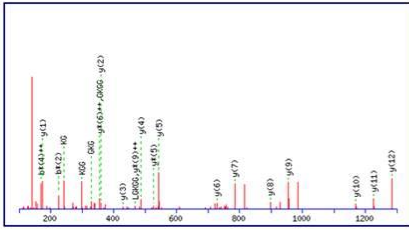


A

MS/MS Fragmentation of GKGGKGLGKGGAKR  
Match to Query 170: 1523.885448 from(762.950000,2+)

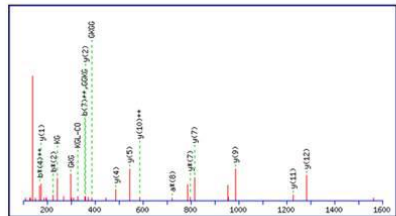


Monoisotopic mass of neutral peptide M<sub>r</sub>(calc): 1523.88  
 Variable modifications:  
 K2 : Propionyl (K)  
 K5 : hydroxy-Butanoyl (K)  
 K9 : Propionyl (K)  
 K13 : Propionyl (K)  
 Ions Score: 83 Expect: 1.3e-006  
 Matches (Bold Red): 31/259 fragment ions using 37 most intense peaks

#	Immon.	a	a <sup>++</sup>	a <sup>+</sup>	a <sup>+++</sup>	b	b <sup>++</sup>	b <sup>+</sup>	b <sup>+++</sup>	Seq.	y	y <sup>++</sup>	y <sup>+</sup>	y <sup>+++</sup>	#
1	30.03	30.03	15.52			58.03	29.52			G					14
2	157.13	214.15	107.58	197.13	99.07	<b>242.15</b>	121.58	<b>225.12</b>	113.07	K	1467.86	734.44	1450.84	725.92	13
3	30.03	271.18	136.09	254.15	127.58	<b>299.17</b>	150.09	282.14	141.58	G	<b>1283.74</b>	642.38	1266.72	633.86	12
4	30.03	328.20	164.60	311.17	156.09	<b>356.19</b>	178.60	339.17	<b>170.09</b>	G	<b>1226.72</b>	613.86	1209.69	605.35	11
5	187.14	542.33	271.67	525.30	263.16	570.32	285.67	553.30	277.15	K	<b>1169.70</b>	585.35	1152.67	576.84	10
6	30.03	599.35	300.18	582.32	291.67	627.35	314.18	610.32	305.66	G	<b>985.58</b>	478.29	938.54	<b>469.77</b>	9
7	86.10	712.44	356.72	695.41	348.21	740.43	370.72	723.40	362.21	L	<b>898.55</b>	449.78	881.52	441.26	8
8	30.03	769.46	385.23	752.43	376.72	797.45	399.23	780.43	390.72	G	<b>785.46</b>	393.24	768.44	384.72	7
9	157.13	953.58	477.29	936.55	468.78	981.57	491.29	964.55	482.78	K	<b>728.44</b>	364.72	711.41	<b>356.21</b>	6
10	30.03	1010.60	505.80	993.57	497.29	1038.59	519.80	1021.57	511.29	G	<b>544.32</b>	272.66	<b>527.29</b>	264.15	5
11	30.03	1067.62	534.31	1050.59	525.80	1095.62	548.31	1078.59	539.80	G	<b>487.30</b>	244.15	470.27	235.64	4
12	44.05	1138.66	569.83	1121.63	561.32	1166.65	583.83	1149.63	575.32	A	<b>430.28</b>	215.64	413.25	207.13	3
13	157.13	1322.78	661.89	1305.75	653.38	1350.77	675.89	1333.75	667.38	K	<b>359.24</b>	180.12	342.21	171.61	2
14	129.11									R	<b>175.12</b>	88.06	158.09	79.55	1

B

MS/MS Fragmentation of GKGGKGLGKGGAKR  
Match to Query 173: 1523.885448 from(762.950000,2+)

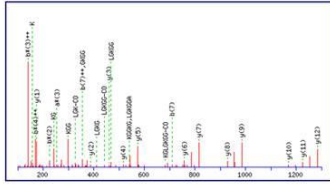


Monoisotopic mass of neutral peptide M<sub>r</sub>(calc): 1523.88  
 Variable modifications:  
 K2 : Propionyl (K)  
 K5 : Propionyl (K)  
 K9 : hydroxy-Butanoyl (K)  
 K13 : Propionyl (K)  
 Ions Score: 51 Expect: 0.0021  
 Matches (Bold Red): 27/259 fragment ions using 27 most intense peaks

#	Immon.	a	a <sup>++</sup>	a <sup>+</sup>	a <sup>+++</sup>	b	b <sup>++</sup>	b <sup>+</sup>	b <sup>+++</sup>	Seq.	y	y <sup>++</sup>	y <sup>+</sup>	y <sup>+++</sup>	#
1	30.03	30.03	15.52			58.03	29.52			G					14
2	157.13	214.15	107.58	197.13	99.07	<b>242.15</b>	121.58	<b>225.12</b>	113.07	K	1467.86	734.44	1450.84	725.92	13
3	30.03	271.18	136.09	254.15	127.58	<b>299.17</b>	150.09	282.14	141.58	G	<b>1283.74</b>	642.38	1266.72	633.86	12
4	30.03	328.20	164.60	311.17	156.09	<b>356.19</b>	178.60	339.17	<b>170.09</b>	G	<b>1226.72</b>	613.86	1209.69	605.35	11
5	157.13	512.32	256.66	495.29	248.15	540.31	270.66	523.29	262.15	K	1169.70	<b>585.35</b>	1152.67	576.84	10
6	30.03	569.34	285.17	552.31	276.66	597.34	<b>299.17</b>	580.31	290.66	G	<b>985.58</b>	493.29	968.55	484.78	9
7	86.10	682.42	341.72	665.40	333.20	710.42	<b>355.71</b>	693.39	347.20	L	928.56	464.78	911.53	456.27	8
8	30.03	739.45	370.23	<b>722.42</b>	361.71	767.44	384.22	750.41	375.71	G	<b>815.47</b>	408.24	<b>798.45</b>	399.73	7
9	187.14	953.58	477.29	936.55	468.78	981.57	491.29	964.55	482.78	K	758.45	379.73	741.43	371.22	6
10	30.03	1010.60	505.80	993.57	497.29	1038.59	519.80	1021.57	511.29	G	<b>544.32</b>	272.66	527.29	264.15	5
11	30.03	1067.62	534.31	1050.59	525.80	1095.62	548.31	1078.59	539.80	G	<b>487.30</b>	244.15	470.27	235.64	4
12	44.05	1138.66	569.83	1121.63	561.32	1166.65	583.83	1149.63	575.32	A	430.28	215.64	413.25	207.13	3
13	157.13	1322.78	661.89	1305.75	653.38	1350.77	675.89	1333.75	667.38	K	<b>359.24</b>	180.12	342.21	171.61	2
14	129.11									R	<b>175.12</b>	88.06	158.09	79.55	1

C

MS/MS Fragmentation of GKGKGLGKGGAKR  
Match to Query 167: 1523.885448 from(762.950000,2+)

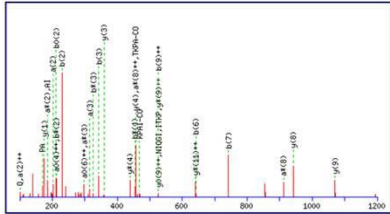


Monoisotopic mass of neutral peptide M<sub>r</sub>(calc): 1523.88  
Variable modifications:  
K2 : Propionyl (K)  
K5 : Propionyl (K)  
K8 : Propionyl (K)  
K13 : hydroxy-Butanoyl (K)  
Ions Score: 70 Expect: 2.9e-005  
Matches (bold red): 59/262 fragment ions using 54 most intense peaks

#	Immun.	a	a <sup>++</sup>	a <sup>*</sup>	a <sup>+++</sup>	b	b <sup>++</sup>	b <sup>*</sup>	b <sup>+++</sup>	Seq.	y	y <sup>++</sup>	y <sup>*</sup>	y <sup>+++</sup>	#
1	30.03	30.03	15.52				58.03	29.52		G					14
2	<b>157.13</b>	214.15	107.58	197.13	99.07	<b>242.15</b>	121.58	<b>225.12</b>	113.07	K	1467.86	734.44	1450.84	725.92	13
3	30.03	271.18	136.09	<b>254.15</b>	127.58	<b>299.17</b>	150.09	282.14	<b>141.58</b>	G	<b>1283.74</b>	642.38	1266.72	633.86	12
4	30.03	328.20	164.60	311.17	156.09	<b>356.19</b>	178.60	339.17	<b>170.09</b>	G	<b>1226.72</b>	613.86	1209.69	605.35	11
5	<b>157.13</b>	512.32	256.66	495.29	248.15	<b>540.31</b>	270.66	523.29	262.15	K	<b>1169.70</b>	585.35	1152.67	576.84	10
6	30.03	569.34	285.17	552.31	276.66	597.34	<b>299.17</b>	580.31	290.66	G	<b>985.58</b>	493.29	968.55	484.78	9
7	86.10	<b>682.42</b>	341.72	665.40	333.20	<b>710.42</b>	<b>355.71</b>	693.39	347.20	L	<b>928.56</b>	464.78	911.53	456.27	8
8	30.03	739.45	370.23	722.42	361.71	767.44	384.22	750.41	375.71	G	<b>815.47</b>	408.24	798.45	399.73	7
9	<b>157.13</b>	923.57	462.29	906.54	453.77	951.56	476.28	934.54	467.77	K	<b>758.45</b>	379.73	741.43	371.22	6
10	30.03	980.59	490.80	963.56	482.28	1008.58	504.80	991.56	496.28	G	<b>574.33</b>	287.67	557.30	279.16	5
11	30.03	1037.61	519.31	1020.58	510.80	1065.61	533.31	1048.58	524.79	G	<b>517.31</b>	259.16	500.28	250.64	4
12	44.05	1108.65	554.83	1091.62	546.31	1136.64	568.82	1119.62	560.31	A	<b>460.29</b>	230.65	443.26	222.13	3
13	187.14	1322.78	661.89	1305.75	653.38	1350.77	675.89	1333.75	667.38	K	<b>389.25</b>	195.13	372.22	186.62	2
14	129.11									R	<b>175.12</b>	88.06	158.09	79.55	1

D

MS/MS Fragmentation of DNIQGITKPAIR  
Match to Query 14944: 1410.785448 from(706.400000,2+)



Monoisotopic mass of neutral peptide M<sub>r</sub>(calc): 1410.78  
Variable modifications:  
K8 : hydroxy-Butanoyl (K)  
Ions Score: 34 Expect: 0.11  
Matches (bold red): 34/263 fragment ions using 35 most intense peaks

#	Immun.	a	a <sup>++</sup>	a <sup>*</sup>	a <sup>+++</sup>	a <sup>0</sup>	a <sup>0++</sup>	b	b <sup>++</sup>	b <sup>*</sup>	b <sup>+++</sup>	b <sup>0</sup>	b <sup>0++</sup>	Seq.	y	y <sup>++</sup>	y <sup>*</sup>	y <sup>+++</sup>	y <sup>0</sup>	y <sup>0++</sup>	#	
1	88.04	88.04	44.52			70.03	35.52	116.03	58.52			98.02	49.52	D								12
2	87.06	<b>202.08</b>	<b>101.54</b>	<b>185.06</b>	93.03	184.07	92.54	<b>230.08</b>	115.54	<b>213.05</b>	107.03	<b>212.07</b>	106.54	N	1296.76	648.89	1279.74	<b>640.37</b>	1278.75	639.88	11	
3	86.10	<b>315.17</b>	158.09	<b>298.14</b>	149.57	297.16	149.08	<b>343.16</b>	172.08	<b>326.13</b>	163.57	325.15	163.08	I	1182.72	591.86	1165.69	583.35	1164.71	582.86	10	
4	<b>101.07</b>	443.22	222.12	426.20	213.60	425.21	<b>215.11</b>	471.22	236.11	<b>454.19</b>	227.60	453.21	227.11	Q	<b>1069.64</b>	535.32	1052.61	<b>526.81</b>	1051.63	<b>526.32</b>	9	
5	30.03	500.25	250.63	483.22	242.11	482.24	241.62	528.24	264.62	511.21	256.11	510.23	255.62	G	<b>941.58</b>	471.29	924.55	462.78	923.57	462.29	8	
6	86.10	613.33	307.17	596.30	298.66	595.32	<b>298.16</b>	<b>641.33</b>	321.17	624.30	312.65	623.31	312.16	I	884.56	442.78	867.53	434.27	866.55	433.78	7	
7	74.06	714.38	357.69	697.35	349.18	696.37	348.69	<b>742.37</b>	371.69	725.35	363.18	724.36	362.68	T	771.47	386.24	754.45	377.73	753.46	377.23	6	
8	187.14	928.51	464.76	<b>911.48</b>	<b>456.25</b>	910.50	455.75	956.50	478.76	939.48	470.24	938.49	469.75	K	670.42	335.72	653.40	327.20			5	
9	70.07	1025.56	513.28	1008.54	504.77	1007.55	504.28	1053.56	<b>527.28</b>	1036.53	518.77	1035.55	518.28	P	<b>456.29</b>	228.65	<b>439.27</b>	220.14			4	
10	44.05	1096.60	548.80	1079.57	540.29	1078.59	539.80	1124.59	562.80	1107.57	554.29	1106.58	553.80	A	<b>359.24</b>	180.12	342.21	171.61			3	
11	86.10	1209.68	605.35	1192.66	596.83	1191.67	596.34	1237.68	619.34	1220.65	610.83	1219.67	610.34	I	288.20	144.61	271.18	136.09			2	
12	129.11													R	<b>175.12</b>	88.06	158.09	79.55			1	

Supplementary information, Figure S3 Spectra of the H4K<sub>hib</sub> peptides identified in the *in vitro* assay.

The reaction mixture was chemically propionylated (*Nat Chem Biol*, 2014, 10,365), tryptic digested, and subjected to MS/MS analysis. (A) Spectrum of H4K8<sub>hib</sub> peptide. (B) Spectrum of H4K12<sub>hib</sub> peptide. (C) Spectrum of H4K16<sub>hib</sub> peptide. (D) Spectrum of H4K31<sub>hib</sub> peptide.