SUPPORTING MATERIAL

Structural Role of RKS Motifs in Chromatin Interactions: An MD Study of HP1 Bound to a Variably Modified Histone Tail

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Table S1: Partial atomic charges for the modified lysine residues used in the simulations.

Atom	Methylated Lysine	Dimethylated Lysine	Trimethylated Lysine
N	-0.34790	-0.34790	-0.34790
H	0.27470	0.27470	0.27470
CA	-0.08182	-0.23140	-0.15146
HA	0.16951	0.18924	0.17620
CB	-0.56172	-0.10388	-0.41312
HB2	0.12855	0.04319	0.11975
HB3	0.12855	0.04319	0.11975
CG	0.38449	0.11841	0.24321
HG2	-0.07328	-0.04165	-0.05375
HG3	-0.07328	-0.04165	-0.05375
CD	0.03657	0.12150	0.11497
HD2	0.06264	0.03292	0.03854
HD3	0.06264	0.03292	0.03854
CE	-0.41503	-0.35400	-0.37340
HE2	0.22364	0.20352	0.20138
HE3	0.22364	0.20352	0.20138
NZ	-0.24031	-0.05523	0.06942
HZ1	0.33648	0.32676	
HZ2	0.33648		
C1	-0.21298	-0.37787	-0.34631
HC11	0.16458	0.19949	0.18775
HC12	0.16458	0.19949	0.18775
HC13	0.16458	0.19949	0.18775
C2		-0.37787	-0.34631
HC21		0.19949	0.18775
HC22		0.19949	0.18775
HC23		0.19949	0.18775
C3			-0.34631
HC31			0.18775
HC32			0.18775
HC33			0.18775
C	0.73410	0.73410	0.73410
O	-0.58940	-0.58940	-0.58940

Table S2: Details of the molecular dynamics runs.

Particle Mesh Ewald grid	128 x 128 x 128
Electrostatics calculation	every 4 timesteps
Non-bonded interactions calculation	every 2 timesteps
Langevin thermostat damping parameter	5/ps
Langevin piston oscillation period	100 fs
Langevin piston damping parameter	50 fs

Table S3: Atoms and dihedral angles selected for clustering of the structures shown in Fig. 2A-B.

	Dihedrals (°)	
Atoms and dihedrals used for clustering	**66%**	**20%**
8CD-8CG-8CB-8CA	-154.77	-153.70
8C-8CA-8CB-8CG	67.83	159.96
9N-8C-8CA-8CB	-108.10	-108.18
9CA-9N-8C-8CA	176.55	-173.85
9C-9CA-9N-8C	-96.51	-80.40
10N-9C-9CA-8N	156.65	-27.18
10CA-10N-9C-9CA	153.92	176.85
10CB-10CA-10N-9C	164.88	158.74

Table S4: Average distance *(Å)* between the Nitrogen atom ζ of K9 (labeled as *N*ζ) and the centroid of each aromatic ring. The star denotes a modified residue.

	H3	**HP1**	**K9$_{me}$**			**K9$_{me2}$**		**K9$_{me3}$**		
			RK*S	RK*S*	GK*S*	RK*S	RK*S*	RK*S	RK*S*	GK*S
residue	K9*	*Y24*	4.80	4.94	4.95	4.57	4.65	4.55	4.60 *(82%)* 6.80 *(18%)*	4.85
	K9*	*W45*	4.65	4.73	4.73 *(75%)* 6.40 *(25%)*	4.58	4.67	4.60	4.76	4.74
	K9*	*Y48*	5.03	5.00	4.96	4.75	4.62	4.62	4.55	4.52

Supplementary Figures and Figure Captions

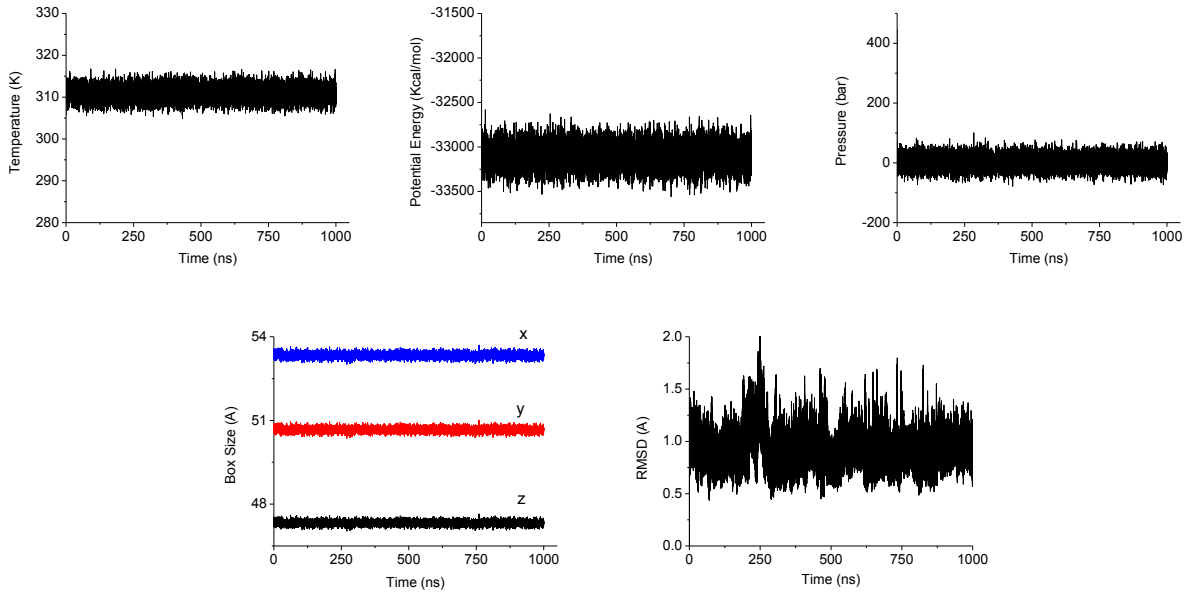

Figure S1: Representative plots of Temperature, Potential energy, Pressure, Size of the simulation box and root-mean-square-deviation (RMSD) of the H3-RKmeS peptide atoms from their initial position during the 1μs trajectories. Simulations of all other H3 peptides exhibit similar behavior.

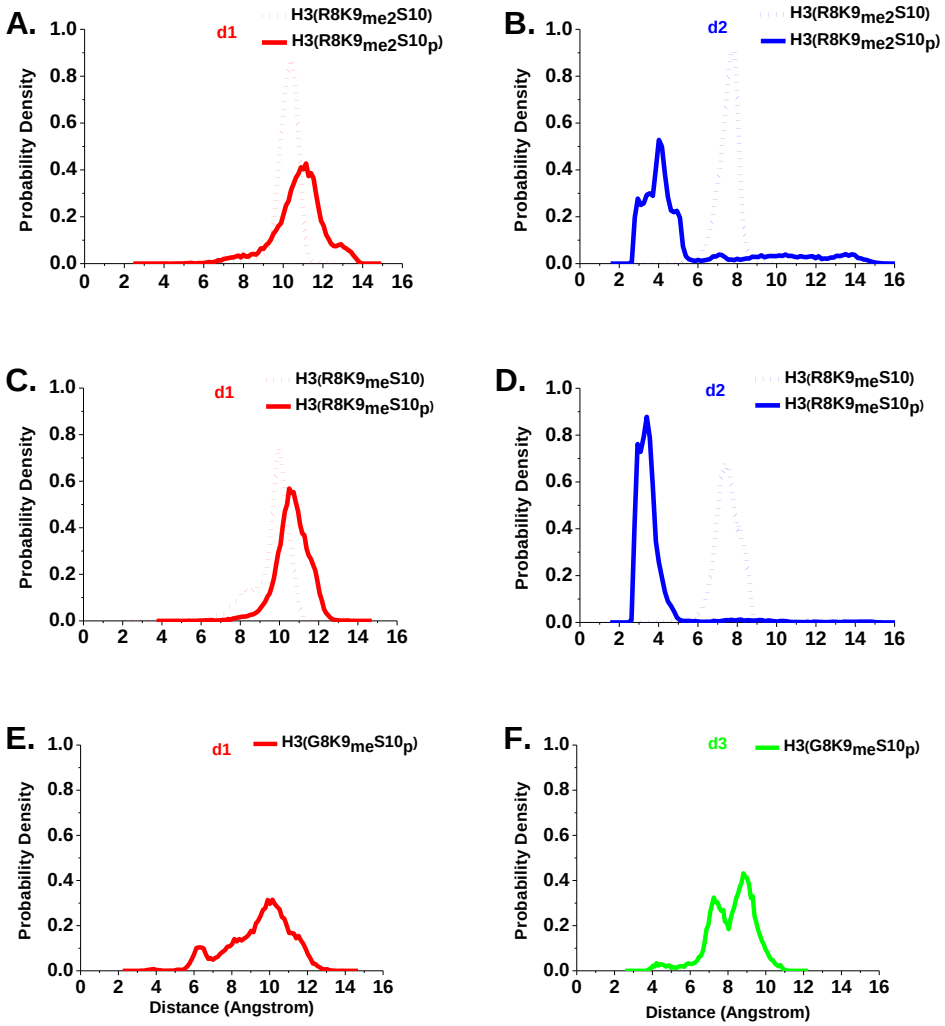

Figure S2: Probability Density Functions (PDF) of distance variables d1, d2 and d3 (defined in the text and shown in Fig. 1, *A* and *C*) for the various forms of the H3 tail. ***A.*** PDF of d1 in the phosphorylated (solid line) and unphosphorylated (dashed line) form of K9-dimethylated H3 tail. ***B.*** PDF of d2 in the phosphorylated (solid line) and unphosphorylated (dashed line) form of K9-dimethylated H3 tail. ***C.*** PDF of d1 in the phosphorylated (solid line) and unphosphorylated (dashed line) form of the K9-monomethylated H3 tail. ***D.*** PDF of d2 in the phosphorylated (solid line) and unphosphorylated (dashed line) form of the K9-monomethylated H3 tail. ***E***. PDF of d1 for the K9-monomethylated and S10-phosphorylated H3 tail, in which R8 has been replaced by G. ***F.*** PDF of d3 for the K9-monomethylated and S10-phosphorylated H3 tail, in which R8 has been replaced by G.

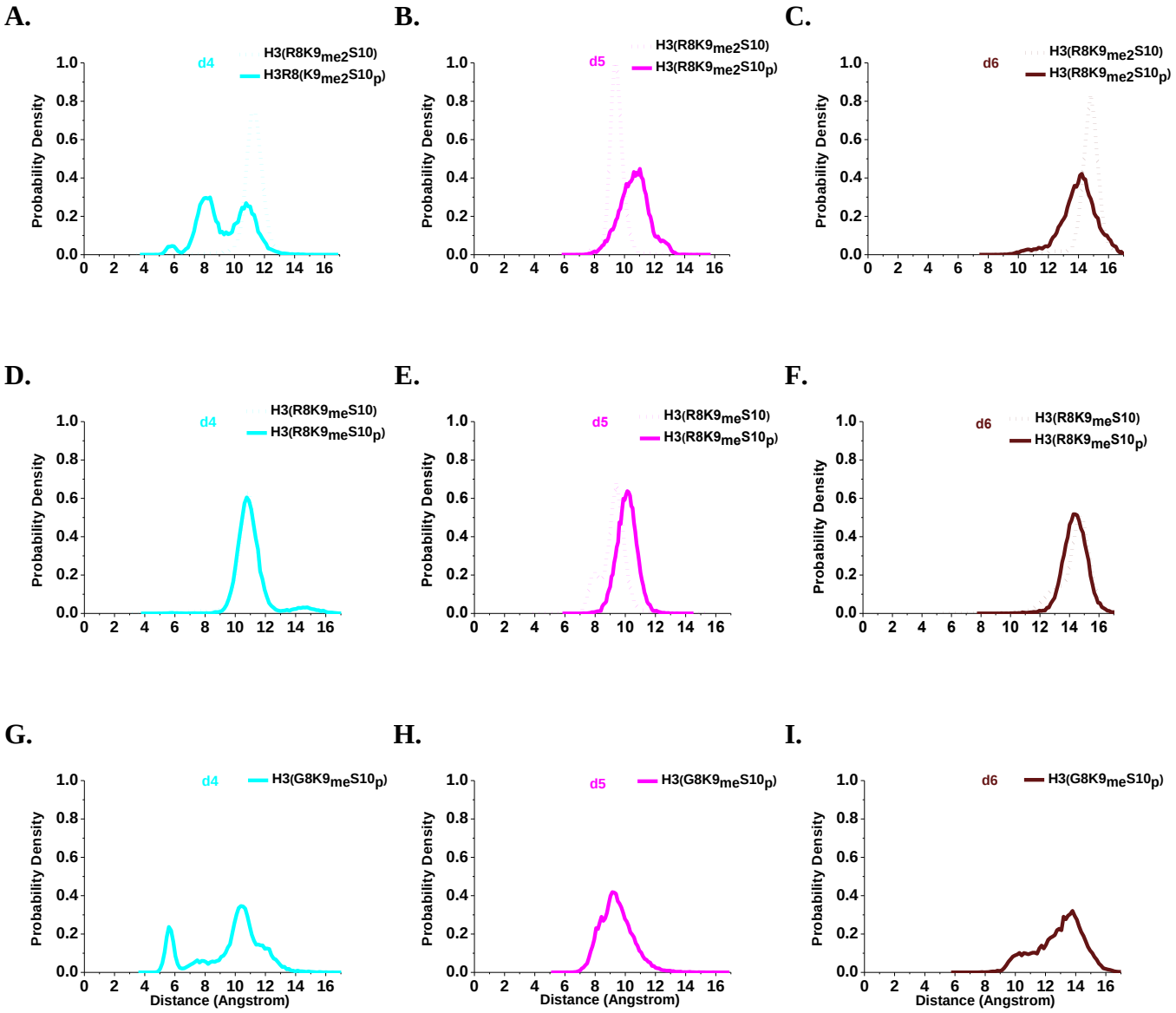

Figure S3: Probability Density Functions (PDFs) of the distance variables d4, d5 and d6 (defined in the text and shown in Fig. 3, *A* and *C*) for the various forms of the H3 tail. ***A-C.*** PDFs of d4 (***A***), d5 (***B***) and d6 (***C***) in the phosphorylated (solid line) and unphosphorylated (dashed line) form of the K9-dimethylated H3 tail. ***D-F***. PDFs of *d4* (***D***), *d5* (***E***) and *d6* (***F***) in the phosphorylated (solid line) and unphosphorylated (dashed line) form of the K9-mono-methylated H3 tail. ***G-I***. PDFs of *d4* (***G***), *d5* (***H***) and *d6* (***I***) in the phosphorylated form of the K9-mono-methylated "mutant" H3 peptide (R8 replaced by G).

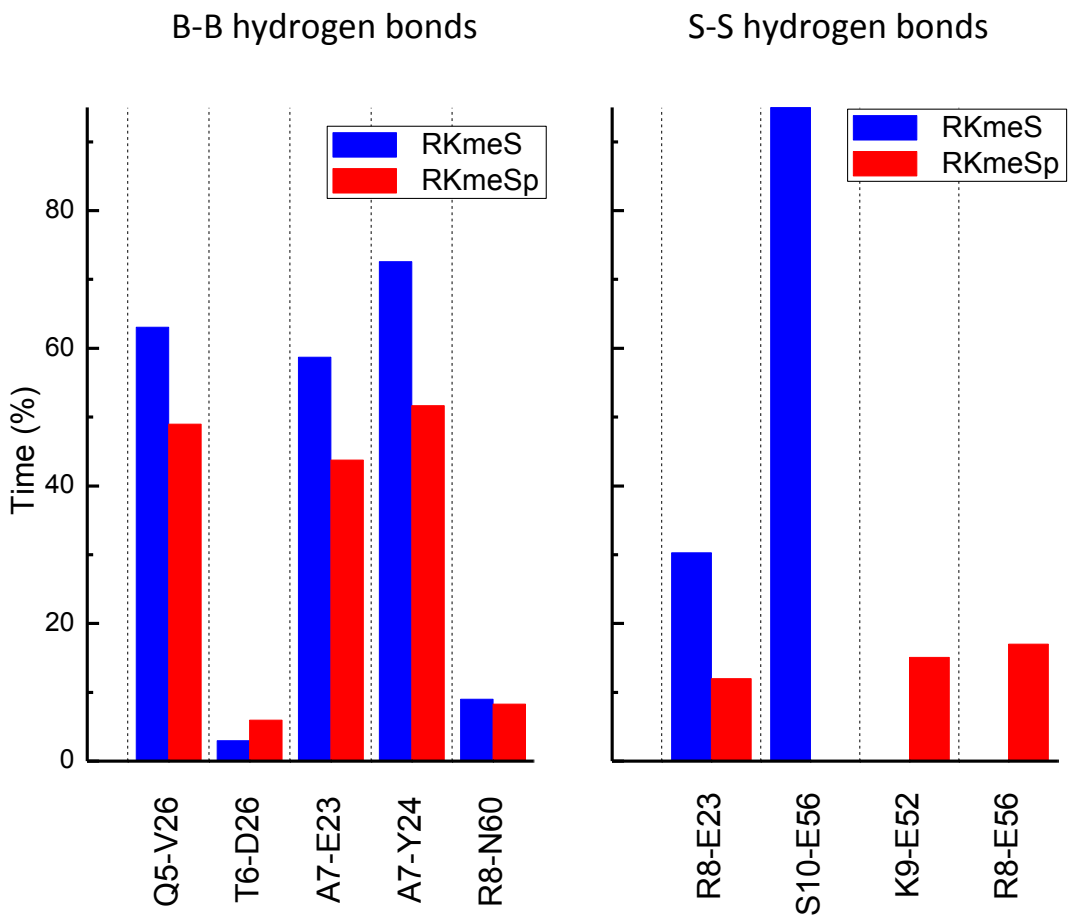

Figure S4: Hydrogen bonding between the H3 oligopeptide dimethylated at K9 and HP1. Vertical bars represent percentage of time during which hydrogen bonds persist for the various aminoacid pairs in the RKme2S *(blue)*and RKme2Sp *(red)* structures for the backbone atoms (B-B) *(left)* and the side chain atoms (S-S) (*right*).