

Supporting information

Melatonin activation by Cytochrome P450 isozymes. How does CYP1A2 compare to CYP1A1?

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1 Methodology

1.1 Primary model preparation

Here is the outline of the steps involved in preparing a structure for a molecular dynamics simulation as starting from a protein databank (PDB) file:

1. Select the PDB file: The first step was to select the enzyme for this study by fetching the data from rcsb.org¹. The enzyme in this study was CYP1A2. We selected the chain A of 2HI4² PDB structure which was the latest released structure and with good resolution.
2. Clean the PDB file: The next step was to remove unnecessary residues (solvent, glycerol, ions) and/or ligands and ensuring that all residues were properly named and had their correct residue types (ATOM or HETATM). This was done using a text editor software and Chimera UCSF.
3. Add missing atoms: In this step, the heme-iron complex was modified to a Compound I (Cpd I) structure. The additional oxygen atom was added over the central iron atom with bond length 1.686 Å, which was a distance taken from a previous study using the quantum mechanics method³. This was customized using Chimera UCSF⁴.
4. Add hydrogen atoms: Once all of the missing atoms were corrected and presented in the PDB file, the hydrogen atoms were added using AmberTools 2018⁵.
5. Prepare the substrate: Melatonin was the substrate of this study. It was fetched from the ML1⁶ PDB and reoptimized using a quantum mechanics method in Gaussian version 16⁷. The optimized structure was converted to a PDB file by the Antechamber⁸ software package provided by AmberTools 2018.

1.2 Substrate docking

Here is the outline of the steps involved in docking a substrate into the active site of the PDB file:

1. Prepare the receptor: The first step was to prepare the receptor, which was the well-prepared structure from section 1.1.
2. Prepare the ligand: The next step was to prepare the ligand, which was the small molecule that will be docked into the receptor, namely melatonin.
3. Select the docking software: In this study, we used AutoDock Vina⁹ which uses a generic algorithm to search for the lowest energy configuration of a small molecule in the active site of a protein.
4. Set up the docking simulation: Once the receptor and ligand have been prepared, the next step was to set up the docking simulation. The size of the simulation box was 20.0626, 23.4102, 21.855 with the center coordinate at 5.74371, 17.7915, 17.8511. The box was placed at the center of the enzyme active site containing CpdI, the I helix, the G helix and the B' helix. The maximum energy difference between each search was set to 2 kcal/mol. The maximum number of binding modes was set to 10. This resulted in 10 binding poses which were further and used for the molecular dynamics simulations.
5. Save different docked substrate poses: The ten low-energy binding poses of melatonin were separately saved as PDB files.

1.3 Molecular dynamics simulation

Here is the outline of the molecular dynamics simulations of the heme-iron complexes:

1. Prepare the mol2 files for non-standard molecules: Amber needs the information about non-standard chemical compounds which was in the mol2 file format. There are atom names, atom types, bond types, bond orders, etc. This was done using Antechamber.
2. Prepare the frcmod files for non-standard molecules: The forcefields for non-standard molecules were not available in Amber. This information can be prepared by converting data from mol2 file using parmchk2 software package provided in AmberTools 2018.
3. Generate forcefield for heme-iron complex: The atoms within 2.5 Å around the central iron atom in the porphyrin are optimized by quantum mechanics and the output was transferred to forcefield parameter. This was generated by MCPB.py¹⁰ that was the python-based software package provided on Amber.
4. Solvate the protein: Once the force field for non-standard residues were generated, the whole system was solvated in water using the TIP3P¹¹ forcefield while the standard amino acids were performed by ff14SB¹² forcefield. The system was neutralized by Na⁺ and Cl⁻. The rectangular solvate-box was set to a size of 10 Å, which was the distance between the box edges and the protein. This can be done using Amber's tleap software package.
5. Minimize the system: Once the system was solvated in water and neutralized, it was minimized to remove any bad contacts between the molecule and solvent. The steepest descent was set to 2000 with a cutoff set to 10 Å.
6. Heat the system: In this step, the system is heated up to bring the system into a more stable state. The system is heated up to a temperature of 310 K during a 10 ns simulation.
7. Run the MD production: Finally, the system was run for 100 ns for the general study and 1000 ns for the comparison of CYP1A1 and CYP1A2. It was run under the constant temperature and pressure condition at 310 K and 1 bar by running through multiple loops of 5 ns.
8. The calculations were run on different intermediates along the mechanisms for O-demethylation and aromatic hydroxylation. The labels of each data were presented in the following figure:



Where:

- A is the intermediate created, including reactant (Re), intermediate 1 (IM1), intermediate 2 (IM2), product (P).
- B is the reaction pathway, including O-demethylation (ODE) or aromatic hydroxylation (HYD).
- C is the model number as taken from the MD simulations that ranges from 1 to 10.

1.4 Data extraction from molecular dynamics simulation

Once the molecular dynamics simulation was finished, all results from multiple loops were combined using the cpptraj software package provided on Amber and pytraj¹³ which was python-based library. The data was in csv format and illustrated by the python libraries named plotly and seaborn. For the geometry visualization, it was presented by Chimera UCSF.

1.5 Data selection of molecular dynamics simulation

Here is the outline of the data selection from the molecular dynamics simulations:

1. Compare the structure at the start and end of the molecular dynamics simulation: Focus on the protein changes and substrate-binding orientations of each system in the snapshots between 1 ns and 100 ns. The results are shown in Figure S1 to S6.
2. Determine the RMSD: The RMSD of each structure was extracted to see the stability of protein, substrate and heme in the molecular dynamics simulations. The results are shown in Figure S7 and S8.
3. Compare the RMSD from different intermediates: Each structure from 90 ns to 100 ns was used as the reference structure to calculate the RMSD of the next systems. For example, the snapshot at 90 ns of reactant was used to calculate the RMSD of 10 models from 1 ns to 100 ns. The RMSD of the comparison from 90 ns to 100 ns were presented in median values. The structure that mostly gave low RMSD was selected as the reference structure for quantum mechanics calculations. That structure was the model 6 of reactant. The results are shown in Figure S9 and S10.
4. Map the substrate in the active site: The substrate orientations were mapped into the quadrants based on the heme plane by measuring the C6 position of the substrate using the pyramid method. From the previous step, the snapshot of the selected reactant at the median that had highest density on x and y axis was selected. The selected snapshot of model 6 reactant was 80.4 ns. The results are shown in Figure S11 and S12.

1.6 Modification of small model for quantum mechanics simulation

The amino acids within 5 Å of the melatonin are reported in Figure S13 with the distributions of amino acids. Those residues are included in the active site cluster model for which quantum mechanics calculation were performed. However, some residues or side chains had to be truncated because of the size limitation of the quantum mechanics calculation. The truncated model had 283 atoms. The result is shown in Figure S14.

1.7 Quantum mechanics simulation

The quantum mechanics calculation was performed with Gaussian version 16 by the following steps:

1. Optimize cluster models and local minima: The modified small model was optimized with basis set 1 (BS1) and the UB3LYP density functional approach. The iron (Fe) atom was described with the LANL2DZ+ECP basis set and the rest of the atoms employed 6-31G*. Calculations were performed at 298.15 K (25°C) in the doublet and quartet spin multiplicities and overall charge -2.
2. Geometry scans for describing specific chemical reaction pathways: The atoms involved in the O-demethylation and aromatic hydroxylation were moved closer to the oxo group by constraint geometry scans were the O-H/C-O distances

were shortened in a stepwise manner. Each step had fixed O-H/C-O distance and applied a full geometry optimization. During this process the electronic energy changed and followed past a maximum and leading to another local minimum. The scan was run until the next local minimum was reached using the computational methods as described in (1).

3. Optimize transition state: The structure at the maximum energy point was reoptimized as a transition state and with basis set BS1 with the same conditions as in (1).
4. Perform better basis set calculations: The optimized structures were subjected to the single point calculation with basis set 2 (BS2) and the UB3LYP density functional. The iron (Fe) atom was described with cc-pVTZ and the rest of the atoms with a 6-311++G* basis set. Energies were calculated at 298.15 K (25°C) in the doublet and quartet spin multiplicities and with overall charge -2.
5. Generate energy data: The optimized structures were confirmed as local minima or first-order saddle points with a frequency calculation with BS1 under the same condition in (1) resulting in energy data including zero-point energy, electronic energy, free energy, etc.
6. The vibrational frequencies obtained from Gaussian were corrected with the Goodvibes¹⁵ software package. The calculated energy was added by the approximation for the quasi-harmonic enthalpy correction. It replaced harmonic energy contributions with a quasi-RRHO vibrational energy term via the Head-Gordon expression at the 298.15 K and 1 atm.

1.8 Chemical reaction determination of quantum mechanics simulation

Here is the outline of the chemical reactions determined by quantum mechanics calculations:

1. Extract energy data: The thermochemistry data was extracted and includes zero-point energy, electronic energy, electronic energy with zero-point energy and free energy. The chemical reaction energies were determined from relative energies which were derived from absolute free energies.
2. Illustrate energy profile: To give the insight of chemical reaction, the energy profile was created from the relative energies from BS2 single point calculation using Chemdraw¹⁴ software.
3. Extract charges and spin densities: The electron configurations and orbitals were determined by charges and spin densities.
4. Extract geometry data: Geometry information presented the positions of the atoms relative to doublet and quartet multiplicities. The geometry data including bond lengths, angles and dihedrals were extracted by a python-based library.
5. Extract imaginary frequencies: The imaginary frequency relates to the activation energy that is required to overcome the energy barrier and initiate a chemical reaction. It was extracted by data mining using python.

2 Reference

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10.5281/zenodo.1435820.

3 Binding poses of CYP1A2 systems from molecular dynamics simulation

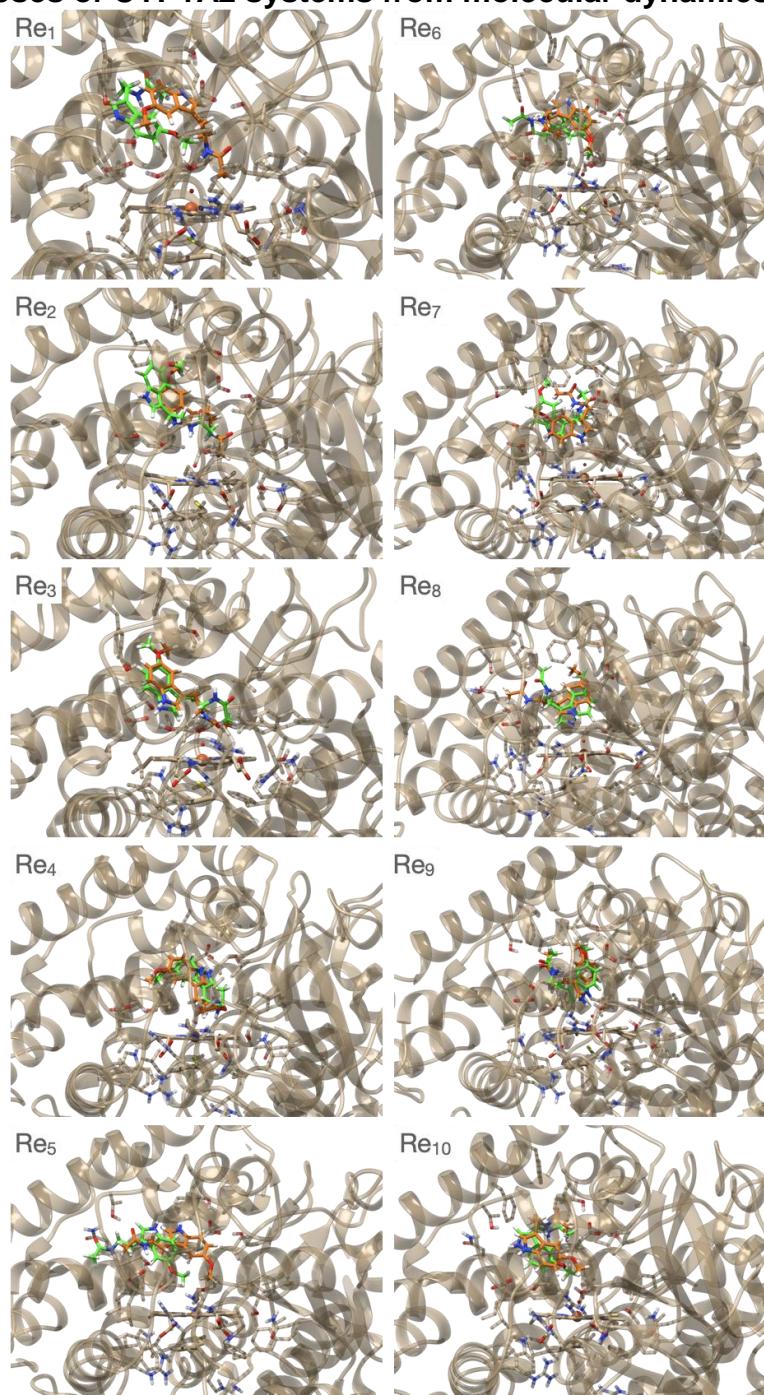


Figure S1 Binding poses of the reactant complex as obtained from 10 MD simulations of 10 different starting orientations of substrate. The substrate orientation structures at 1 and 100 ns are presented in orange and green.

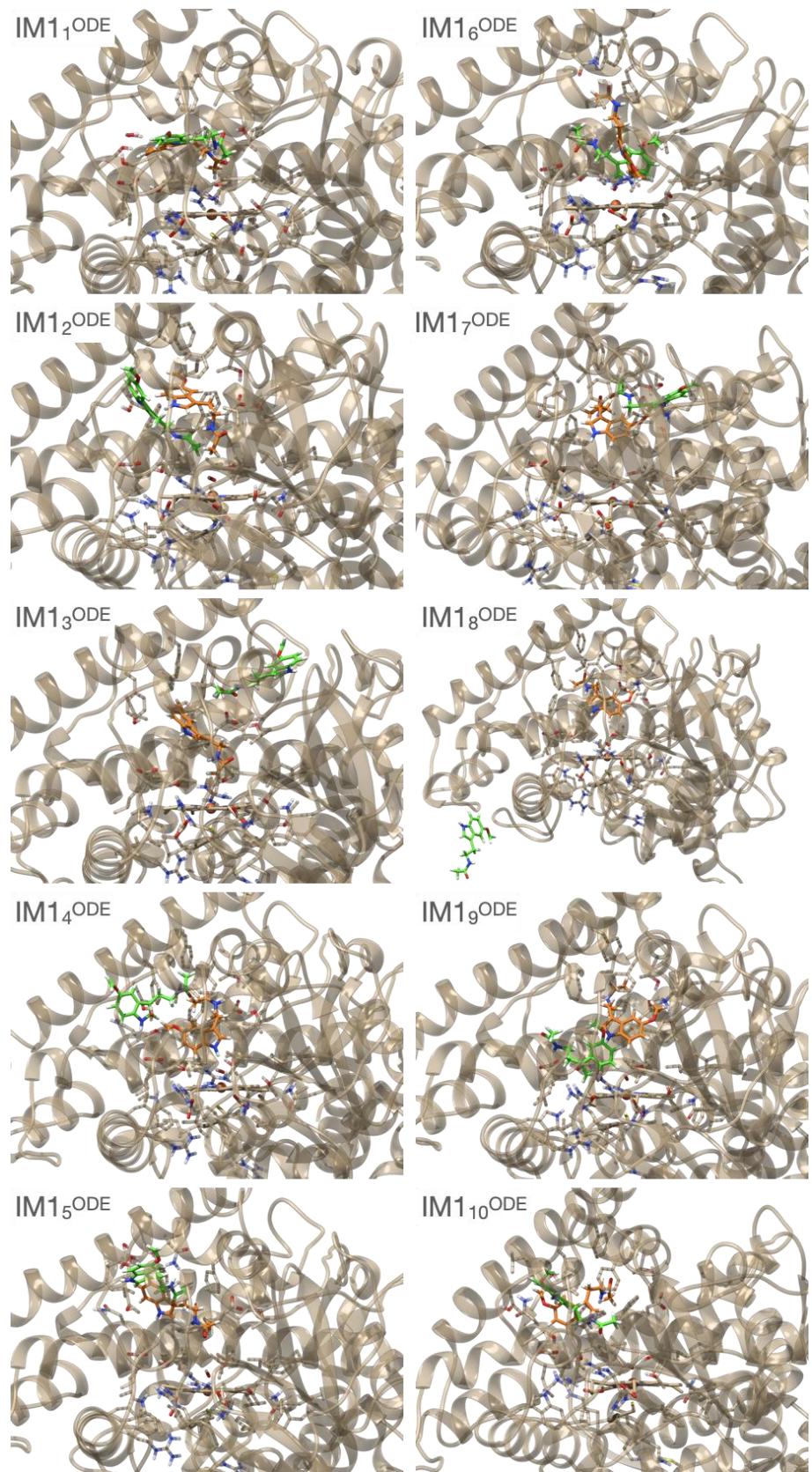


Figure S2 Binding poses of the substrate in intermediate-1 along the O-demethylation pathway in 10 models as obtained from MD simulations. Structures at 1 and 100 ns are presented in orange and green.

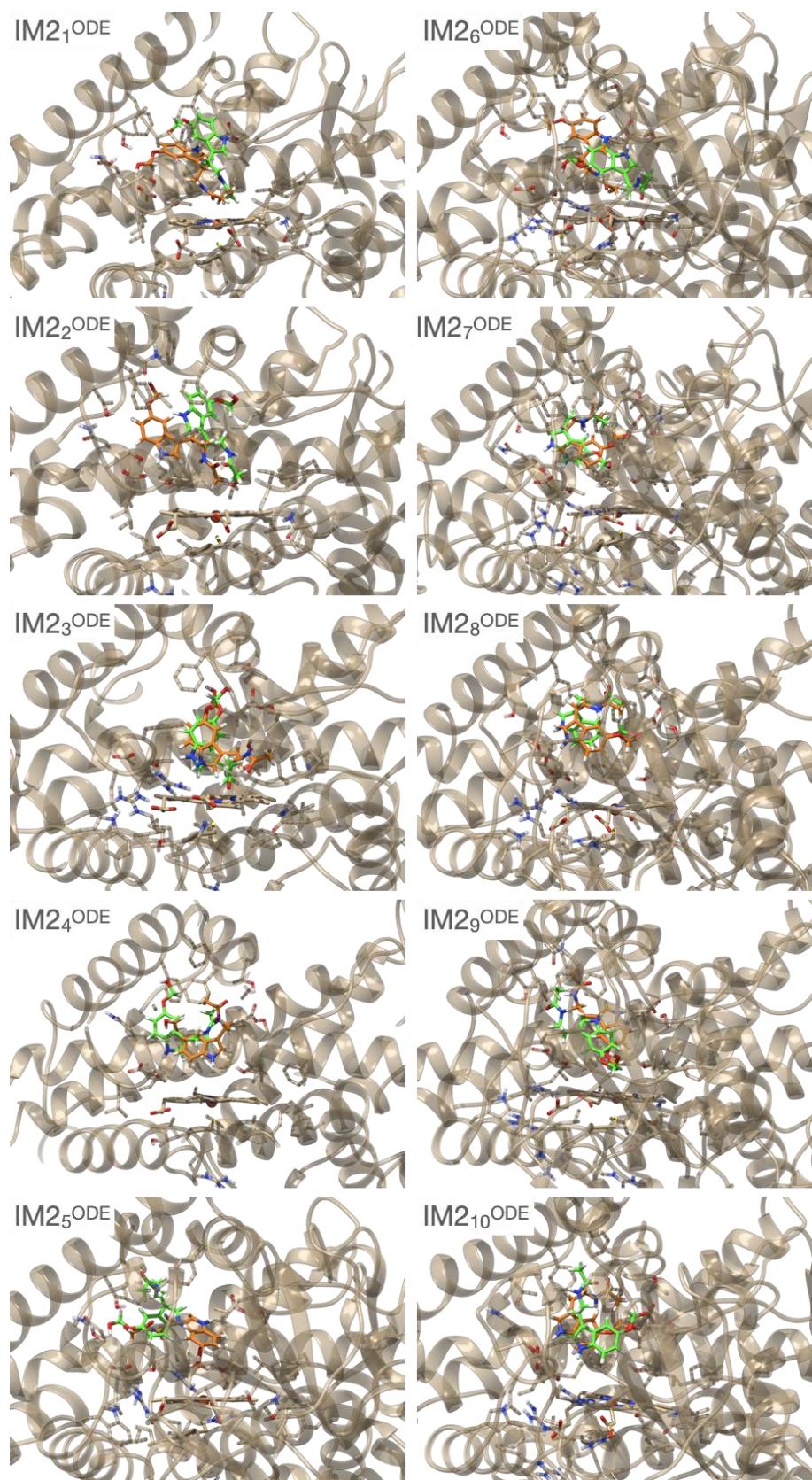


Figure S3 Binding poses of the substrate in intermediate-2 along the O-demethylation pathway in 10 models as obtained from MD simulations. Structures at 1 and 100 ns are presented in orange and green.

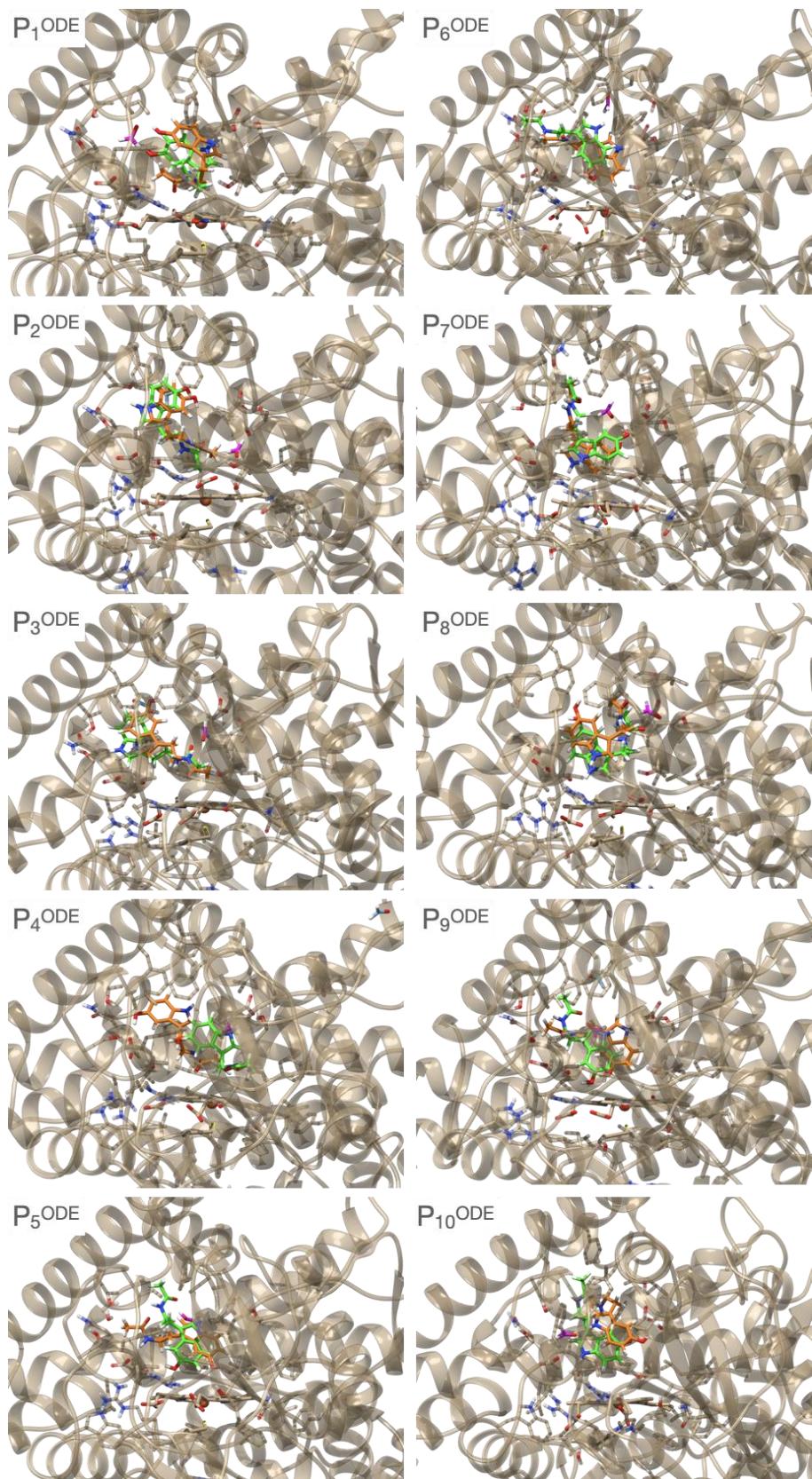


Figure S4 Binding poses of the substrate in product along the O-demethylation pathway in 10 models as obtained from MD simulations. Structures at 1 and 100 ns are presented in orange and green.

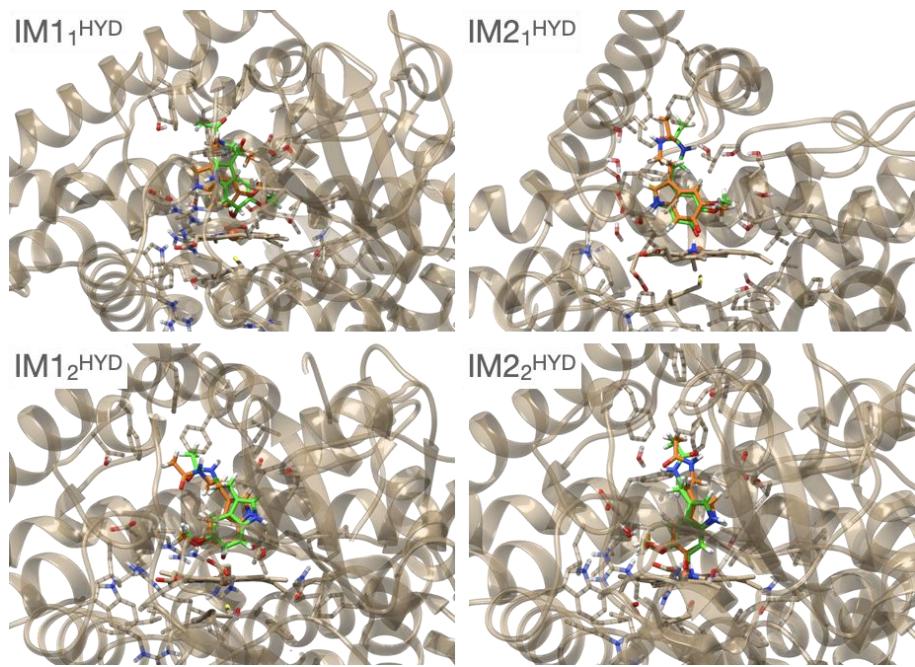


Figure S5 Binding poses of the substrate in intermediate-1 and intermediate-2 along the aromatic hydroxylation pathway in 2 models as obtained from MD simulations. Structures at 1 and 100 ns are presented in orange and green.

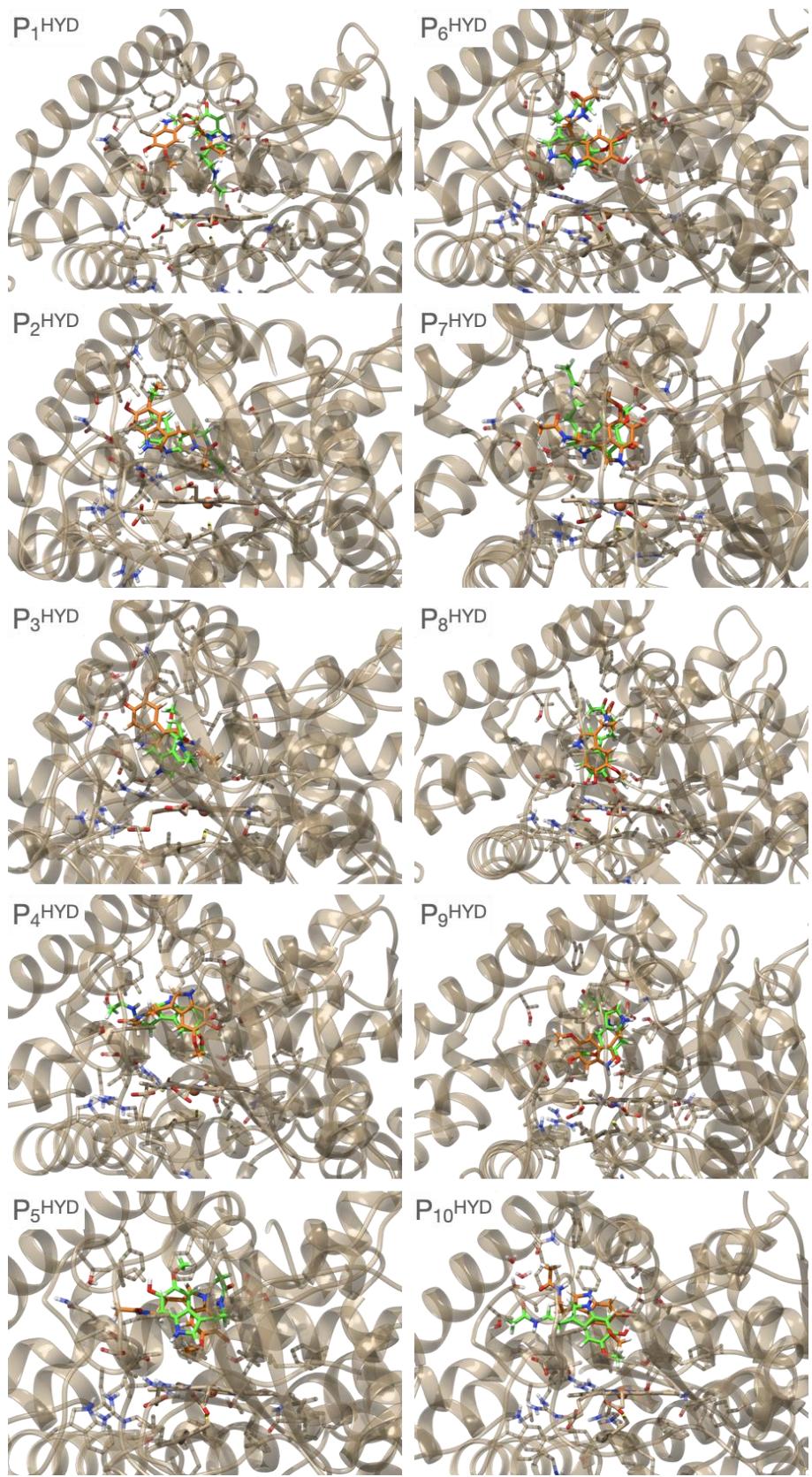


Figure S6 Binding poses of the substrate in product along the aromatic hydroxylation pathway in 10 models as obtained from MD simulations. Structures at 1 and 100 ns are presented in orange and green.

4 RMSD of CYP1A1 systems

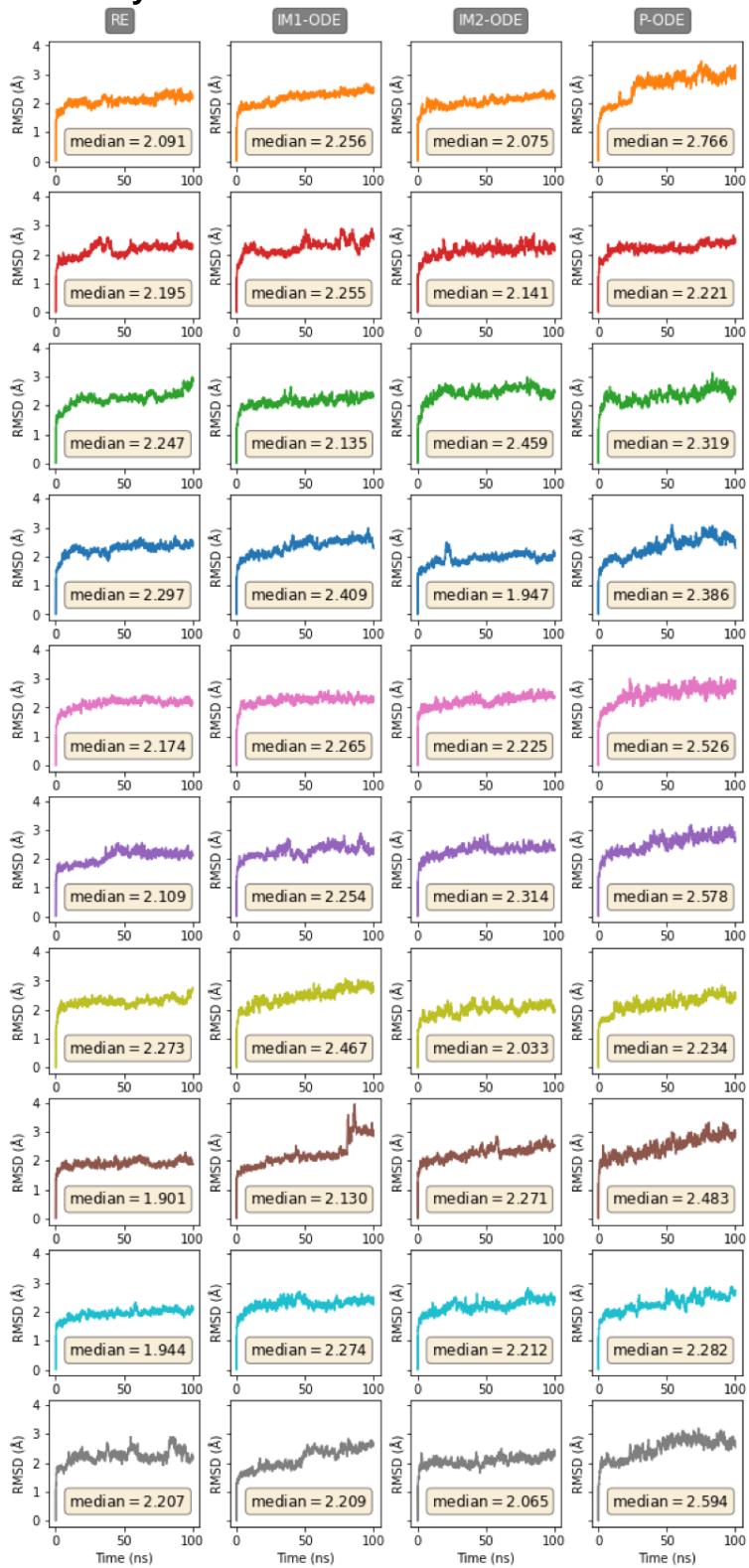


Figure S7 RMSD values of the protein for the MD simulations of various intermediates along the pathway for O-demethylation in 100 ns.

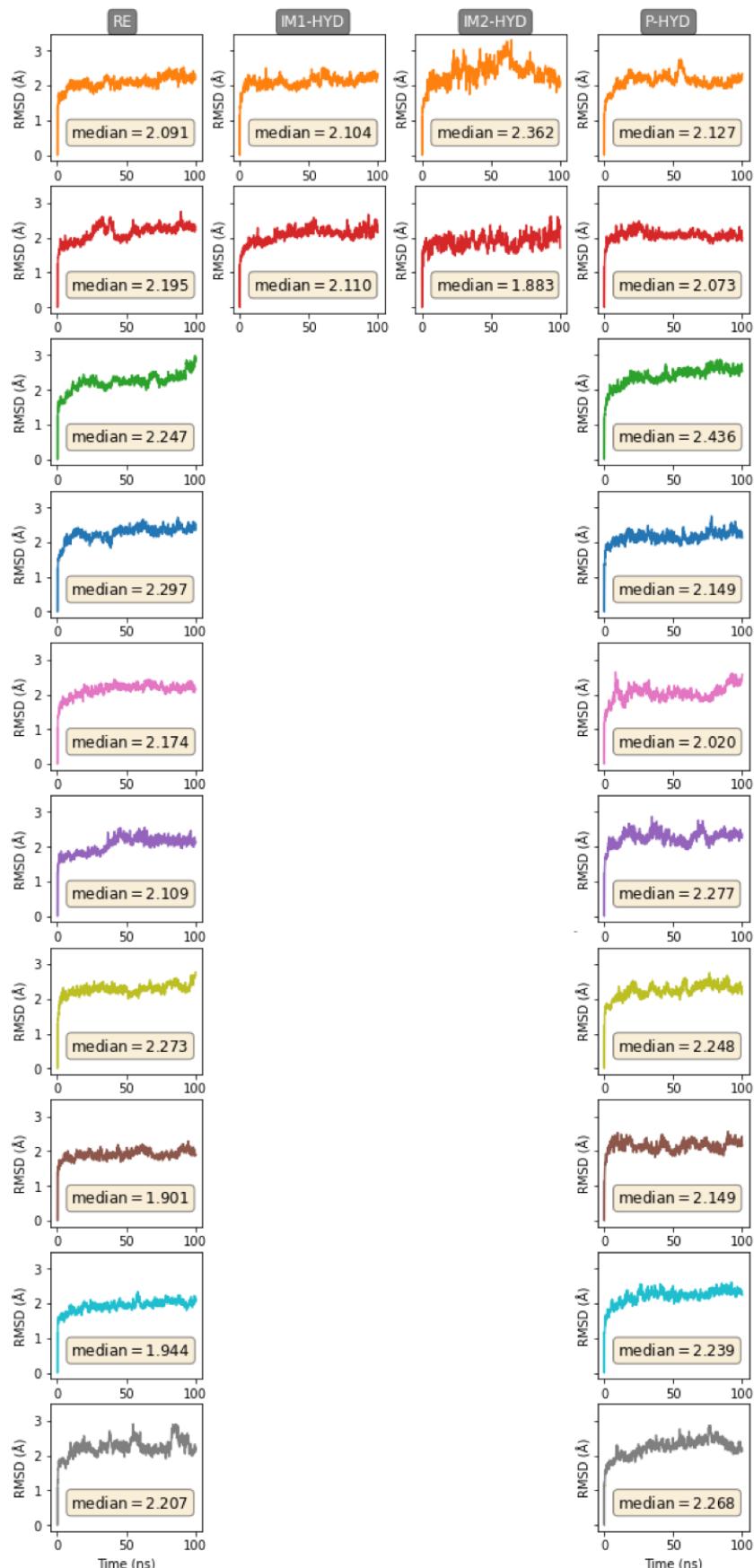


Figure S8 RMSD values of the protein for the MD simulations of various intermediates along the pathway for aromatic hydroxylation in 100 ns.

5 RMSD matching for different CYP1A2 systems

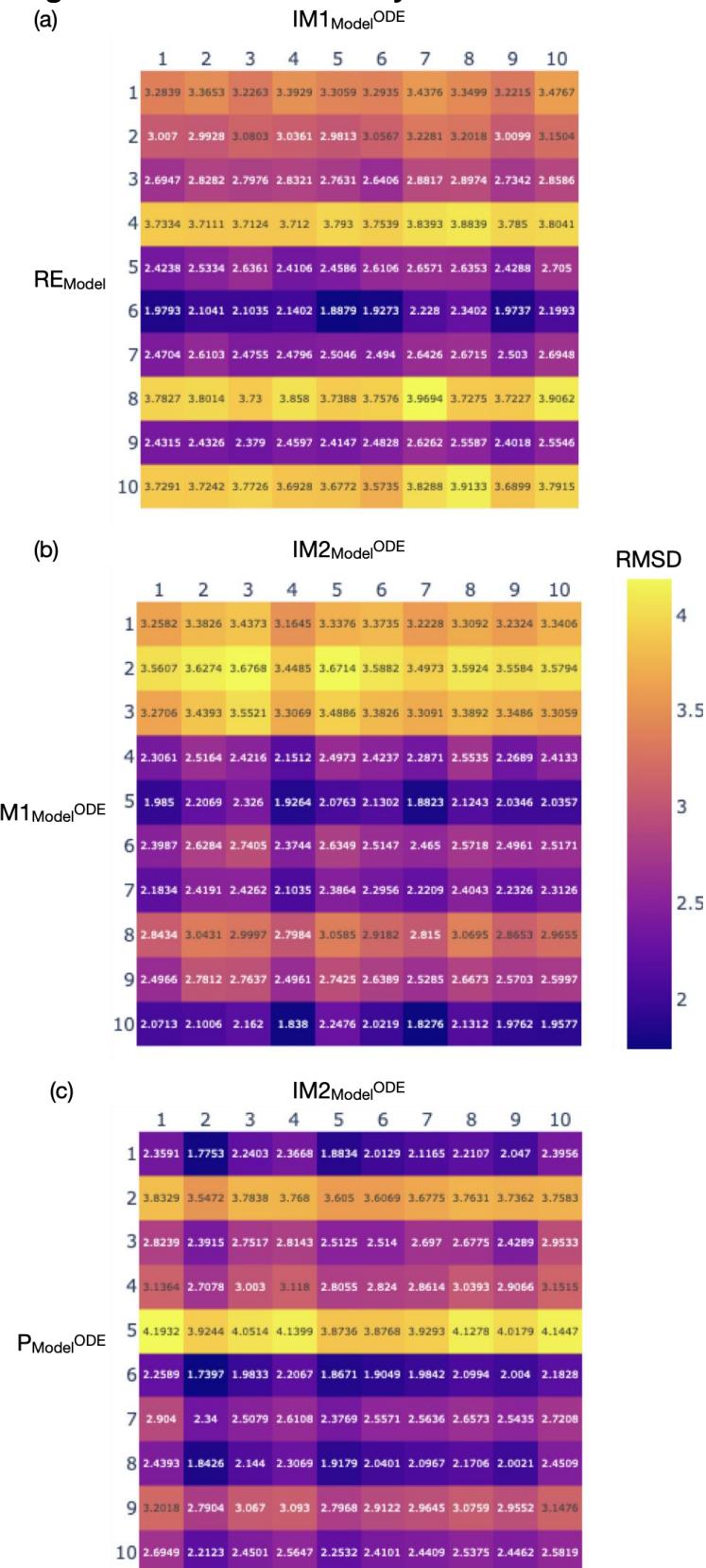


Figure S9 RMSD comparison of intermediate states for O-demethylation during a 100 ns MD simulation.

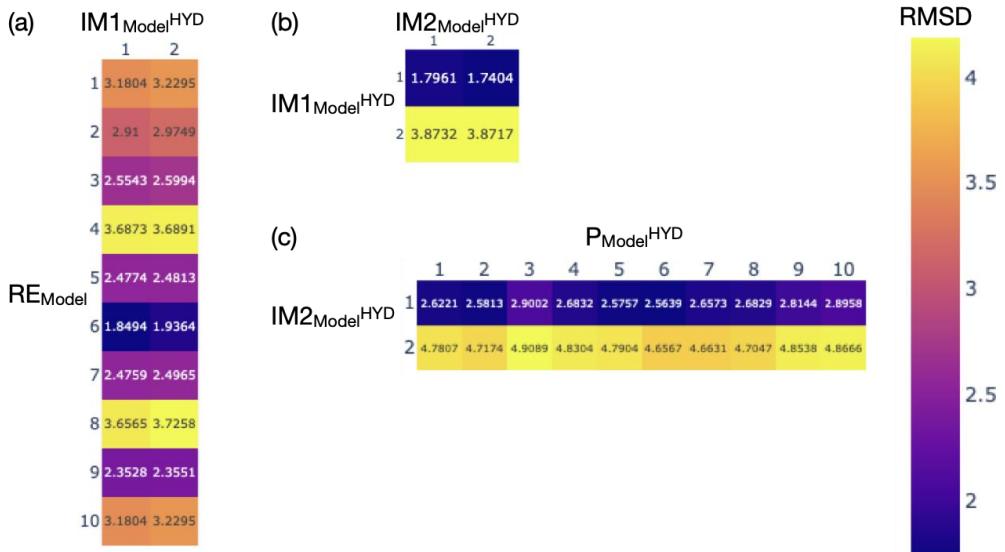


Figure S10 RMSD comparison of intermediate states for aromatic hydroxylation during a 100 ns MD simulation.

6 Substrate location in the active sites

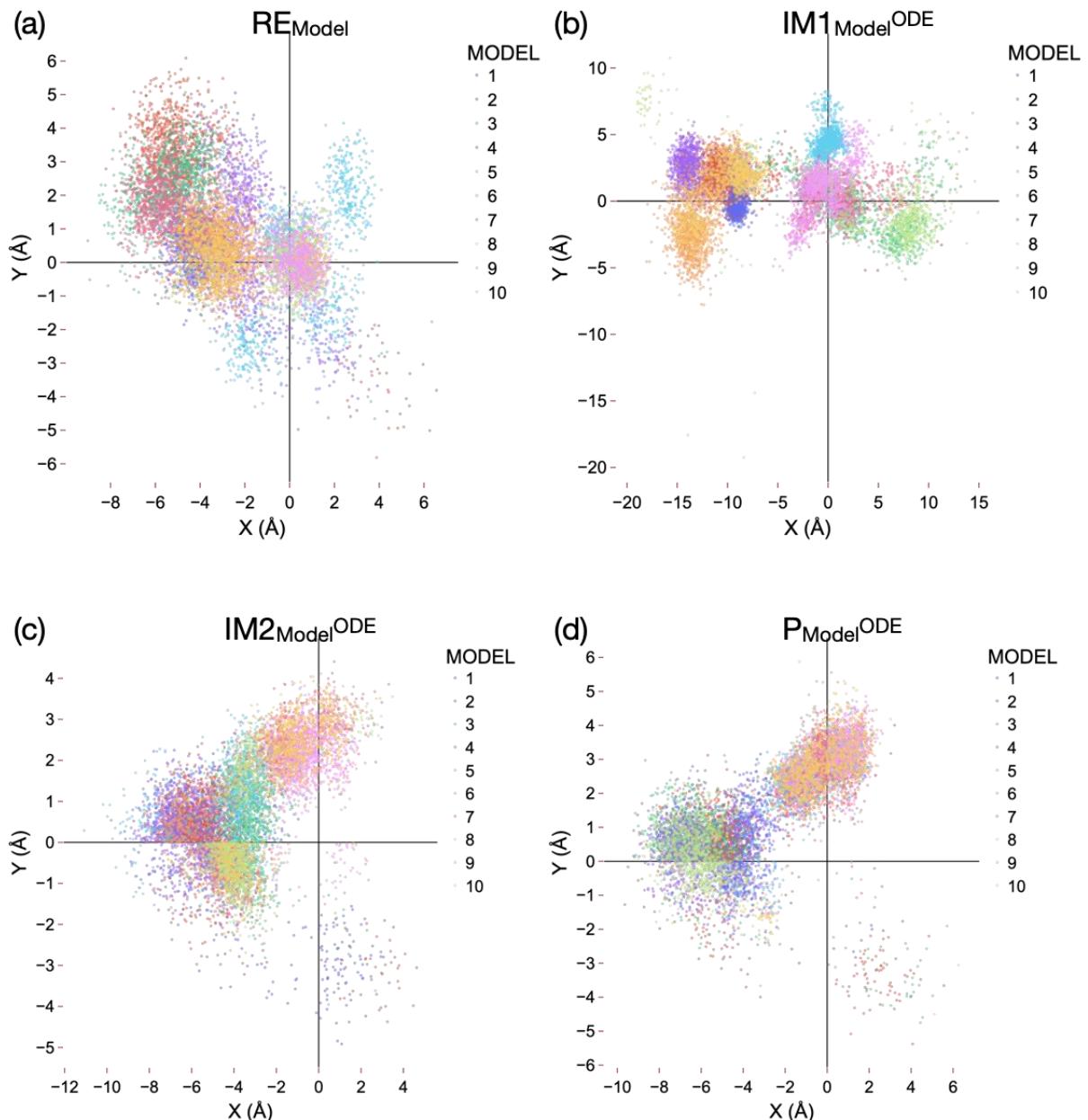


Figure S11 Substrate mapping in the active site in intermediates along the mechanism for O-demethylation as obtained from a 100 ns MD simulation.

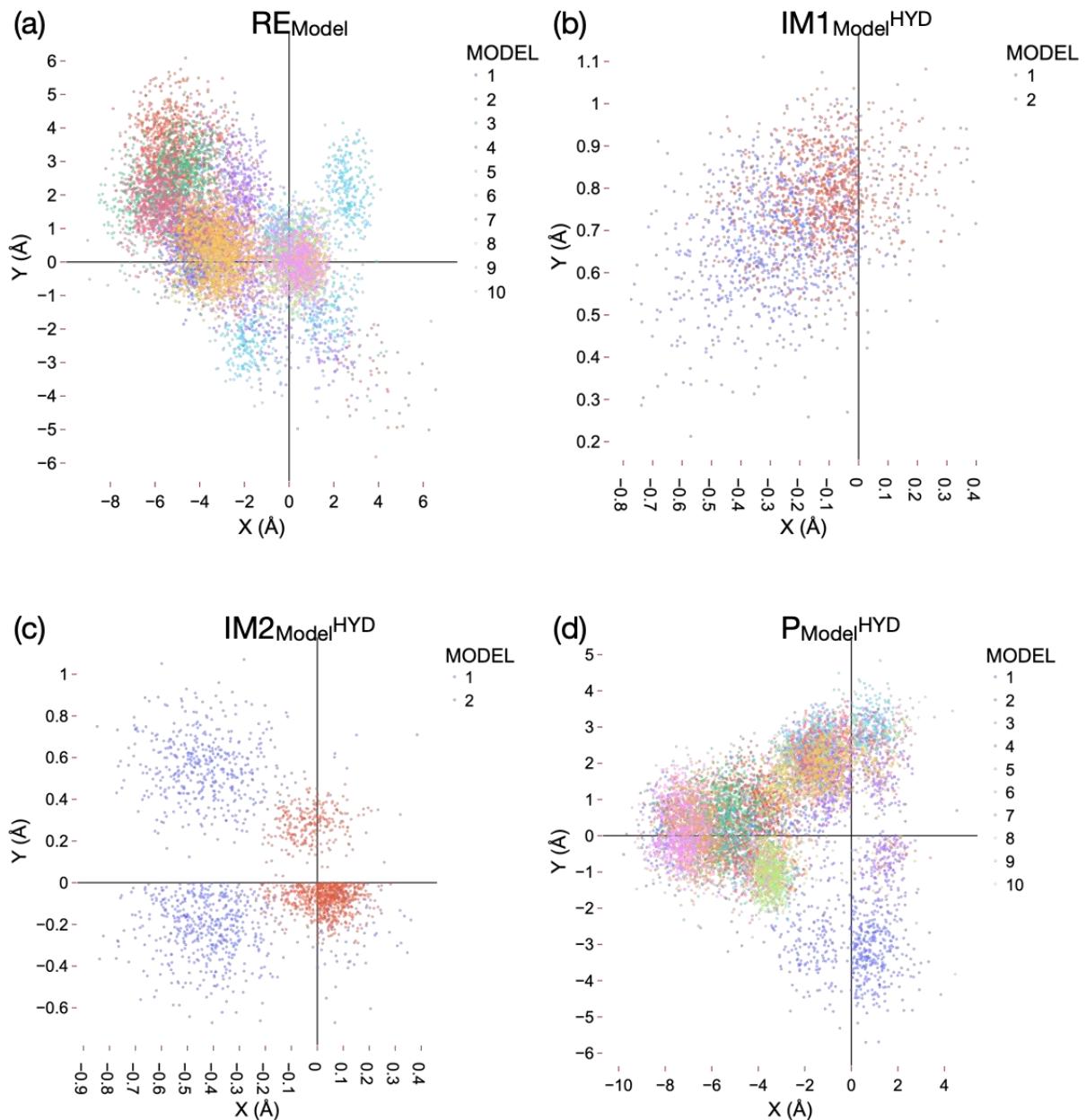


Figure S12 Substrate mapping in the active site in intermediates along the mechanism for aromatic hydroxylation as obtained from a 100 ns MD simulation.

7 Amino acid distributions

(a)

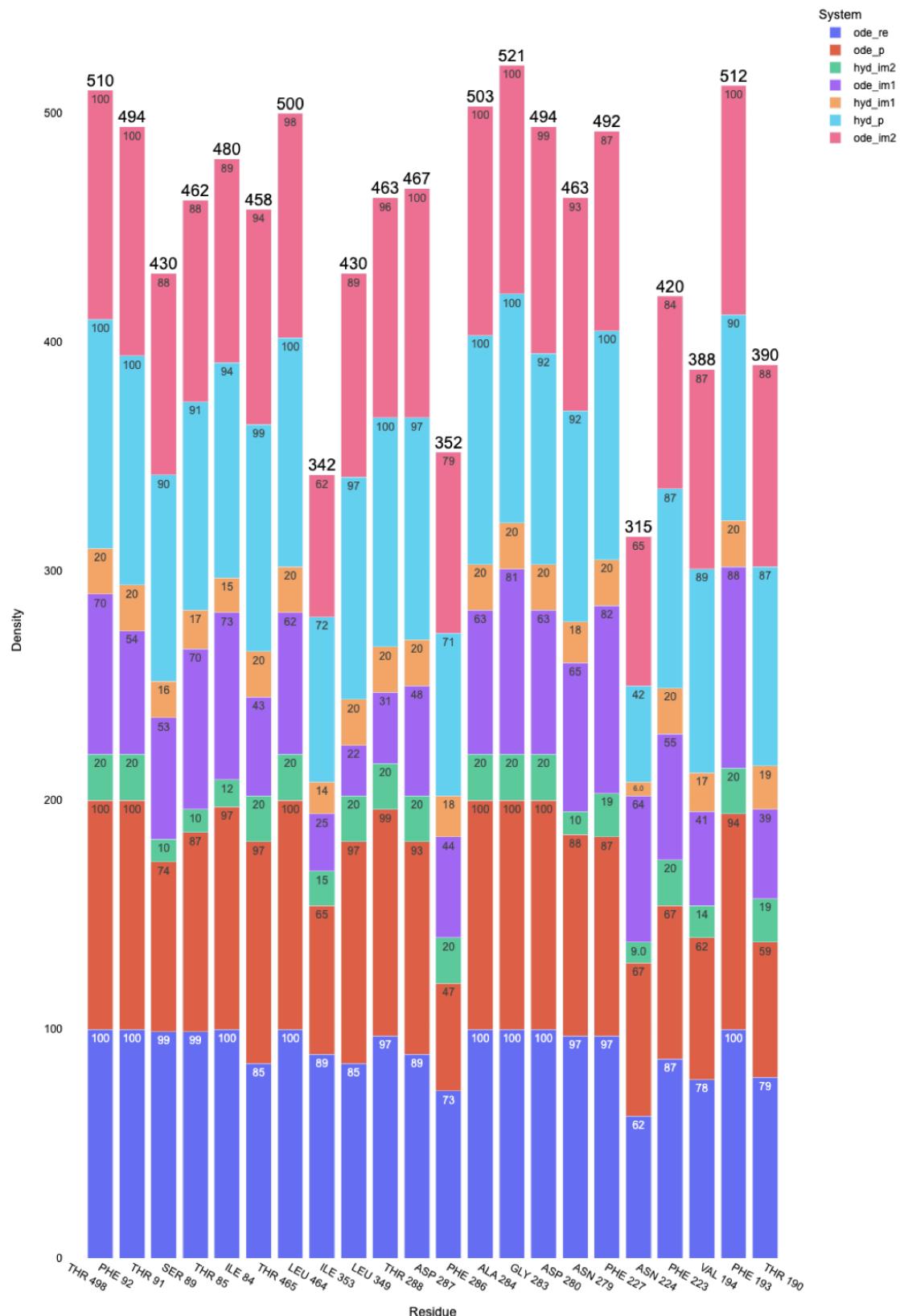


Figure S13 Amino acid distributions within 5 Å around the melatonin as derived from snapshot structures from MD simulations.

8 Linear interaction energy of 10 reactant models of CYP1A2

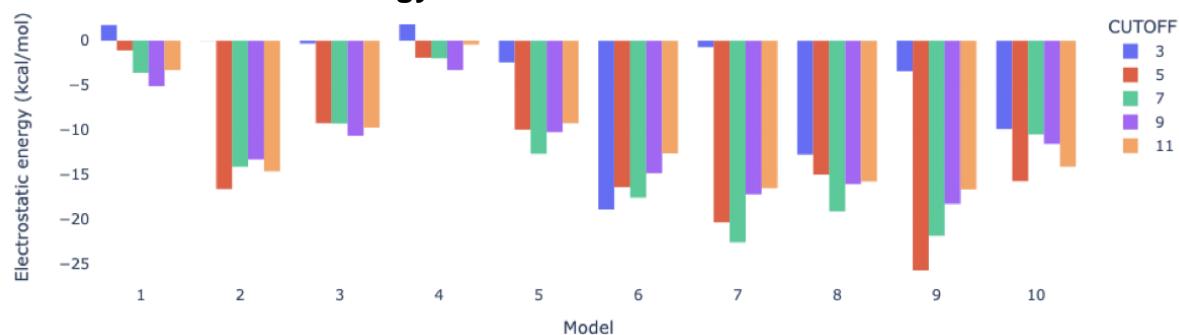


Figure S14 Linear interaction energy of the 10 reactant models as obtained from the CYP1A2 MD simulations.

9 Active site cluster of quantum mechanics simulation

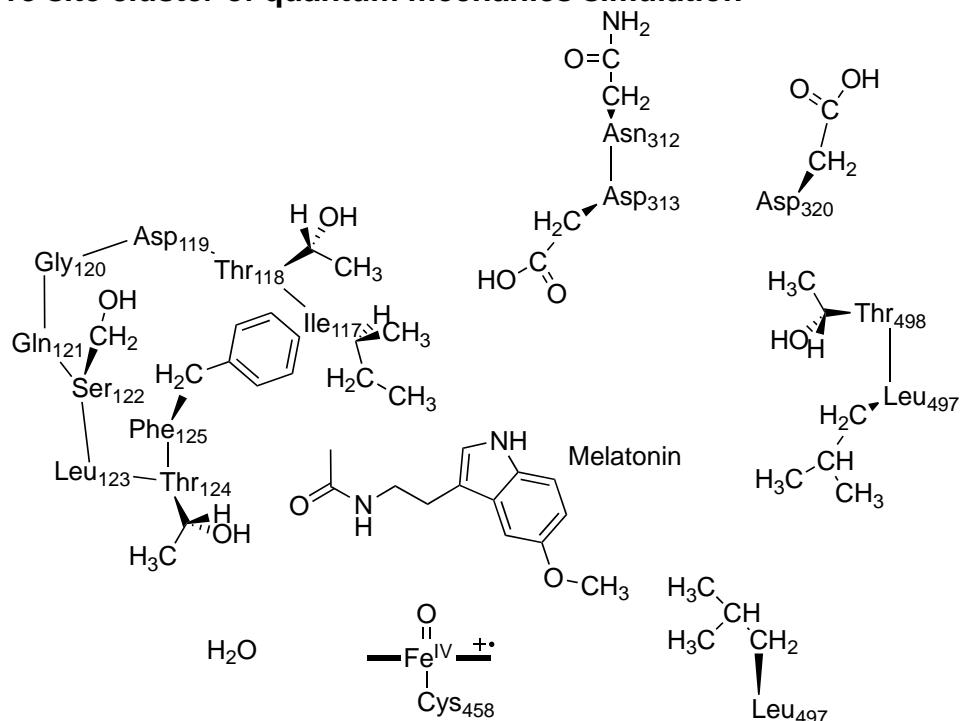


Figure S15 QM active site cluster of 283 atoms used in this work. It was selected from RE₆ at the lowest total energy (80.4 ns).

10 CYP1A1 molecular dynamics Simulation in 1 μ s

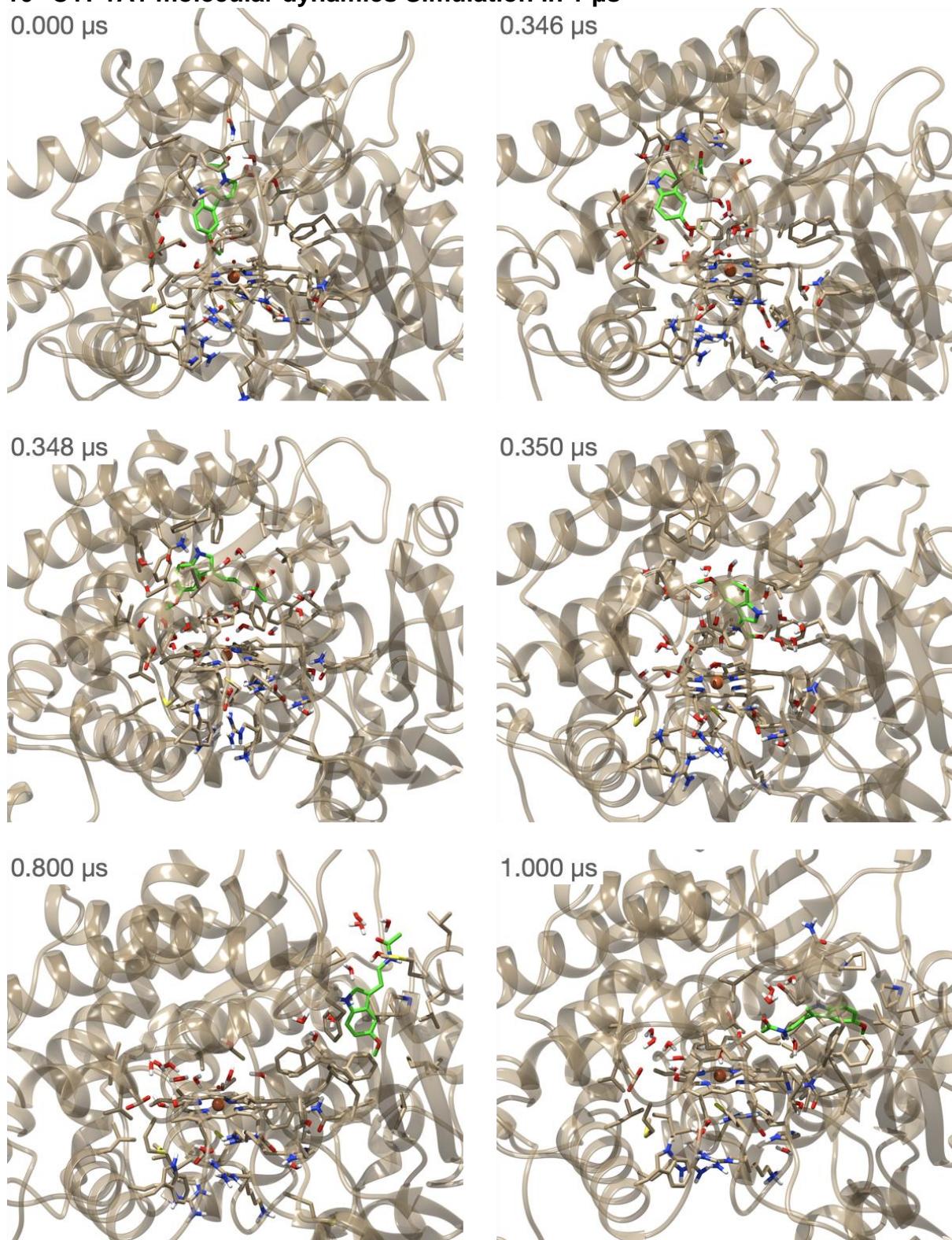


Figure S16 Binding poses of the substrate in the reactant complex in CYP1A1 as obtained from MD simulations. The starting structure was taken from Model 3 of Mokkawes et al (J. Phys. Chem. A 2022).

11 Model 6 Reactant of CYP1A2 molecular dynamics Simulation in 1 μ s

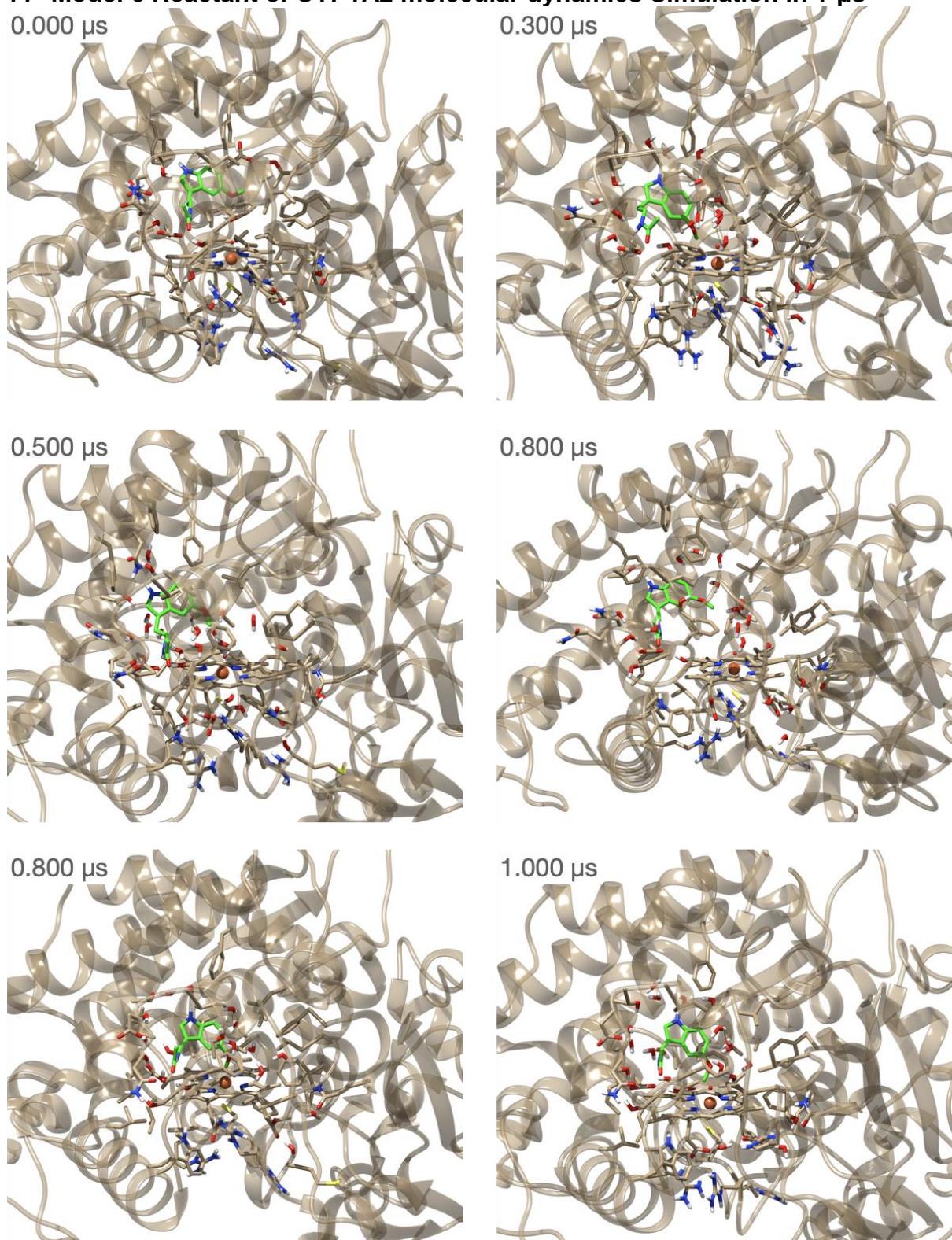
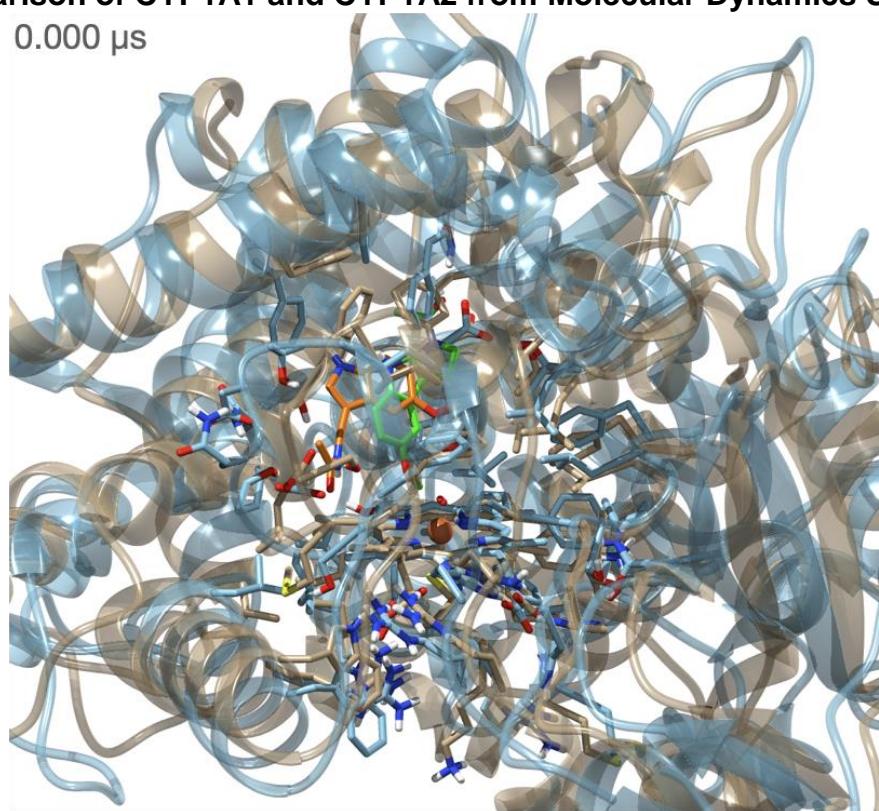


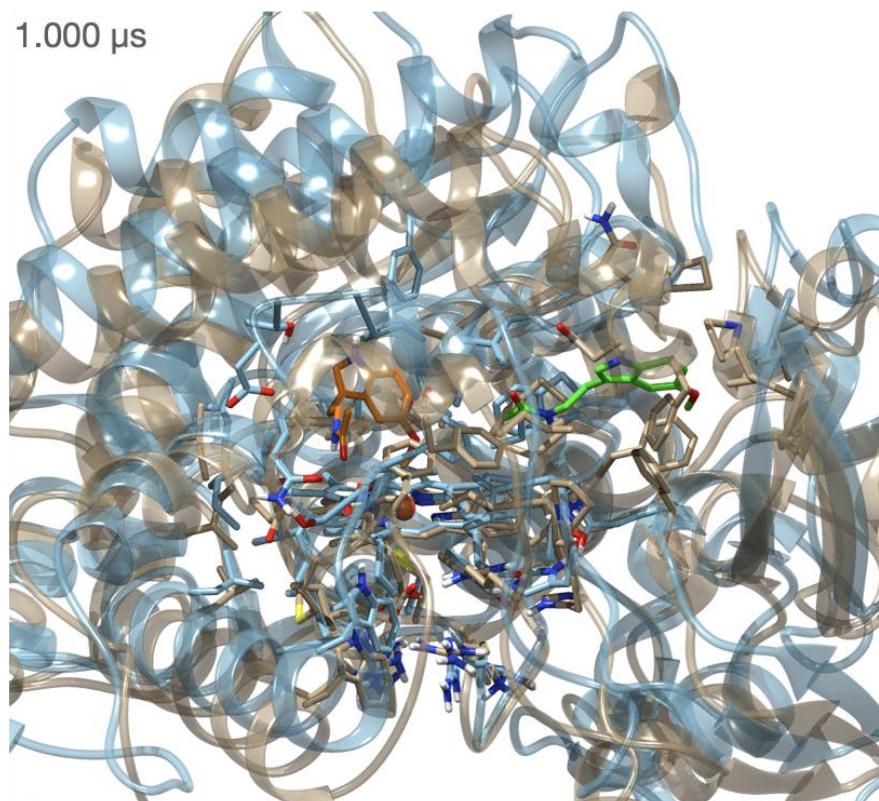
Figure S17 Binding poses of the substrate in the reactant complex in CYP1A1 as obtained from MD simulations. The starting structure was taken from Model 6 of the reactant complex discussed above.

12 Comparison of CYP1A1 and CYP1A2 from Molecular Dynamics Simulation

0.000 μ s



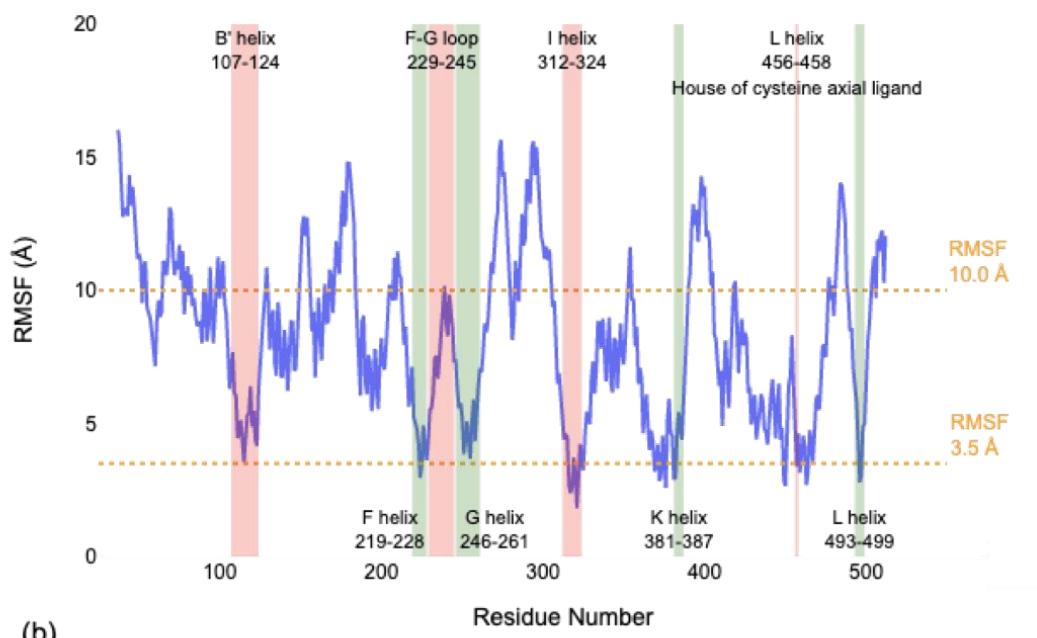
1.000 μ s



CYP1A1 Melatonin^{CYP1A1}
CYP1A2 Melatonin^{CYP1A2}

Figure S18 Overlay of the start and end point structures of the 1 μ s MD simulation for CYP1A1 and CYP1A2.

(a)



(b)

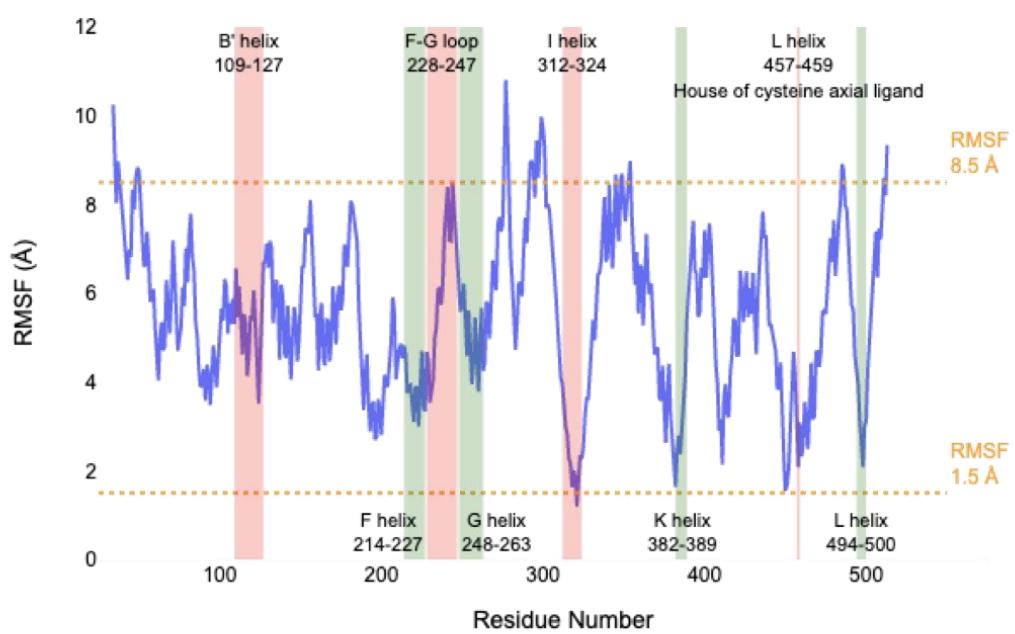


Figure S19 RMSF values of the 1 μ s MD simulation for CYP1A1 and CYP1A2.

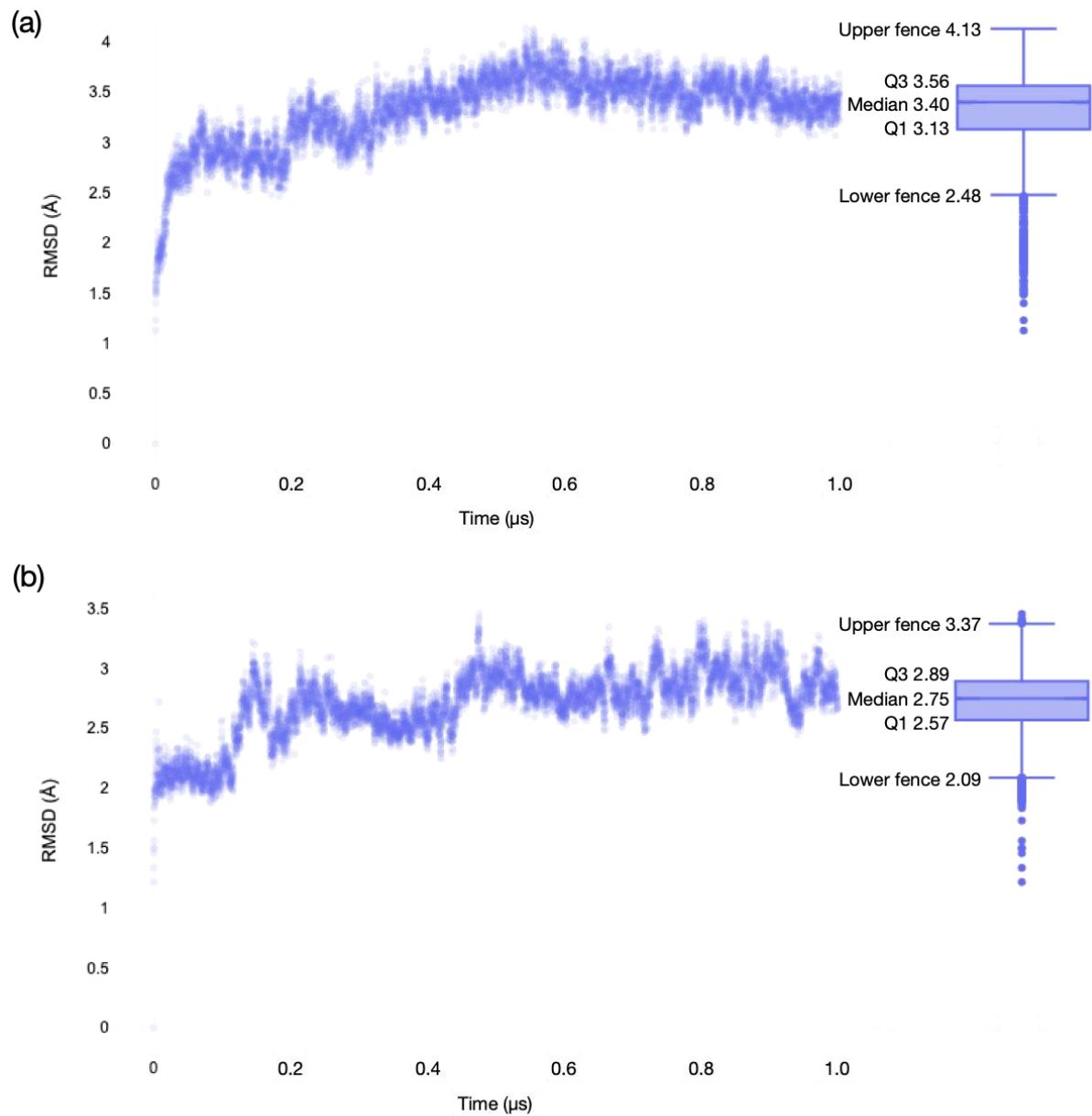


Figure S20 Enzyme RMSD values of the 1 μs MD simulations for CYP1A1 and CYP1A2.

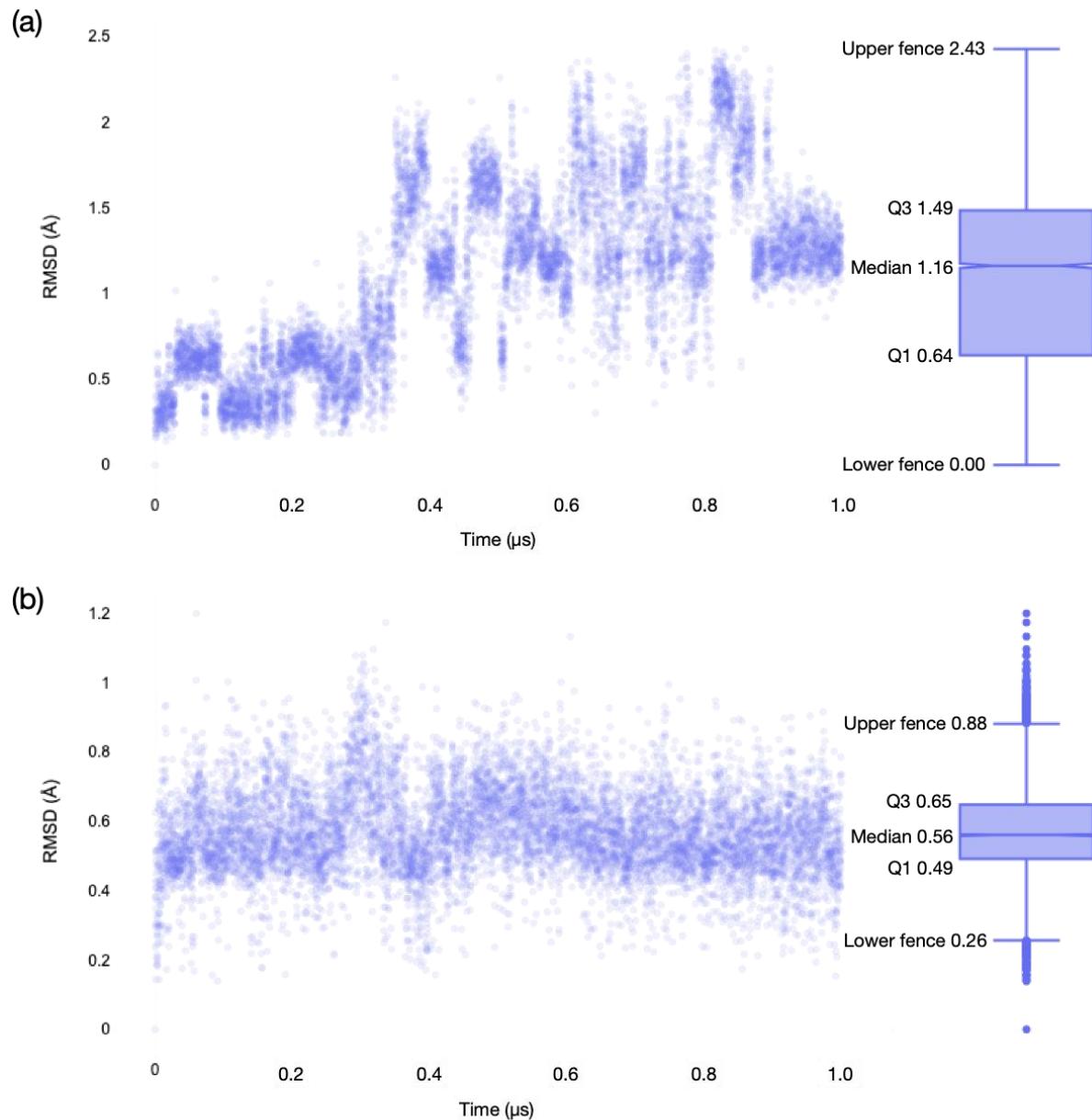
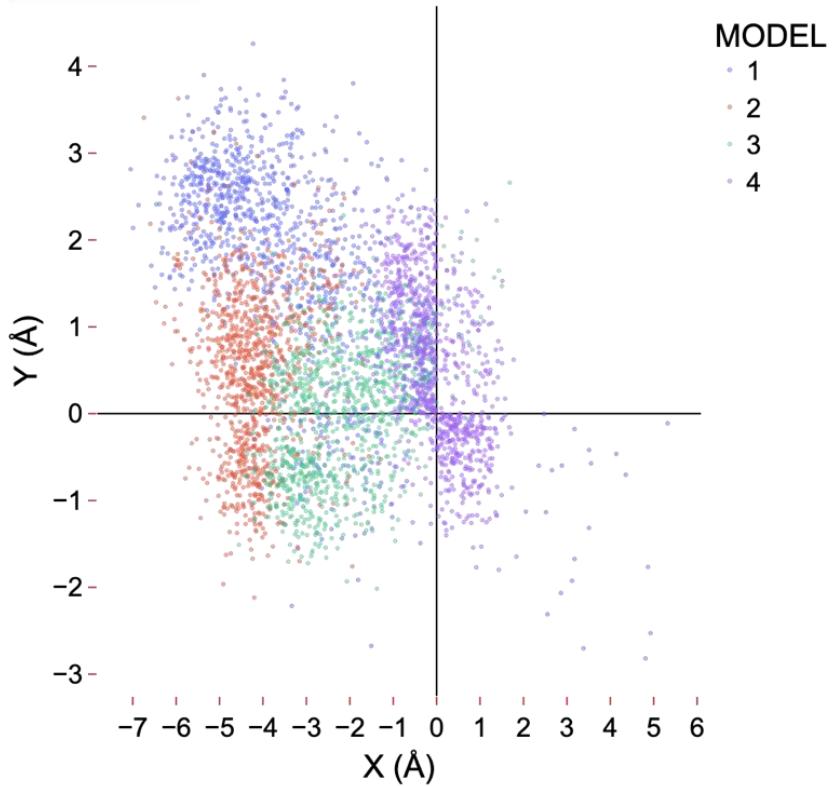


Figure S21 Substrate melatonin RMSD values of the 1 μ s MD simulations for CYP1A1 and CYP1A2.

(a) CYP1A1



(b) CYP1A2

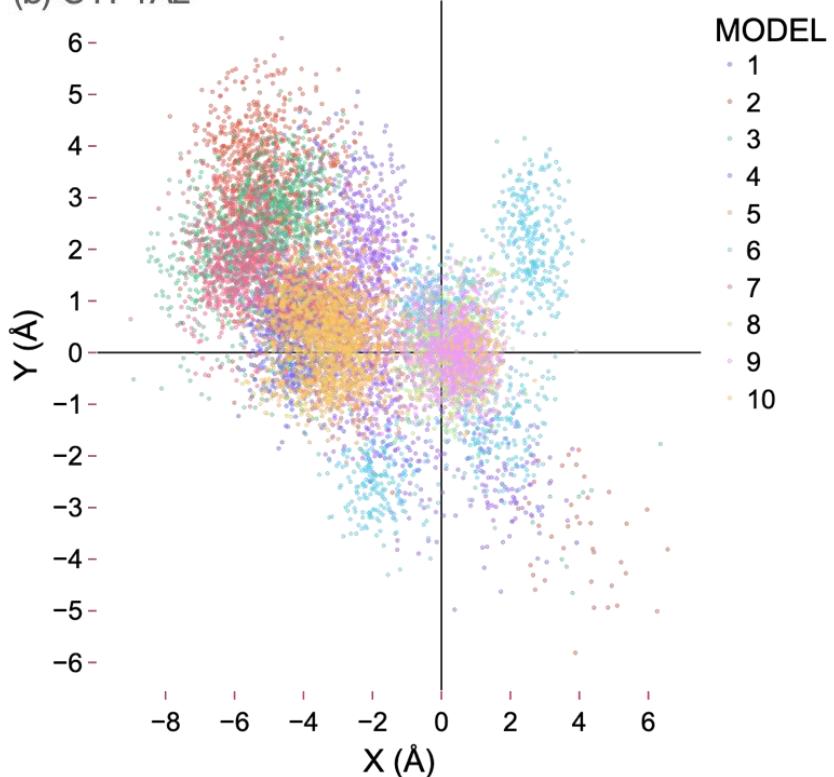


Figure S22 Substrate mapping of the reactant models as obtained from MD simulations for CYP1A1 and CYP1A2.

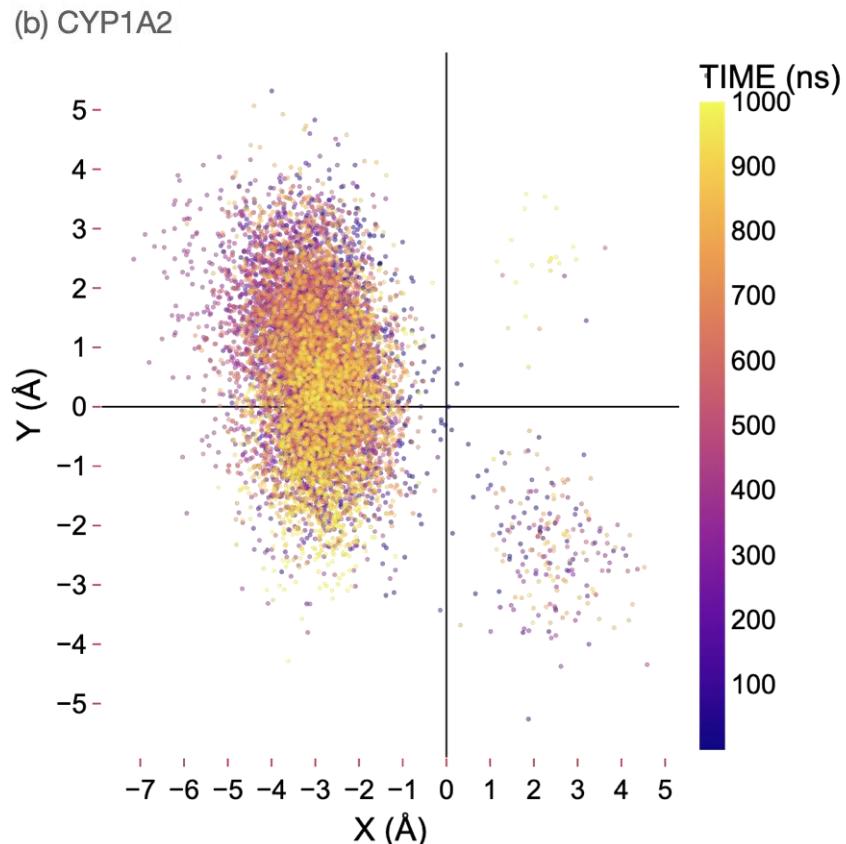
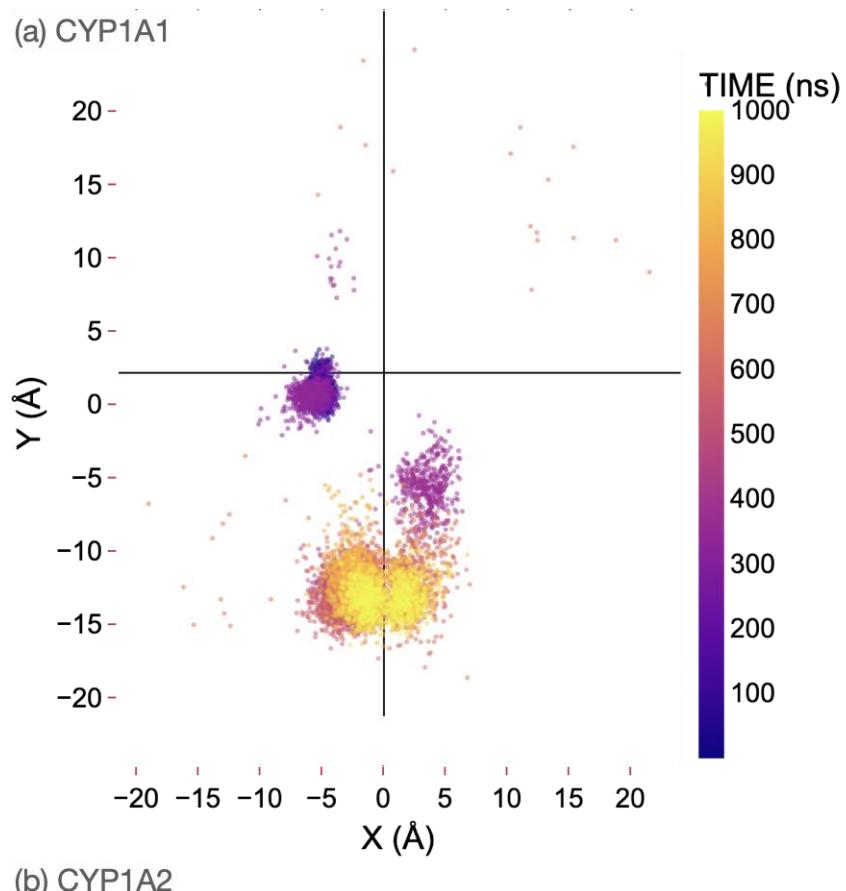


Figure S23 Substrate mapping of reactant models as obtained from the 1 μ s MD simulations for CYP1A1 and CYP1A2.

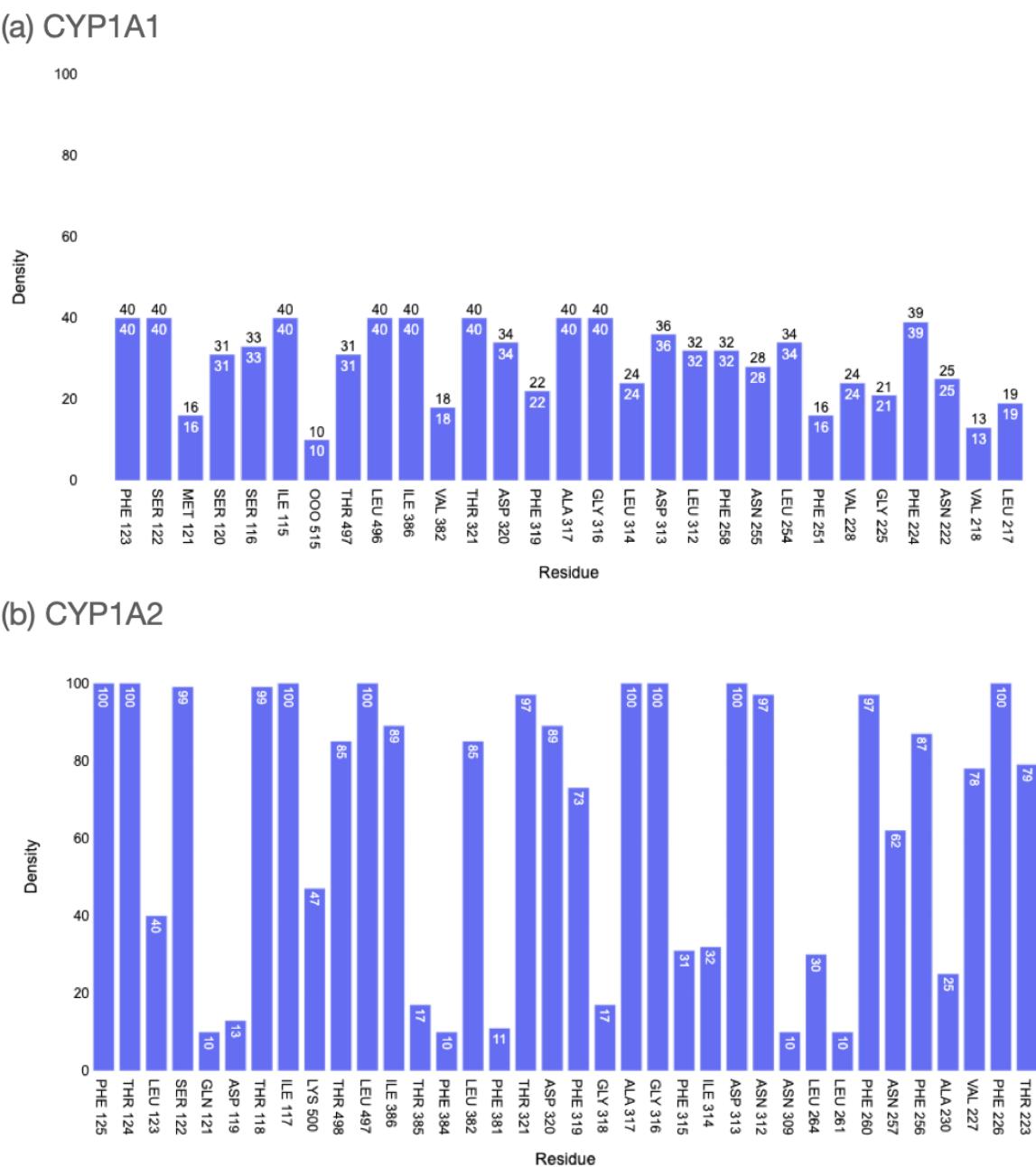
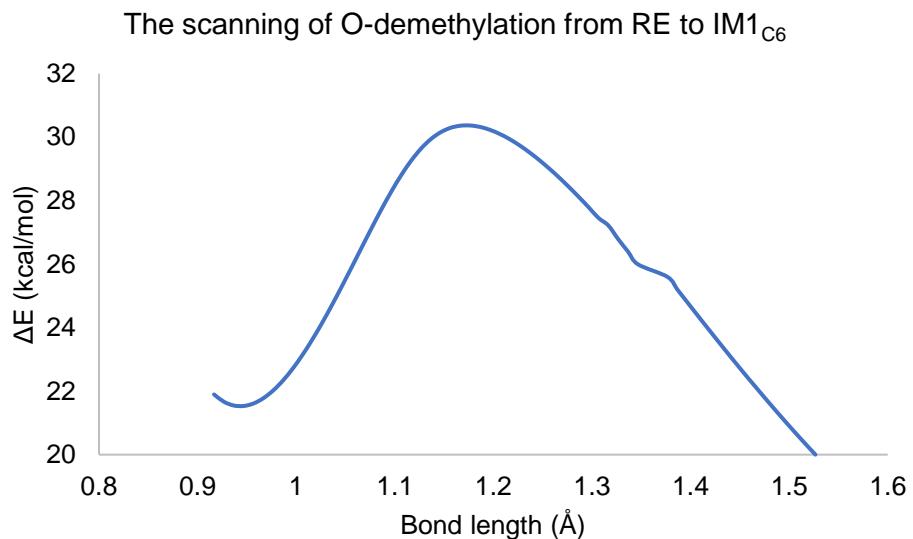


Figure S24 Statistical distribution of amino acid residues positioned within 5 Å from melatonin substrate in the molecular dynamics simulations of the 4 reactant models of CYP1A1 and the 10 models of CYP1A2. Densities in %.

13 Scanning from quantum mechanics calculation

(a)



(b)

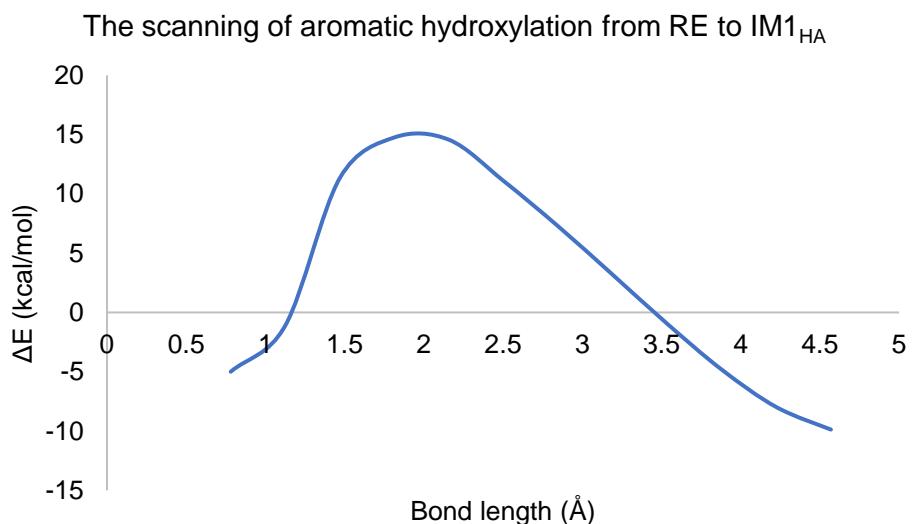


Figure S25 Scanning from quantum mechanics calculations. Each data point represents a full geometry optimization with one degree of freedom (the reaction coordinate) fixed.

14 Energy data from quantum mechanics calculation

Table S1 Absolute energies in kcal mol⁻¹ of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through aromatic hydroxylation in the doublet and quartet spin states.

CAL	Energy	State					MUL
		RE	TS1 _{C6}	IM1 _{C6}	IM2 _{C6}	P _{C6}	
BS1	ZPE	1484.549	1484.561	1485.443	1487.218	1486.983	2
	E	-4556160.818	-4556145.862	-4556183.697	-4556187.538	-4556209.768	
	E+ZPE	-4554676.269	-4554661.300	-4554698.254	-4554700.320	-4554722.785	
	G	-4554821.969	-4554802.893	-4554844.535	-4554837.643	-4554863.286	
BS2_SOL	E	-5273049.633	-5273045.172	-5273078.670	-5273081.505	-5273101.941	
	E+ZPE	-5271565.084	-5271560.611	-5271593.227	-5271594.287	-5271614.958	
	G	-5271710.784	-5271702.204	-5271739.508	-5271731.610	-5271755.460	
BS1	ZPE	1484.553	1484.803	1483.859	1485.341	1484.780	4
	E	-4556160.729	-4556142.630	-4556170.360	-4556182.697	-4556197.669	
	E+ZPE	-4554676.176	-4554657.827	-4554686.501	-4554697.356	-4554712.888	
	G	-4554822.331	-4554798.990	-4554834.141	-4554838.216	-4554855.959	
BS2_SOL	E	-5273050.261	-5273041.624	-5273064.116	-5273077.967	-5273099.272	
	E+ZPE	-5271565.708	-5271556.821	-5271580.257	-5271592.626	-5271614.492	
	G	-5271711.863	-5271697.984	-5271727.898	-5271733.487	-5271757.562	

Table S2 Absolute energies in kcal mol⁻¹ of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through O-demethylation in the doublet and quartet spin state.

CAL	Energy	State				MUL
		RE	TS1 _{HA}	IM1 _{HA}	IM2	
BS1	ZPE	1484.549	1479.619	1481.210	1485.942	2
	E	-4556160.818	-4556124.127	-4556132.678	-4556197.813	
	E+ZPE	-4554676.269	-4554644.508	-4554651.467	-4554711.871	
	G	-4554821.969	-4554788.926	-4554796.824	-4554858.175	
BS2_SOL	E	-5273049.633	-5273028.234	-5273033.757	-5273098.633	
	E+ZPE	-5271565.084	-5271548.615	-5271552.546	-5271612.691	
	G	-5271710.784	-5271693.032	-5271697.903	-5271758.995	
BS1	ZPE	1484.553	1480.201	1482.271	1485.373	4
	E	-4556160.729	-4556123.845	-4556132.951	-4556198.009	
	E+ZPE	-4554676.176	-4554643.643	-4554650.680	-4554712.636	
	G	-4554822.331	-4554787.634	-4554797.893	-4554859.431	
BS2_SOL	E	-5273050.261	-5273026.520	-5273039.291	-5273102.415	
	E+ZPE	-5271565.708	-5271546.319	-5271557.020	-5271617.042	
	G	-5271711.863	-5271690.309	-5271704.233	-5271763.837	

Table S3 Relative energies in kcal mol⁻¹ of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through aromatic hydroxylation in the doublet and quartet spin state.

CAL	Energy	State					MUL
		RE	TS1 _{C6}	IM1 _{C6}	IM2 _{C6}	P _{C6}	
BS1	ΔE	0.000	14.957	-22.878	-26.720	-48.950	2
	Δ(E+ZPE)	0.000	14.969	-21.984	-24.051	-46.515	
	ΔG	0.000	19.076	-22.566	-15.674	-41.317	
BS2_SOL	ΔE	0.000	4.461	-29.037	-31.872	-52.308	2
	Δ(E+ZPE)	0.000	4.474	-28.143	-29.202	-49.874	
	ΔG	0.000	8.580	-28.724	-20.825	-44.676	
BS1	ΔE	0.089	18.188	-9.541	-21.879	-36.850	4
	Δ(E+ZPE)	0.093	18.443	-10.232	-21.086	-36.619	
	ΔG	-0.362	22.979	-12.172	-16.247	-33.990	
BS2_SOL	ΔE	-0.628	8.009	-14.483	-28.334	-49.639	4
	Δ(E+ZPE)	-0.624	8.264	-15.173	-27.541	-49.407	
	ΔG	-1.079	12.800	-17.114	-22.703	-46.778	

Table S4 Relative energies in kcal mol⁻¹ of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through O-demethylation in the doublet and quartet spin state.

CAL	Energy	State				MUL
		RE	TS1 _{HA}	IM1 _{HA}	IM2	
BS1	ΔE	0.000	36.691	28.141	-36.995	2
	Δ(E+ZPE)	0.000	31.761	24.802	-35.602	
	ΔG	0.000	33.043	25.145	-36.206	
BS2_SOL	ΔE	0.000	21.399	15.876	-49.000	2
	Δ(E+ZPE)	0.000	16.470	12.538	-47.606	
	ΔG	0.000	17.752	12.881	-48.211	
BS1	ΔE	0.089	36.973	27.868	-37.191	4
	Δ(E+ZPE)	0.093	32.626	25.590	-36.366	
	ΔG	-0.362	34.335	24.076	-37.462	
BS2_SOL	ΔE	-0.628	23.113	10.343	-52.782	4
	Δ(E+ZPE)	-0.624	18.766	8.065	-51.957	
	ΔG	-1.079	20.475	6.551	-53.053	

15 Energy data from quantum mechanics calculation with RRHO fixed

Table S5 Absolute energies with the correction in kcal mol⁻¹ of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through aromatic hydroxylation in the doublet and quartet spin states.

CAL	Energy	State					MUL
		RE	TS1 _{C6}	IM1 _{C6}	IM2 _{C6}	P _{C6}	
BS1	G	-4554821.969	-4554802.893	-4554844.535	-4554837.643	-4554863.286	2
	G _{Cr}	-4554812.403	-4554794.879	-4554833.892	-4554831.325	-4554855.788	
BS2_SOL	G	-5271710.784	-5271702.204	-5271739.508	-5271731.610	-5271755.460	4
	G _{Cr}	-5271701.218	-5271694.190	-5271728.866	-5271725.291	-5271747.961	
BS1	G	-4554822.331	-4554798.990	-4554834.141	-4554838.216	-4554855.959	2
	G _{Cr}	-4554812.723	-4554791.224	-4554823.373	-4554831.325	-4554847.950	
BS2_SOL	G	-5271711.863	-5271697.984	-5271727.898	-5271733.487	-5271757.562	4
	G _{Cr}	-5271701.226	-5271690.218	-5271717.129	-5271725.291	-5271749.553	

G_{Cr} is free energy with rrho correction

Table S6 Absolute energies with the correction in kcal mol⁻¹ of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through O-demethylation in the doublet and quartet spin state.

CAL	Energy	State				MUL
		RE	TS1 _{HA}	IM1 _{HA}	IM2	
BS1	G	-4554821.969	-4554788.926	-4554796.824	-4554858.175	2
	G _{Cr}	-4554812.403	-4554779.557	-4554787.152	-4554848.087	
BS2_SOL	G	-5271710.784	-5271693.032	-5271697.903	-5271758.995	4
	G _{Cr}	-5271701.218	-5271683.664	-5271688.230	-5271748.906	
BS1	G	-4554822.331	-4554787.634	-4554797.893	-4554859.431	2
	G _{Cr}	-4554812.723	-4554778.606	-4554787.607	-4554849.552	
BS2_SOL	G	-5271711.863	-5271690.309	-5271704.233	-5271763.837	4
	G _{Cr}	-5271702.666	-5271681.282	-5271693.947	-5271753.959	

G_{Cr} is free energy with rrho correction

Table S7 Relative energies with the correction in kcal mol⁻¹ of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through aromatic hydroxylation in the doublet and quartet spin state.

CAL	Energy	State				MUL
		RE	TS1 _{C6}	IM1 _{C6}	IM2 _{C6}	
BS1	ΔG	0.000	19.076	-22.566	-15.674	-41.317
	ΔG _{Cr}	0.000	17.524	-21.490	-18.922	-43.385
BS2_SOL	ΔG	0.000	8.580	-28.724	-20.826	-44.676
	ΔG _{Cr}	0.000	7.029	-27.647	-24.073	-46.743
BS1	ΔG	-0.362	22.979	-12.172	-16.247	-33.990
	ΔG _{Cr}	-0.320	21.179	-10.970	-18.922	-35.547
BS2_SOL	ΔG	-1.079	12.800	-17.114	-22.703	-46.778
	ΔG _{Cr}	-0.008	11.000	-15.911	-24.073	-48.335

G_{Cr} is free energy with rrho correction

Table S8 Relative energies with the correction in kcal mol⁻¹ of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through O-demethylation in the doublet and quartet spin state.

CAL	Energy	State				MUL
		RE	TS1 _{HA}	IM1 _{HA}	IM2	
BS1	G	0.000	33.043	25.145	-36.206	2
	G _{Cr}	0.000	32.846	25.251	-35.684	
BS2_SOL	G	0.000	17.752	12.881	-48.211	2
	G _{Cr}	0.000	17.555	12.988	-47.688	
BS1	G	-0.362	34.335	24.076	-37.462	4
	G _{Cr}	-0.320	33.796	24.796	-37.150	
BS2_SOL	G	-1.079	20.475	6.551	-53.053	4
	G _{Cr}	-1.448	19.936	7.272	-52.740	

G_{Cr} is free energy with rrho correction

16 Group charges and spin densities from quantum mechanics simulation

Table S9 Group charges (doublet/quartet) of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through aromatic hydroxylation in the doublet and quartet spin state.

Residue group	Composition	RE	TS1 _{C6}	IM1 _{C6}	IM2 _{C6}	P _{C6}
Compound I	Fe	0.479 / 0.471	0.413 / 0.432	0.274 / 0.461	0.264 / 0.428	0.238 / 0.447
	O	-0.422 / -0.418	-0.46 / -0.479	-0.557 / -0.546	-0.562 / -0.57	0.158 / 0.158
	Fe-O*	0.058 / 0.053	-0.047 / -0.047	-0.284 / -0.085	-0.298 / -0.142	0.396 / 0.605
	Por*	-0.054 / -0.047	-0.346 / -0.375	-0.559 / -0.519	-0.55 / -0.748	-0.417 / -0.52
	S	-0.189 / -0.191	-0.207 / -0.164	-0.177 / -0.349	-0.17 / -0.15	0.554 / 0.554
	S-H*	-0.101 / -0.104	-0.13 / -0.083	-0.111 / -0.277	-0.109 / -0.08	1.008 / 1.002
Melatonin	C ₆ *	-0.199 / -0.199	-0.129 / -0.082	0.102 / 0.103	0.101 / 0.098	0.323 / 0.326
	H _{C6} *	0.16 / 0.16	0.179 / 0.169	0.141 / 0.118	0.148 / 0.15	0.174 / 0.173
	The rest atoms*	-0.04 / -0.04	0.28 / 0.229	0.497 / 0.416	0.5 / 0.514	-0.832 / -0.941
Environment*		-1.824 / -1.824	-1.808 / -1.813	-1.787 / -1.755	-1.792 / -1.793	-2.651 / -2.644
Total**		-2 / -2	-2 / -2	-2 / -2	-2 / -2	-2 / -2

** is the summation of *

Table S10 Group charges (doublet/quartet) of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through O-demethylation in the doublet and quartet spin state.

Residue group	Composition	RE	TS1 _{HA}	IM1 _{HA}	IM2
Compound I	Fe	0.479 / 0.471	0.356 / 0.415	0.324 / 0.405	0.306 / 0.49
	O	-0.422 / -0.418	-0.6 / -0.596	-0.696 / -0.678	0.164 / 0.165
	Fe-O*	0.058 / 0.053	-0.244 / -0.181	-0.372 / -0.273	0.47 / 0.656
	Por*	-0.054 / -0.047	-0.102 / -0.247	-0.075 / -0.283	-0.36 / -0.376
	S	-0.189 / -0.191	-0.185 / -0.104	-0.184 / -0.091	0.534 / 0.534
	S-H*	-0.101 / -0.104	-0.101 / -0.023	-0.1 / -0.001	0.985 / 0.983
Melatonin	C _{Meth} *	-0.236 / -0.235	-0.246 / -0.262	-0.164 / -0.173	0.158 / 0.158
	H _{CMeth} *	0.219 / 0.218	0.389 / 0.396	0.412 / 0.441	0.154 / 0.153
	The rest atoms*	-0.061 / -0.061	0.12 / 0.129	0.112 / 0.103	-0.883 / -1.053
Environment*		-1.824 / -1.824	-1.816 / -1.812	-1.813 / -1.815	-2.523 / -2.521
Total**		-2 / -2	-2 / -2	-2 / -2	-2 / -2

** is the summation of *

Table S11 Group spin densities (doublet/quartet) of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through aromatic hydroxylation in the doublet and quartet spin state.

Residue group	Composition	RE	TS1 _{C6}	IM1 _{C6}	IM2 _{C6}	P _{C6}
Compound I	Fe	1.239 / 1.113	1.418 / 1.44	0.992 / 2.573	0.988 / 2.988	1.123 / 3.253
	O	0.856 / 0.902	0.352 / 0.722	0.044 / 0.103	0.05 / 0.026	0 / 0
	Fe-O*	2.095 / 2.015	1.769 / 2.162	1.036 / 2.677	1.039 / 3.015	1.123 / 3.253
	Por*	-0.593 / 0.527	-0.21 / -0.002	-0.076 / -0.058	-0.075 / 0.14	-0.089 / -0.083
	S	-0.518 / 0.468	-0.172 / 0.233	0.035 / 0.358	0.037 / -0.163	0 / 0
	S-H*	-0.503 / 0.455	-0.167 / 0.229	0.033 / 0.35	0.034 / -0.163	0 / -0.001
Melatonin	C ₆ *	0 / 0.001	-0.12 / 0.046	-0.003 / -0.001	-0.004 / -0.003	0.001 / -0.003
	H _{C6} *	-0.001 / -0.001	0.006 / 0.009	0 / 0.009	0 / 0.001	0 / 0
	The rest atoms*	0.002 / 0.002	-0.277 / 0.555	0.011 / 0.023	0.005 / 0.01	-0.027 / -0.166
Environment*		0 / 0.001	-0.001 / 0.002	0 / 0	0 / 0	-0.008 / 0
Total**		1 / 3	1 / 3	1 / 3	1 / 3	1 / 3

** is the summation of *

Table S12 Group spin densities (doublet/quartet) of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through O-demethylation in the doublet and quartet spin state.

Residue group	Composition	RE	TS1 _{HA}	IM1 _{HA}	IM2
Compound I	Fe	1.239 / 1.113	1.022 / 1.295	0.992 / 1.755	1.222 / 2.49
	O	0.856 / 0.902	0.498 / 0.633	0.093 / 0.271	0 / 0
	Fe-O*	2.095 / 2.015	1.52 / 1.929	1.085 / 2.025	1.222 / 2.49
	Por*	-0.593 / 0.527	-0.616 / 0.157	-0.702 / -0.095	-0.14 / 0.06
	S	-0.518 / 0.468	-0.319 / 0.35	-0.325 / 0.096	0 / 0
	S-H*	-0.503 / 0.455	-0.311 / 0.342	-0.317 / 0.096	0 / -0.002
Melatonin	C _{Meth} *	0.002 / 0.003	0.386 / 0.424	0.848 / 0.846	0 / 0
	H _{Meth} *	-0.001 / -0.002	-0.059 / -0.037	0.009 / 0.021	0 / 0
	The rest atoms*	0 / 0.001	0.079 / 0.185	0.074 / 0.102	-0.081 / 0.458
Environment*		0 / 0.001	0.001 / 0	0.004 / 0.005	-0.001 / -0.006
Total**		1 / 3	1 / 3	1 / 3	1 / 3

** is the summation of *

17 Imaginary frequency data from quantum mechanics simulation

Table S13 Imaginary frequencies for the metabolism of melatonin by CYP1A2 through aromatic hydroxylation and O-demethylation.

State	Imaginary frequency
^{2,4} TS1 _{C₆}	i240.44 (Doublet), i410.71 (Quartet)
^{2,4} TS1 _{HA}	i1602.02 (Doublet), i1775.64 (Quartet)

18 Geometry data from quantum mechanics simulation

Table S14 Geometry data (doublet/quartet) of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through aromatic hydroxylation in the doublet and quartet spin state. Bond length is Å and angle is degree.

Composition	RE	TS1 _{C₆}	IM1 _{C₆}	IM2 _{C₆}	P _{C₆}
Fe-O _{CpdI}	1.629 / 1.631	1.679 / 1.715	1.922 / 1.933	1.927 / 1.920	2.356 / 2.595
Fe-S _{CpdI}	2.575 / 2.562	2.434 / 2.403	2.322 / 2.353	2.318 / 2.393	2.242 / 2.339
C ₆ -O _{CpdI}	3.743 / 3.739	2.052 / 1.887	1.387 / 1.388	1.326 / 1.326	1.387 / 1.376
H _{C₆} -C ₆	1.086 / 1.086	1.082 / 1.083	1.110 / 1.110	2.163 / 2.188	1.939 / 1.936
H _{C₆} -N _{CpdI}	5.239 / 5.232	2.467 / 2.624	2.415 / 2.419	1.043 / 1.028	3.058 / 2.998
H _{C₆} -O _{CpdI}	2.888 / 2.875	2.275 / 2.192	2.081 / 2.083	2.304 / 2.241	0.974 / 0.973
	156.966 /	126.974 /	126.884 /	129.186 /	132.156 /
C ₆ -O _{CpdI} -Fe	157.370	136.423	126.325	135.900	134.214
	150.798 /	136.892 /	118.091 /	144.397 /	85.503 /
C ₆ -H _{C₆} -N _{CpdI}	151.609	126.877	118.952	154.665	91.265

Table S15 Geometry data (doublet/quartet) of UB3LYP/BS1 optimized geometries for the metabolism of melatonin by CYP1A2 through O-demethylation in the doublet and quartet spin state. Bond lengths are in Å and angles in degrees.

Composition	RE	TS1 _{HA}	IM1 _{HA}	IM2
Fe-O _{CpdI}	1.629 / 1.631	1.755 / 1.731	1.812 / 1.786	3.381 / 3.469
Fe-S _{CpdI}	2.575 / 2.562	2.440 / 2.383	2.431 / 2.329	2.236 / 2.429
H _{C_{Meth}} -O _{CpdI}	2.369 / 2.384	1.175 / 1.187	0.975 / 0.981	0.974 / 0.973
H _{C_{Meth}} -C _{Meth}	1.095 / 1.095	1.354 / 1.375	2.397 / 2.311	1.956 / 1.956
O _{CpdI} -C _{Meth}	3.434 / 3.447	2.518 / 2.543	3.203 / 3.201	1.392 / 1.391
H _{C_{Meth}} -O _{CpdI} -Fe	141.215 / 141.387	116.977 / 123.516	105.949 / 109.607	52.602 / 50.622
H _{C_{Meth}} -O _{CpdI} -C _{Meth}	5.201 / 5.249	5.796 / 7.641	29.018 / 20.837	110.352 / 110.408

Geometry coordination

RE^2

6	2.719429000	26.652134000	18.197097000	1	13.149229000	21.168805000	16.763309000
6	6.914258000	28.986224000	17.600464000	1	12.864275000	22.898007000	16.641065000
6	8.246242000	27.743597000	22.080341000	1	11.199740000	20.691062000	15.313475000
6	3.995356000	25.526777000	22.719282000	1	10.239006000	22.535301000	13.889383000
6	3.762419000	27.387266000	17.638455000	1	11.035713000	23.730806000	14.928752000
6	3.740216000	27.936450000	16.304209000	1	9.765837000	22.682604000	15.585843000
6	4.919257000	28.595863000	16.136177000	6	12.318094000	21.374705000	13.191980000
6	5.656350000	28.444798000	17.367701000	6	13.516101000	20.838152000	14.385790000
6	7.647384000	28.859436000	18.774646000	8	13.288551000	22.567043000	14.072729000
6	8.965251000	29.405061000	18.981516000	6	7.991776000	17.698789000	22.263219000
6	9.345168000	29.042877000	20.239120000	6	6.842099000	17.043199000	23.034438000
6	8.254377000	28.284798000	20.798845000	6	9.364850000	17.153861000	22.677970000
6	7.193276000	27.053154000	22.657769000	6	9.619123000	15.668889000	22.349634000
6	7.193392000	26.529791000	24.001216000	1	9.581225000	15.378354000	20.841233000
6	5.994287000	25.912276000	24.184313000	1	10.959091000	15.218017000	22.951007000
6	5.260658000	26.051140000	22.950534000	1	7.955231000	18.773781000	22.489131000
6	3.281004000	25.636337000	21.532648000	1	9.461221000	17.290314000	23.761258000
6	1.959191000	25.085805000	21.325685000	1	10.145501000	17.765745000	22.202905000
6	1.612470000	25.394537000	20.046995000	1	8.823144000	15.086343000	22.833520000
6	2.716039000	26.139694000	19.485943000	1	9.786404000	14.319107000	20.641570000
7	4.934576000	27.705250000	18.267928000	1	8.605860000	15.610626000	20.400979000
7	7.229084000	28.187478000	19.894371000	1	10.337734000	15.969773000	20.307421000
7	5.998589000	26.763135000	22.036001000	1	11.141242000	14.151985000	22.764375000
7	3.724662000	26.280868000	20.405725000	7	10.982353000	15.378902000	24.035614000
26	5.526787000	27.105857000	20.096882000	6	11.795758000	15.778972000	22.512053000
1	1.830607000	26.467448000	17.598152000	6	4.362560000	16.448852000	22.917352000
1	7.367920000	29.551414000	16.791271000	8	3.353716000	17.543178000	23.007050000
1	9.135098000	27.891781000	22.686671000	6	3.007932000	18.022218000	21.713735000
1	3.521530000	24.983391000	23.531190000	1	3.771609000	18.737867000	23.858475000
6	5.964538000	19.705412000	17.895444000	1	5.642051000	17.298687000	21.391058000
6	6.717509000	20.689617000	17.231338000	1	4.682244000	16.062151000	23.909993000
6	6.004312000	21.056151000	19.962521000	1	2.468797000	17.070989000	23.466349000
6	5.626964000	19.903837000	19.265427000	1	2.493947000	17.290205000	21.255322000
6	5.370920000	18.454370000	17.482556000	1	5.975564000	19.469891000	23.896402000
6	7.080236000	21.833589000	17.930746000	1	4.647137000	19.232627000	23.424696000
6	6.729316000	22.028952000	19.280816000	1	4.027423000	18.427186000	24.877655000
7	4.901286000	18.800909000	19.677557000	6	6.0457092000	13.274868000	20.490400000
1	4.268204000	18.754995000	20.481929000	8	-0.722758000	13.591736000	21.411606000
6	5.441718000	17.860302000	16.101551000	6	-1.640776000	12.793870000	21.599877000
6	4.823474000	16.458563000	15.935891000	6	0.689881000	14.282582000	19.337864000
7	3.375573000	16.400289000	16.067781000	8	1.612459000	12.973803000	19.655371000
6	2.553555000	16.697494000	15.030028000	8	2.278681000	15.904575000	20.860798000
6	1.066599000	16.576013000	15.333335000	1	1.368982000	13.201900000	21.097332000
8	2.966439000	17.029342000	13.913223000	1	1.123501000	13.762005000	18.478140000
1	2.963618000	16.155484000	16.969067000	1	-0.270619000	14.696806000	19.002009000
8	7.834616000	22.813075000	17.274561000	7	-0.663627000	14.820501000	22.012440000
6	7.055542000	23.915884000	16.785547000	6	-1.826339000	15.403834000	22.664818000
1	4.069237000	17.104164000	18.700914000	6	-1.715682000	15.325144000	24.197981000
1	7.002188000	20.586050000	16.187884000	8	-0.441012000	15.864698000	24.605671000
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1	7.014142000	22.954522000	19.771685000	1	0.076182000	15.439508000	21.651313000
1	4.957147000	18.524745000	15.373134000	1	-2.724027000	14.877429000	22.324586000
1	6.495118000	17.791667000	15.785114000	1	-2.460422000	15.998802000	24.638777000
1	5.252482000	15.771302000	16.674387000	1	0.206370000	15.444349000	24.007190000
1	5.078061000	16.090643000	14.936777000	1	-1.741043000	13.921926000	25.845734000
1	0.582698000	17.535903000	15.127360000	1	-1.187998000	13.220187000	24.302536000
1	0.865209000	16.284419000	16.367512000	1	-2.904289000	13.542456000	24.547808000
1	0.626206000	15.838944000	14.652366000	6	-1.839109000	21.837563000	23.183140000
1	7.750453000	24.571316000	16.252910000	6	-1.427845000	23.279500000	22.868775000
1	6.586670000	24.466028000	17.608576000	8	-0.360566000	23.738220000	23.298004000
1	6.285796000	23.556797000	16.090978000	6	-0.608516000	20.918356000	23.182693000
16	4.695043000	29.291461000	21.1757779000	6	-0.968512000	19.449168000	23.342938000
6	11.206169000	24.106885000	18.798917000	8	-1.831852000	18.909810000	22.632028000
8	12.394192000	24.158595000	19.123916000	7	-0.302218000	18.766947000	24.297686000
7	10.663209000	23.038644000	18.155575000	1	-2.309209000	21.820765000	24.176223000
6	11.452903000	21.846350000	17.880141000	1	0.092656000	21.255399000	23.950933000
6	12.358852000	21.923624000	16.634882000	1	-0.084216000	21.013434000	22.223762000
6	11.691546000	21.675970000	15.267328000	1	0.450795000	19.212349000	24.800672000
6	10.619345000	22.715027000	14.903375000	1	-0.392907000	17.747221000	24.375201000
6	12.763764000	21.606647000	14.167314000	7	-2.299446000	23.980593000	22.119351000
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6	-1.793710000	20.726255000	19.072973000	1	-1.203485000	26.265637000	15.178481000
6	-2.899195000	19.695520000	18.985275000	1	-1.293508000	23.744935000	16.817295000
8	-3.581853000	19.550642000	17.934178000	1	-1.094611000	26.034659000	17.683838000
6	-0.370199000	20.126468000	19.201987000	1	1.206567000	23.606103000	16.687193000
6	0.640788000	21.288835000	19.266690000	1	1.180403000	24.683647000	15.268678000
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1	-0.316907000	19.575041000	20.152285000	6	-2.852182000	21.939241000	11.913321000
1	0.602684000	21.842700000	18.317678000	6	-1.638123000	21.064632000	12.192070000
1	0.313475000	21.999743000	20.039546000	6	-1.807480000	19.670464000	12.149211000
1	0.920583000	18.678487000	18.196160000	6	-0.356594000	21.564530000	12.458192000
1	-0.093026000	19.633033000	17.092490000	6	-0.737918000	18.802751000	12.371367000
1	-0.790071000	18.311435000	18.057632000	6	0.714661000	20.691889000	12.685847000
1	2.723201000	21.744004000	19.684090000	6	0.537508000	19.308202000	12.645914000
1	2.145440000	20.259965000	20.469289000	1	-3.408757000	23.918360000	14.021974000
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6	-5.525704000	18.143945000	20.061197000	1	-0.898086000	17.727400000	12.334317000
8	-5.965043000	18.450341000	18.729225000	1	1.697733000	21.104090000	12.901090000
6	-5.920923000	19.256894000	21.031196000	1	1.368788000	18.632815000	12.837456000
1	-2.528811000	19.097158000	20.907336000	1	8.014852000	26.629603000	24.699489000
1	-3.847919000	17.373848000	21.136961000	1	5.625538000	25.398177000	25.062807000
1	-6.068021000	17.229189000	20.334227000	1	9.515694000	29.982655000	18.249520000
1	-5.267238000	19.022605000	18.335758000	1	10.272222000	29.261560000	20.753793000
1	-6.999492000	19.426105000	20.964952000	1	2.919049000	27.821900000	15.608600000
1	-5.413604000	20.194813000	20.785168000	1	5.272062000	29.139865000	15.269023000
1	-5.664617000	18.991359000	22.064101000	1	1.375347000	24.559090000	22.073026000
7	-4.394139000	16.242025000	18.246887000	1	0.712885000	25.163429000	19.490396000
6	-4.005092000	15.407015000	17.127394000	1	7.831137000	17.616833000	21.182425000
6	-4.223735000	16.072409000	15.760379000	1	4.034712000	15.629138000	22.309672000
8	-4.425957000	15.394515000	14.760426000	1	0.249988000	12.281498000	20.082933000
1	-5.253339000	16.783837000	18.169227000	1	-1.887243000	16.456568000	22.374354000
1	-2.943815000	15.166761000	17.249070000	1	-2.588113000	21.471880000	22.473716000
7	-4.190212000	17.442924000	15.757362000	1	-1.540935000	25.883653000	22.501625000
6	-4.283148000	18.199716000	14.523119000	1	-3.592166000	23.932789000	11.639195000
6	-5.497037000	19.127764000	14.410898000	1	-1.994216000	21.383453000	19.929499000
8	-5.523990000	20.021626000	13.562838000	1	12.089507000	21.670478000	18.753875000
1	-3.858655000	17.938305000	16.581811000	1	-5.378887000	27.247136000	16.729126000
1	-3.391132000	18.814285000	14.370855000	1	-8.589977000	19.093086000	15.558698000
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7	-6.534694000	18.874223000	15.250346000	1	-2.455058000	27.737460000	17.490551000
6	-7.733151000	19.702326000	15.264429000	1	-3.046535000	26.825686000	18.612741000
6	-7.632527000	20.868654000	16.265001000	1	-4.562502000	14.464966000	17.110706000
8	-8.344753000	20.920072000	17.264995000	6	4.717514000	17.963691000	18.586427000
1	-6.382695000	18.231929000	16.017384000	8	6.221110000	25.725059000	19.581913000
1	-7.879013000	20.077228000	14.247126000	6	10.245257000	25.251964000	19.091701000
7	-6.676175000	21.779252000	15.956446000	1	10.345082000	25.531218000	20.144873000
6	-6.314477000	22.856443000	16.858128000	1	10.532590000	26.128120000	18.499448000
6	-5.905131000	24.078197000	16.023974000	1	9.196526000	25.017707000	18.888955000
8	-5.866878000	24.038427000	14.789330000	1	3.694683000	28.753017000	21.909191000
6	-5.168654000	22.439054000	17.798070000				
8	-3.961697000	22.275098000	17.046739000				
1	-6.199529000	21.738532000	15.061121000				
1	-7.193077000	23.095800000	17.467276000	6	2.735608000	26.641516000	18.209257000
1	-5.007219000	23.176153000	18.591845000	6	6.941482000	28.953093000	17.611260000
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7	-5.576818000	25.180628000	16.741746000	6	3.784695000	27.365966000	17.647435000
6	-5.149680000	26.395403000	16.084371000	6	3.773430000	27.896362000	16.305083000
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8	-3.196870000	27.577679000	15.362098000	6	5.684466000	28.411038000	17.374408000
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6	8.255281000	28.293513000	20.830010000	6	9.582266000	15.379203000	20.836003000
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6	1.958463000	25.090261000	21.341330000	1	9.787496000	14.320037000	20.635974000
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26	5.535222000	27.106054000	20.123615000	6	4.438774000	16.447189000	22.912621000
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1	6.561071000	24.464231000	17.596996000	8	-0.364868000	23.740978000	23.311340000
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16	4.684698000	29.294602000	21.148596000	6	-0.970399000	19.451133000	23.347360000
6	11.179979000	24.124581000	18.775323000	8	-1.835501000	18.911852000	22.638467000
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7	10.637623000	23.054629000	18.134263000	1	-2.310940000	21.821234000	24.183194000
6	11.427996000	21.862129000	17.861465000	1	0.091197000	21.257463000	23.954155000
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6	11.668915000	21.688990000	15.249157000	1	0.452300000	19.214242000	24.801824000
6	10.597069000	22.727790000	14.883488000	1	-0.391503000	17.748789000	24.377217000
6	12.741966000	21.618684000	14.150001000	7	-2.299029000	23.982737000	22.124808000
1	9.646059000	23.048889000	17.904137000	6	-2.055879000	25.357675000	21.701023000
1	10.733440000	21.018009000	17.776568000	6	-3.373124000	26.057398000	21.330956000
1	13.125278000	21.183033000	16.746873000	6	-4.180133000	25.287988000	20.259766000
1	12.840749000	22.912170000	16.622612000	8	-4.416629000	25.892658000	19.162975000
1	11.177011000	20.704159000	15.295839000	8	-4.544175000	24.120638000	20.548539000
1	10.217573000	22.547267000	13.869319000	1	-3.157218000	23.580202000	21.736626000
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1	13.493983000	20.850170000	14.369531000	6	-1.795127000	20.722895000	19.081902000
1	13.267036000	22.578905000	14.055211000	6	-2.900501000	19.692366000	18.992250000
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8	6.970949000	16.675887000	24.198854000	6	0.639578000	21.285184000	19.269518000
6	9.366772000	17.154236000	22.673330000	6	-0.069981000	19.129718000	18.075158000
6	9.620787000	15.669343000	22.344468000	6	2.081949000	20.861165000	19.555989000

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1	3.870738000	23.165694000	24.175468000	8	7.345036000	19.090813000	12.223830000
1	10.819930000	27.284642000	19.995124000	1	6.568314000	18.587183000	15.269719000
1	10.534090000	25.831284000	22.265103000	8	11.777230000	23.863475000	18.420865000
1	4.906814000	28.650822000	15.704232000	6	12.624344000	24.003201000	17.281987000
1	7.576175000	28.847561000	16.156880000	1	5.992599000	20.596615000	16.314566000
1	0.604581000	25.027705000	20.554285000	1	10.803100000	22.476618000	16.306814000
1	0.842962000	26.471521000	18.270792000	1	7.570158000	23.326919000	20.121368000
1	8.049431000	18.513751000	20.960962000	1	10.109498000	23.283330000	20.335110000
1	4.354321000	16.434576000	22.260451000	1	8.118551000	21.272129000	14.003544000
1	1.360840000	12.871248000	19.729771000	1	9.692302000	21.110753000	14.763452000

1	8.921802000	18.8056666000	15.626721000	1	1.424826000	17.186363000	20.817573000
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1	12.992954000	23.030281000	16.934482000	6	-0.824076000	21.634181000	23.518006000
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8	15.650312000	21.032628000	20.957201000	7	-0.033950000	19.457948000	22.754634000
7	13.611823000	21.809350000	20.307318000	1	-1.502522000	23.679910000	23.585809000
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6	12.186137000	19.347531000	18.797474000	1	0.880252000	19.728482000	23.122374000
6	13.092018000	19.864545000	17.668130000	1	-0.078084000	18.641897000	22.143849000
6	11.696823000	17.927626000	18.469015000	7	-1.903969000	23.859695000	20.746265000
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1	4.171189000	19.698866000	25.670562000	1	-0.574164000	21.534803000	18.096706000
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1	4.206527000	18.010635000	23.852744000	1	-2.836695000	19.263875000	16.022023000
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6	0.701971000	16.317262000	18.669239000	1	-7.428807000	18.573915000	15.944125000
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1	-0.588439000	23.286731000	12.994642000	6	7.079359000	19.474324000	16.322571000
1	2.265013000	19.385013000	14.175501000	6	6.904713000	17.947460000	16.164596000
1	1.861704000	23.492672000	12.966717000	7	5.636261000	17.443422000	16.668761000
1	3.306912000	21.544728000	13.554395000	6	4.477915000	17.665500000	15.996003000
1	3.985807000	23.499677000	25.643589000	6	3.210577000	17.179624000	16.682325000
1	1.559267000	24.482463000	24.955375000	8	4.441492000	18.217794000	14.892014000
1	9.979669000	25.428242000	21.669588000	1	5.616327000	17.040094000	17.605294000
1	8.939820000	24.476089000	23.954580000	8	4.276260000	24.272222000	17.646636000
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1	8.684068000	15.912997000	19.015847000	1	6.330497000	19.955268000	15.683717000
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1	2.897435000	13.018421000	20.806342000	1	7.701192000	17.413400000	16.693786000
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1	-3.994251000	21.680057000	12.926155000	1	2.707329000	16.454401000	16.034308000
1	-2.582467000	22.5558111000	18.908111000	1	3.615234000	25.237602000	16.014013000
1	10.093852000	22.529084000	20.805991000	1	3.688051000	23.463526000	15.814087000
1	-7.726373000	26.678666000	15.485335000	1	5.193771000	24.434103000	15.779781000
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1	8.761182000	22.662637000	16.471208000	1	8.967077000	21.453985000	18.554148000
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6	2.720607000	27.500144000	19.510217000	1	9.219956000	10.949905000	19.265575000
7	5.615665000	26.842924000	19.643418000	1	10.772625000	11.715164000	19.656355000
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7	1.836773000	16.311669000	21.323064000	1	-4.344255000	18.217767000	17.146506000
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1	-0.317770000	23.327421000	18.641222000	6	0.262867000	20.076892000	13.936430000
1	0.451458000	19.636460000	17.960435000	6	-0.014274000	22.365099000	13.236824000
1	-0.704833000	20.092431000	16.704995000	6	1.650647000	20.218876000	13.922547000
1	-1.227552000	19.127683000	18.111180000	6	1.378223000	22.512235000	13.220790000
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1	1.596607000	21.938058000	19.623543000	1	-4.012533000	22.902945000	14.741746000
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6	8.806150000	24.820040000	21.928939000	6	7.859683000	13.172520000	18.415684000
6	8.216536000	24.311147000	23.049649000	6	9.876656000	11.760853000	18.990146000
6	6.817034000	24.680374000	22.993769000	1	10.287197000	16.419218000	19.048375000
6	4.493012000	24.720894000	23.920053000	1	11.031009000	14.118274000	19.549270000
6	3.518265000	24.380240000	24.928104000	1	10.380123000	14.274943000	17.913925000
6	2.310542000	24.855720000	24.497504000	1	8.875583000	13.119961000	20.310419000
6	2.531329000	25.490606000	23.219298000	1	7.229349000	12.292066000	18.595971000
6	1.705700000	26.683044000	21.186874000	1	7.260931000	14.058678000	18.651926000
6	0.642151000	27.236063000	20.370863000	1	8.089040000	13.196115000	17.341001000
6	1.238149000	27.771420000	19.267612000	1	9.228243000	10.904503000	19.213954000
6	2.664775000	27.545675000	19.408242000	1	10.778821000	11.672394000	19.607974000
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TS1²_{HA}							
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6 2.610405000 25.668942000 17.298244000

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6	-6.706757000	19.027292000	20.093471000	1	-3.938743000	17.644035000	11.888312000
8	-7.308873000	19.349758000	18.831529000	1	-0.514301000	20.240035000	12.050548000
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6	-6.534513000	15.777333000	17.285905000	1	8.716099000	18.970157000	23.478757000
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6	-7.142492000	23.523893000	16.954380000	1	11.025054000	23.999198000	19.719086000
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6	1.073077000	24.998639000	19.439181000	1	10.796473000	19.647656000	22.567161000
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1	-1.900504000	26.176243000	14.711571000				
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1	0.234493000	23.152750000	15.910920000	7	7.158571000	26.568159000	18.988754000
1	0.240427000	24.303646000	14.550690000	7	5.692159000	25.689879000	21.284162000
1	-0.758084000	22.844487000	14.471033000	7	3.355648000	25.417720000	19.685290000
7	-3.858088000	24.199434000	13.135221000	26	5.223272000	26.010889000	19.351035000
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6	-4.728558000	22.390844000	11.610848000	1	7.482499000	27.586264000	15.763570000
6	-3.697228000	21.277394000	11.715687000	1	8.981120000	26.349296000	21.848286000
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6	7.008045000	19.767518000	19.607787000	1	9.134148000	14.313634000	23.999181000
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6	9.363086000	19.059612000	19.805590000	1	6.296800000	14.891035000	21.052650000
6	7.997454000	18.803257000	19.965783000	1	5.424290000	14.189397000	23.099497000
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6	4.378010000	19.793636000	19.597705000	6	2.511466000	14.848377000	19.834143000
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1	11.344585000	24.285129000	13.986160000	1	-1.889633000	25.140003000	20.453647000
1	10.325409000	23.615183000	15.263933000	1	-3.189335000	27.215899000	20.669098000
1	10.702201000	22.177155000	11.820733000	1	-4.132471000	26.536104000	22.014793000
1	11.936674000	20.967059000	12.216951000	6	-2.494449000	20.435716000	17.969020000
1	12.361672000	22.687184000	12.174361000	6	-3.804871000	19.720630000	18.243064000
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6	9.312322000	16.842932000	24.552067000	6	-1.276504000	20.112965000	18.854503000
8	9.986402000	16.262397000	25.403907000	6	-0.141511000	21.126680000	18.585113000
6	10.028131000	19.130081000	25.421775000	6	-0.797513000	18.663978000	18.649449000
6	11.368904000	19.296610000	24.678423000	6	0.888081000	21.195516000	19.718605000
6	11.242420000	20.163762000	23.416102000	1	-2.263420000	20.236244000	16.914860000
6	12.428960000	19.880613000	25.625124000	1	-1.569604000	20.242019000	19.907116000
1	8.011086000	18.386555000	25.230407000	1	0.354661000	20.860917000	17.642138000
1	10.214972000	18.615427000	26.371287000	1	-0.570968000	22.128516000	18.446233000
1	9.627472000	20.125483000	25.663583000	1	-0.014627000	18.407384000	19.372114000
1	11.706519000	18.293977000	24.380672000	1	-0.362371000	18.539112000	17.651276000
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1	10.532706000	19.753081000	22.690218000	1	1.700598000	21.884860000	19.465249000
1	10.903219000	21.177062000	23.671046000	1	0.426193000	21.562302000	20.643464000
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6	8.932719000	14.7944489000	23.201081000	8	-4.016125000	16.271278000	18.964991000
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8	7.387332000	14.883534000	21.404539000	8	-7.061072000	19.266035000	18.648004000
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