

Fine-tuning the Photosynthetic Light Harvesting Apparatus by Molecular Simulations

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The natural photosynthetic process is fine-tuned for optimal yield under low light conditions. This comes at a cost. To avoid the deleterious effects of excess light, plants and algae have developed photoprotective mechanisms as well-orchestrated responses that negatively influence productivity (crop yield and biomass). Non-Photochemical Quenching (NPQ) of chlorophyll fluorescence is a major such response at the first line of fire that dissipates the excess light energy as heat. In search for sustainable and carbon-neutral productivity, two of our society's grand challenges, there is a major interest in elucidating the fine-details of NPQ, in order to improve productivity with increased CO₂ uptake. NPQ seems to be the ideal target for fine-tuning, as it limits the excitation energy that the photosynthetic apparatus receives from light harvesting complexes (LHC) of Photosystem II (PSII) and considerably decelerates CO₂ assimilation. The major LHCII antenna switches between two states within the thylakoid membranes of higher plants, i.e., a light-harvesting and a photoprotective mode. The switch balances the amount of absorbed irradiation and the ability of the apparatus to use this energy. It is co-regulated by a pH gradient (Δ pH) across the thylakoid membrane and the interaction with the PSII subunit S (PsbS) that is proposed to induce LHCII aggregation. The crystal structures of the associated proteins refer only to the quenched states, whereas an all-atom structure at the light harvesting mode is elusive and this hinders the study of NPQ switch. Using molecular simulations and quantum calculations, we aim at addressing important NPQ components: (a) the effect of Δ pH and PsbS on LHCII conformation and aggregation, (b) the excitation energy flow within LHCII under light harvesting and quenched states.

Due to the high computational cost associated with the methodology employed for systems with up to 260.000 atoms (Parallel-Tempering Metadynamics at the Well-Tempered Ensemble, PTmetaD-WTE), Tier-0 and Tier-1 HPC resources were granted at two separate projects (PRACE 18-2018194641 "CDynLHCII - Clustering Dynamics of the major LHCII of PSII under Photoprotection", and 15-DECI0328, "LHCPSSalt - LHCII-PsbS complex conformations under varying salt content"). The scientific challenges were to re-produce native protein states under NPQ, and to identify for the first time, at all-atom resolution, both the protein scaffold dynamics that lead to these states, and the site of switch within LHCII. Our results demonstrate the aggregation potential of LHCII and, consistent with the experimental literature, reveal the role of Δ pH-PsbS at atomic resolution. Diffusion, aggregation, and subsequent conformational changes of the integral membrane protein LHCII are highly regulated under NPQ, whereas the light-harvesting potential of LHCII is tuned via allosteric regulation of the excitonic coupling in chlorophyll-carotenoid pairs within. Our original findings may help addressing the agricultural crisis in the future and redesign photosynthesis, as a big scientific challenge. The elucidation of the details of the NPQ mechanism promises to provide the tools for identification, breeding, and molecular engineering of crop plants. Plants with a greater capacity for photoprotective thermal dissipation are protected against decrease in productivity caused also by unfavourable environmental conditions.