

Supplementary Information

The Intricate Effects of alpha-Amino and Lysine Modifications on Arginine Methylation on the N-terminal Tail of Histone H4

Melody D. Fulton^{‡†}, Jing Zhang^{‡†}, Maomao He[†], Meng-Chiao Ho[‡], Y. George Zheng^{†*}

[‡] These authors made equal contribution to this work

[†]Department of Pharmaceutical and Biomedical Sciences, College of Pharmacy, The University
of Georgia, Athens, Georgia 30602, United States.

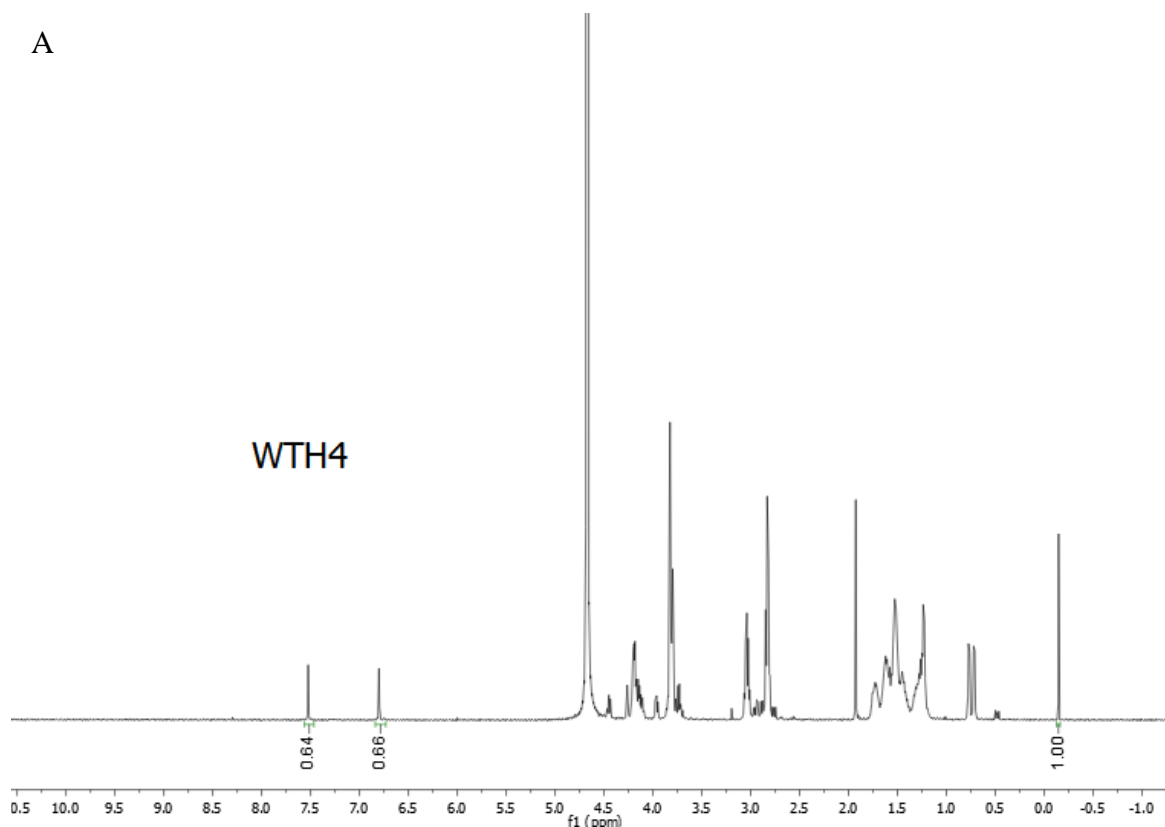
[‡]Institute of Biological Chemistry, Academia Sinica, Taipei 115, Taiwan

*** To whom correspondence should be addressed:** Y. G. Zheng, Tel: (706) 542-0277; Fax:
(706) 542-5358; Email: yzheng@uga.edu

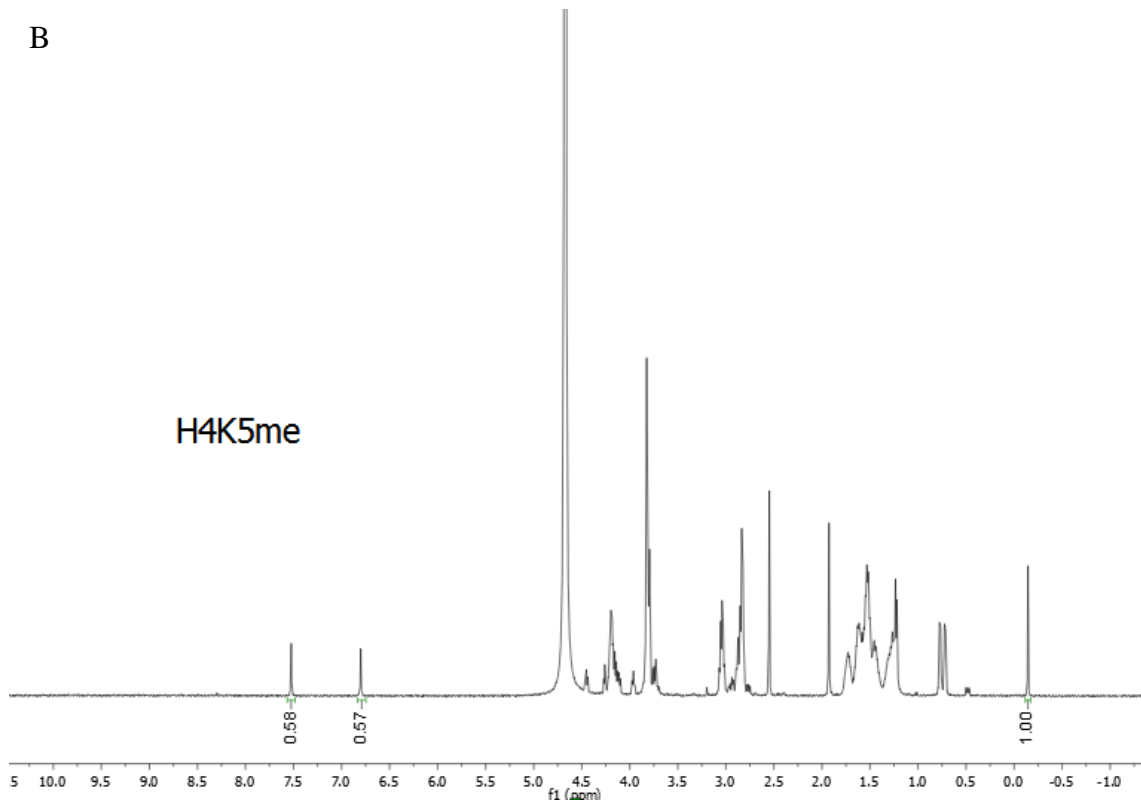
Table S1. Calibration of histone H4(1-20) peptide concentration by ^1H NMR. Adjustment factor = Mol (NMR) / Mol (weight).

Peptide	Code for NMR Spectrum	Adjustment Factor
Ac-H4(1-20)	WTH4	0.64
Ac-H4(1-20)K5 _{me}	H4K5me	0.58
Ac-H4(1-20)K5 _{ac}	H4K5ac	0.65
Ac-H4(1-20)K5 _{pr}	H4K5pr	0.64
Ac-H4(1-20)K5 _{bu}	H4K5bu	0.65
Ac-H4(1-20)K5 _{cr}	H4K5cr	0.77
Ac-H4(1-20)K5 _{hib}	H4K5hib	0.67

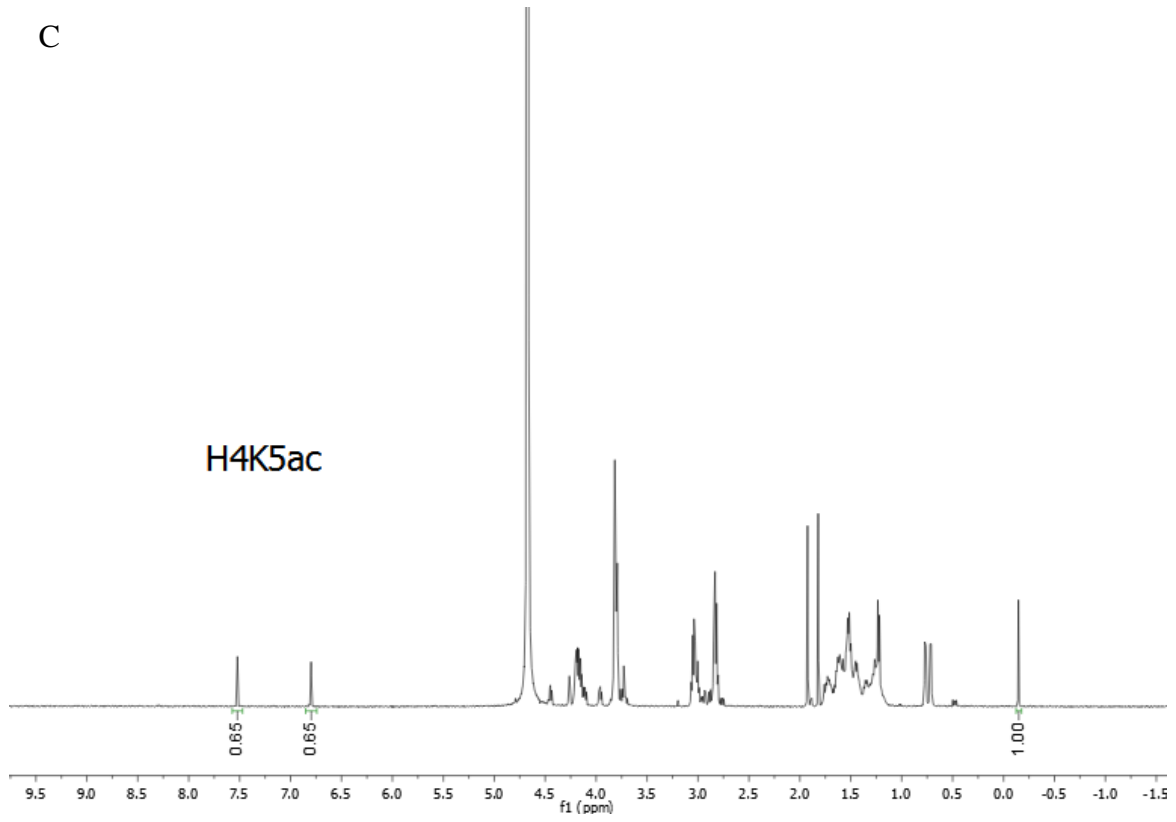
Figure S1. NMR spectra of the histone H4(1-20) peptides. A) Ac-H4(1-20), code WTH4. B) Ac-H4(1-20)K5_{me}, code H4K5me. C) Ac-H4(1-20)K5_{ac}, code H4K5ac. D) Ac-H4(1-20)K5_{pr}, code H4K5pr. E) Ac-H4(1-20)K5_{bu}, code H4K5bu. F) Ac-H4(1-20)K5_{cr}, code H4K5cr. G) Ac-H4(1-20)K5_{hib}, code H4K5hib.



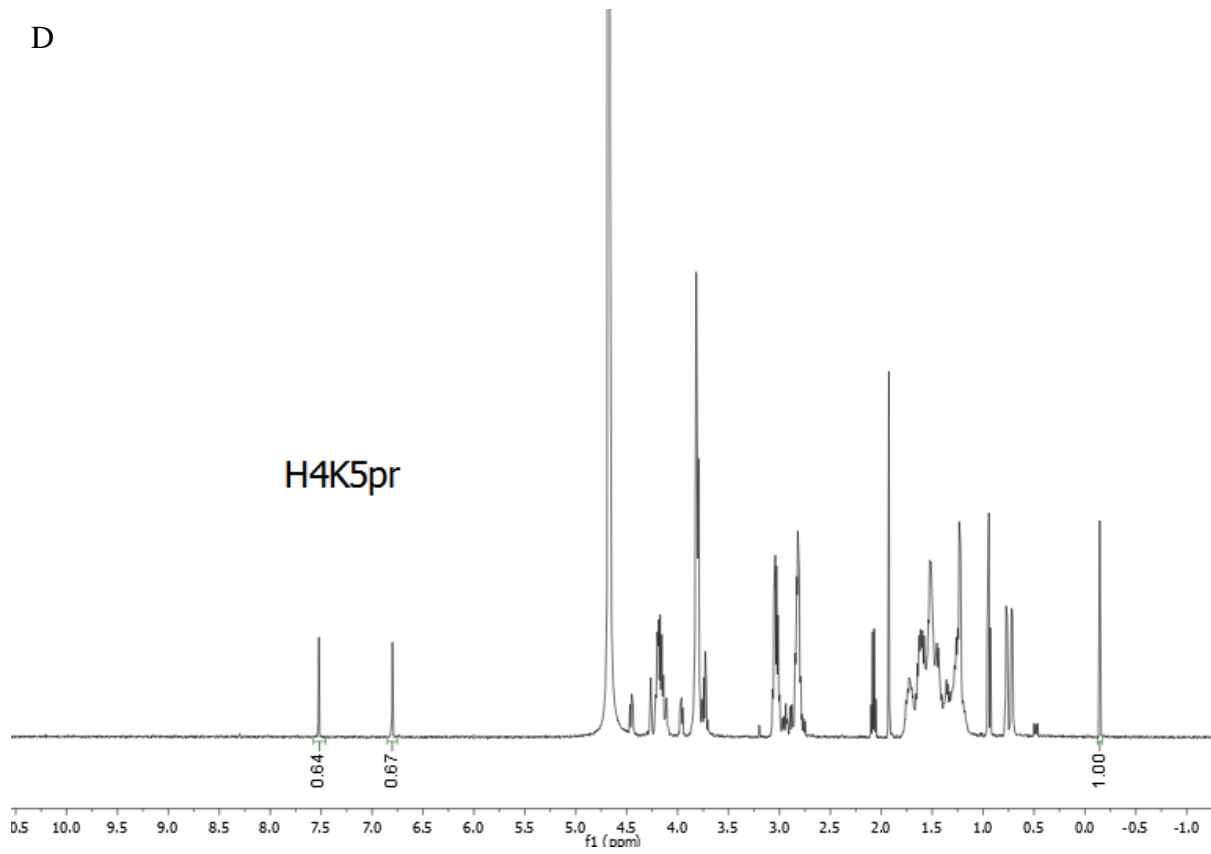
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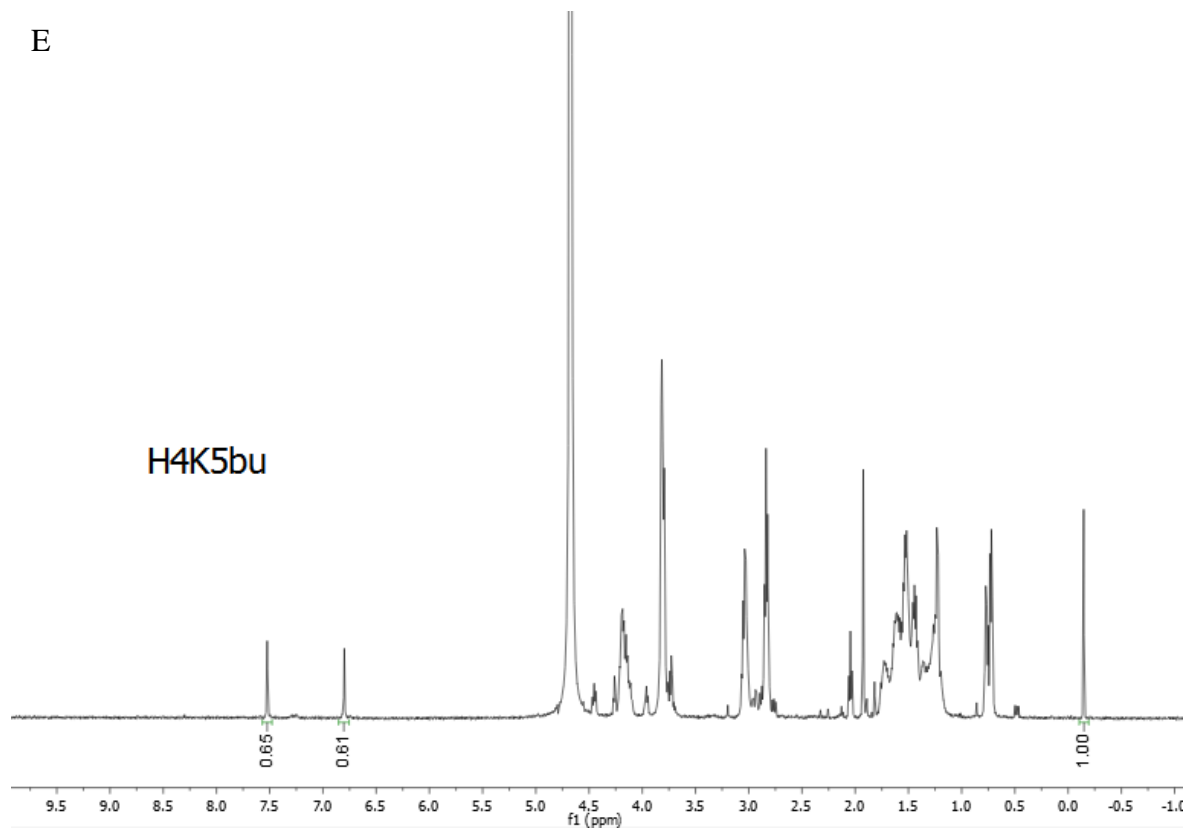
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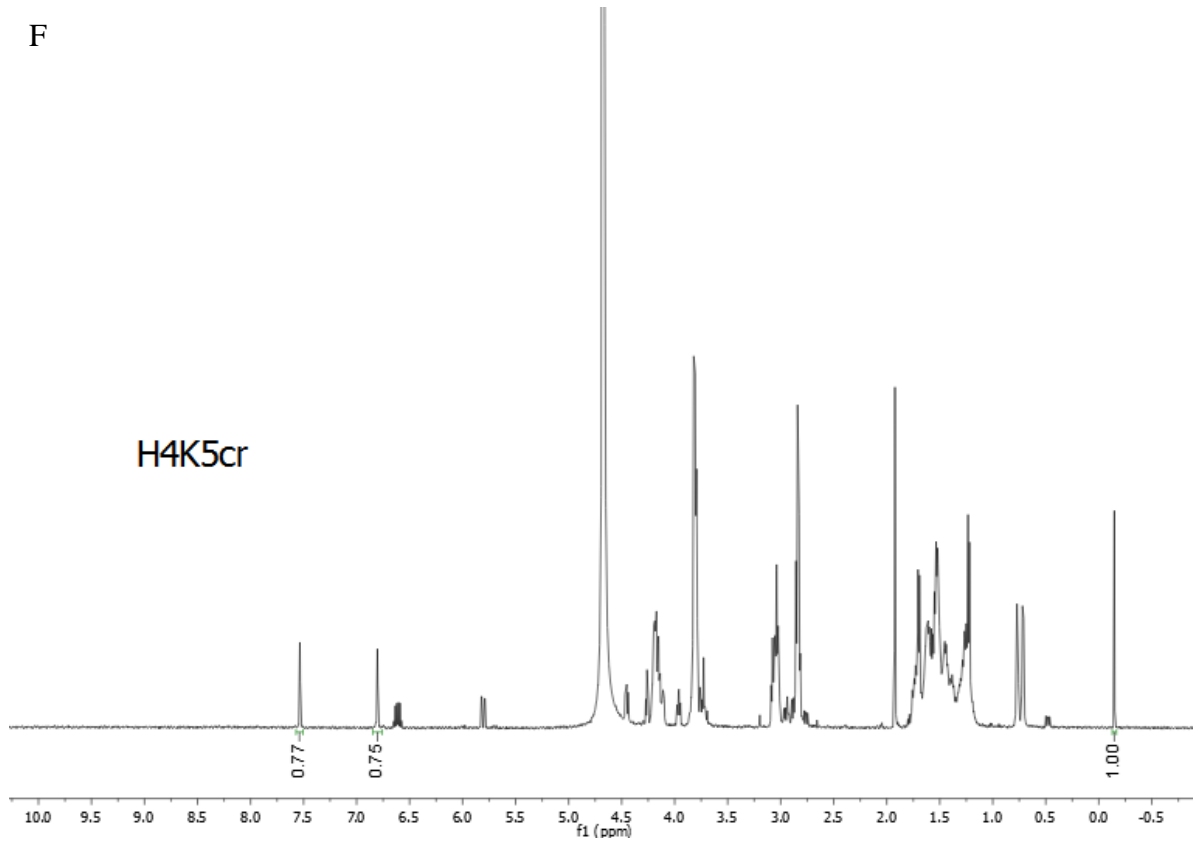
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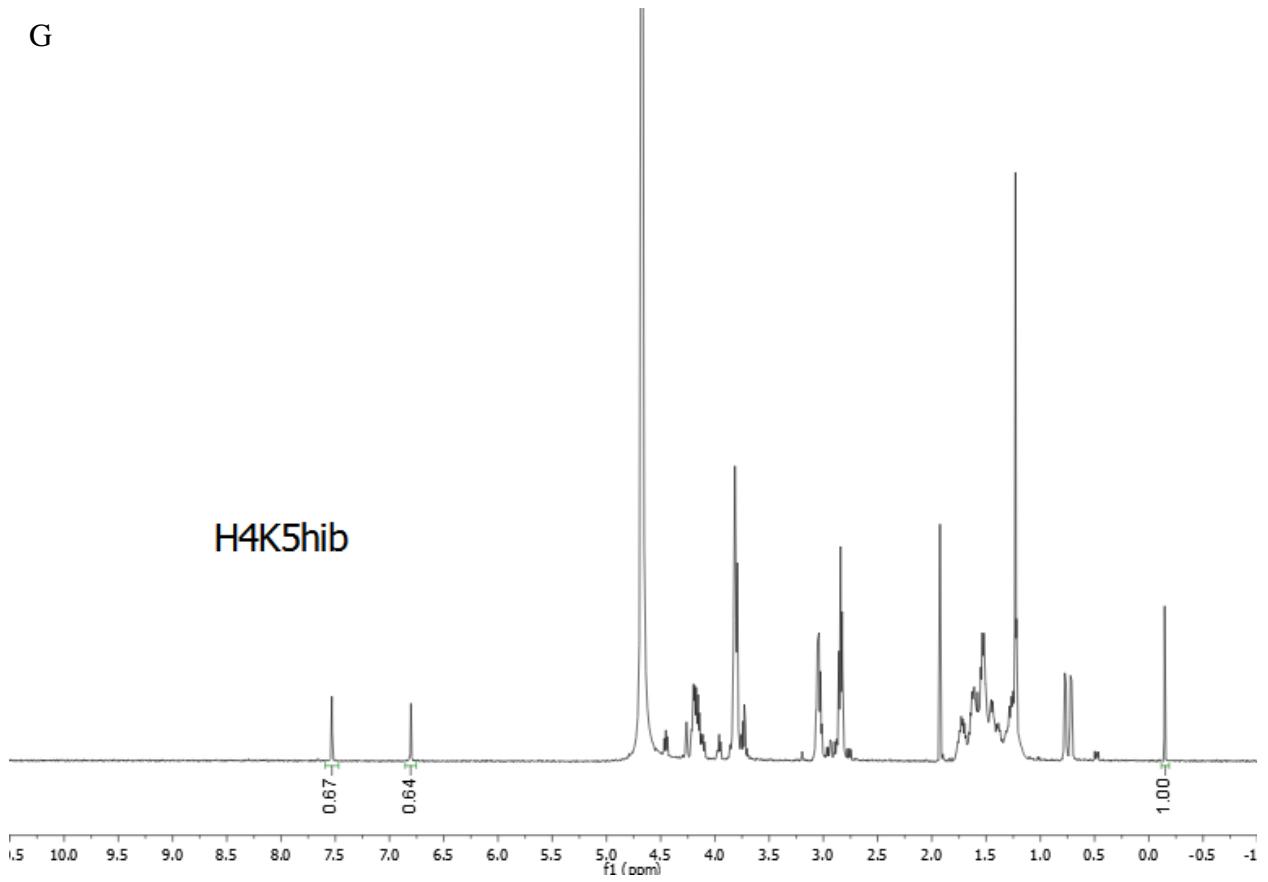
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F



G



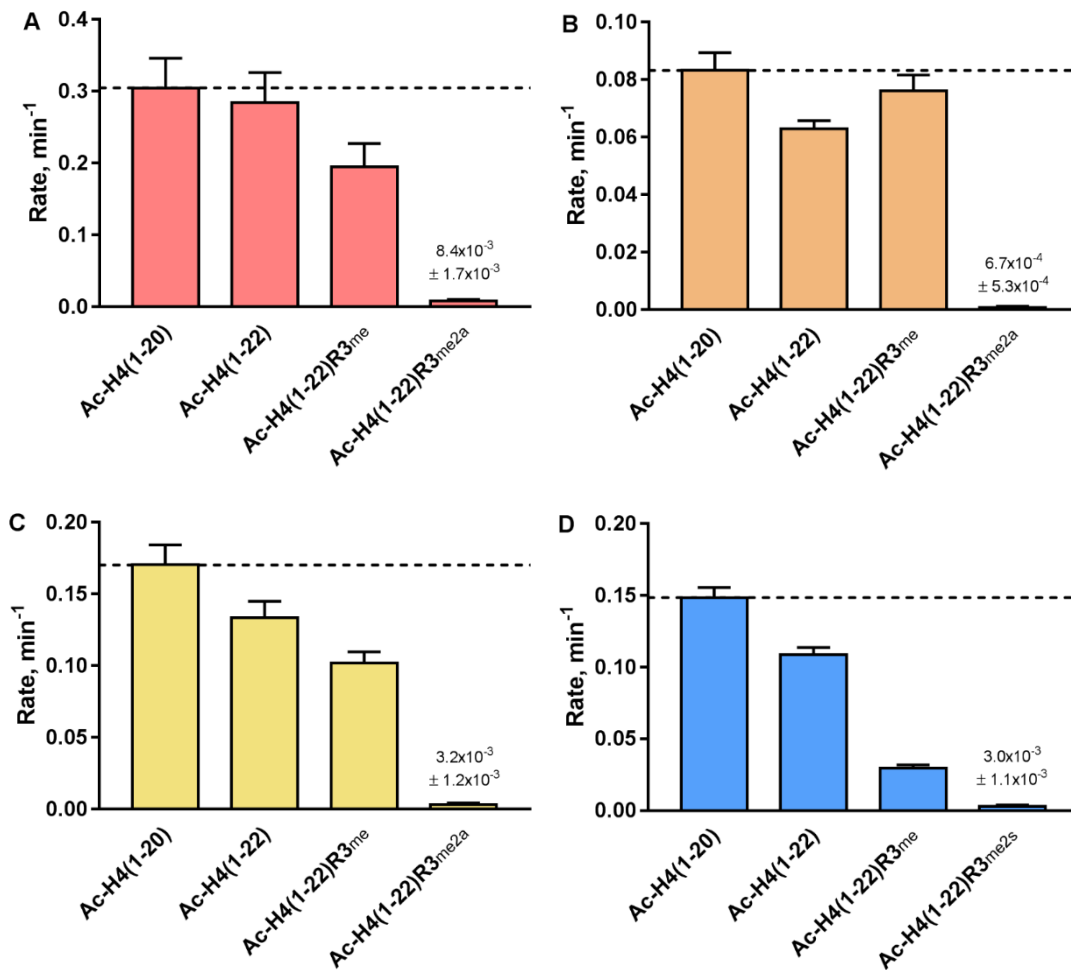


Figure S2. The impact of monomethylated (MMA) and dimethylated (ADMA or SDMA) H4R3 on PRMT activity. Single-point radioactive methylation assay performed with 0.05 μM PRMT, 5 μM [¹⁴C]SAM, and 10 μM of peptide substrate at 30 °C over a period of 35 min. Average rates of arginine methylation by A) PRMT1, B) PRMT3, C) PRMT8, and D) PRMT5-MEP50 with peptide substrates Ac-H4(1-20), Ac-H4(1-22), Ac-H4(1-22)R3^{me} (MMA), Ac-H4(1-22)R3^{me2a} (ADMA), and Ac-H4(1-22)R3^{me2s} (SDMA) are presented in each bar graph. Error bars represent standard deviation.