Supplementary material

table S1 Calculated accessibility surfaces for the charge substituting residues (see sequence alignment in figure S2) shown in the two Peridinin-Distance-Plots (figure S5 and figure S6). [a] and calculated pKa values [b]

PCP	HSPCP						MFPCP					
Contacting residue	Glu-136	**Glu-202**	Lys-143	Arg-113	Lys-284	Arg-150	Lys-124	Glu-133	Asp-109	Glu-101	Lys-147	Lys-288
Contacted Per-	611*	**621***	613*	613*	623* 624*	614*	611	611	613	613 614	614	622
AccSRes [Å²]	19.5	**12.1**	18	20.2	26.2	30.4	13	17.8	20.8	17.9	21.7	36
AccSResVac [Å²]	52.5	**49.5**	64.2	75.2	66.3	73.2	65	52.6	42.5	51.3	65.7	64.8
RAccSRes [%]	37	**24**	28	27	40	42	20	34	49	35	33	55.5
AccSFG [Å²]	12	**6.7**	10.6	18.5	11.7	21.5	6.2	16.4	13.6	12.3	4.5	12.6
AccSFGVac [Å²]	25.5	**19.4**	19	39.8	19	36.7	19.3	26.2	24.2	25.3	17.6	17.1
RAccSFG [%]	47	**35**	56	47	62	59	32	63	56	49	26	73.6
IRD [Å]							2.9			2.9		
pKa	4.8	**3.9**	10.2	12.1	10.2	11.9	10	4.1	4	3.3	10.1	10.2
Location	Surf	**Bur**	Surf	Surf	Surf	Surf	Surf	Surf	Surf	Surf	Surf	Surf

[a] Accessible surface (AccS) values of residues (Res) and functional groups (FG) calculated with WHAT IF. The relative accessibility (RAcc) is the accessibility within the protein divided by the accessibility in a tripeptide G-X-G in vacuum (Vac). IRD is the distance between the functional groups of two residues.

[b] The pKa values for the residues were calculated with Propka 2.0. The Propka 2.0 server also indicates whether the residue is classified as surface exposed (Surf) or buried (Bur) within the protein.

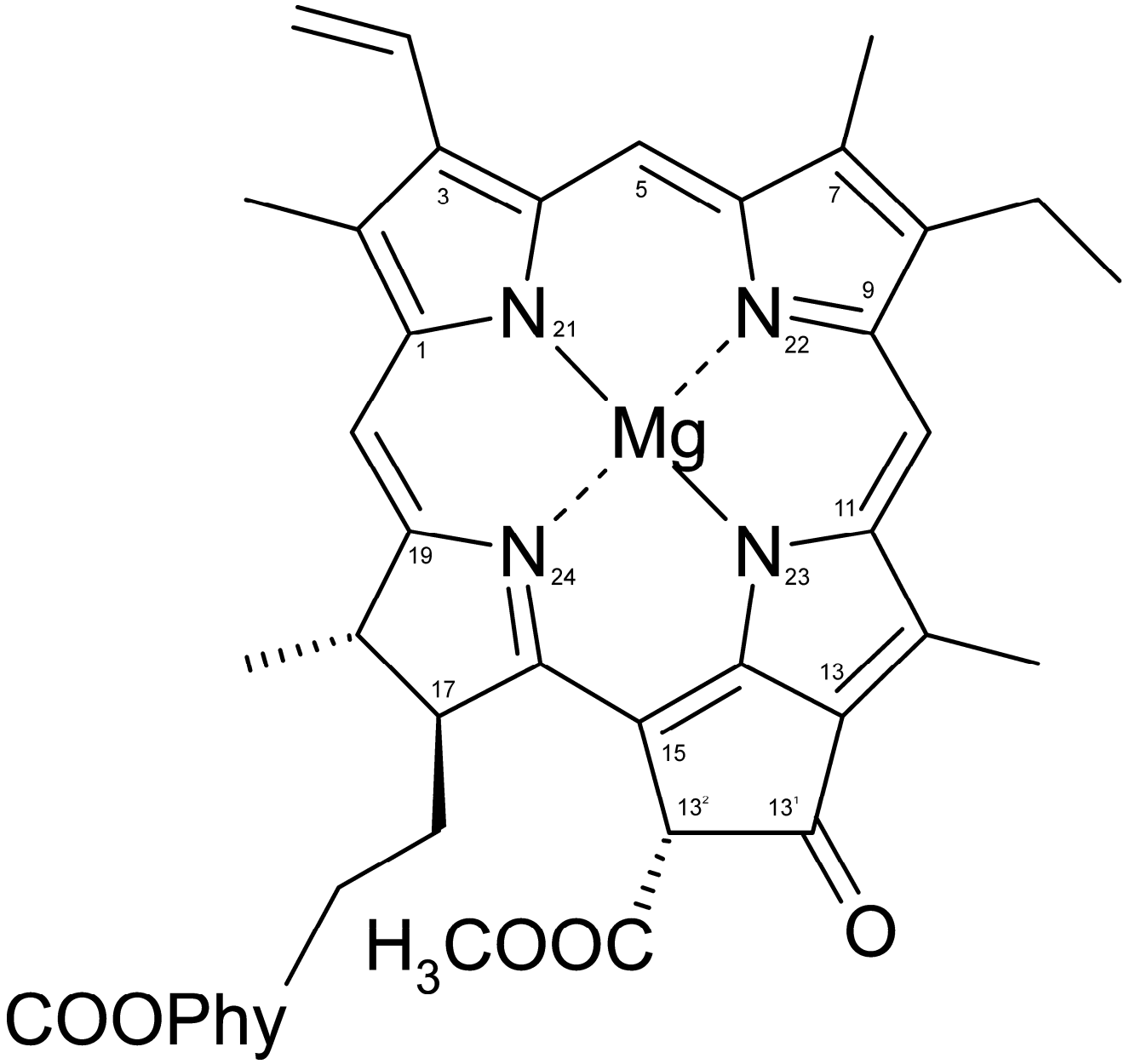

figure S1 Structure of Chl-*a*. Atom numbering is according to [Scheer, H. (2006) An overview of Chlorophylls and Bacteriochlorophylls: Biochemistry, Biophysics, Functions and Applications, in *Chlorophylls and Bacteriochlorophylls.* (Grimm, B., Porra, R. J., Rüdiger, W., and Scheer, H., Eds.), pp 1-26. Springer, Dordrecht, Netherlands.]. Phy = phytol tail.

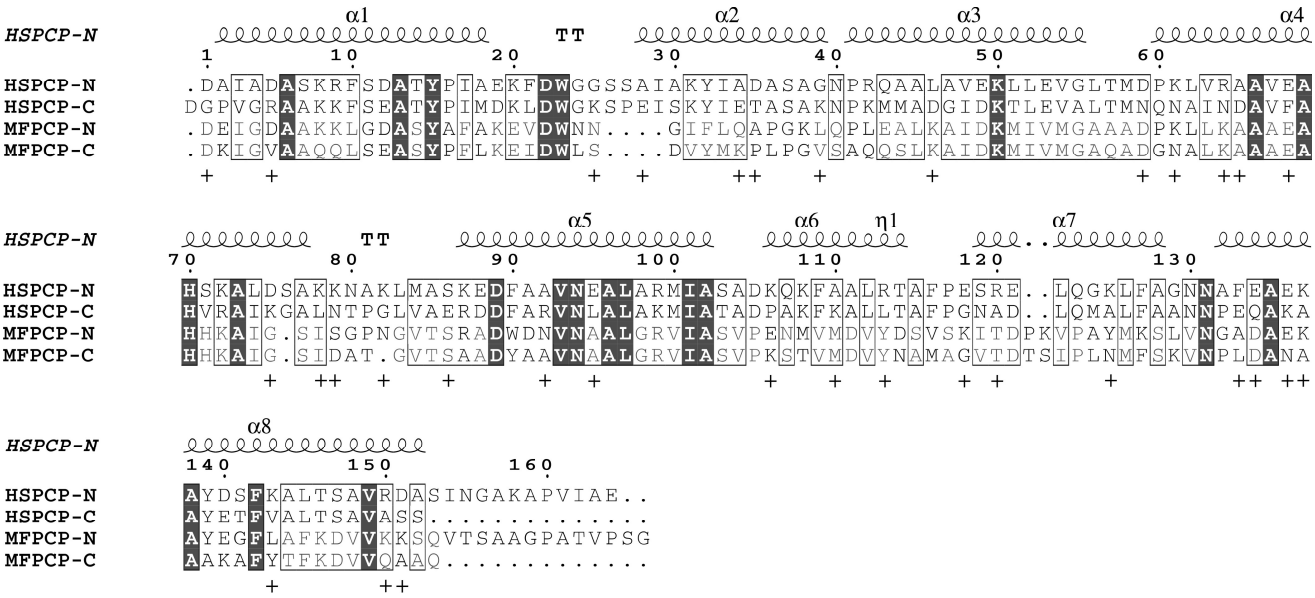

figure S2 Sequence alignment of MFPCP and HSPCP domains. Plus symbols indicate charge differences between the HSPCP-N and HSPCP-C domain residues (= Charge substituting residues).

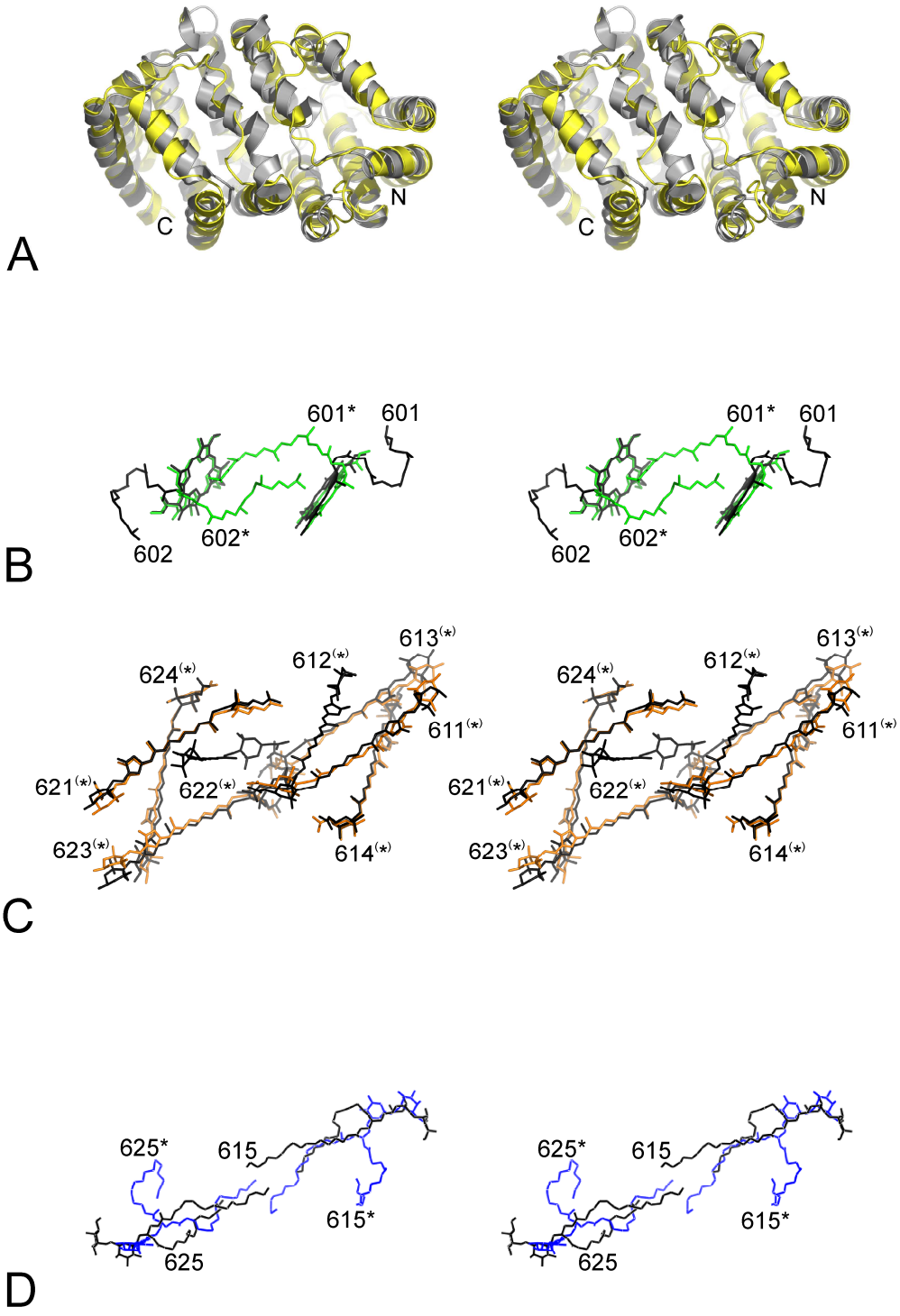

figure S3 Superimposition of HSPCP and MFPCP , showing only the protein (grey and yellow ribbons) (A), the chlorophylls (green and black sticks) (B), the peridinins (orange and black sticks) (C) and the lipids (blue and black sticks) (D).

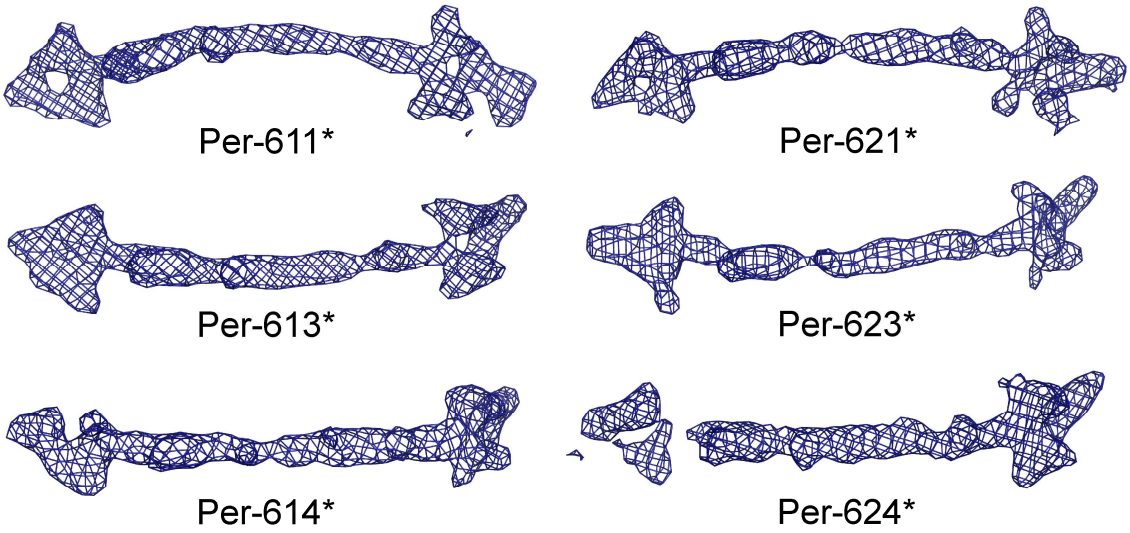

figure S4 Peridinin omit difference maps at σ-level 2.5. View is comparable to the bottom one in figure 7.

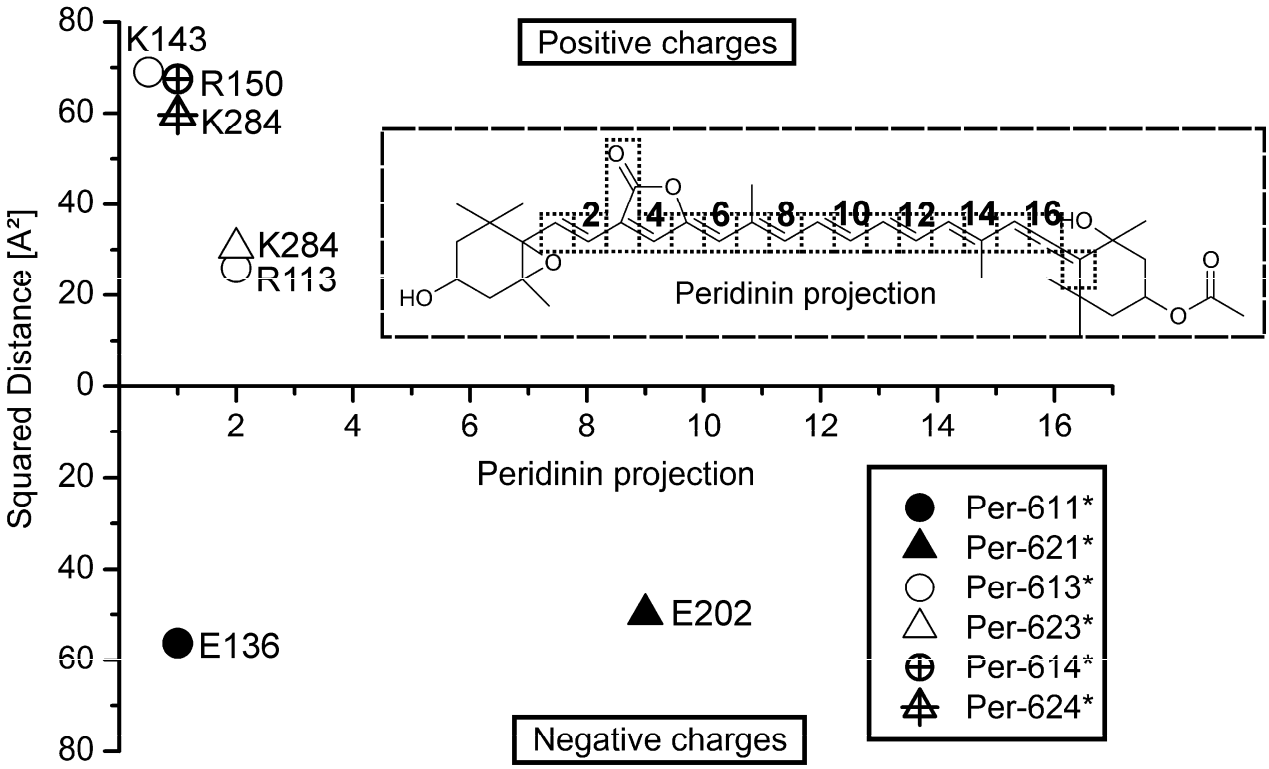

figure S5 This figure sums up the information of figure 8 in a single diagram. It represents a plot of the distance (r^2 because of Coulombic law) of substituted point charges against a 1D projection of peridinin (see inlet for numbering). Positively charged residues are shown above the x-axis, negatively charged residues below. Data point of Per-613*-K143 shifted from 1 to 0.5 for better illustration.

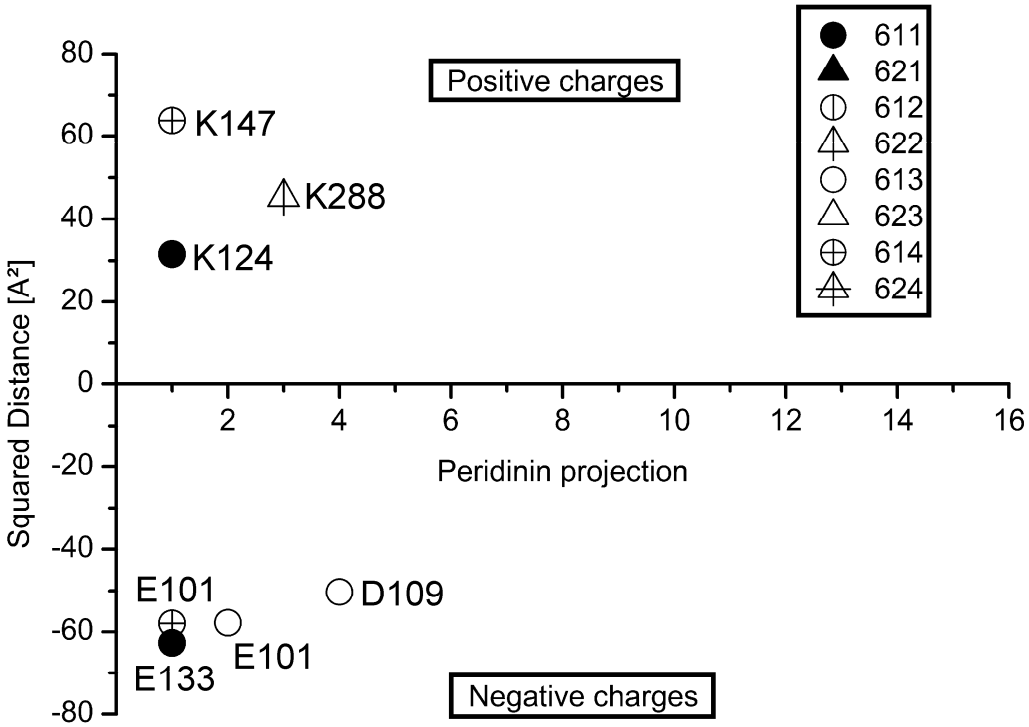

figure S6 Peridinin-Distance-Plot for MFPCP. In MFPCP 23 mutations include a charge substitution (apparent in figure S2). Only 6 residues within these 23 mutations have their charged functional groups (carboxygroup of Glu/Asp, guanidinogroup of Arg and Ammoniumgroup of Lys) within ~ 8 Å of the conjugated π-system of a peridinin molecule. An asymmetry of the peridinin pair Per-613/Per-623 is apparent: Only Per-613 is coordinated by two negatively charged residues, Glu-101 and Asp-109, at position 2 and 4 of the peridinin projection (numbering depicted in figure S5). But the charged functional groups of both residues (Glu-101: 12.3 Å²; Asp-109: 13.6 Å²) are as accessible to the solvent as the group of E-136 (12 Å²) in HSPCP (table S1). The effect of the positively charged Lys-288 onto Per-622 is weakened by its accessibility to the solvent (table S1: Relative accessibility of functional group – 73.6%). The glutamate and lysine residues at the binding site of Per-611 (Glu-133 and Lys-124) and Per-614 (Glu-101 and Lys-147) make salt bridges (distance between charged groups ~3 Å, listed in table S1) which results in neutralization of charges. None of the plotted residues is classified as buried by the Propka 2.0 server (table S1).

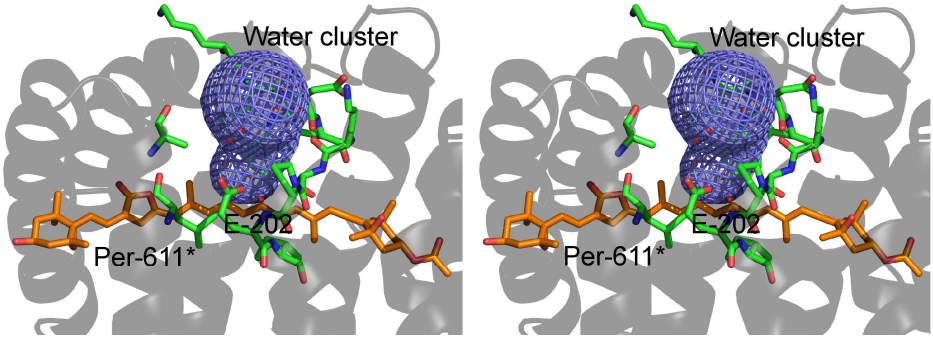

figure S7 Water cluster (red spheres) within a calculated cavity (blue mesh) around Per-611*.

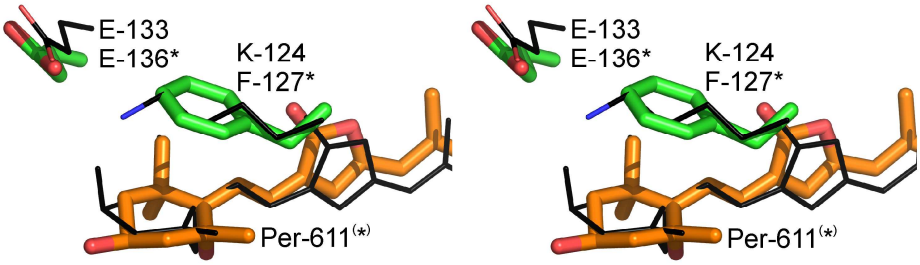

figure S8 Stereoview of the coordination of Per-611* in HSPCP (orange and green) superimposed by the corresponding residues in MFPCP (black).

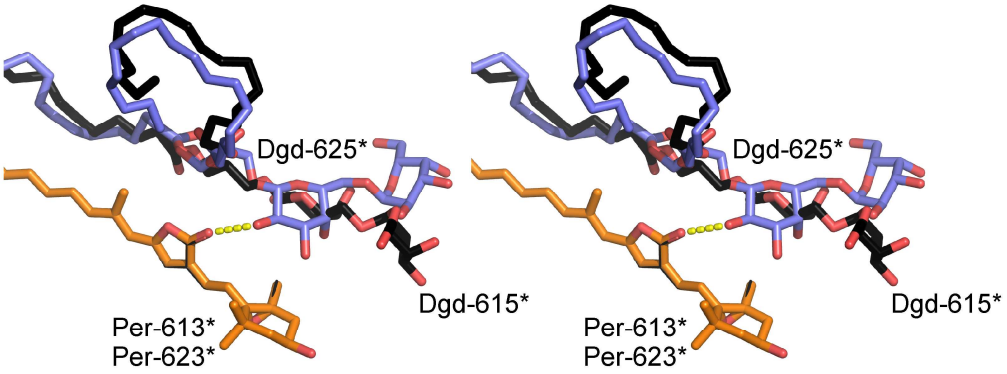

figure S9 H-bond of Dgd-625* (blue) to the CCG of Per-623* (orange) (stereo view). Superimposed is the Dgd-615* position in the C-domain (black).

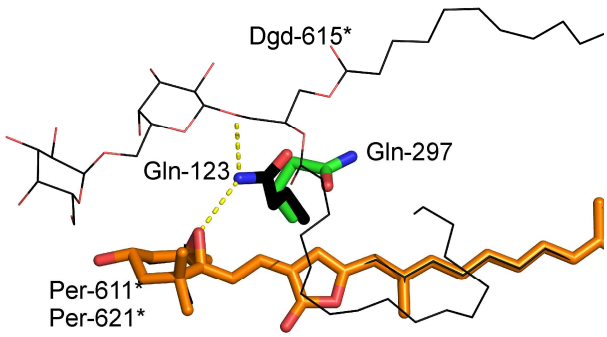

figure S10 Positioning of Gln123 in N-domain (black) and Gln297 in C-domain (green) in proximity to Per-611*/Per621*.