SUPPLEMENTARY INFORMATION

Structural Motifs for CTD Kinase Specificity on RNA Polymerase II during Eukaryotic Transcription

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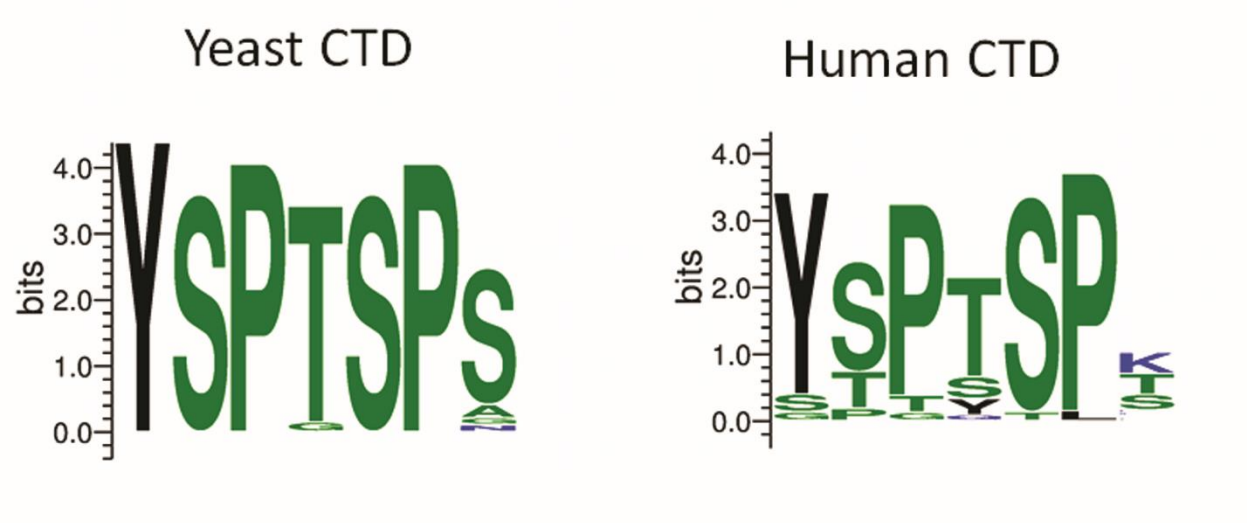
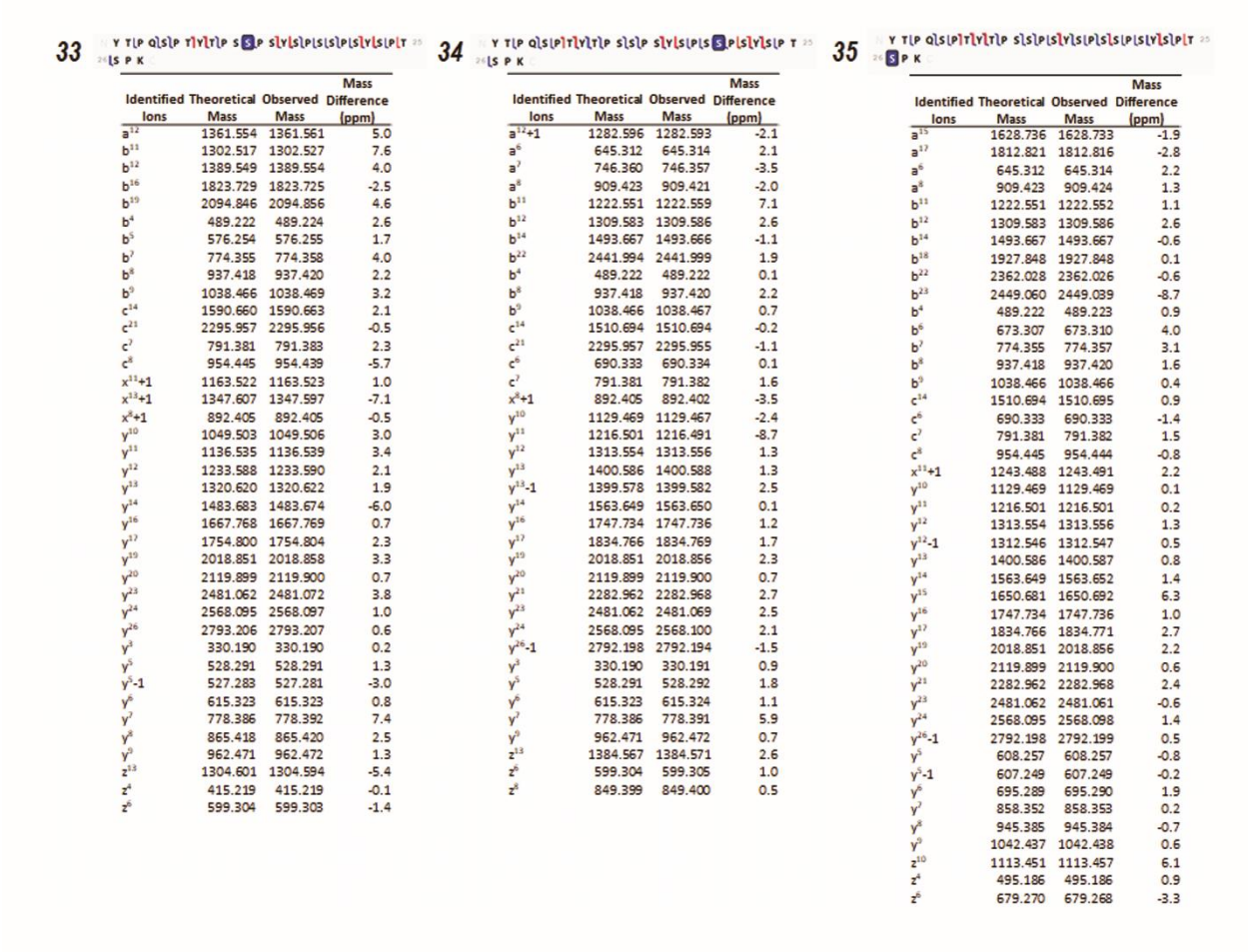


Supplementary Figure 1: Sequence logos of the conservation of Yeast and Human CTD. (LogOdds Logo NCBI: Yu, Y.-K.; Capra, J. A.; Stojmirović, A.; Landsman, D.; Altschul, S. F. Log-Odds Sequence Logos. Bioinformatics 2015, 31 (3), 324–331. https://doi.org/10.1093/bioinformatics/btu634.)



33

Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{12}	1361.554	1361.561	5.0
b^{11}	1302.517	1302.527	7.6
b^{12}	1389.549	1389.554	4.0
b^{16}	1823.729	1823.725	-2.5
b^{19}	2094.846	2094.856	4.6
b^{4}	489.222	489.224	2.6
b^{5}	576.254	576.255	1.7
b^{7}	774.355	774.358	4.0
b^{8}	937.418	937.420	2.2
b^{9}	1038.466	1038.469	3.2
c^{14}	1590.660	1590.663	2.1
c^{21}	2295.957	2295.956	-0.5
c^{7}	791.381	791.383	2.3
c^{8}	954.445	954.439	-5.7
x^{11}+1	1163.522	1163.523	1.0
x^{13}+1	1347.607	1347.597	-7.1
x^{8}+1	892.405	892.405	-0.5
y^{10}	1049.503	1049.506	3.0
y^{11}	1136.535	1136.539	3.4
y^{12}	1233.588	1233.590	2.1
y^{13}	1320.620	1320.622	1.9
y^{14}	1483.683	1483.674	-6.0
y^{16}	1667.768	1667.769	0.7
y^{17}	1754.800	1754.804	2.3
y^{19}	2018.851	2018.858	3.3
y^{20}	2119.899	2119.900	0.7
y^{23}	2481.062	2481.072	3.8
y^{24}	2568.095	2568.097	1.0
y^{26}	2793.206	2793.207	0.6
y^{3}	330.190	330.190	0.2
y^{5}	528.291	528.291	1.3
y^{5}-1	527.283	527.281	-3.0
y^{6}	615.323	615.323	0.8
y^{7}	778.386	778.392	7.4
y^{8}	865.418	865.420	2.5
y^{9}	962.471	962.472	1.3
z^{13}	1304.601	1304.594	-5.4
z^{4}	415.219	415.219	-0.1
z^{6}	599.304	599.303	-1.4

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Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{12}+1	1282.596	1282.593	-2.1
a^{6}	645.312	645.314	2.1
a^{7}	746.360	746.357	-3.5
a^{8}	909.423	909.421	-2.0
b^{11}	1222.551	1222.559	7.1
b^{12}	1309.583	1309.586	2.6
b^{14}	1493.667	1493.666	-1.1
b^{22}	2441.994	2441.999	1.9
b^{4}	489.222	489.222	0.1
b^{8}	937.418	937.420	2.2
b^{9}	1038.466	1038.467	0.7
c^{14}	1510.694	1510.694	-0.2
c^{21}	2295.957	2295.955	-1.1
c^{6}	690.333	690.334	0.1
c^{7}	791.381	791.382	1.6
x^{8}+1	892.405	892.402	-3.5
y^{10}	1129.469	1129.467	-2.4
y^{11}	1216.501	1216.491	-8.7
y^{12}	1313.554	1313.556	1.3
y^{13}	1400.586	1400.588	1.3
y^{13}-1	1399.578	1399.582	2.5
y^{14}	1563.649	1563.650	0.1
y^{16}	1747.734	1747.736	1.2
y^{17}	1834.766	1834.769	1.7
y^{19}	2018.851	2018.856	2.3
y^{20}	2119.899	2119.900	0.7
y^{21}	2282.962	2282.968	2.7
y^{23}	2481.062	2481.069	2.5
y^{24}	2568.095	2568.100	2.1
y^{26}-1	2792.198	2792.194	-1.5
y^{3}	330.190	330.191	0.9
y^{5}	528.291	528.292	1.8
y^{6}	615.323	615.324	1.1
y^{7}	778.386	778.391	5.9
y^{9}	962.471	962.472	0.7
z^{13}	1384.567	1384.571	2.6
z^{6}	599.304	599.305	1.0
z^{8}	849.399	849.400	0.5

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Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{15}	1628.736	1628.733	-1.9
a^{17}	1812.821	1812.816	-2.8
a^{6}	645.312	645.314	2.2
a^{8}	909.423	909.424	1.3
b^{11}	1222.551	1222.552	1.1
b^{12}	1309.583	1309.586	2.6
b^{14}	1493.667	1493.667	-0.6
b^{18}	1927.848	1927.848	0.1
b^{22}	2362.028	2362.026	-0.6
b^{23}	2449.060	2449.039	-8.7
b^{4}	489.222	489.223	0.9
b^{6}	673.307	673.310	4.0
b^{7}	774.355	774.357	3.1
b^{8}	937.418	937.420	1.6
b^{9}	1038.466	1038.466	0.4
c^{14}	1510.694	1510.695	0.9
c^{6}	690.333	690.333	-1.4
c^{7}	791.381	791.382	1.5
c^{8}	954.445	954.444	-0.8
x^{11}+1	1243.488	1243.491	2.2
y^{10}	1129.469	1129.469	0.1
y^{11}	1216.501	1216.501	0.2
y^{12}	1313.554	1313.556	1.3
y^{12}-1	1312.546	1312.547	0.5
y^{13}	1400.586	1400.587	0.8
y^{14}	1563.649	1563.652	1.4
y^{15}	1650.681	1650.692	6.3
y^{16}	1747.734	1747.736	1.0
y^{17}	1834.766	1834.771	2.7
y^{19}	2018.851	2018.856	2.2
y^{20}	2119.899	2119.900	0.6
y^{21}	2282.962	2282.968	2.4
y^{23}	2481.062	2481.061	-0.6
y^{24}	2568.095	2568.098	1.4
y^{26}-1	2792.198	2792.199	0.5
y^{5}	608.257	608.257	-0.8
y^{5}-1	607.249	607.249	-0.2
y^{6}	695.289	695.290	1.9
y^{7}	858.352	858.353	0.2
y^{8}	945.385	945.384	-0.7
y^{9}	1042.437	1042.438	0.6
z^{10}	1113.451	1113.457	6.1
z^{4}	495.186	495.186	0.9
z^{6}	679.270	679.268	-3.3

Supplementary Figure 2: Lists of fragment ions for peptides analyzed in Figure 1B by UVPD-MS for repeats 33,34 and 35. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.

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Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{12}+1	1335.551	1335.554	2.0
a^{6}+1	605.294	605.293	-1.6
a^{8}+1	869.405	869.403	-1.8
a^{2}	222.100	222.101	1.3
b^{12}	1362.538	1362.539	1.0
b^{4}	448.196	448.196	1.1
b^{8}	896.392	896.393	2.0
c^{2}	267.122	267.122	2.0
c^{4}	465.222	465.223	1.3
c^{6}	649.307	649.306	-2.1
x^{13}+1	1469.620	1469.620	0.1
x^{6}+1	736.292	736.288	-4.9
x^{7}+1	899.355	899.354	-1.7
x^{8}+1	1000.403	1000.400	-2.9
x^{10}	1183.480	1183.482	1.4
x^{11}	1284.528	1284.531	2.6
y^{10}	1157.501	1157.503	2.2
y^{11}	1258.548	1258.551	2.3
y^{12}	1355.601	1355.603	1.4
y^{13}	1442.633	1442.634	0.9
y^{2}	243.158	243.159	1.5
y^{7}	872.368	872.370	2.0
y^{8}	973.416	973.418	2.5
y^{9}	1070.469	1070.461	-7.5
z^{11}	1242.529	1242.533	2.5
z^{13}	1426.614	1426.619	3.6
z^{8}	957.397	957.392	-5.2

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Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{6}+1	685.260	685.261	0.9
a^{9}+1	1036.403	1036.399	-3.5
a^{10}	1132.448	1132.447	-0.9
a^{12}	1334.543	1334.544	0.3
a^{13}	1431.596	1431.593	-2.1
a^{8}	948.363	948.358	-4.9
b^{12}	1362.538	1362.541	2.0
b^{7}	813.295	813.295	0.4
b^{8}	976.358	976.359	1.5
c^{11}	1278.517	1278.517	0.2
c^{2}	267.122	267.122	2.1
c^{7}	830.321	830.321	-0.5
c^{8}	993.384	993.387	2.7
x^{13}+1	1469.620	1469.618	-1.2
x^{8}+1	920.437	920.435	-1.3
x^{11}	1284.528	1284.531	2.4
x^{4}	471.233	471.233	0.9
x^{7}	818.381	818.380	-1.3
y^{10}	1157.501	1157.504	2.9
y^{11}	1258.548	1258.548	-0.3
y^{12}	1355.601	1355.602	0.9
y^{13}	1442.633	1442.634	0.9
y^{2}	243.158	243.159	1.4
y^{3}	344.206	344.206	0.9
y^{5}	542.306	542.307	0.5
y^{6}	629.338	629.340	1.9
y^{7}	792.402	792.403	1.7
y^{8}	893.449	893.450	1.1
y^{9}	990.502	990.504	1.9
z^{11}	1242.529	1242.530	0.1
z^{13}	1426.614	1426.616	1.1
z^{4}	429.235	429.235	0.8
z^{6}	613.320	613.320	1.1
z^{8}	877.431	877.431	0.3

Supplementary Figure 3: Lists of fragment ions for peptides analyzed in Figure 1B by UVPD-MS for repeats 41 and 42. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.

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Y S|P T|S|P|T|V|S|P|T|S|P|V|V|T|P|T|S|P K

Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{14}+1	1517.657	1517.659	2.0
a^{9}+1	956.437	956.440	3.6
a^{15}	1679.712	1679.713	0.5
a^{7}	705.333	705.334	0.5
a^{8}	868.397	868.393	-3.7
b^{11}	1181.524	1181.521	-3.0
b^{14}	1544.644	1544.649	3.3
b^{15}	1707.707	1707.706	-0.3
b^{16}	1808.755	1808.759	2.6
b^{2}	250.095	250.096	1.2
b^{4}	448.196	448.196	0.5
b^{7}	733.328	733.329	1.0
b^{8}	896.392	896.393	1.6
b^{9}	983.424	983.425	1.8
c^{14}	1561.670	1561.677	4.6
c^{2}	267.122	267.122	2.0
c^{7}	750.355	750.357	2.8
x^{11}+1	1283.556	1283.555	-0.7
x^{4}+1	458.225	458.225	0.1
x^{4}	457.217	457.218	0.8
x^{7}	818.381	818.383	2.5
x^{8}	917.449	917.451	1.3
y^{10}	1155.521	1155.524	2.4
y^{11}	1256.569	1256.565	-3.5
y^{12}	1353.622	1353.623	1.3
y^{13}	1440.654	1440.657	2.1
y^{14}	1603.717	1603.722	3.0
y^{15}	1704.765	1704.752	-7.5
y^{16}	1801.818	1801.821	1.9
y^{19}	2086.950	2086.954	2.0
y^{2}	243.158	243.159	1.2
y^{3}	330.190	330.191	1.3
y^{5}	528.291	528.291	1.2
y^{6}	629.338	629.339	1.4
y^{7}	792.402	792.403	1.3
y^{8}	891.470	891.476	6.4
y^{9}	988.523	988.524	1.5
z^{11}	1240.550	1240.549	-0.8
z^{4}	415.219	415.219	-0.5
z^{6}	613.320	613.320	0.6
z^{8}	875.451	875.453	2.1

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Y S|P T|S|P T|V|S|P|T|S|P|V|V|T|P T S|P K

Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{13}+1	1338.622	1338.621	-0.6
a^{9}+1	956.437	956.427	-9.6
a^{12}	1240.561	1240.553	-6.7
a^{8}	868.397	868.397	0.7
b^{11}	1181.524	1181.522	-1.7
b^{12}	1268.556	1268.557	0.9
b^{14}	1464.677	1464.681	2.8
b^{15}	1627.741	1627.743	1.3
b^{16}	1728.788	1728.790	0.7
b^{2}	250.095	250.096	0.6
b^{4}	448.196	448.196	-0.1
b^{7}	733.328	733.330	2.6
b^{8}	896.392	896.393	1.4
b^{9}	983.424	983.425	1.3
c^{14}	1481.704	1481.706	1.8
c^{2}	267.122	267.122	1.6
c^{7}	750.355	750.356	2.3
c^{8}	913.418	913.420	2.0
c^{9}	1000.450	1000.450	0.4
y^{10}	1155.521	1155.523	1.7
y^{11}	1256.569	1256.570	0.7
y^{12}	1353.622	1353.623	0.8
y^{13}	1440.654	1440.656	1.8
y^{14}	1603.717	1603.722	2.8
y^{16}	1801.818	1801.820	1.3
y^{19}	2086.950	2086.950	0.2
y^{2}	243.158	243.159	0.7
y^{3}	608.257	608.257	0.2
y^{6}	709.305	709.305	0.6
y^{7}	872.368	872.369	0.9
y^{8}	971.437	971.437	0.1
y^{9}	1068.489	1068.491	1.4
z^{11}	1240.550	1240.553	2.1
z^{6}	693.286	693.286	-0.5

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Y S|P T S|P T|V|S|P|T|S|P|V|V|T|P|T|S|P K

Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{14}+1	1517.657	1517.660	2.5
b^{11}	1261.490	1261.492	1.2
b^{12}	1348.522	1348.524	1.4
b^{14}	1544.644	1544.646	1.8
b^{15}	1707.707	1707.709	1.2
b^{16}	1808.755	1808.757	1.4
b^{4}	448.196	448.196	0.1
b^{7}	813.295	813.295	0.6
b^{8}	976.358	976.359	0.9
b^{9}	1063.390	1063.391	1.4
c^{14}	1561.670	1561.672	1.3
c^{2}	267.122	267.122	1.8
c^{7}	830.321	830.322	0.8
c^{9}	1080.416	1080.415	-0.8
x^{11}+1	1203.590	1203.592	2.3
x^{2}+1	270.145	270.145	0.1
x^{7}+1	819.389	819.389	0.4
x^{8}	917.449	917.453	3.4
y^{10}	1075.555	1075.557	1.7
y^{11}	1176.603	1176.606	3.2
y^{12}	1273.655	1273.657	1.1
y^{13}	1360.687	1360.690	2.1
y^{14}	1523.751	1523.750	-0.6
y^{16}	1721.851	1721.853	1.1
y^{17}	1888.850	1888.843	-3.4
y^{19}	2086.950	2086.952	0.8
y^{2}	243.158	243.159	1.1
y^{3}	330.190	330.191	1.0
y^{5}	528.291	528.291	1.1
y^{6}	629.338	629.339	1.4
y^{7}	792.402	792.403	1.1
y^{8}	891.470	891.473	2.7
y^{9}	988.523	988.525	1.6
z^{11}	1160.584	1160.583	-0.8
z^{4}	415.219	415.219	-0.9
z^{6}	613.320	613.320	0.2
z^{8}	875.451	875.453	2.3

Supplementary Figure 4: Lists of fragment ions for peptides analyzed in Figure 1B by UVPD-MS for repeats 43,44 and 45. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.

46/48 N Y S P T S P T Y S P T S P K C

Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
$a^{12}+1$	1321.535	1321.536	0.8
$a^{9}+1$	956.437	956.437	-0.1
a^{8}	868.397	868.399	2.5
b^{4}	448.196	448.196	1.2
b^{5}	535.228	535.228	1.1
b^{8}	896.392	896.390	-1.4
c^{1}	180.090	180.090	3.0
c^{2}	267.122	267.122	2.3
c^{9}	1000.450	1000.453	3.0
$x^{12}+1$	1368.572	1368.583	7.9
$x^{13}+1$	1455.604	1455.608	2.6
$x^{5}+1$	635.244	635.245	0.7
$x^{7}+1$	885.340	885.340	0.8
$x^{8}+1$	986.387	986.384	-3.2
x^{11}	1270.512	1270.515	2.7
x^{6}	721.268	721.269	1.2
y^{10}	1143.485	1143.488	2.5
y^{11}	1244.533	1244.535	1.5
y^{12}	1341.585	1341.587	1.4
y^{2}	243.158	243.159	1.6
y^{4}	511.204	511.206	2.6
y^{5}	608.257	608.258	0.8
y^{7}	858.352	858.354	1.2
y^{8}	959.400	959.403	3.4
y^{9}	1056.453	1056.454	1.4
z^{11}	1228.514	1228.516	1.9
z^{13}	1412.599	1412.601	1.5
z^{4}	495.186	495.186	0.4
z^{6}	679.270	679.270	0.1
z^{8}	943.381	943.383	2.0

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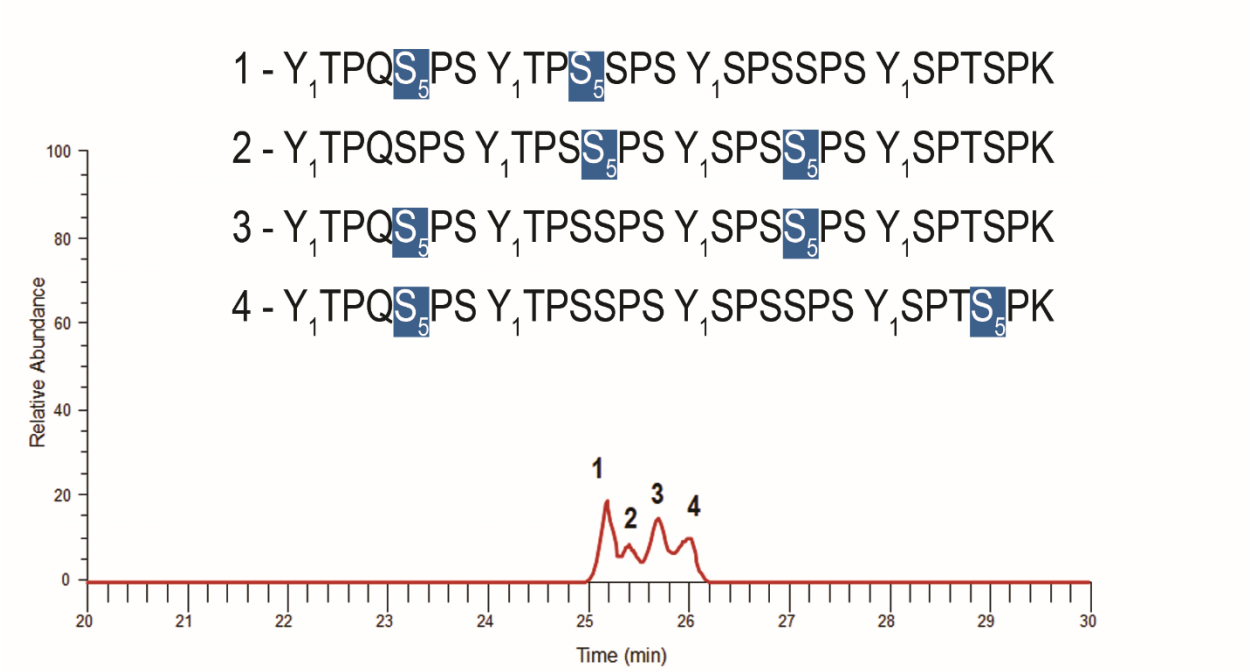
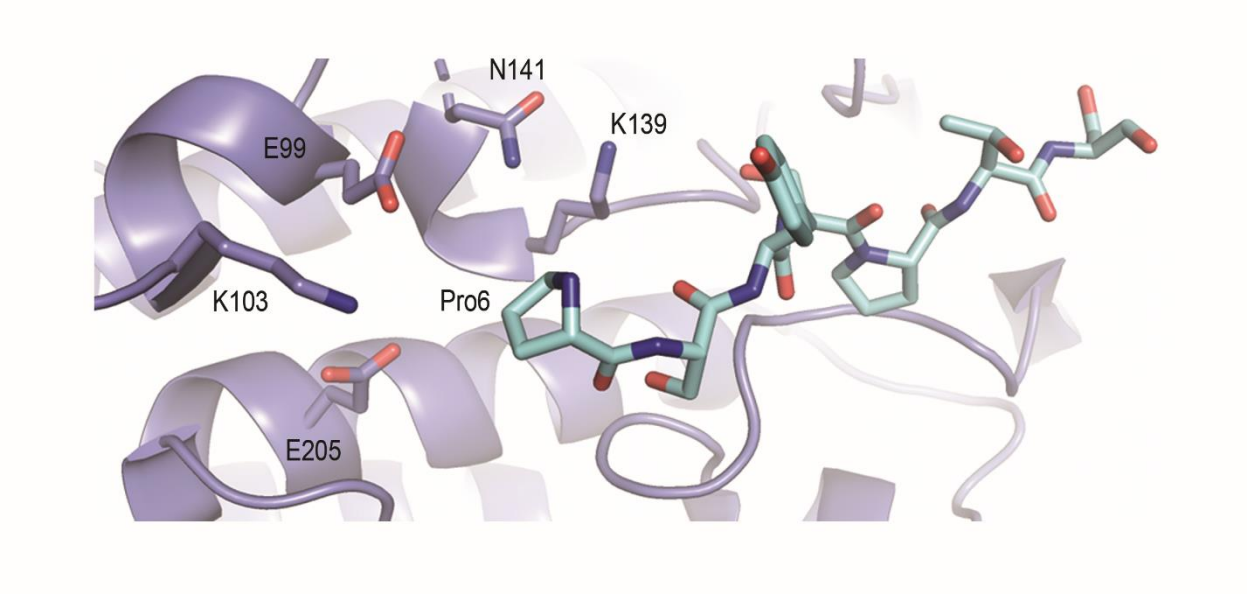
Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
$a^{6}+1$	685.260	685.260	-0.2
$a^{9}+1$	1036.403	1036.401	-1.5
a^{10}	1132.448	1132.450	1.5
a^{12}	1320.528	1320.526	-0.9
a^{13}	1417.580	1417.577	-2.3
a^{8}	948.363	948.361	-2.5
b^{12}	1348.522	1348.523	0.4
b^{7}	813.295	813.295	0.1
b^{8}	976.358	976.358	0.5
c^{11}	1278.517	1278.517	-0.1
c^{6}	729.273	729.274	1.3
c^{8}	993.384	993.386	1.5
$x^{13}+1$	1455.604	1455.604	-0.7
$x^{4}+1$	458.225	458.225	0.4
$x^{6}+1$	642.310	642.309	-1.8
x^{10}	1169.464	1169.463	-1.3
x^{11}	1270.512	1270.514	1.9
x^{4}	457.217	457.217	-0.1
x^{7}	804.365	804.366	0.8
x^{8}	905.413	905.414	1.1
y^{10}	1143.485	1143.489	3.4
y^{11}	1244.533	1244.532	-0.9
y^{12}	1341.585	1341.586	0.8
y^{13}	1428.617	1428.619	1.4
y^{2}	243.158	243.159	1.5
y^{3}	330.190	330.191	1.1
y^{5}	528.291	528.291	1.1
y^{6}	615.323	615.324	1.5
y^{7}	778.386	778.386	0.3
y^{8}	879.434	879.436	2.7
y^{9}	976.487	976.489	2.2
z^{11}	1228.514	1228.516	1.7
z^{13}	1412.599	1412.600	1.3
z^{4}	415.219	415.219	0.1
z^{6}	599.304	599.304	0.5
z^{8}	863.415	863.416	1.3

Supplementary Figure 5: Lists of fragment ions for peptides analyzed in Figure 1B by UVPD-MS for repeats 46/48 to 47/49. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.



Supplementary Figure 6: LC-MS/MS trace obtained for doubly phosphorylated peptides of T4Q variation in human distal CTD. The peak numbers correspond to the indicated sites (blue highlights) of phosphorylation identified by MS/MS.



Supplementary Figure 7: The modeling of CTD with Ser2 in the active site places Pro6 in a hydrophilic environment.

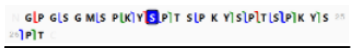
2

Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{+12}	1118.530	1118.531	0.8
a^{+15}	1403.663	1403.670	5.2
a^{+18}	1781.853	1781.851	-1.4
a^{+26}	2719.248	2719.249	0.6
a^{+8}	643.287	643.288	0.7
a^{13}	1218.570	1218.576	4.6
a^{17}	1693.813	1693.816	1.8
a^{20}	1978.946	1978.954	4.1
a^{22}	2163.031	2163.028	-1.0
a^{24}	2454.189	2454.190	0.6
a^{9}	770.375	770.377	3.2
b^{13}	1246.565	1246.568	2.3
b^{14}	1333.597	1333.596	-0.9
b^{16}	1558.745	1558.757	8.0
b^{18}	1808.840	1808.825	-8.6
b^{20}	2006.941	2006.939	-0.7
b^{21}	2093.973	2093.972	-0.6
b^{23}	2319.121	2319.118	-1.1
b^{9}	798.369	798.365	-6.0
c^{10}	978.459	978.459	-0.1
c^{12}	1162.544	1162.545	1.3
c^{13}	1263.592	1263.591	-0.5
c^{15}	1447.676	1447.677	0.6
c^{16}	1575.771	1575.772	0.7
c^{17}	1738.835	1738.834	-0.3
c^{26}	2763.261	2763.257	-1.7
c^{6}	503.216	503.217	1.8
c^{9}	815.396	815.392	-4.4
x^{17}	1929.840	1929.839	-0.4
x^{19}	2220.998	2221.007	4.2
y^{10}	1143.485	1143.480	-4.1
y^{11}	1306.548	1306.561	9.5
y^{13}-1	1530.688	1530.682	-4.1
y^{14}	1618.728	1618.722	-3.8
y^{16}-1	1815.821	1815.820	-0.4
y^{20}	2292.072	2292.072	0.2
y^{21}	2379.104	2379.108	1.9
y^{25}	2711.219	2711.232	4.8
y^{9}-1	1055.445	1055.444	-0.6
z^{25}	2695.200	2695.223	8.5
z^{26}	2792.253	2792.274	7.6

3

Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{9}	770.375	770.370	-5.3
a^{26}	2718.240	2718.253	4.8
a^{17}	1693.813	1693.815	1.2
a^{8}+1	643.287	643.286	-1.9
a^{25}+1	2622.195	2622.199	1.3
a^{22}+1	2244.005	2244.004	-0.2
a^{19}+1	1958.872	1958.874	0.8
a^{18}+1	1861.820	1861.818	-0.9
a^{21}+1	2146.952	2146.963	5.3
a^{11}+1	1021.478	1021.468	-9.9
a^{12}+1	1118.530	1118.531	0.1
b^{14}	1333.597	1333.595	-1.9
b^{11}	1048.465	1048.464	-0.7
b^{21}	2173.939	2173.934	-2.3
b^{23}	2399.087	2399.076	-4.4
c^{13}	1263.592	1263.585	-4.9
c^{12}	1162.544	1162.544	0.4
c^{19}	2002.886	2002.879	-3.2
x^{17}	1929.840	1929.839	-0.4
x^{19}	2220.998	2221.005	3.0
y^{6}	691.354	691.354	0.3
y^{7}	778.386	778.388	2.1
y^{10}	1143.485	1143.482	-2.8
y^{13}	1531.696	1531.693	-1.9
y^{16}-1	1815.821	1815.819	-0.8
z^{8}	863.415	863.412	-4.0
z^{17}	1887.842	1887.831	-5.9

4



Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{26}	2718.240	2718.252	4.2
a^{17}	1773.780	1773.781	0.9
a^{24}	2534.155	2534.160	2.0
a^{22}	2242.997	2243.000	1.5
a^{12}+1	1198.497	1198.497	-0.1
a^{18}+1	1861.820	1861.824	2.6
a^{19}+1	1958.872	1958.878	2.9
a^{21}+1	2146.952	2146.954	1.0
b^{25}	2649.182	2649.191	3.4
b^{9}	798.369	798.364	-6.7
b^{21}	2173.939	2173.946	3.4
c^{19}	2002.886	2002.875	-5.6
c^{10}	978.459	978.460	1.1
x^{19}	2220.998	2221.012	6.4
x^{8}+1	906.421	906.415	-6.8
y^{6}	691.354	691.354	-0.8
y^{7}	778.386	778.388	2.6
y^{9}	976.487	976.486	-1.1
y^{13}	1451.730	1451.727	-1.5
y^{21}	2379.104	2379.105	0.7
y^{24}-1	2653.190	2653.182	-2.9
y^{6}-1	690.346	690.342	-6.7
y^{16}-1	1735.854	1735.852	-1.2
z^{17}	1887.842	1887.843	0.5
z^{26}	2792.253	2792.267	5.1

Supplementary Figure 8: Lists of fragment ions for mono-phosphorylated peptides (m/z of 956.11) analyzed in Figure 6A by UVPD-MS. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.

2

Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
$a^{11}+1$	1022.425	1022.416	-9.5
$a^{12}+1$	1119.478	1119.486	6.8
$a^{13}+1$	1404.611	1404.615	2.8
$a^{23}+1$	2374.943	2374.953	4.5
$a^{8}+1$	643.287	643.286	-1.6
a^{10}	934.385	934.383	-3.1
a^{13}	1219.518	1219.509	-7.4
a^{15}	1403.603	1403.61	5.2
a^{17}	1695.709	1695.72	6.5
a^{21}	2147.839	2147.839	-0.2
a^{25}	2624.03	2624.057	10
a^{26}	2721.083	2721.107	8.8
b^{10}	962.38	962.383	3
b^{11}	1049.412	1049.417	4.5
b^{13}	1247.513	1247.517	3.6
b^{14}	1334.545	1334.551	4.3
b^{16}	1560.64	1560.645	2.9
b^{17}	1723.704	1723.708	2.3
b^{19}	2088.802	2088.804	0.8
b^{21}	2175.834	2175.843	4.1
b^{24}	2564.993	2565.005	4.5
b^{6}	486.19	486.191	2.3
b^{7}	573.222	573.223	2.6
b^{9}	799.317	799.322	5.8
c^{13}	1264.539	1264.543	2.9
c^{16}	1577.667	1577.673	4.2
c^{23}	2418.956	2418.962	2.3
c^{9}	816.343	816.346	3.4
$x^{4}+1$	493.193	493.192	-2
$x^{9}+1$	1004.421	1004.427	5.6
y^{11}	1307.496	1307.501	4
y^{13}	1533.591	1533.598	4.5
$y^{13}-1$	1532.583	1532.585	0.8
y^{14}	1620.623	1620.63	4.2
y^{16}	1818.724	1818.73	3.7
$y^{16}-1$	1817.716	1817.72	2.5
y^{17}	1905.756	1905.765	4.7
y^{18}	2068.819	2068.818	-0.6
$y^{20}-1$	2293.907	2293.918	5
y^{21}	2381.946	2381.95	1.6
$y^{23}-1$	2569.001	2569.005	1.6
y^{3}	303.143	303.144	2.8
y^{4}	466.206	466.207	1.5
y^{6}	692.302	692.304	3.2
y^{7}	779.334	779.338	5.1
z^{5}	579.23	579.233	4.7

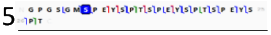
3

Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
$a^{11}+1$	1022.425	1022.42	-5.2
$a^{12}+1$	1119.478	1119.482	3.5
$a^{13}+1$	1404.611	1404.615	3.2
$a^{23}+1$	2374.943	2374.953	4.4
$a^{26}+1$	2722.091	2722.11	7.1
a^{10}	934.385	934.383	-3
a^{17}	1695.709	1695.717	5.1
a^{18}	1782.741	1782.746	3
a^{21}	2147.839	2147.837	-1.2
a^{9}	771.322	771.318	-5.9
b^{10}	962.38	962.383	2.5
b^{11}	1049.412	1049.416	3.8
b^{13}	1247.513	1247.517	3.4
b^{14}	1334.545	1334.55	3.6
b^{16}	1560.64	1560.648	5.1
b^{17}	1723.704	1723.708	2.5
b^{18}	1810.736	1810.743	4.1
b^{20}	2008.836	2008.851	7.4
b^{21}	2175.834	2175.842	3.7
b^{24}	2564.993	2565.003	4
b^{25}	2652.025	2652.035	3.8
b^{6}	486.19	486.191	2.1
c^{16}	1577.667	1577.673	4.3
c^{23}	2418.956	2418.965	3.9
c^{9}	816.343	816.346	2.8
$x^{4}+1$	493.193	493.193	0.1
y^{11}	1307.496	1307.499	2.4
y^{13}	1533.591	1533.597	4
y^{14}	1620.623	1620.633	6.2
$y^{16}-1$	1817.716	1817.719	1.6
y^{17}	1905.756	1905.764	4.2
y^{18}	2068.819	2068.821	1
$y^{20}-1$	2293.907	2293.917	4.4
y^{21}	2381.946	2381.95	1.3
$y^{23}-1$	2569.001	2569.004	1.2
y^{3}	303.143	303.144	2.3
y^{6}	692.302	692.304	2.9
y^{7}	859.3	859.301	0.6
y^{8}	960.348	960.345	-2.4
y^{9}	1057.401	1057.404	2.9

4

Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
$a^{10}+1$	935.393	935.384	-9.6
$a^{12}+1$	1119.478	1119.483	4.6
$a^{16}+1$	1613.619	1613.628	5.5
$a^{19}+1$	1960.768	1960.780	6.3
$a^{22}+1$	2245.900	2245.908	3.8
$a^{23}+1$	2374.943	2374.950	3.2
a^{13}	1219.518	1219.507	-8.6
a^{21}	2147.839	2147.840	0.4
a^{25}	2624.030	2624.044	5.1
a^{26}	2721.083	2721.095	4.6
a^{9}	771.322	771.321	-1.4
b^{10}	962.380	962.383	2.3
b^{11}	1049.412	1049.416	3.8
b^{13}	1247.513	1247.515	1.9
b^{16}	1640.607	1640.612	3.2
b^{17}	1803.670	1803.678	4.3
b^{18}	1890.702	1890.710	4.4
b^{20}	2088.802	2088.805	1.2
b^{21}	2175.834	2175.841	3.1
b^{23}	2401.930	2401.948	7.6
b^{24}	2564.993	2565.003	4
b^{6}	486.190	486.191	2
b^{9}	799.317	799.320	3.8
c^{16}	1657.633	1657.637	2.5
c^{9}	816.343	816.346	3
$x^{11}+1$	1254.517	1254.524	5.9
$x^{9}+1$	1004.421	1004.429	7.3
y^{10}	1064.466	1064.470	3.8
y^{13}	1453.625	1453.634	6
$y^{13}-1$	1452.617	1452.609	-5.4
$y^{13}-1$	1452.617	1452.621	2.6
y^{14}	1620.623	1620.636	7.6
y^{17}	1905.756	1905.764	4.2
y^{18}	2068.819	2068.828	4.3
y^{20}	2294.914	2294.924	4.3
y^{21}	2381.946	2381.951	1.8
$y^{23}-1$	2569.001	2568.998	-1.1
y^{3}	303.143	303.144	2.5
y^{4}	466.206	466.207	1.3
y^{6}	692.302	692.304	3.2
y^{7}	779.334	779.337	4
y^{9}	977.434	977.437	2.7

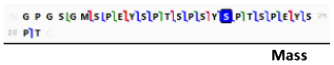
5



Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
$a^{12}+1$	1199.444	1199.452	5.9
$a^{14}+1$	1387.524	1387.518	-4.7
$a^{18}+1$	1863.715	1863.715	0.2
$a^{21}+1$	2148.847	2148.859	5.5
$a^{26}+1$	2722.091	2722.112	7.7
$a^{6}+1$	459.203	459.205	4.5
a^{10}	1014.352	1014.356	4.4
a^{11}	1101.384	1101.385	0.7
a^{17}	1775.675	1775.681	3.4
b^{10}	1042.347	1042.350	3.5
b^{11}	1129.379	1129.382	2.5
b^{13}	1327.479	1327.480	0.9
b^{14}	1414.511	1414.517	3.8
b^{16}	1640.607	1640.612	3.4
b^{17}	1803.670	1803.678	4.6
b^{18}	1890.702	1890.712	5.4
b^{20}	2088.802	2088.819	8
b^{21}	2175.834	2175.841	3.2
b^{23}	2401.930	2401.947	7.2
b^{24}	2564.993	2565.003	3.9
b^{25}	2652.025	2652.036	4.1
b^{6}	486.190	486.191	1.9
b^{9}	879.283	879.284	1.1
c^{10}	1059.373	1059.367	-6
c^{13}	1344.505	1344.515	7.2
c^{16}	1657.633	1657.637	2.5
c^{9}	896.310	896.310	0.7
$x^{11}+1$	1254.517	1254.522	4.1
x^{8}	906.361	906.359	-1.4
y^{10}	1064.466	1064.470	4
y^{11}	1227.530	1227.536	5.6
y^{12}	1356.572	1356.562	-7.3
y^{13}	1453.625	1453.631	4.4
y^{14}	1540.657	1540.663	3.7
y^{16}	1738.757	1738.763	3
$y^{16}-1$	1737.750	1737.752	1.2
y^{17}	1825.789	1825.795	2.8
$y^{20}-1$	2213.940	2213.935	-2.2
y^{21}	2381.946	2381.947	0.2
$y^{23}-1$	2569.001	2569.007	2.4
y^{3}	303.143	303.144	2.7
y^{6}	692.302	692.304	3.1
y^{7}	779.334	779.337	4.5
y^{9}	977.434	977.437	2.9

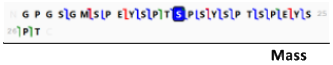
Supplementary Figure 9: Lists of fragment ions for mono-phosphorylated peptides (m/z of 957.05) analyzed in Figure 6B by UVPD-MS. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.

2



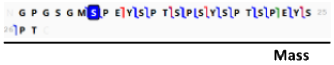
Identified Ions	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{11}+1	1022.425	1022.426	0.6
a^{15}+1	1404.611	1404.613	2.0
a^{19}+1	1918.757	1918.762	2.6
a^{21}+1	2106.837	2106.840	1.5
a^{22}+1	2203.889	2203.897	3.2
a^{26}+1	2680.080	2680.093	4.9
a^{6}+1	459.203	459.204	3.4
a^{8}+1	643.287	643.290	4.4
a^{9}+1	772.330	772.332	2.9
a^{12}	1118.470	1118.461	-8.4
a^{15}	1403.603	1403.608	3.5
a^{17}	1653.698	1653.703	3.0
a^{9}	771.322	771.322	-0.2
b^{10}	962.380	962.382	2.1
b^{11}	1049.412	1049.415	2.5
b^{13}	1247.513	1247.516	2.8
b^{14}	1334.545	1334.551	4.4
b^{16}	1518.630	1518.637	5.0
b^{17}	1681.693	1681.699	3.6
b^{20}	2046.792	2046.810	8.8
b^{21}	2133.824	2133.831	3.2
b^{23}	2359.919	2359.930	4.7
b^{24}	2522.983	2522.994	4.4
b^{26}	2707.067	2707.086	7.0
b^{6}	486.190	486.191	3.4
c^{13}	1264.539	1264.531	-6.6
c^{16}	1535.656	1535.662	3.9
c^{23}	2376.945	2376.948	1.0
c^{6}	503.216	503.217	2.8
c^{9}	816.343	816.346	3.8
x^{19}+1	2182.838	2182.840	0.8
x^{23}+1	2554.985	2554.981	-1.5
x^{4}+1	493.193	493.195	2.7
y^{10}	1144.433	1144.437	4.1
y^{12}	1394.528	1394.539	7.8
y^{13}	1491.581	1491.585	2.8
y^{13}-1	1490.573	1490.568	-3.2
y^{14}	1578.613	1578.618	3.1
y^{16}	1776.713	1776.719	3.5
y^{16}-1	1775.705	1775.710	2.5
y^{17}	1863.745	1863.753	4.3
y^{18}	2026.808	2026.809	0.2
y^{20}-1	2251.896	2251.904	3.6
y^{21}	2339.936	2339.947	5.0
y^{3}	303.143	303.144	2.8
y^{4}	466.206	466.208	3.3
y^{5}	595.249	595.250	2.2
y^{6}	692.302	692.304	3.4
y^{7}	779.334	779.337	4.0
y^{9}	977.434	977.437	3.0
z^{18}	2010.790	2010.807	8.6

3



Identified Ionsx	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{11}+1	1022.425	1022.426	0.6
a^{12}+1	1119.478	1119.479	0.5
a^{18}+1	1821.704	1821.706	0.8
a^{21}+1	2106.837	2106.847	4.9
a^{22}+1	2203.889	2203.895	2.4
a^{26}+1	2680.080	2680.087	2.6
a^{10}	934.385	934.391	5.8
a^{13}	1219.518	1219.520	2.0
a^{14}	1386.516	1386.520	2.8
a^{25}	2582.020	2582.030	3.9
b^{10}	962.380	962.383	3.2
b^{11}	1049.412	1049.416	3.7
b^{13}	1247.513	1247.517	3.5
b^{16}	1598.596	1598.586	-6.2
b^{17}	1761.659	1761.668	5.2
b^{18}	1848.691	1848.700	4.6
b^{20}	2046.792	2046.805	6.3
b^{21}	2133.824	2133.830	3.1
b^{24}	2522.983	2522.995	4.8
b^{4}	298.128	298.129	5.3
b^{6}	486.190	486.191	3.3
c^{16}	1615.622	1615.630	4.5
c^{23}	2376.945	2376.951	2.3
c^{6}	503.216	503.219	5.4
c^{9}	816.343	816.347	4.3
x^{16}+1	1803.700	1803.695	-3.0
x^{23}+1	2554.985	2554.981	-1.6
y^{10}	1064.466	1064.471	4.7
y^{11}	1227.530	1227.533	2.9
y^{12}	1314.562	1314.560	-0.8
y^{13}	1411.614	1411.621	4.4
y^{14}	1578.613	1578.621	5.0
y^{16}	1776.713	1776.720	3.8
y^{17}	1863.745	1863.754	4.8
y^{20}-1	2251.896	2251.903	3.3
y^{21}	2339.936	2339.949	5.4
y^{23}-1	2526.990	2527.006	6.3
y^{3}	303.143	303.144	3.0
y^{4}	466.206	466.208	4.4
y^{5}	595.249	595.251	3.5
y^{6}	692.302	692.304	3.5
y^{7}	779.334	779.337	4.6
y^{9}	977.434	977.438	3.4
z^{18}	2010.790	2010.808	9.2

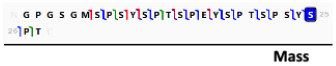
4



Identified Ionsx	Theoretical Mass	Observed Mass	Mass Difference (ppm)
a^{18}+1	1821.704	1821.692	-6.7
a^{22}+1	2203.889	2203.900	4.7
a^{23}+1	2332.932	2332.950	7.5
a^{17}	1733.664	1733.672	4.2
a^{21}	2105.829	2105.831	1.1
a^{25}	2582.020	2582.030	3.9
b^{10}	1042.347	1042.349	2.1
b^{11}	1129.379	1129.381	2.1
b^{13}	1327.479	1327.488	6.6
b^{14}	1414.511	1414.514	2.0
b^{17}	1761.659	1761.663	2.2
b^{18}	1848.691	1848.701	5.0
b^{20}	2046.792	2046.802	5.2
b^{21}	2133.824	2133.829	2.3
b^{23}	2359.919	2359.940	8.7
b^{24}	2522.983	2522.992	3.6
b^{25}	2610.015	2610.026	4.3
b^{6}	486.190	486.191	3.4
c^{13}	1344.505	1344.505	-0.3
c^{16}	1615.622	1615.629	4.0
c^{24}	2540.009	2540.011	0.8
c^{9}	896.310	896.312	3.1
y^{10}	1064.466	1064.470	3.6
y^{11}	1227.530	1227.533	2.9
y^{12}	1314.562	1314.549	-9.4
y^{13}	1411.614	1411.621	4.7
y^{13}-1	1410.607	1410.615	5.9
y^{14}	1498.646	1498.654	5.0
y^{16}	1696.747	1696.743	-2.0
y^{17}	1783.779	1783.785	3.7
y^{20}-1	2171.930	2171.936	3.0
y^{21}	2339.936	2339.949	5.7
y^{3}	303.143	303.144	2.9
y^{3}	303.143	303.144	2.9
y^{4}	466.206	466.207	2.2
y^{4}	466.206	466.207	2.2
y^{6}	692.302	692.304	3.7
y^{7}	779.334	779.337	3.7
y^{9}	977.434	977.437	3.4

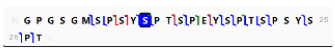
Supplementary Figure 10: Lists of fragment ions for mono-phosphorylated peptides (m/z of 943.40) analyzed in Figure 6C by UVPD-MS. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.

2



Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{12}+1	1077.468	1077.475	6.8
a^{15}+1	1362.600	1362.606	4.6
a^{26}+1	2638.070	2638.077	2.9
a^{8}+1	643.287	643.290	4.0
a^{11}	979.407	979.409	1.7
a^{13}	1177.507	1177.500	-6.0
a^{17}	1653.698	1653.705	4.3
a^{18}	1740.730	1740.739	5.4
a^{25}	2540.009	2540.023	5.4
b^{10}	920.370	920.374	4.1
b^{11}	1007.402	1007.406	4.3
b^{13}	1205.502	1205.507	4.0
b^{14}	1292.534	1292.540	4.3
b^{15}	1389.587	1389.591	3.0
b^{16}	1518.630	1518.637	4.5
b^{17}	1681.693	1681.699	3.8
b^{18}	1768.725	1768.733	4.3
b^{20}	1966.825	1966.835	4.7
b^{21}	2053.857	2053.866	3.9
b^{23}	2237.942	2237.958	6.9
b^{24}	2401.006	2401.016	4.1
b^{25}	2568.004	2568.017	5.2
b^{6}	486.190	486.191	3.6
b^{7}	573.222	573.224	3.6
b^{9}	757.306	757.311	5.6
c^{10}	937.396	937.399	3.0
c^{13}	1222.529	1222.531	1.7
c^{16}	1535.656	1535.663	4.5
c^{6}	503.216	503.218	3.1
c^{9}	774.333	774.336	4.3
x^{19}+1	2140.828	2140.834	3.0
y^{10}	1102.422	1102.427	4.4
y^{11}	1265.485	1265.494	7.2
y^{13}	1491.581	1491.586	3.8
Y^{13}-1	1490.573	1490.565	-5.3
y^{14}	1578.613	1578.617	2.9
y^{16}	1776.713	1776.719	3.5
y^{17}	1863.745	1863.749	2.3
y^{20}	2210.893	2210.898	2.1
y^{4}	546.173	546.175	4.3
y^{6}	730.257	730.260	3.1
y^{7}	817.290	817.292	3.5
y^{9}	1015.390	1015.394	3.7

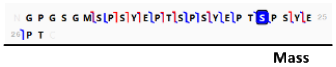
3



Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{11}+1	1060.381	1060.382	0.5
a^{14}+1	1345.514	1345.506	-5.8
a^{15}+1	1442.566	1442.574	5.6
a^{16}+1	1571.609	1571.611	1.2
a^{18}+1	1821.704	1821.707	1.2
a^{19}+1	1918.757	1918.766	4.4
a^{21}+1	2106.837	2106.839	1.0
a^{26}+1	2638.070	2638.079	3.6
a^{6}+1	459.203	459.206	7.7
a^{8}+1	643.287	643.289	2.5
a^{13}	1257.474	1257.482	6.6
a^{17}	1733.664	1733.678	7.7
a^{25}	2540.009	2540.017	3.2
b^{10}	920.370	920.373	3.3
b^{14}	1372.501	1372.506	3.8
b^{16}	1598.596	1598.598	1.0
b^{17}	1761.659	1761.666	3.6
b^{18}	1848.691	1848.697	3.3
b^{21}	2133.824	2133.830	3.0
b^{24}	2480.972	2480.979	2.7
b^{25}	2568.004	2568.015	4.1
b^{26}	2665.057	2665.076	7.0
b^{6}	486.190	486.191	3.0
b^{9}	757.306	757.308	2.6
c^{13}	1302.495	1302.500	4.1
c^{16}	1615.622	1615.628	3.7
c^{8}	687.301	687.306	8.0
c^{9}	774.333	774.335	3.3
x^{9}+1	962.411	962.417	6.3
x^{8}	864.350	864.351	0.6
y^{10}	1022.456	1022.460	4.0
y^{11}	1185.519	1185.525	5.3
y^{13}	1411.614	1411.619	3.4
y^{14}	1498.646	1498.650	2.5
Y^{16}-1	1695.739	1695.741	1.2
y^{17}	1863.745	1863.749	2.0
y^{20}	2210.893	2210.898	2.1
y^{21}	2297.925	2297.935	4.3
y^{3}	303.143	303.144	2.6
y^{6}	650.291	650.293	3.4
y^{7}	737.323	737.326	3.4
y^{8}	838.371	838.373	2.0
y^{9}	935.424	935.427	3.3
y^{9}-1	934.416	934.420	4.6
z^{19}	2097.822	2097.816	-3.0

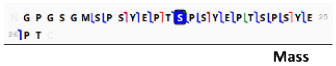
Supplementary Figure 11: Lists of fragment ions for mono-phosphorylated peptides (m/z of 929.39) analyzed in Figure 6D by UVPD-MS. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.

2



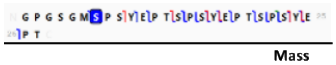
Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{10}+1	893.383	893.384	1.1
a^{11}+1	1022.425	1022.427	1.3
a^{13}	1219.518	1219.507	-9.3
b^{10}	920.370	920.372	2.4
b^{11}	1049.412	1049.415	2.9
b^{12}	1146.465	1146.463	-1.7
b^{13}	1247.513	1247.517	3.1
b^{14}	1334.545	1334.550	4.1
b^{15}	1431.598	1431.603	3.7
b^{16}	1518.630	1518.637	4.8
b^{17}	1681.693	1681.700	3.9
b^{18}	1810.736	1810.742	3.7
b^{20}	2008.836	2008.846	4.8
b^{21}	2175.834	2175.847	5.8
b^{24}	2522.983	2522.992	3.6
b^{25}	2652.025	2652.035	3.6
b^{6}	486.190	486.191	2.2
c^{10}	937.396	937.400	3.6
c^{12}	1163.491	1163.491	-0.1
c^{13}	1264.539	1264.543	3.4
c^{16}	1535.656	1535.662	4.1
c^{23}	2376.945	2376.938	-3.2
c^{6}	503.216	503.218	4.1
c^{8}	687.301	687.302	1.9
c^{9}	774.333	774.335	3.0
y^{10}	1186.443	1186.448	3.9
y^{11}	1349.506	1349.512	4.5
y^{13}	1533.591	1533.599	5.0
y^{13}	1533.591	1533.595	2.8
y^{13}+1	1532.583	1532.588	3.1
y^{14}	1620.623	1620.631	4.5
y^{16}	1818.724	1818.729	3.0
y^{20}	2294.914	2294.922	3.4
y^{21}	2381.946	2381.953	2.6
y^{3}	345.154	345.154	2.1
y^{4}	508.217	508.218	2.7
y^{5}	692.302	692.304	3.2
y^{7}	859.300	859.301	1.6
y^{9}	1057.401	1057.404	2.8
z^{3}	329.135	329.138	9.9

3



Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{11}+1	1022.425	1022.419	-6.4
a^{14}+1	1387.524	1387.516	-5.8
a^{10}	892.375	892.379	4.5
a^{13}	1219.518	1219.508	-8.0
a^{25}	2624.030	2624.044	5.2
b^{10}	920.370	920.372	2.2
b^{11}	1049.412	1049.416	3.6
b^{13}	1247.513	1247.516	2.8
b^{16}	1598.596	1598.604	4.7
b^{17}	1761.659	1761.666	3.7
b^{18}	1890.702	1890.708	3.2
b^{20}	2088.802	2088.803	0.2
b^{24}	2522.983	2522.990	2.9
b^{25}	2652.025	2652.035	3.8
b^{9}	757.307	757.309	3.6
c^{12}	1163.491	1163.490	-1.6
c^{16}	1615.622	1615.629	4.2
c^{23}	2376.945	2376.946	0.1
c^{9}	774.333	774.335	3.0
x^{8}	906.361	906.367	6.8
x^{9}	1003.413	1003.405	-8.2
y^{10}	1106.477	1106.483	5.3
y^{10}	1106.477	1106.480	2.8
y^{12}	1356.572	1356.567	-3.4
y^{13}	1453.625	1453.635	7.1
y^{13}-1	1452.617	1452.613	-2.5
y^{14}	1620.623	1620.630	4.1
y^{16}	1818.724	1818.730	3.2
y^{16}-1	1817.716	1817.718	1.4
y^{20}	2294.914	2294.925	4.6
y^{21}	2381.946	2381.952	2.2
y^{3}	345.154	345.154	2.2
y^{5}	595.249	595.251	3.8
y^{6}	692.302	692.304	2.9
y^{7}	779.334	779.337	3.8
y^{9}	977.434	977.437	2.7

4



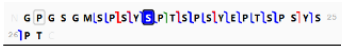
Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{10}+1	973.349	973.348	-1.6
a^{11}+1	1102.392	1102.388	-3.7
a^{22}	2244.892	2244.913	9.1
b^{10}	1000.336	1000.339	2.4
b^{11}	1129.379	1129.381	1.9
b^{14}	1414.511	1414.513	1.6
b^{16}	1598.596	1598.610	8.7
b^{17}	1761.659	1761.665	3.4
b^{18}	1890.702	1890.706	2.0
b^{20}	2088.802	2088.803	0.1
b^{23}	2359.919	2359.937	7.5
b^{24}	2522.983	2522.988	2.3
b^{25}	2652.025	2652.033	3.2
b^{6}	486.190	486.191	2.6
b^{7}	653.188	653.189	2.1
c^{13}	1344.505	1344.513	5.5
c^{16}	1615.622	1615.628	3.4
c^{23}	2376.945	2376.950	2.1
c^{9}	854.299	854.301	1.8
y^{10}	1106.477	1106.479	2.2
y^{11}	1269.540	1269.543	2.4
y^{12}	1356.572	1356.574	1.6
y^{13}	1453.625	1453.630	3.4
y^{14}	1540.657	1540.663	3.7
y^{16}	1738.757	1738.762	2.4
y^{21}	2381.946	2381.950	1.4
y^{3}	345.154	345.154	1.9
y^{5}	595.249	595.250	1.9
y^{6}	692.302	692.304	2.5
y^{7}	779.334	779.336	3.3
y^{9}	977.434	977.437	2.5
z^{10}	1090.458	1090.465	6.3
z^{3}	329.135	329.138	9.9

Supplementary Figure 12: Lists of fragment ions for mono-phosphorylated peptides (m/z of 957.05) analyzed in Figure 6E by UVPD-MS. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.

2

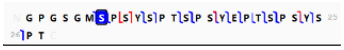
Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{11}+1	980.415	980.411	-3.9
a^{14}+1	1265.547	1265.539	-6.3
a^{8}+1	643.287	643.288	1.1
a^{10}	892.375	892.366	-10.2
a^{12}	1076.460	1076.455	-4.4
a^{13}	1177.507	1177.497	-9.0
a^{18}	1740.730	1740.718	-6.8
a^{21}	2025.863	2025.872	4.8
b^{11}	1007.402	1007.402	0.4
b^{13}	1205.502	1205.505	2.6
b^{14}	1292.534	1292.538	2.6
b^{16}	1476.619	1476.627	5.5
b^{17}	1639.682	1639.687	2.6
b^{18}	1768.725	1768.726	0.8
b^{20}	1966.825	1966.832	3.3
b^{24}	2401.006	2401.011	2.1
b^{25}	2568.004	2568.013	3.4
b^{9}	757.307	757.310	4.5
c^{10}	937.396	937.400	4.0
c^{16}	1493.645	1493.651	3.7
c^{23}	2254.969	2254.976	3.2
c^{8}	687.301	687.302	2.3
c^{9}	774.333	774.335	2.2
y^{10}	1144.433	1144.435	2.6
y^{11}	1307.496	1307.500	3.3
y^{13}	1491.581	1491.583	1.6
y^{13}	1491.581	1491.586	3.8
y^{14}	1578.613	1578.615	1.5
y^{16}	1776.713	1776.719	3.0
y^{17}	1863.745	1863.749	2.1
y^{20}	2210.893	2210.901	3.7
y^{21}	2297.925	2297.933	3.5
y^{3}	383.109	383.110	1.2
y^{6}	730.258	730.259	1.8
y^{7}	817.290	817.292	2.6
y^{9}	1015.390	1015.392	1.9

3



Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{12}+1	1157.434	1157.441	6.1
a^{14}+1	1345.514	1345.508	-4.1
b^{10}	920.370	920.372	2.3
b^{13}	1285.469	1285.471	2.2
b^{14}	1372.501	1372.505	3.5
b^{16}	1556.585	1556.588	1.4
b^{17}	1719.649	1719.653	2.5
b^{18}	1848.691	1848.698	3.5
b^{20}	2046.792	2046.797	2.7
b^{21}	2133.824	2133.830	2.7
b^{24}	2480.972	2480.977	1.9
b^{25}	2568.004	2568.013	3.7
b^{9}	757.307	757.309	2.9
c^{13}	1302.495	1302.501	4.7
c^{16}	1573.612	1573.616	2.9
c^{23}	2334.935	2334.940	2.1
c^{8}	687.301	687.303	3.2
c^{9}	774.333	774.335	2.2
x^{8}+1	865.358	865.359	0.7
y^{10}	1064.466	1064.469	2.7
y^{11}	1227.530	1227.531	1.5
y^{12}	1314.562	1314.560	-1.6
y^{13}	1411.614	1411.619	3.2
y^{14}	1498.646	1498.653	4.2
y^{16}-1	1695.739	1695.741	1.3
y^{17}	1863.745	1863.749	2.1
y^{18}	2026.808	2026.820	5.5
y^{20}	2210.893	2210.900	3.2
y^{21}	2297.925	2297.933	3.4
y^{6}	650.291	650.293	2.7
y^{7}	737.323	737.325	2.7
y^{8}	838.371	838.372	0.7
y^{9}	935.424	935.425	1.4
z^{19}	2097.822	2097.811	-5.0

4



Identified Ions	Theoretical mass	Observed mass	Mass Difference (ppm)
a^{10}+1	973.349	973.357	8.1
b^{10}	1000.336	1000.338	2.0
b^{11}	1087.368	1087.370	1.4
b^{13}	1285.469	1285.472	2.6
b^{14}	1372.501	1372.505	2.9
b^{16}	1556.585	1556.595	6.2
b^{17}	1719.649	1719.653	2.5
b^{18}	1848.691	1848.698	3.7
b^{20}	2046.792	2046.798	3.2
b^{21}	2133.824	2133.829	2.5
b^{24}	2480.972	2480.978	2.6
b^{25}	2568.004	2568.013	3.7
b^{6}	486.190	486.191	1.7
c^{16}	1573.612	1573.617	3.0
c^{23}	2334.935	2334.941	2.6
c^{9}	854.299	854.302	3.1
y^{10}	1064.466	1064.469	2.7
y^{11}	1227.530	1227.531	1.3
y^{13}	1411.614	1411.617	1.9
y^{14}	1498.646	1498.651	3.3
y^{17}	1783.779	1783.784	2.9
y^{20}	2130.927	2130.943	7.3
y^{21}	2297.925	2297.935	4.3
y^{4}	466.206	466.207	1.8
y^{6}	650.291	650.293	3.0
y^{7}	737.323	737.326	3.3
y^{8}	838.371	838.369	-2.4
y^{9}	935.424	935.426	3.0
z^{19}	2017.855	2017.843	-6.0

Supplementary Figure 13: Lists of fragment ions for mono-phosphorylated peptides (m/z of 929.05) analyzed in Figure 6F by UVPD-MS. In each case, the 3+ charge state was selected, and UVPD was performed using 2 pulses (1.5 mJ per pulse). The identified site of phosphorylation is shaded in blue in the sequence map above each table.