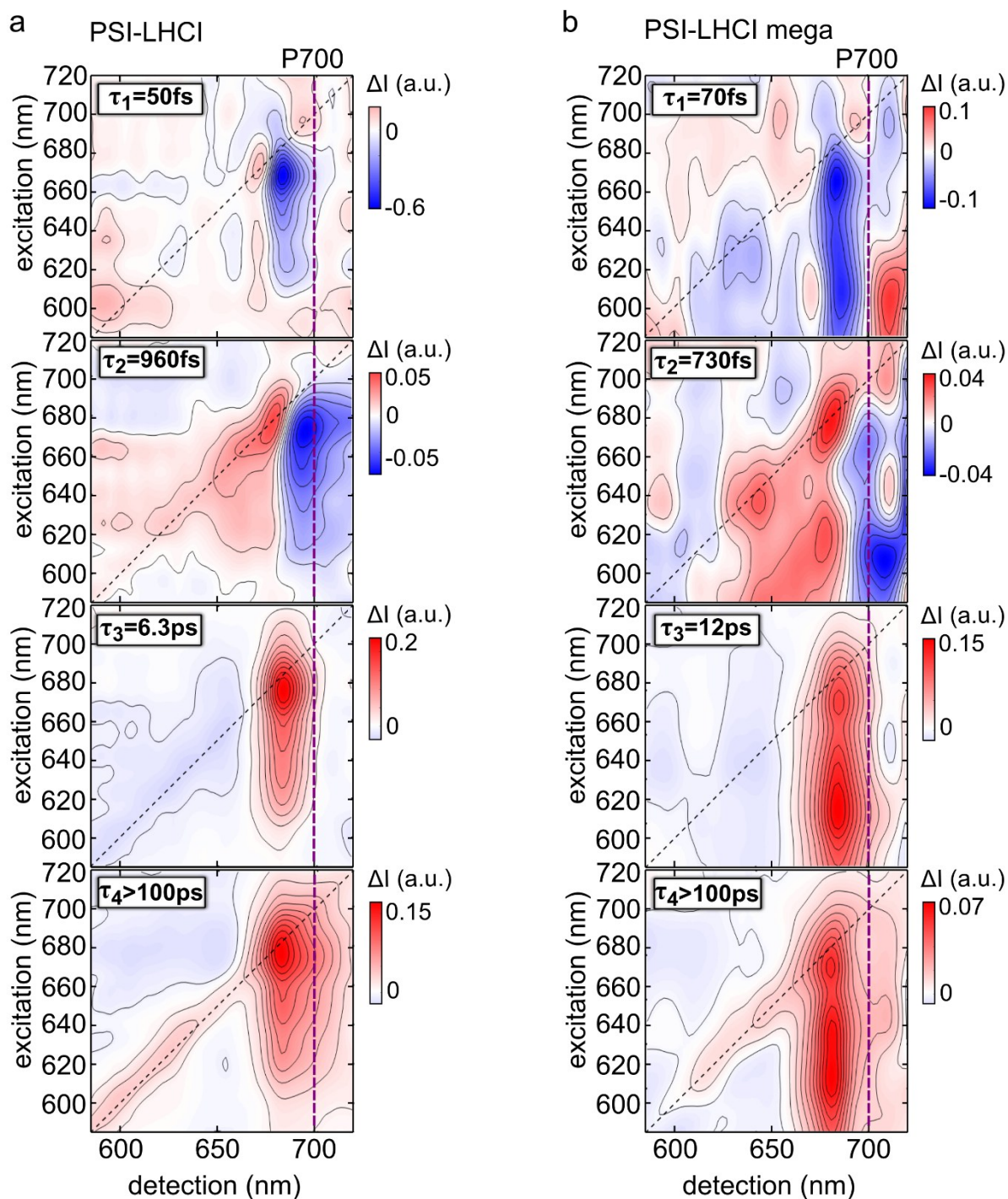
Supplementary Fig. 15. Structural comparison of *P. oceanica* PSI-LHCI and PSI-LHCI-mega complexes. Superimposition of the structures of PSI-LHCI (orange) and PSI-LHCI-mega (grey). The position of the flexible loop of PsaK (residues 71-101) is highlighted by the blue circle.



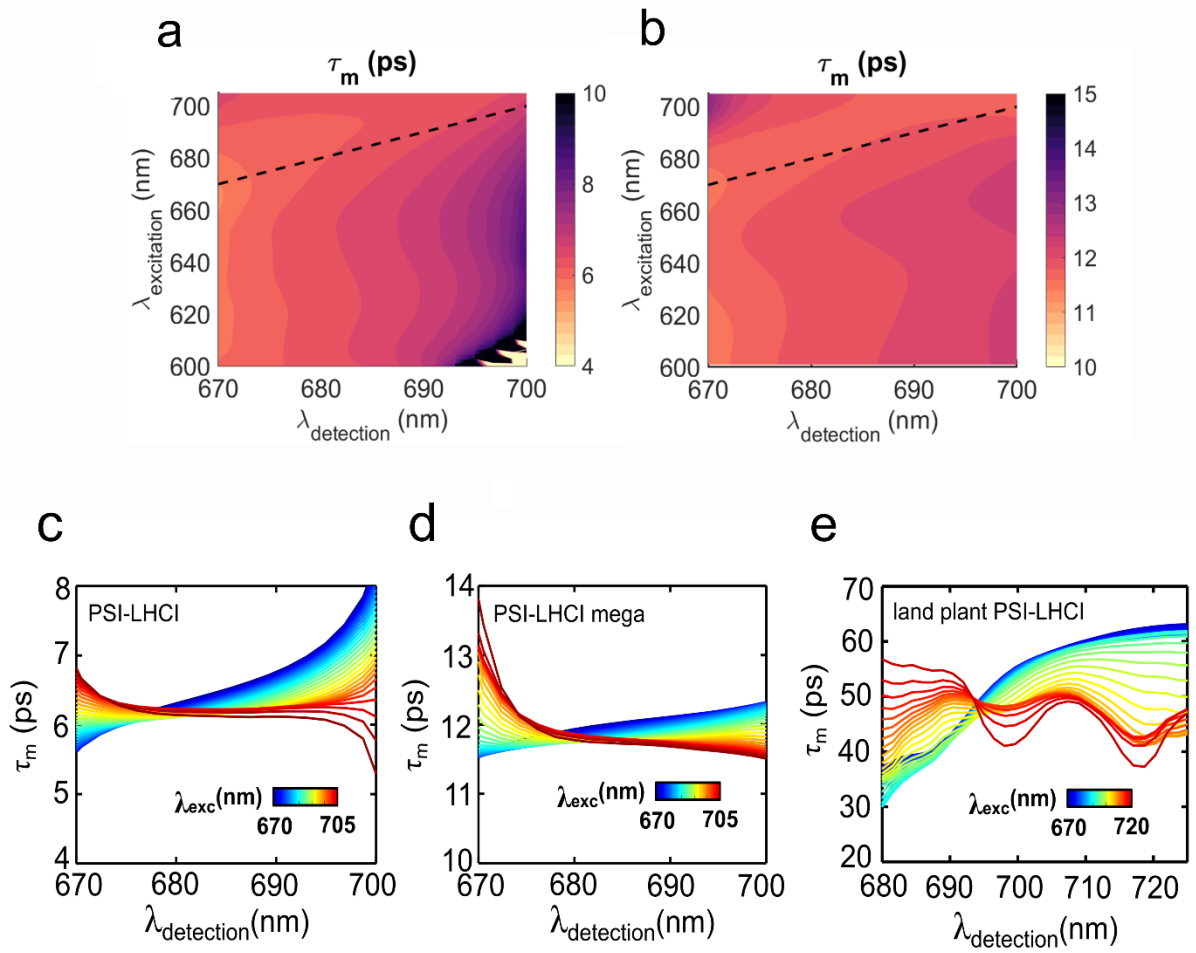
Supplementary Fig. 17. 2DES results of *P. oceanica* PSI-LHCI. 2DES maps at four different waiting times: $t_2=50$ fs, 500 fs, 1 ps, and 5 ps.



Supplementary Fig. 18. 2DES results of *P. oceanica* PSI-LHCI-mega. 2DES maps at four different waiting times: $t_2=50$ fs, 300 fs, 1 ps, and 5 ps.

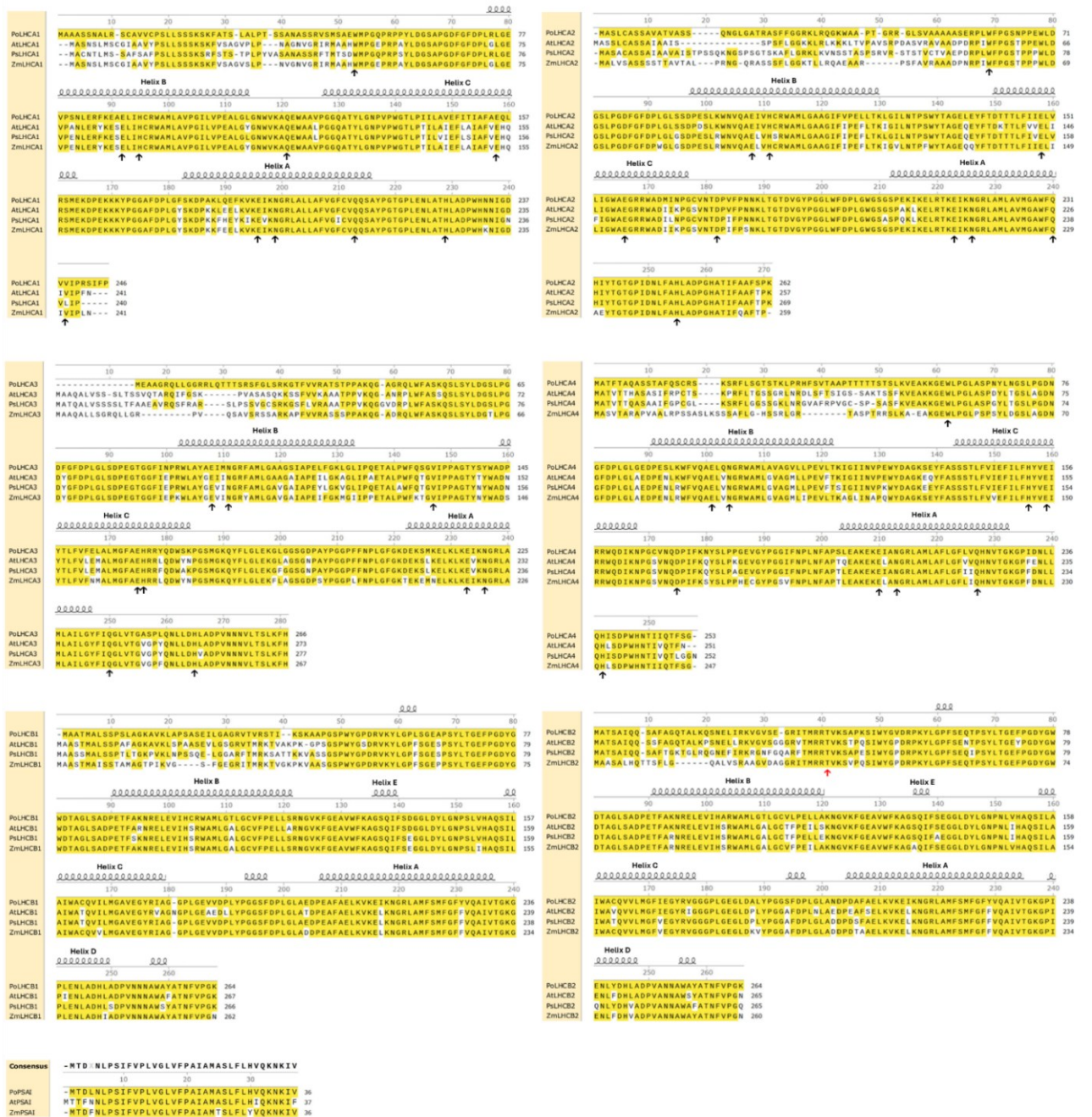


Supplementary Fig. 19. 2DES global analysis results of *P. oceanica* complexes. (a) 2D-DAS maps of PSI-LHCI; the temporal evolution can be described with four time constants: $\tau_1 = 50 \text{ fs}$, $\tau_2 = 960 \text{ fs}$, $\tau_3 = 6.3 \text{ ps}$, and a non-decaying component (limited by the temporal window adopted in the experiment). (b) 2D-DAS maps of PSI-LHCI-mega; the temporal evolution can be described with four time constants: $\tau_1 = 70 \text{ fs}$, $\tau_2 = 730 \text{ fs}$, $\tau_3 = 12 \text{ ps}$, and a non-decaying component (limited by the temporal window adopted in the experiment). The dashed violet line indicates the absorption of the P700.

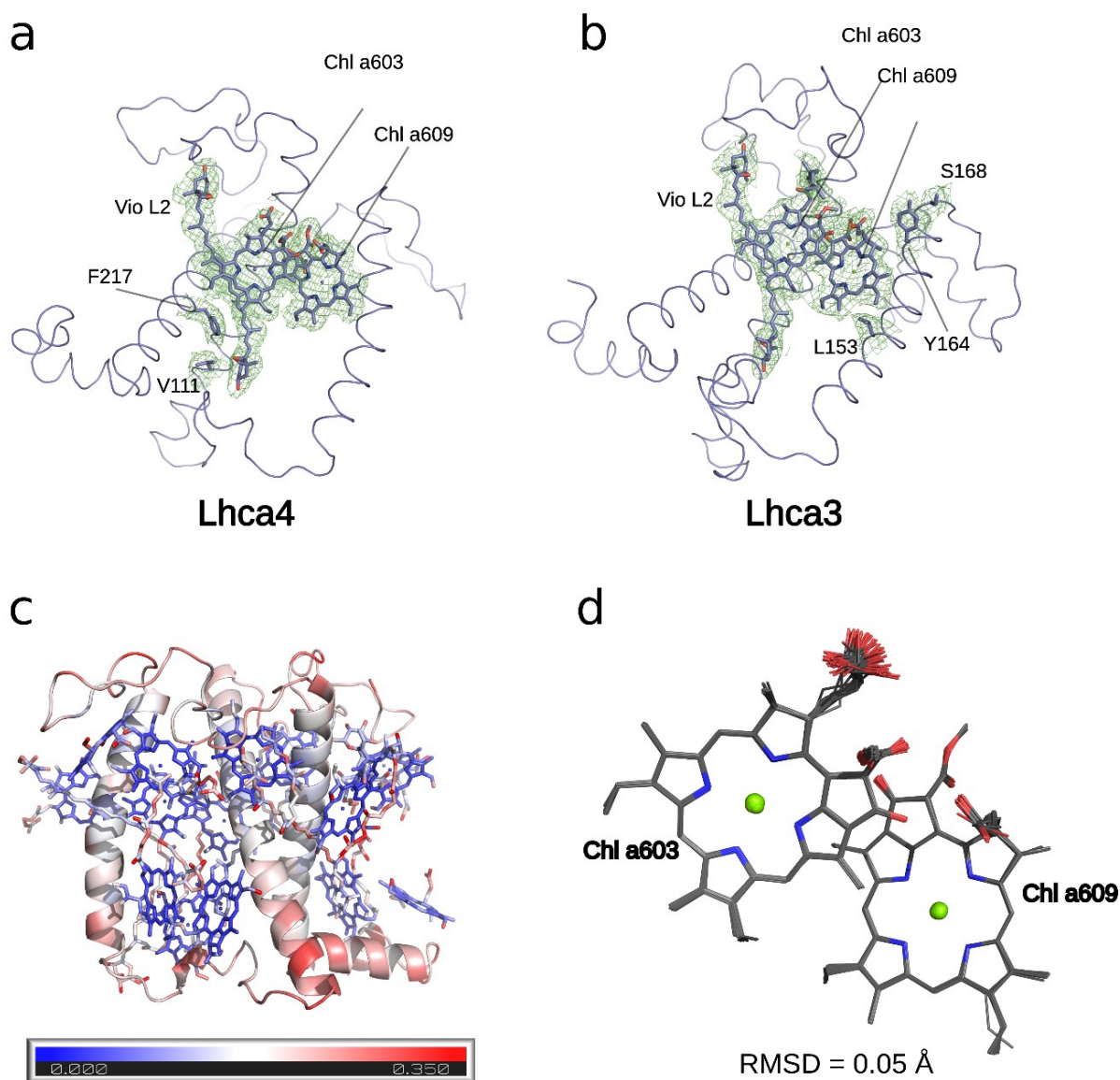


Supplementary Fig. 20. Mean decay lifetime (τ_m) from 2DES analysis. Mean decay lifetime maps of the PSI-LHCI (a) and PSI-LHCI-mega (b) from *P. oceanica*. (c-d) Horizontal slice of the mean decay lifetime maps obtained from a and b, and from land plant PSI-LHCI (e) obtained from ¹⁶. The cuts are obtained by exciting in the range from 670 nm (blue) to 705 nm (red) for the *P. oceanica* supercomplexes and from 670 nm to 720 nm for the land plant PSI-LHCI. See Supplementary Methods for details.

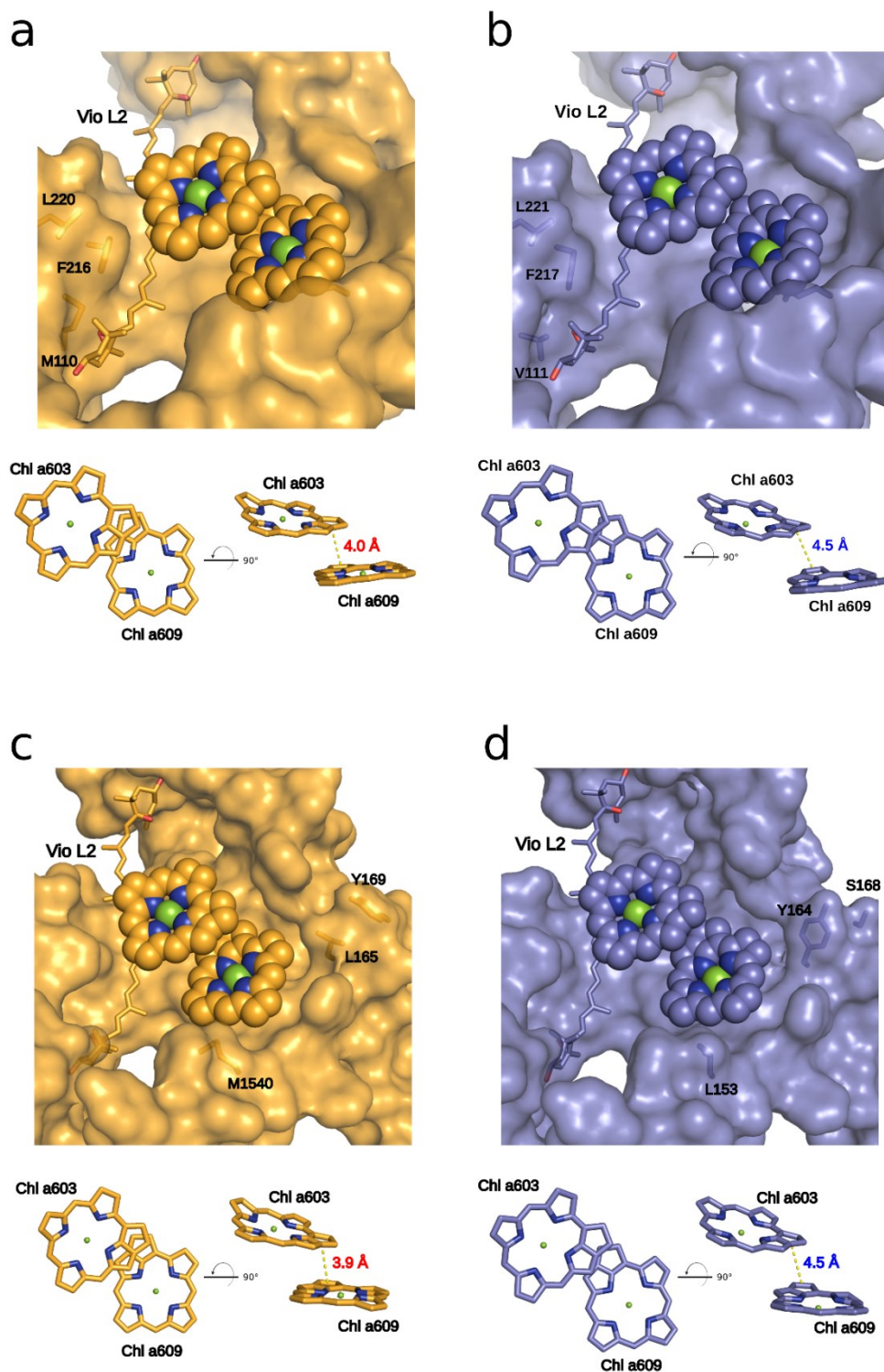
In *P. oceanica* supercomplexes, we observe similar trends, where the mean decay increases with increasing wavelength, as already observed in the photosystems with low-energy Chl forms. The peculiar aspect that differentiates the *P. oceanica* supercomplexes with respect to the land plants PSI-LHCI is how the value of τ_m changed as a function of the detection wavelength. The horizontal cuts of the maps (c-e) emphasized this aspect. In particular, by exciting around 670/690 nm (absorption of Chls bulk), the mean decay slowly increases upon increasing the detection wavelength. Specifically, the mean decay increases by 35% for *Po*PSI-LHCI and by 6% for the PSI-LHCI-mega, showing a weak wavelength dependence. Differently, in the land plant PSI-LHCI, the mean decay increases by almost 77%, indicating a preferential equilibration path between bulk Chls and low-energy Chl forms, instead of scattering to the PSI core. This demonstrates the energy-trapping limitations imposed by the presence of low-energy Chl forms.



Supplementary Fig. 21. Sequence alignments of LHCs and PSAI proteins. The amino acid sequences of LHCA1, LHCA2, LHCA3, LHCA4, LHCB1, LHCB2, and PSAI from *P. oceanica* (*Po*), *A. thaliana* (*At*), *P. sativum* (*Po*), and *Z. mays* (*Zm*) were aligned with Clustal Omega (<https://www.ebi.ac.uk/jdispatcher/msa/clustalo>). Chl-binding residues of LHCI subunits are indicated with a black arrow, while the phosphorylated Threonine in LHCB2 is indicated with a red arrow.



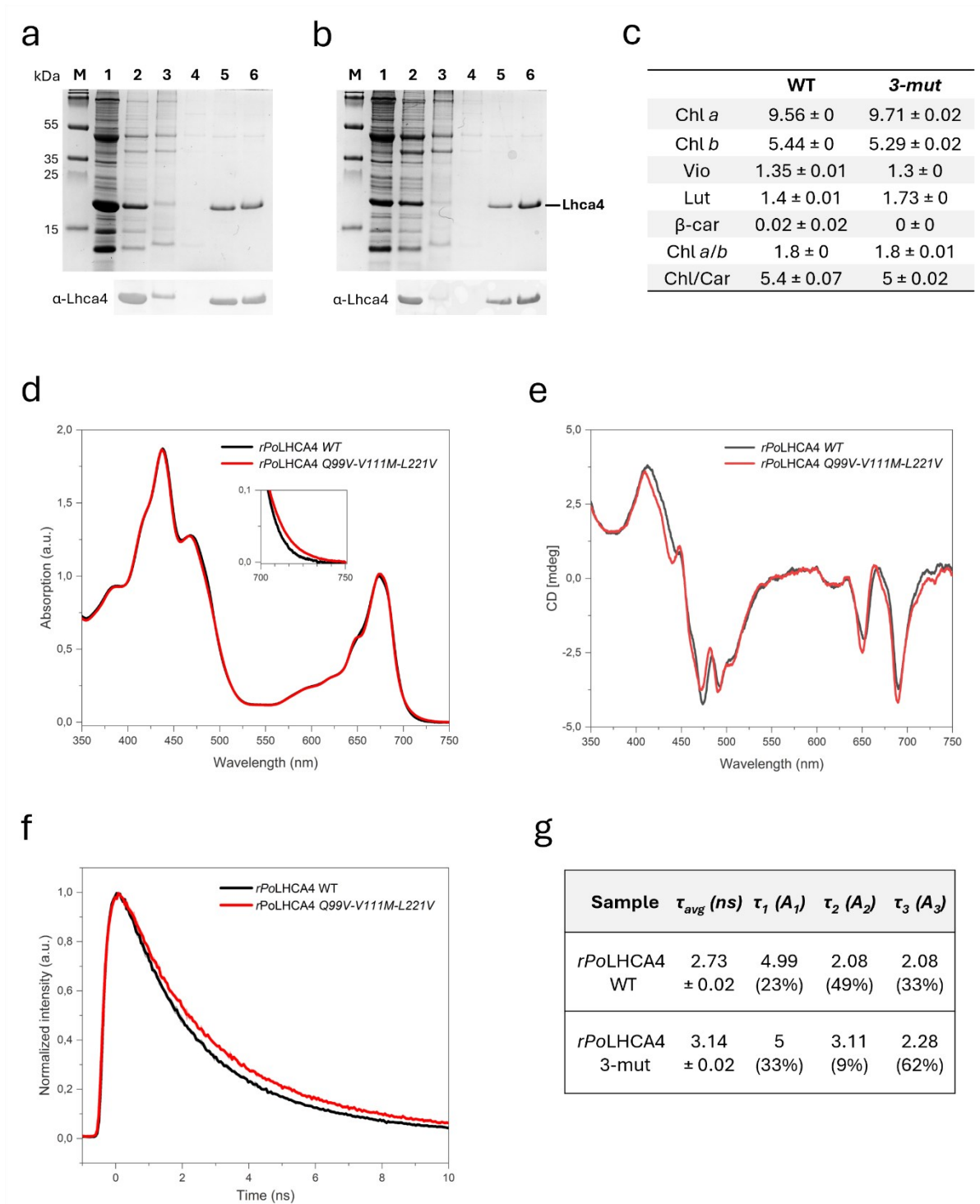
Supplementary Fig. 22. Structure of the chlorophyll *a*603-*a*609 clusters in *P. oceanica* Lhca3 and Lhca4. The pigments (chlorophyll (Chl) *a*603, *a*609, and Violaxanthin (Vio) L2) and relevant side chains in Lhca4 (a) and Lhca3 (b) are shown in sticks and are surrounded by the density maps contoured at 3.5 σ level. The local resolution of the map around the *a*603-*a*609 clusters is ~2.84 Å for Lhca4 and ~3.26 Å for Lhca3. (c) The average model of Lhcb4 obtained from 100 independent runs of random coordinate perturbation with phenix.pdbtools (RMSD 0.5 Å) and refinement with Servalcat¹⁸. The model is color-coded according to the root mean square fluctuation (RMSF), ranging from 0 (blue) to 0.35 Å (red). (d) The 100 refined models of Chl *a*603 and *a*609 displayed with approximately the same orientation as in Supplementary Figure 23. The mean RMSD of the positions of the atoms in the chlorin rings is 0.05 Å.



Supplementary Fig. 23. Conformation of the low-energy Chl form cluster in *P. oceanica* and *Z. mays*. Chlorine rings of the Chls a603 and a609 in *Z. mays* (orange) and *P. oceanica* (lavender) Lhca4 (**a-b**) and Lhca3 (**c-d**) are shown as spheres with van der Waals radius, Violaxanthin L2 and relevant side chains in sticks, and the protein surface in transparency. The relative positions and orientations of the two Chl rings in the different structures are shown in the lower panels, along with the distance between the E-ring centers of the two Chls.

PSI-LHCI	<i>A. thaliana</i> WT (PDB: 9GBI)	<i>Z. mays</i> (PDB: 5ZJI)	<i>P. oceanica</i> (PDB: 9SIL)	<i>A. thaliana</i> <i>a603-NH</i> (PDB: 9GC2)
Lhca3	4.1 Å	3.9 Å	4.5 Å	4.5 Å
Lhca4	4.3 Å	4.0 Å	4.5 Å	4.8 Å
77K emission peaks	732 nm	736 nm	722 nm	721 nm

Supplementary Fig. 24. Correlation between the E-ring overlap distance of Chls *a603-a609* in Lhca3 and Lhca4 from PSI-LHCI Cryo-EM structures, with their 77K fluorescence emission peaks. Distances (in Ångstroms, Å) were measured using PyMOL, as described for Supplementary Figure 23. 77K fluorescence emission data for PSI-LHCI are from Figure 1f, with the maximum emission peak reported in nanometers (nm).



Supplementary Fig. 25. Spectroscopic and biochemical characterization of *P. oceanica* (*Po*) recombinant (r) LHCA4 WT and Q99V-V111M-L221V mutant. Recombinant proteins carry an N-terminal His-tag. (a) *rPoLHCA4* WT and (b) *rPoLHCA4* Q99V-V111M-L221V mutant, Coomassie-staining SDS-PAGE (top) and immunoblot (bottom) of fractions collected during protein purification. Lane M: molecular weight marker; 1: inclusion bodies (IB) solubilized with 2% SDS; 2: IB solubilized with 2% LDS; 3: IMAC flow-through; 4:

IMAC wash; 5: IMAC eluate; 6: sucrose gradient band. (c) HPLC pigment quantification of *rPoLHCA4* WT and Q99V-V111M-L221V mutant (3-mut), (n=3). Chl *a*, Chlorophyll *a*; Chl *b*, Chlorophyll *b*; Vio, Violaxanthin; Lut, Lutein; β -car, β -carotene. (d) Room-temperature absorption spectra normalized to the 550-750nm region. (e) Circular Dichroism (CD) spectra. (f) Fluorescence traces decay of *rPoLHCA4* WT and Q99V-V111M-L221V mutant (3-mut), (n=3), upon excitation at 440 nm. (d) Fitting parameters derived from the fluorescence lifetime data in panel f (n=3), including the average lifetime (τ_{avg}) and the triexponential fit components (lifetimes τ and amplitudes *A*). Details of the procedure are described in the Methods section.

>*PoLHCA1*

MAAASSNALRSCAVVCPSLLSSSKSKFATSLALPTSSANASSRVSMSEWMPGQPRPPYLDG
SAPGDFGFDPLRLGEVPSNLERFKEAELIHCRAWMLAVPGILVPEALGLGNWVKAQEWAAV
PGGQATYLGPNVPWGTLPIILAVEFITIAFAEQLRSMEKDPEKKKYPGGAFDPLGFSKDKPAKL
QEFKVKEIKNGRLALLAFVGFCVQQSAYPGTGPLENLATHLADPWHNNIGDVVIPRSIFP

>*PoLHCA2*

MASLCASSAVATVASSQNGLGATRASFFGGRKLRQGWAAPTGRRGLSVAAAAASERPLW
FPGSNPPEWLDGSLPGDFGFDPLGLSSDPESLKWNVQAEIVHCRWAMLGAAGIFVPELLTKL
GILNTPSWYTAGELEYFTDTTTTLFIHELVLIGWAEGRRWADMINPGCVNTDPVFPNNKLTGTD
VGYPGGLWFDPLGWGSGSPEKIKELRTKEIKNGRLAMLAVMGAWFQHIYTGTPIDNLFH
LADPGHATIFAAFSK

>*PoLHCA3*

MEAAGRQLLGGRRQLTTTTSRFGLSRKGTFFVRATSTPPAKQGAGRQLWFASKQSLSYLDG
SLPGDFGFDPLGLSDPEGTGGFINPRWLAYAEIMNGRFAMLGAAGSIAPELFGKLGLIPQETA
LPWFQSGVIPAGTYSYWADPYTLFVFELALMGFAEHRRYQDWSKPGSMGKQYFLGLEKGL
GGSGDPAYPGGPFNPLGFGKDEKSMKELKLKEIKNGRLAMLAILGYFIQGLVTGASPLQNL
LDHLADPVNNNVLTSKLFH

>*PoLHCA4*

MATFTAQASSTAFQSCRSKSRFLSGTSTKLPRHFSVTAAPTTTTTSTSLKVEAKKGEWLPGLA
SPNYLNGSLPGDNGFDPLGLGEDPESLKWFVQAEIQNGRWAMLAVAGVLLPEVLTKIGIINV
PEWYDAGKSEYFASSTLFFVIEFILFHYVEIRRWQDIKNPGCVNQDPIFKNYSLPPGEVGYPGG
IFNPLNFAPSLEAKEKEIANGRLAMLAFGLFLVQHNVTKGKPIDNLLQHISDPWHNTIIQTFSG

>*PoLHCB1*

MAATMALSSPSLAGKAVKLAPSASEILGAGRVTVRSTIKSKAAPGSPWYGPDRVKYLGPLSG
EAPSYLTGEFPGDYGWDTAGLSADPETFAKNRELEVIHCRWAMLGTLGCVPELLSRNGVKF
GEAVWFKAGSQIFSDGGLDYLGNPSLVHAQSILAIWACQVILMGAVEGYRIAGGPLGEVVDP
LYPGGSFDPLGLAEDPEAFELKVKEIKNGRLAMFSMFGFYVQAIVTGKGPLENLADHLADP
VNNNAWAYATNFVPGK

>PoLHCB2

MATSAIQQSAFAGQTALKQSNELIRKVGVSEGRITMRRTVKSAPKSIWYGVDRPKYLGPFSE
QTPSYLTGEFPGDYGWDTAGLSADPETFAKNRELEVIHARWAMLGTLGCVLPELLAKNGVK
FGEAVWFKAGSQIFSEGGLDYLGNPNLVHAQSILAIWACQVVLMGFIEGYRVGGGPLGEGLD
ALYPGGSFDPLGLANDPDFAELKVKEIKNGRLAMFSMFGFYVQAIVTGKGPIENLYDHLAD
PVANNAWAYATNFVPGK

>PoPSAA

MIIRSPEPEVKIVVDRDPVKTSFEEWARPGHFSRTIAKGPDTTTWIWNLHADAHDFDSHTSDL
EEISRKVFSAHFGQLSIIFLWLSGMYFHGARFSNYEAWLSDPHTHIGPSAQVVWPIVGQEILNGD
VGGGFRGIQITSGFFQLWRASGITSELQLYCTAIGALVFASLMLFAGWFWHYHKAAPKLAWFQ
DVESMLNHHLAGLLGLGSLWAGHQIHISLPINQFLDAGVDPKEIPLPHEFILNRDLLAQLYPS
FAEGATPFFTLNWSKYADFLTFRGGLDPITGGLWLSDIAHHHLAIAILFLIAGHMYRTNWGIG
HGLKDILEAHKGPFQTGGHKGGLYEILTTSWHAQLSLNLAMLGSLTIVVAHHMYSMPYPYL
AIDYGTQLSLFTHHMWIGGFLIVGAAAHA AIFMVRDYDPTTRYNDLLDRVLRHRDAIISHLN
WACIFLGFHSFGLYIHNDTMSALGRPQDMFSDTAIQLQPIFAQWVQNTHALAPGTTAPGATA
STSLTWGGGELVAVGGKVALLPIPLGTADFLVHHIHAFTIHVTVLILLKGVL FARSSRLIPDKA
NLGFRFPCDGPGRGGTCQVSAWDHVFLGLFWMYNAISVVFHFSWKMQSDVWGTISDQGV
VTHITGGNFAQSSITINGWLRDFLWAQASQVIQS YGSSLSAYGLFFLGAHFVWAFSLMFLFSG
RGYWQELIESIVWAHNKLKVAPATQPRALSIVQGRAVGVTYLLGGIATTWAFFLARIIVG

>PoPSAB

MALRFPFRSQGLAQDPTTRRIWFGIATAHDFESHDDITEERLYQNI FASHFGQLAIIFLWTSGN
LFHVAWQGNFESWVQDPLHVRPIAHAIWDPHFGQPAVEAFTRGGALGPVNIAYSGVYQWW
YTIGLRTNEDLYTGALFLLFLSAISLIAGWLHLQPKWKPSVSWFKNAESRLNHHLSGLFGVSS
LAWTGHLVHVAIPASRGEYVRWNNFLDVL PYPQGLGPLFTGQWNLYAQNPDSGSHLFGTSQ
GAGTAILTLLGGFHPQTQSLWLT DIAHHHLAIAFIFLVAGHMYRTNFGIGHSIKDLLEAHIPPG
GRLGRGHKGGLYDTINNSIHFQLGLALASLG VVTSLVAQHMYSLPAYAFIAQDFTTQAALYTH
HQYIAGFIMTGAFAHGAIFFIRDYNPEQNEDNVLARMLDHKEAIISHL SWASLFLGFHTLGLY
VHNDVMLAFGTPEKQILIEPIFAQWIQSAHGKTSYGF DVLLSSTNGPAFNAGRNIWLPGWLN
AVNENSNSLFLTIGPGDFLVHHAIALGLHTTTLIL VKGALDARGSKLMPDKKDFGYSFPCDGP
GRGGTCDISAWDAFYLA VFWMLNTIGWVTFYWHWKHITLWQGNVSQFNESSTYLMGWLR
DYLWLNSSQLINGYNPFGMNSLSVWAWMFLFGHLVWATGFMFLISWRGYWQELIETLAWA
HERTPLANLIRWRDKPVALSIVQARLVGLAHFSVGYIFTYAAFLIASTSGKFG

>PoPSAC

MSHSVKIYDTCIGCTQCVRACPTDVLEMIPWDGCKAKQIASAPRTEDCVGCKRCESACPTDF
LSVRVYLYHETTRSMALAY

>PoPSAD

MASQASLFTPATLTPKLPATSSTTTPRTLAPWKSISTPKPLSLRTTRLSATQEATTEAPVGFTPP
ELDPNTPSPIFGGSTGGLLRKAQVEEFYVITWTSPEQCLALGTRLRSKYKIDYQFYRVFPNG
EVQYLHPKDGIIYPEKVNAGRLGVGLNMRSIGKNVSPIEVKFTGKQPYDL

>PoPSAE

MATSSMASAASSFVLPSAPPKNMTNLRISAVFFPKNNRW SHRGTVVVQAASGEETAPVAEEK
RPAADPPAAAKPKPEKIGPKRGTKVKILRRESYWFNGVGSVVTVDQDPNSRYPVVVRFNKV
NYYNVFTNNYAFDEILEVK

>PoPSAF

MASLTTPSSTLSKRLHHPKLHHSKLLKPLTISCSSSITPSPQTPEASNNNNNSQLKAFSA AVALSSI
LLSSPIVPPASADIAGLTPCKESKAFKREKQSLKKLESSLKKYAPDSAPALAIKASMEKTKRR
FDNYGKQGLLCGSDGLPHLIVSGDQRHWGEFITPGILFLYIAGWIGWVGRSYLIAIRDEKKPT
MKEIHDVPLASKLIWRGFIWPVAAYRELINGDLVVDDADVSVT

>PoPSAG

MASSTIFSAPTLLRNNNSSTTTTHLITPSHHSSFQGLRPLKPTDAFLIRASTTTTAPKSKAIRPAGV
RAELNPQVVICLSTGLSLFLGRFVFFNFQRENVAKQGLPKQNGISHFEAGDERAKEYVGLLKS
NDPVGFNIVDVLAWGSIGHIVAYYVLATSSNGYDPNFFG

>PoPSAH

MALSLSIVASVQP VATVRGLGGSSLNGTKLAVRPQSRRMAPRTSLRSSRAVVAKYGDKSVYF
DLEDLPNTTGQWDLYGSDAPSPYNSLQSKFFETFAAPFTKRGLLLKFLILGGGFTLAYYSSTA
SGDILPIKKGPQLPELGPDRKK

>PoPSAI

MTDLNLPSIFVPLVGLVFPAIAMASFLHVQKNKIV

>PoPSAJ

MRDIKTYLSVAPVLTTLWFGSLAGLLIEINRLFPDALSFPPF

>PoPSAK

MATQLSSTATVMSLPQFIGLRTKPSMATLPVVRQGRGALGARCGDFIGSSTNIIMVTTTSLM
LFAGRFG LAPSANRKATAGLKLEVRDSGLQTGDPAGFTLADTLACGVVGHIMGVGIVLGLK
SIGAI

>PoPSAL

MATASASPMLKSGFHSGGLLAPRGISGSPLRALLPSKRRTCLTVRAIQAEKPTTFQVIEPINGD
PFIGSLETPVTSSPLVAWYLSNLPAYRTAVSPLLRGIEVGLAHGYLLVGPFVITGPLRNTPYHG
AAGSLAAGGLVVILSICLTMYGISSFKEGEPSCAPGLTLTGRKKEADKLQTADGWAQFTGGF
FFGGISGVIWAYFLLYVLDLPYYIK

>PoPSAO

MAAMATTSTTVAGLLPLSRPSTKASLSSGFIKTQVATRNP LSQLASGGKFTCFERDWLRRD
FNVIGFGLIGWLAPSSIPVIGNSLTGLFFSSIGTELAHWPTGPALSSPFWLWMVTWHFGLFVC
LTFGQIGFKGRNEGYF

Supplementary Fig. 26. Amino acid sequences of the LHCI, LHCII, and PSI core subunits in the structure of PSI-LHCI and PSI-LHCI-mega complexes of *P. oceanica*.

Supplementary Table 1. Summarization of the structural models of *P. oceanica* PSI-LHCI supercomplex (PDB: 9S1L).

Subunits	Residues	Chlorophylls	Carotenoids	Lipids	Others
PsaA	750 (27-750)	46 Chl <i>a</i>	6 β -car	2 LHG; 1 LMT 1 LHG; 2 LMT; 1	1 PQN; 1 SF4
PsaB	734 (2-734)	40 Chl <i>a</i>	8 β -car	DGD	1 PQN
PsaC	81 (2-81)				2 SF4
PsaD	203 (63-202)				
PsaE	142 (79-141)				
PsaF	237 (79-233)	2 Chl <i>a</i>	1 β -car	2 LMG	
PsaG	168 (70-167)	3 Chl <i>a</i>	1 β -car	2 LMG	
PsaH	148 (55-148)	1 Chl <i>a</i>	1 β -car		
PsaI	36 (3-34)		1 β -car		
PsaJ	42 (2-41)	1 Chl <i>a</i>	1 β -car		
PsaK	129 (47-124)	3 Chl <i>a</i>	2 β -car		
PsaL	215 (62-213)	3 Chl <i>a</i>	3 β -car		
Lhca1	246 (49-243)	12 Chl <i>a</i> ; 2 Chl <i>b</i>	2 Lut; 1 Vio; 1 β -car	2 LHG; 1 LMG	
Lhca2	253 (55-252)	9 Chl <i>a</i> ; 5 Chl <i>b</i>	1 Lut; 1 Vio; 2 β -car	2 LMG; 1 LMT	
Lhca3	266 (48-260)	12 Chl <i>a</i> ; 1 Chl <i>b</i>	1 Lut; 1 Vio; 1 β -car		
Lhca4	262 (56-258)	11 Chl <i>a</i> ; 4 Chl <i>b</i>	1 Lut; 1 Vio; 2 β -car	2 LMG; 1 LMT	
Total		155	39	20	5

β -car, β -carotene; Lut, Lutein; Vio, Violaxanthin; LHG, 1,2-dipalmitoyl-phosphatidyl-glycerole; LMG, 1,2 Distearoyl-Monogalactosyl-Diglyceride; LMT, β -Dodecyl-Maltoside; DGD; Digalactosyl-Diacyl-Glycerol; PQN phylloquinone; SF4, Fe₂S₂ iron-sulfur cluster.

Supplementary Table 2. Summarization of the structural models of *P. oceanica* PSI-LHCI-mega complex (PDB: 9S1M).

Subunits	Residues	Chlorophylls	Carotenoids	Lipids	Others
PsaA	750 (27-750)	46 Chl <i>a</i>	6 β -car	2 LHG; 1 LMT	1 PQN; 1 SF4
PsaB	734 (2-734)	40 Chl <i>a</i>	8 β -car	1 LHG; 2 LMT; 1 DGD	1 PQN
PsaC	81 (2-81)				2 SF4
PsaD	203 (63-202)				
PsaE	142 (79-141)				
PsaF	237 (79-233)	2 Chl <i>a</i>	1 β -car	2 LMG	
PsaG	168 (70-167)	3 Chl <i>a</i>	1 β -car	2 LMG	
PsaH	148 (55-148)	1 Chl <i>a</i>	1 β -car		
PsaI	36 (3-34)		1 β -car		
PsaJ	42 (2-41)	1 Chl <i>a</i>	1 β -car		
PsaK	129 (47-126)	4 Chl <i>a</i>	2 β -car		
PsaL	215 (53-214)	3 Chl <i>a</i>	3 β -car		
PsaO	142 (50-138)	3 Chl <i>a</i>	2 β -car		
Lhca1	246 (49-246)	12 Chl <i>a</i> ; 2 Chl <i>b</i>	2 Lut; 1 Vio; 1 β -car	2 LHG; 1 LMG	
Lhca2	253 (56-258)	9 Chl <i>a</i> ; 5 Chl <i>b</i>	1 Lut; 1 Vio	1 LHG	
Lhca3	266 (48-260)	12 Chl <i>a</i> ; 1 Chl <i>b</i>	1 Lut; 1 Vio; 1 β -car		
Lhca4	262 (55-252)	11 Chl <i>a</i> ; 4 Chl <i>b</i>	1 Lut; 1 Vio; 2 β -car	2 LMG; 1 LMT	
Lhca1_s	246 (49-243)	12 Chl <i>a</i> ; 2 Chl <i>b</i>	2 Lut; 1 Vio; 1 β -car	1 LHG; 1 LMG	
Lhca4₆	262 (55-252)	11 Chl <i>a</i> ; 4 Chl <i>b</i>	1 Lut; 1 Vio; 2 β -car	2 LMG; 1 LMT	
Lhcb1_x	264 (46-263)	8 Chl <i>a</i> ; 6 Chl <i>b</i>	2 Lut; 1 Vio; 1 Neo	1 LHG	
Lhcb1_y	264 (37-263)	8 Chl <i>a</i> ; 6 Chl <i>b</i>	2 Lut; 1 Vio; 1 Neo	1 LHG	
Lhcb2_z	264 (46-263)	8 Chl <i>a</i> ; 6 Chl <i>b</i>	2 Lut; 1 Vio 1 Neo	1 LHG	
Total		230	59	26	5

β -car, β -carotene; Lut, Lutein; Vio, Violaxanthin; Neo, Neoxanthin; LHG, 1,2-dipalmitoyl-phosphatidyl-glycerole; LMG, 1,2 Distearoyl-Monogalactosyl-Diglyceride; LMT, β -Dodecyl-Maltoside; DGD; Digalactosyl-Diacyl-Glycerol; PQN phyloquinone; SF4, Fe₂S₂ iron-sulfur cluster.

Supplementary Table 3. FRET rate for intra-subunit preferential EET pathways in PSI-LHCI-mega. Only the strongest pathways are shown, using a threshold of < 10 picoseconds (ps) for EET between Chls within the same subunit. Protein chain subunits are indicated in parentheses. CLA: Chl *a*; CHL: Chl *b*.

	s^{-1}	ps
CLA604 (1) - CLA606 (1)	6.7E+12	0.15
CLA609 (1) - CLA603 (1)	4.3E+12	0.23
CLA611 (1) - CLA612 (1)	2.6E+12	0.38
CLA609 (1) - CLA608 (1)	1.5E+12	0.67
CHL607 (1) - CLA606 (1)	9.2E+11	1.09
CHL601 (1) - CLA611 (1)	4.7E+11	2.13
CLA602 (1) - CLA603 (1)	4.7E+11	2.13
CLA606 (1) - CLA608 (1)	1.2E+11	8.33
CLA603 (2) - CLA609 (2)	4.9E+12	0.20
CHL606 (2) - CLA604 (2)	3.6E+12	0.28
CLA611 (2) - CLA612 (2)	2.2E+12	0.45
CHL606 (2) - CHL607 (2)	1.3E+12	0.77
CHL608 (2) - CHL609 (2)	8.8E+11	1.14
CLA603 (2) - CLA602 (2)	5.6E+11	1.79
CHL601 (2) - CLA611 (2)	4.2E+11	2.38
CHL608 (2) - CLA612 (2)	2.5E+11	4.00
CLA604 (3) - CLA606 (3)	8.2E+12	0.12
CLA603 (3) - CLA609 (3)	3.8E+12	0.26
CLA609 (3) - CLA615 (3)	2.1E+12	0.48
CLA604 (3) - CLA605 (3)	1.0E+12	1.00
CHL608 (3) - CLA609 (3)	9.1E+11	1.10
CHL608 (3) - CLA610 (3)	4.2E+11	2.38
CLA603 (3) - CLA602 (3)	3.9E+11	2.56
CLA609 (3) - CLA606 (3)	1.2E+11	8.33
CLA603 (4) - CLA609 (4)	3.5E+12	0.29
CHL606 (4) - CLA604 (4)	2.7E+12	0.37
CHL606 (4) - CHL607 (4)	1.7E+12	0.59
CLA601 (4) - CLA611 (4)	1.1E+12	0.91
CHL608 (4) - CLA609 (4)	8.2E+11	1.22
CLA601 (4) - CLA602 (4)	5.8E+11	1.72
CHL608 (4) - CLA610 (4)	2.2E+11	4.55
CLA603 (4) - CLA609 (4)	3.5E+12	0.29
CLA604 (5) - CLA606 (5)	9.4E+12	0.11
CHL607 (5) - CLA606 (5)	7.7E+12	0.13
CLA603 (5) - CLA609 (5)	4.5E+12	0.22
CLA611 (5) - CLA612 (5)	2.8E+12	0.36
CLA608 (5) - CLA609 (5)	1.1E+12	0.91
CLA601 (5) - CLA602 (5)	3.3E+11	3.03
CLA610 (5) - CLA608 (5)	3.2E+11	3.13
CLA608 (5) - CLA606 (5)	1.2E+11	8.33
CHL606 (6) - CLA604 (6)	4.9E+12	0.20
CLA603 (6) - CLA609 (6)	3.9E+12	0.26
CLA612 (6) - CLA611 (6)	2.3E+12	0.44
CLA601 (6) - CLA611 (6)	1.5E+12	0.67
CHL606 (6) - CHL607 (6)	1.4E+12	0.71

CHL608 (6) - CLA609 (6)	7.3E+11	1.37
CLA601 (6) - CLA602 (6)	5.5E+11	1.82
CHL608 (6) - CLA610 (6)	2.4E+11	4.17
CLA307 (L) - CLA306 (L)	5.6E+11	1.79
CLA307 (L) - CLA308 (L)	2.9E+11	3.45
CLA306 (L) - CLA308 (L)	1.2E+11	8.33
CLA200 (O) - CLA204 (O)	1.8E+11	5.56
CHL606 (Y) - CLA604 (Y)	5.3E+12	0.19
CLA612 (Y) - CLA611 (Y)	3.0E+12	0.33
CHL606 (Y) - CHL607 (Y)	1.5E+12	0.67
CHL609 (Y) - CLA603 (Y)	1.5E+12	0.67
CHL608 (Y) - CHL609 (Y)	9.4E+11	1.06
CHL601 (Y) - CLA611 (Y)	5.2E+11	1.92
CHL608 (Y) - CLA610 (Y)	3.5E+11	2.86
CLA603 (Y) - CLA602 (Y)	3.5E+11	2.86
CLA610 (Y) - CLA612 (Y)	3.0E+11	3.33
CHL607 (Y) - CLA604 (Y)	2.3E+11	4.35
CLA602 (Y) - CLA611 (Y)	1.3E+11	7.69
CLA613 (Y) - CLA614 (Y)	1.2E+11	8.33
CHL606 (X) - CLA604 (X)	6.8E+12	0.15
CLA611 (X) - CLA612 (X)	4.7E+12	0.21
CHL608 (X) - CHL609 (X)	3.9E+12	0.26
CHL606 (X) - CHL607 (X)	2.1E+12	0.48
CLA602 (X) - CLA611 (X)	1.7E+12	0.59
CHL609 (X) - CLA603 (X)	1.1E+12	0.91
CHL601 (X) - CLA611 (X)	4.8E+11	2.08
CHL601 (X) - CLA611 (X)	4.8E+11	2.08
CLA602 (X) - CLA603 (X)	3.7E+11	2.70
CHL608 (X) - CLA610 (X)	3.2E+11	3.13
CLA610 (X) - CLA612 (X)	2.7E+11	3.70
CHL601 (X) - CLA602 (X)	2.2E+11	4.55
CHL607 (X) - CLA604 (X)	2.2E+11	4.55
CLA611 (Z) - CLA612 (Z)	3.0E+12	0.33
CHL606 (Z) - CHL607 (Z)	2.0E+12	0.50
CHL609 (Z) - CLA603 (Z)	9.2E+11	1.09
CHL605 (Z) - CHL609 (Z)	7.9E+11	1.27
CHL601 (Z) - CLA611 (Z)	7.5E+11	1.33
CHL606 (Z) - CLA604 (Z)	3.7E+11	2.70
CLA602 (Z) - CLA603 (Z)	2.1E+11	4.76
CLA610 (Z) - CLA612 (Z)	2.1E+11	4.76
CHL606 (Z) - CHL608 (Z)	1.8E+11	5.56
CLA613 (Z) - CLA614 (Z)	1.8E+11	5.56
CHL607 (Z) - CLA604 (Z)	1.7E+11	5.88
CHL601 (Z) - CLA602 (Z)	1.4E+11	7.14
CHL605 (Z) - CHL606 (Z)	1.1E+11	9.09
CHL606 (Z) - CHL609 (Z)	1.1E+11	9.09
CLA611 (Z) - CLA602 (Z)	1.0E+11	10.00

Supplementary Table 4. FRET rate for inter-subunit preferential EET pathways in PSI-LHCI-mega. Only the strongest pathways are shown, using a threshold of < 100 ps for EET between Chls belonging to different subunits. Protein chain subunits are indicated in parentheses.

	s^{-1}	ps
CLA617 (1) - CLA615 (4)	3.8E+11	2.63
CLA603 (1) - CLA840 (B)	2.6E+11	3.85
CHL607 (1) - CLA223 (G)	2.4E+11	4.17
CLA603 (1) - CLA821 (B)	1.4E+11	7.14
CLA609 (1) - CLA821 (B)	1.4E+11	7.14
CLA613 (1) - CLA615 (4)	1.0E+11	10.00
CLA609 (1) - CLA820 (B)	8.6E+10	11.63
CLA606 (1) - CLA223 (G)	7.0E+10	14.29
CHL601 (1) - CHL616 (4)	6.9E+10	14.49
CLA617 (1) - CLA317 (F)	6.5E+10	15.38
CHL601 (1) - CLA615 (4)	4.8E+10	20.83
CLA602 (1) - CLA840 (B)	4.0E+10	25.00
CLA609 (1) - CLA840 (B)	4.0E+10	25.00
CLA603 (1) - CLA820 (B)	3.3E+10	30.30
CHL607 (2) - CLA102 (J)	8.8E+10	11.36
CHL616 (2) - CLA601 (4)	7.2E+10	13.89
CHL601 (2) - CLA615 (3)	6.2E+10	16.13
CLA614 (2) - CLA606 (3)	6.1E+10	16.39
CHL616 (2) - CLA611 (4)	5.3E+10	18.87
CLA609 (2) - CLA601 (4)	3.6E+10	27.78
CHL606 (2) - CLA614 (4)	2.5E+10	40.00
CLA603 (3) - CLA813 (A)	7.8E+10	12.82
CLA603 (3) - CLA811 (A)	4.7E+10	21.28
CLA602 (3) - CLA813 (A)	4.6E+10	21.74
CLA613 (3) - CLA202 (K)	4.3E+10	23.26
CHL616 (4) - CHL601 (1)	6.9E+10	14.49
CHL607 (4) - CLA617 (1)	4.5E+10	22.22
CHL606 (4) - CLA614 (1)	4.0E+10	25.00
CHL616 (4) - CLA611 (1)	3.9E+10	25.64
CLA601 (4) - CLA609 (2)	3.6E+10	27.78
CLA617 (5) - CLA615 (6)	3.6E+11	2.78
CLA613 (5) - CLA615 (6)	9.4E+10	10.64
CHL601 (5) - CHL616 (6)	6.4E+10	15.63
CHL601 (5) - CLA615 (6)	3.2E+10	31.25
CLA603 (5) - CLA209 (H)	1.8E+10	55.56
CLA602 (5) - CLA209 (H)	1.6E+10	62.50
CHL607 (6) - CLA617 (5)	5.6E+10	17.86
CHL606 (6) - CLA614 (5)	3.8E+10	26.32
CHL616 (6) - CLA611 (5)	3.3E+10	30.30
CLA209 (H) - CLA306 (L)	4.5E+11	2.22
CLA102 (J) - CLA316 (F)	4.5E+10	22.22
CLA102 (J) - CLA808 (A)	1.3E+10	76.92
CLA307 (L) - CLA843 (A)	2.6E+11	3.85
CLA307 (L) - CLA834 (A)	1.7E+11	5.88

CLA308 (L) - CLA834 (A)	1.4E+11	7.14
CLA203 (O) - CLA824 (A)	1.0E+12	1.00
CLA203 (O) - CLA823 (A)	1.1E+11	9.09
CLA203 (O) - CLA825 (A)	3.8E+10	26.32
CLA204 (O) - CLA857 (A)	2.3E+10	43.48
CLA200 (O) - CLA834 (A)	2.2E+10	45.45
CLA200 (O) - CLA839 (A)	1.5E+10	66.67
CLA614 (Y) - CLA204 (O)	6.4E+10	15.63
CLA613 (Y) - CLA603 (Z)	2.6E+10	38.46
CHL601 (Y) - CLA203 (O)	2.2E+10	45.45
CHL605 (Y) - CLA606 (S)	2.0E+10	50.00
CHL601 (Y) - CHL609 (Z)	1.5E+10	66.67
CHL601 (Y) - CHL605 (Z)	1.4E+10	71.43
CLA611 (Y) - CLA203 (O)	1.3E+10	76.92
CHL606 (Y) - CHL601 (X)	9.4E+09	106.40
CLA602 (X) - CLA603 (Y)	6.3E+10	15.87
CHL609 (X) - CLA602 (Z)	2.2E+10	45.45
CLA614 (X) - CLA603 (Y)	1.7E+10	58.82
CHL607 (X) - CLA613 (Z)	1.5E+10	66.67
CHL601 (X) - CHL608 (Y)	1.1E+10	90.91
CLA602 (Z) - CLA603 (X)	7.3E+10	13.70
CLA603 (Z) - CLA602 (Y)	6.8E+10	14.71
CLA613 (Z) - CLA603 (X)	2.4E+10	41.67
CHL607 (Z) - CLA614 (Y)	1.9E+10	52.63
CHL609 (Z) - CLA602 (Y)	1.8E+10	55.56
CHL601 (Z) - CHL608 (X)	1.5E+10	66.67

Supplementary Table 5. Cryo-EM data collection, refinement, and validation statistics.

	PSI-LHCI (EMD-54455, EMD-54379 ^a , PDB 9S1L)	L-PSI-LHCI-LHCII (EMD-54456, EMD-54439 ^a , PDB 9S1M)	PSI-LHCI PSI core (EMD-54380) ^b	PSI-LHCI LHCI belt (EMD-54396) ^c	L-PSI-LHCI-LHCII PSI core (EMD-54441) ^d	L-PSI-LHCI-LHCII LHCI belt (EMD-54443) ^e	L-PSI-LHCI-LHCII Lhca1 ₅ -4 ₆ -LHCII (EMD-54444) ^f
Data collection and processing							
Magnification	130,000X	130,000X	130,000X	130,000X	130,000X	130,000X	130,000X
Voltage (kV)	200	200	200	200	200	200	200
Total electron dose (e-/Å)	45	45	45	45	45	45	45
Nominal defocus range (μm)	-0.75 – -2.5	-0.75 – -2.5	-0.75 – -2.5	-0.75 – -2.5	-0.75 – -2.5	-0.75 – -2.5	-0.75 – -2.5
Pixel size (Å)	0.92	0.92	0.92	0.92	0.92	0.92	0.92
Number of particles in final reconstruction	120603	37677	120603	120603	37677	37677	22405
Symmetry imposed	C1	C1	C1	C1	C1	C1	C1
Map resolution (Å)	2.83	3.31	2.79	3.06	3.23	3.5	3.63
Map sharpening B factor (Å ²)	74.5	62.8	71.9	82.1	58.8	55.9	51.3
Model Refinement and Validation							
Initial model used (PDB code)	5ZJI	9S1L and 5ZJI					
<u>Model composition</u>							
<i>Protein residues</i>	3174	4358					
<i>Ligands</i>	215	318					
<u>R.m.s. deviations</u>							
<i>Bond lengths (Å)</i>	0.003	0.004					
<i>Bond angles (°)</i>	0.631	0.650					
MolProbity score	1.45	1.70					
Clash score	5.29	8.45					
Rotamers outliers (%)	0.93	0.91					
<u>Ramachandran plot</u>							
<i>Favored (%)</i>	97.1	96.4					
<i>Allowed (%)</i>	2.9	3.6					
<i>Disallowed (%)</i>	0	0					
Mean B factor (Å ²)							
<i>Protein</i>	68.07	100.15					
<i>Ligands</i>	64.04	93.10					
CC (model vs data)							
<i>Mask</i>	0.83	0.85					
<i>Box</i>	0.68	0.75					

^{a)} The two EMDB codes belong to the combined and original map, respectively. ^{b)} Focused map of PSI core of PSI-LHCI complex. ^{c)} Focused map of the LHCI belt of the PSI-LHCI complex. ^{d)} Focused map of the PSI core of the L-PSI-LHCI-LHCII complex. ^{e)} Focused map of the LHCI belt of L-PSI-LHCI-LHCII. ^{f)} Focused map of Lhca1₅-Lhca4₆-LHCII of L-PSI-LHCI-LHCII complex. L-PSI-LHCI-LHCII refers to PSI-LHCI-mega

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