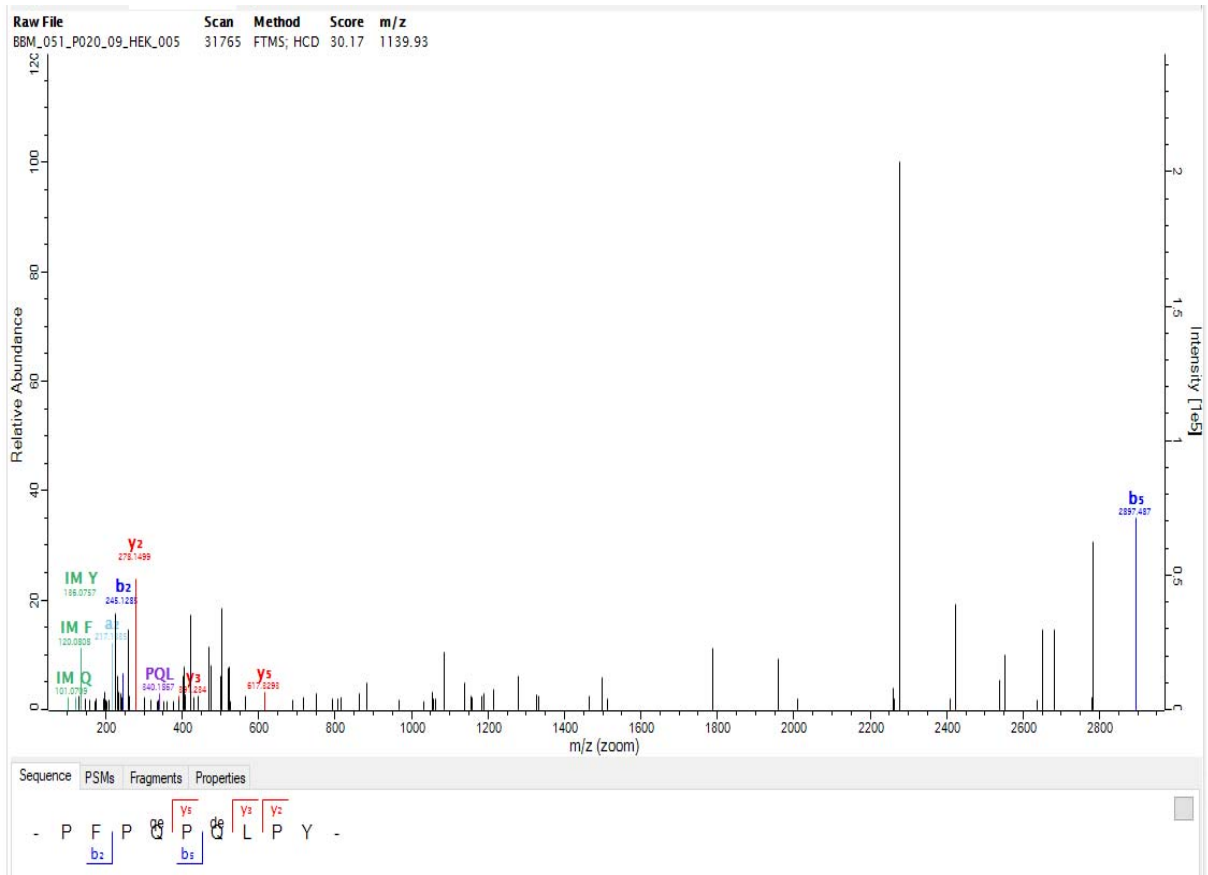
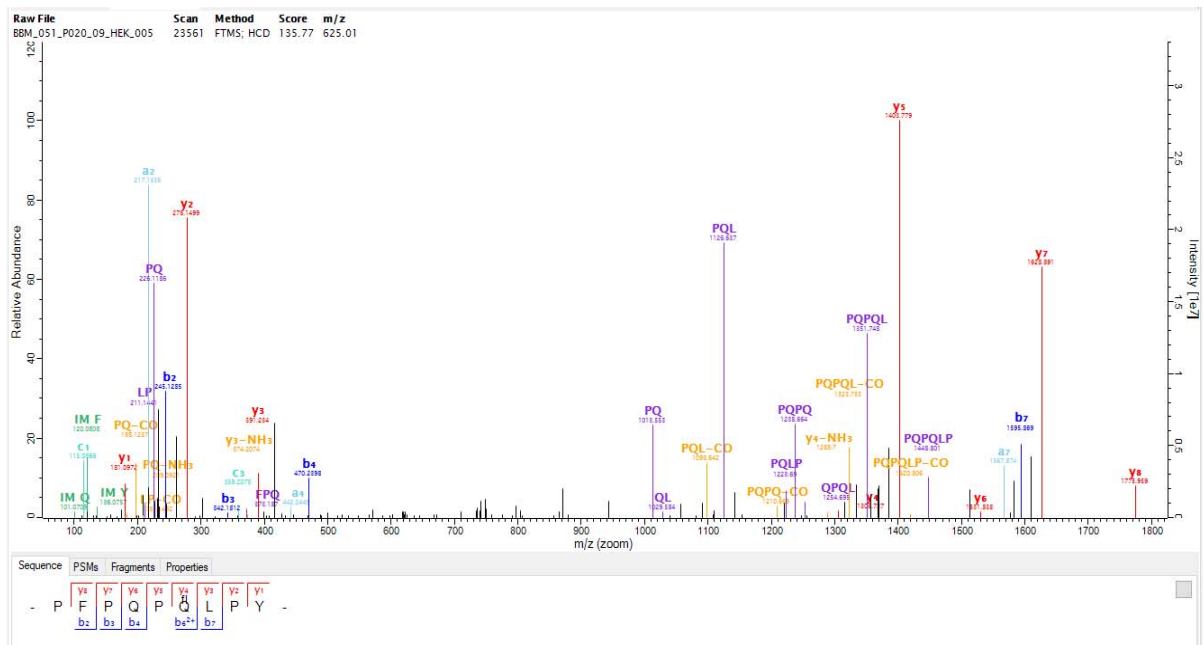


A



B



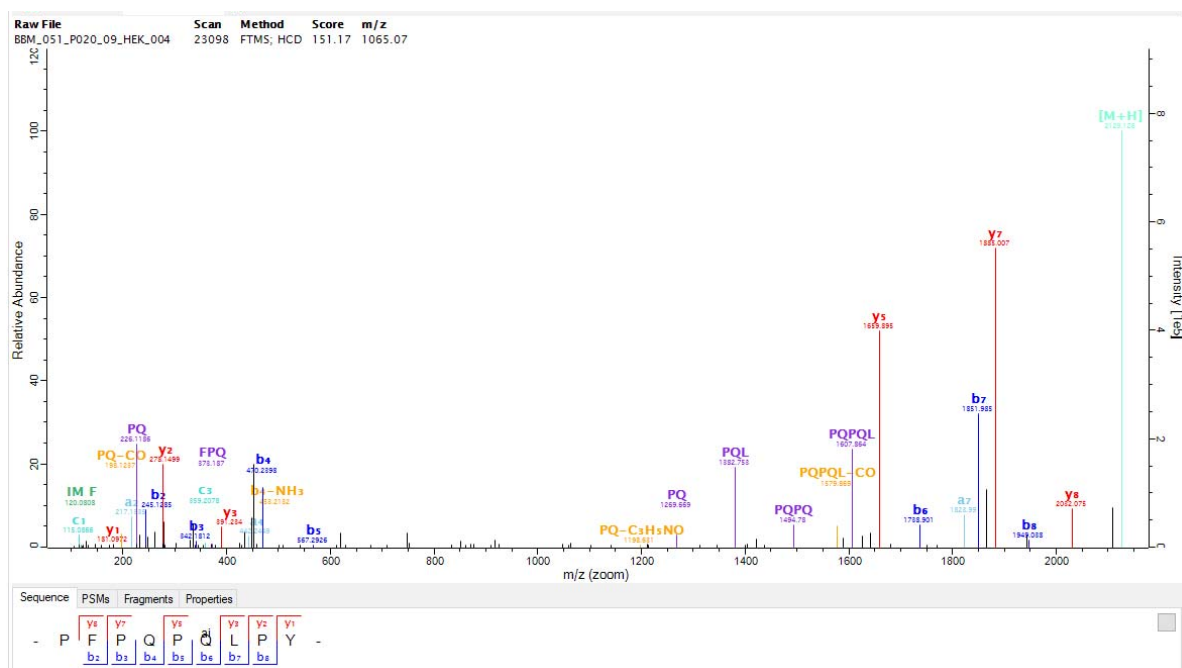
Raw File  
BNL\_051\_PO20\_09\_HEK\_006  
15863 FTMS, HCD 79.47 659.68

Sequence PSMs Fragments Properties

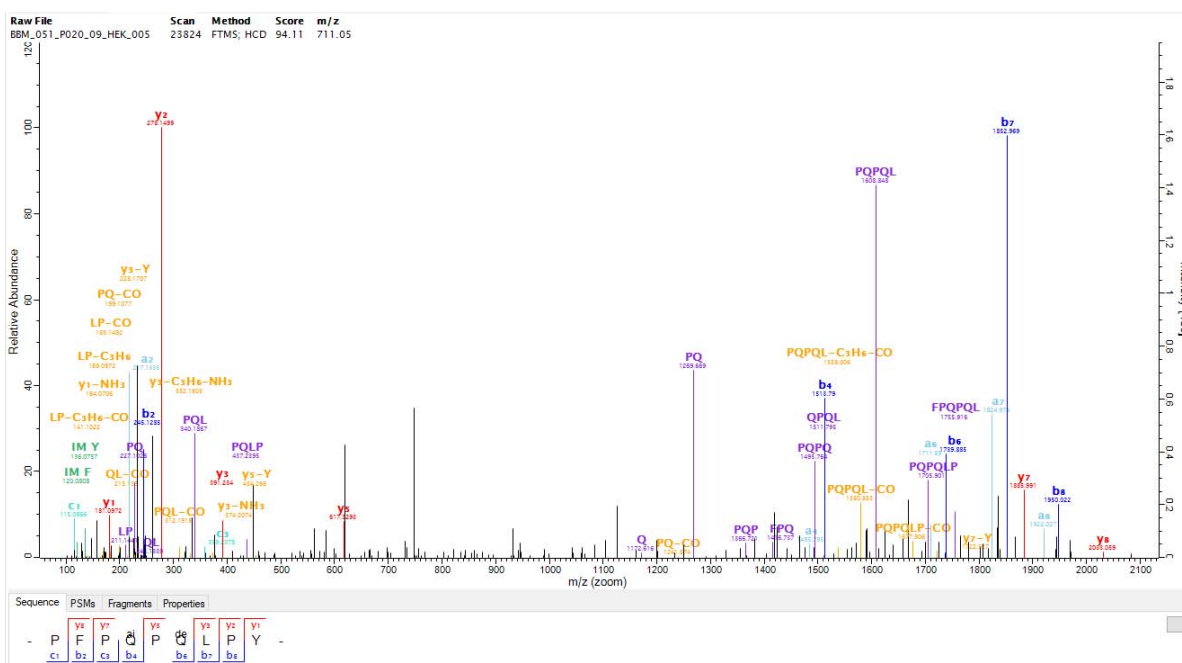
- P F P Q P L P Y -

b2 b2 b4

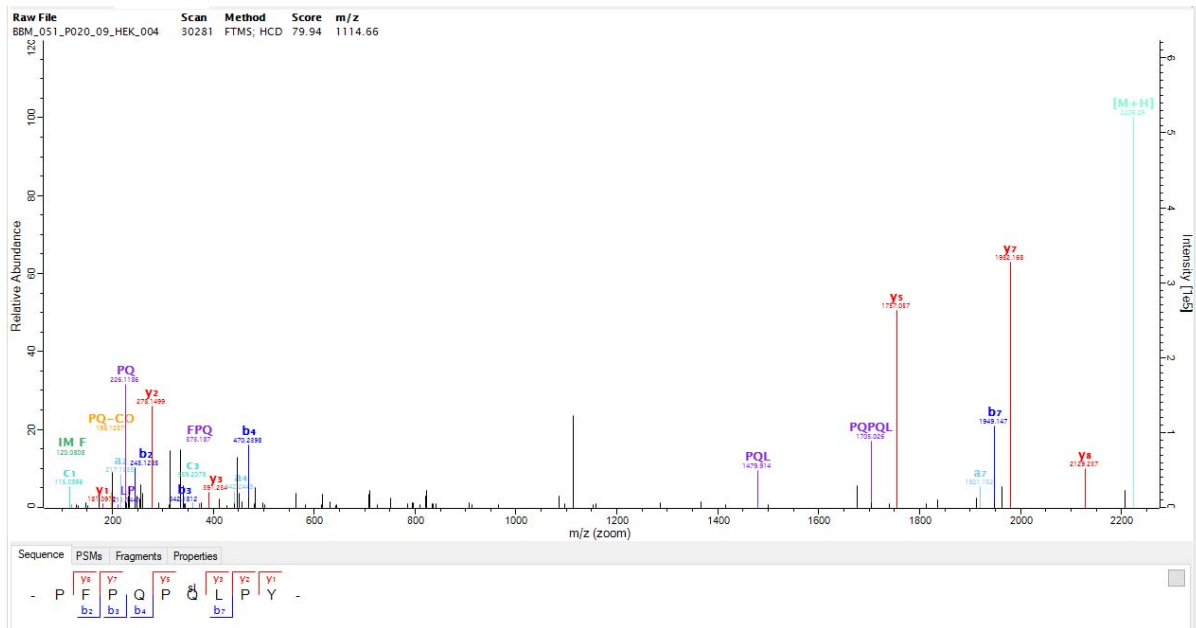
E



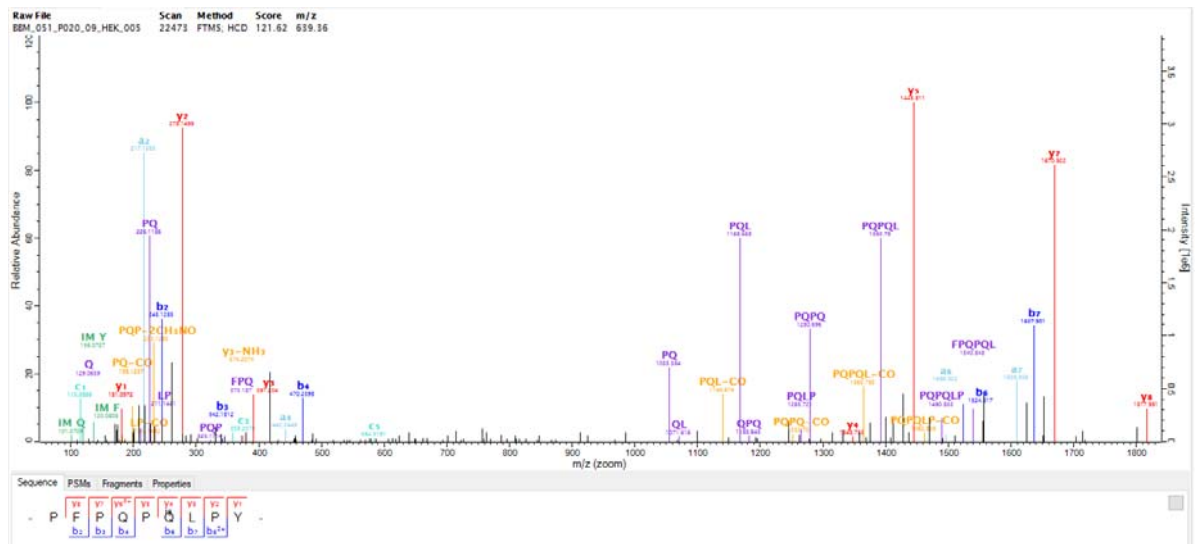
F



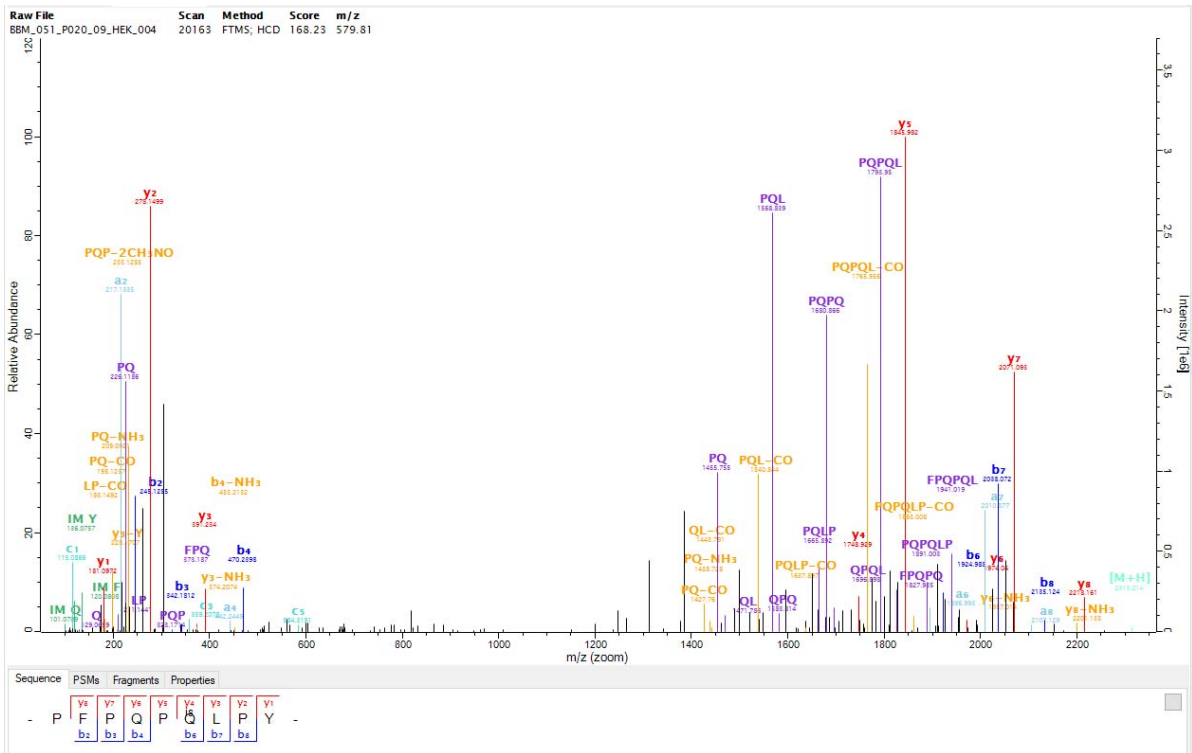
G



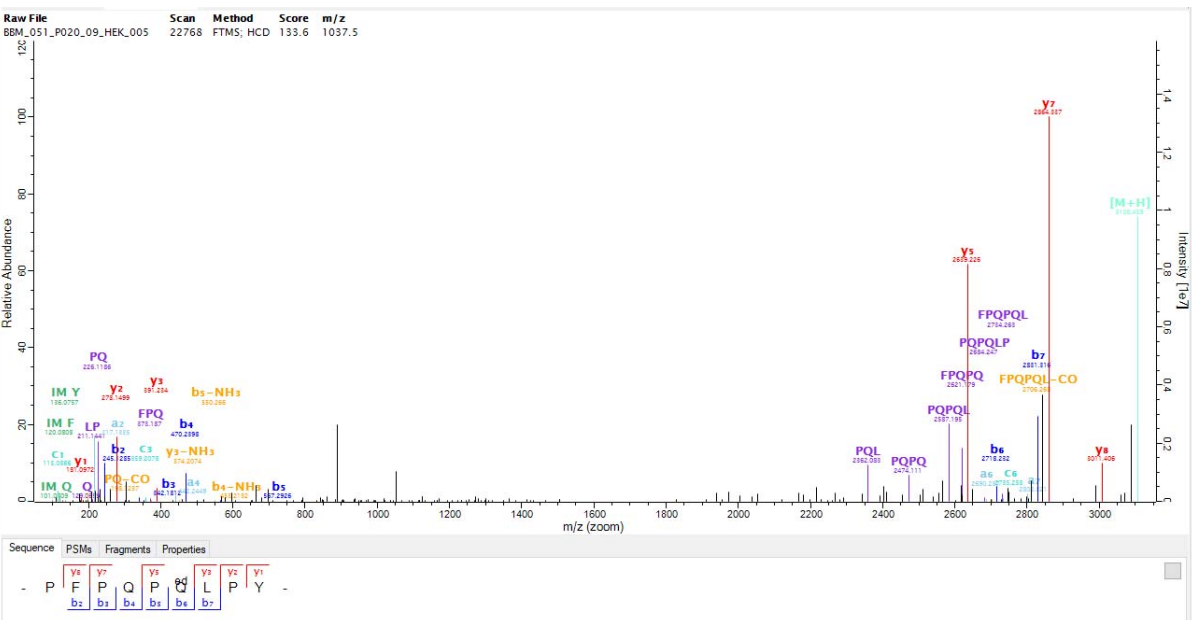
H



I



J



Raw File Scan Method Score m/z

BBM\_051\_P020\_09\_HEK\_006 22860 FTMS; HCD 119.29 1037.83

Relative Abundance

m/z (zoom)

Intensity [a.u.]

Sequence PSMs Fragments Properties

- P F P P P Y -

c1 b2 b2 b4 b6 b7 b8

Raw File Scan Method Score m/z  
BBM\_051\_P020\_09\_HEK\_006 21736 FTMS, HCD 85.21 878.42

Relative Abundance

m/z (zoom)

Sequence PSMs Fragments Properties

- P 

Y6	Y7
F	P
Q	Q
b2	b4

Y6	Y7
L	P
b7	Y

 -

Raw File Scan Method Score m/z  
 BM\_051\_PO20\_09\_HEK\_005 24109 FTMS, HCD 133.99 1218.11

Relative Abundance

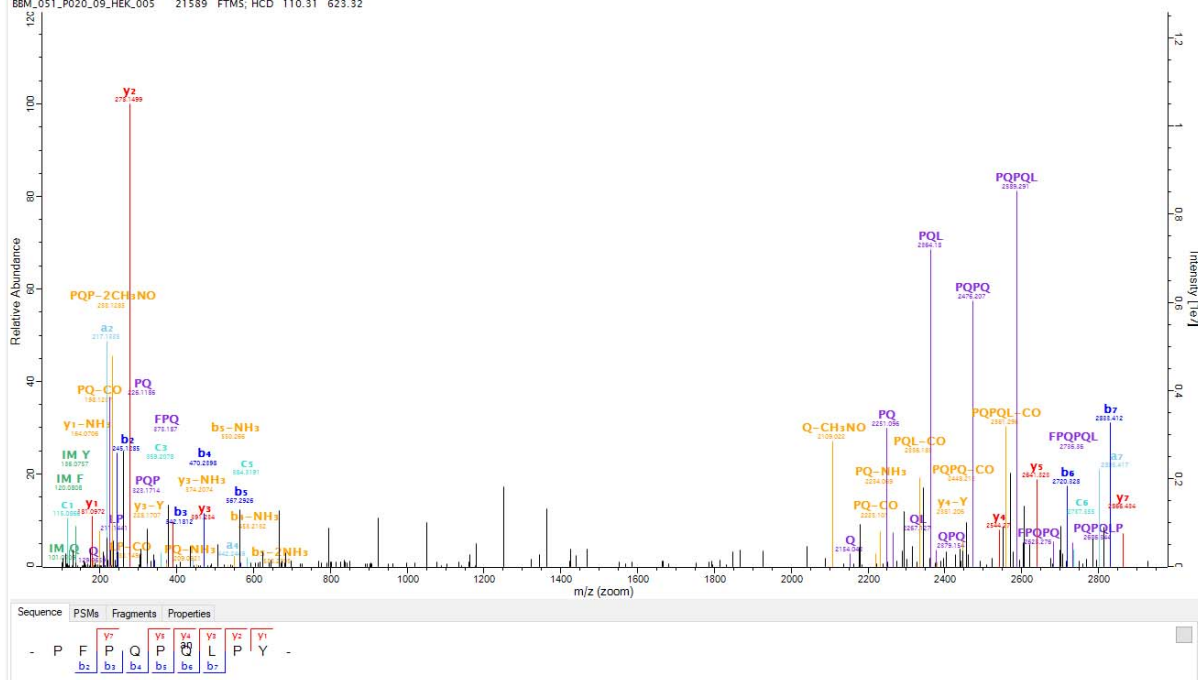
m/z (zoom)

Sequence PSMs Fragments Properties

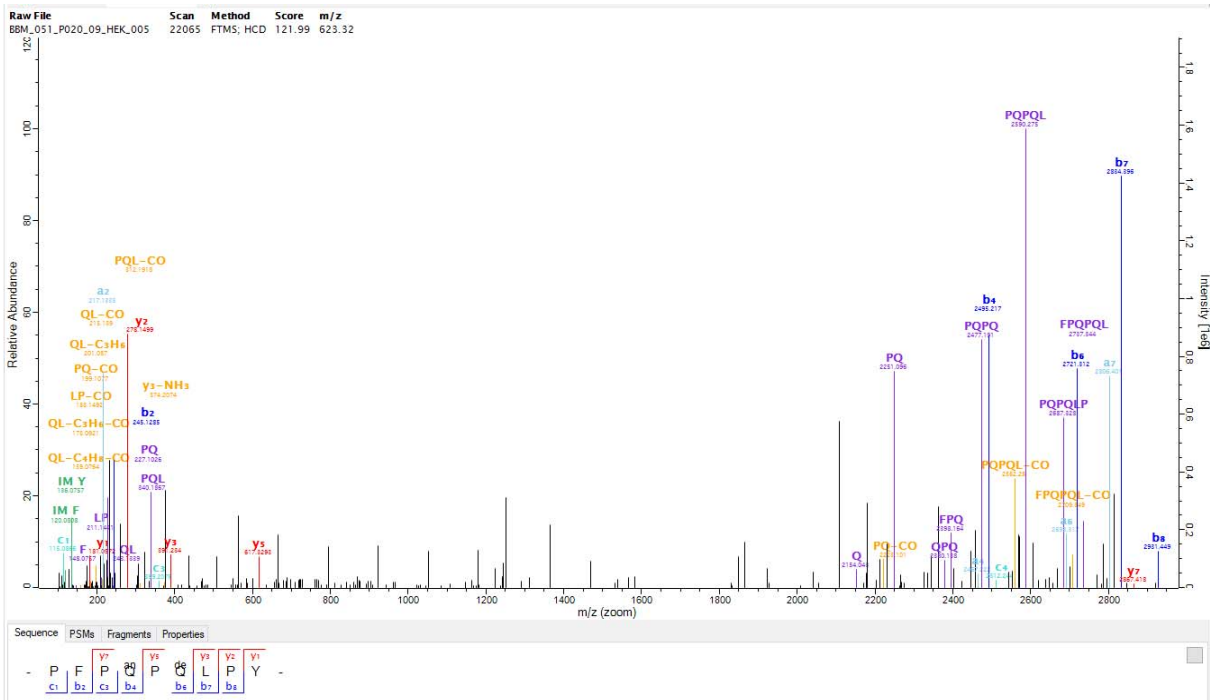
- P F P Q P L P Y -

Y7 b2 Y5 Y1 Y1 Y2  
 b2 b2 b4 L b7

Raw File	Scan	Method	Score	m/z
BBM_051_P020_09_HEK_005	21589	FTMS; HCD	110.31	623.32



P





Raw File Scan Method Score m/z  
BBM\_051\_P020\_09\_HEK\_005 21782 FTMS; HCD 53.17 552.55

Relative Abundance

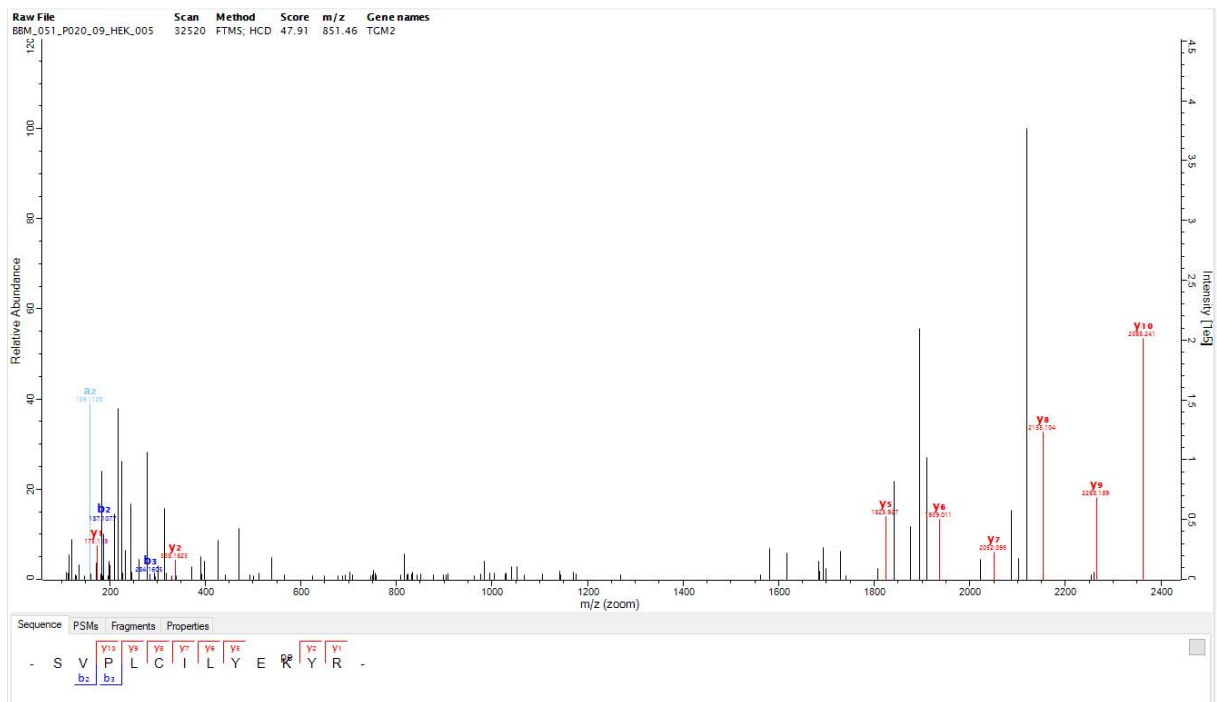
m/z (zoom)

Sequence PSMs Fragments Properties

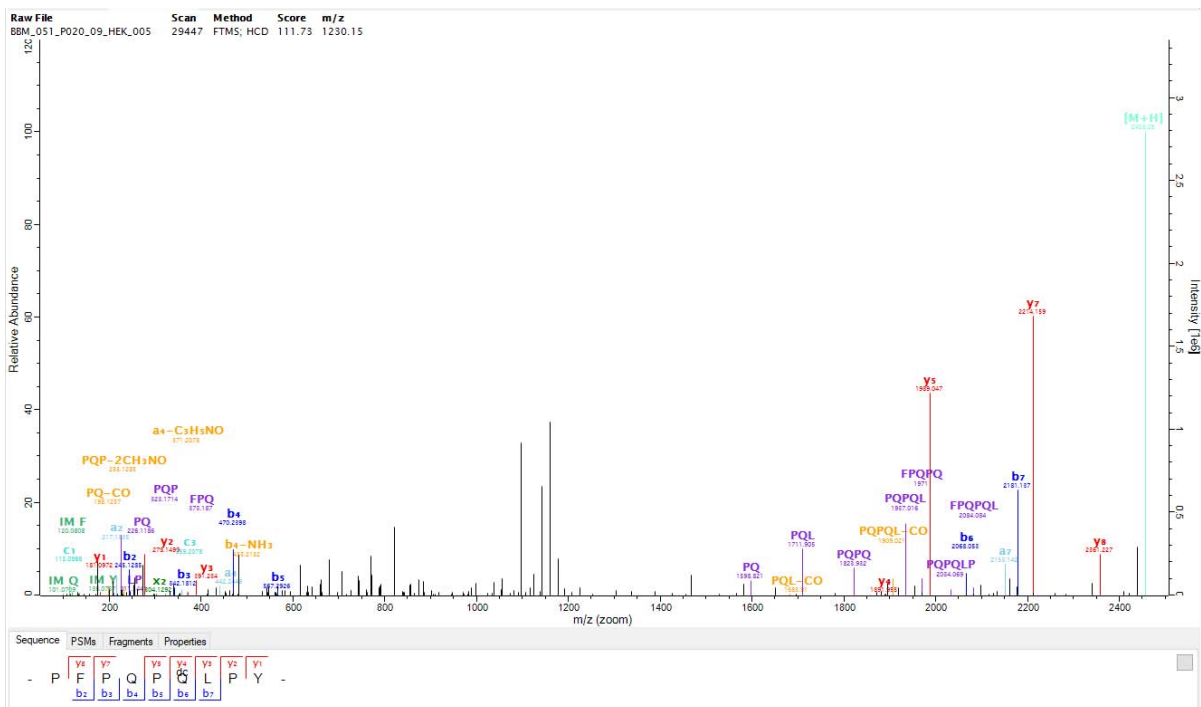
- P F P P P L P Y -

c1 b2 b2 b6 b7

S



T



New File

Scan Method Score m/z  
BBM\_051\_P020\_09\_HEK\_004 26646 FTMS, HCD 110.81 695.62

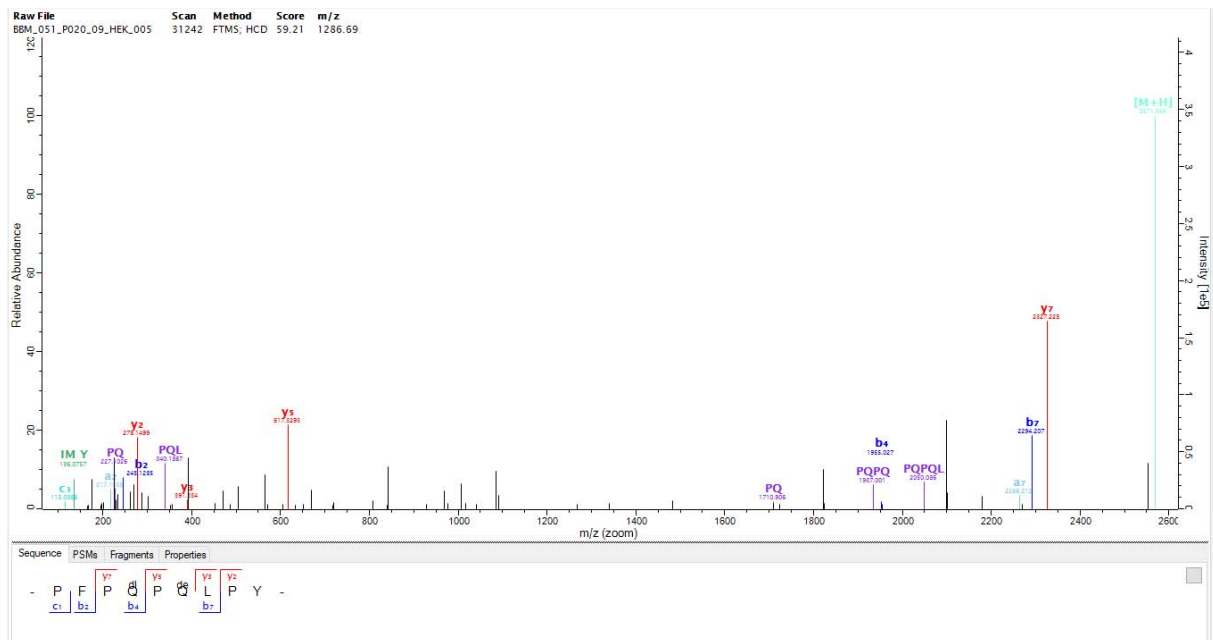
Sequence PSMs Fragments Properties

- P F P Q P L P Y -

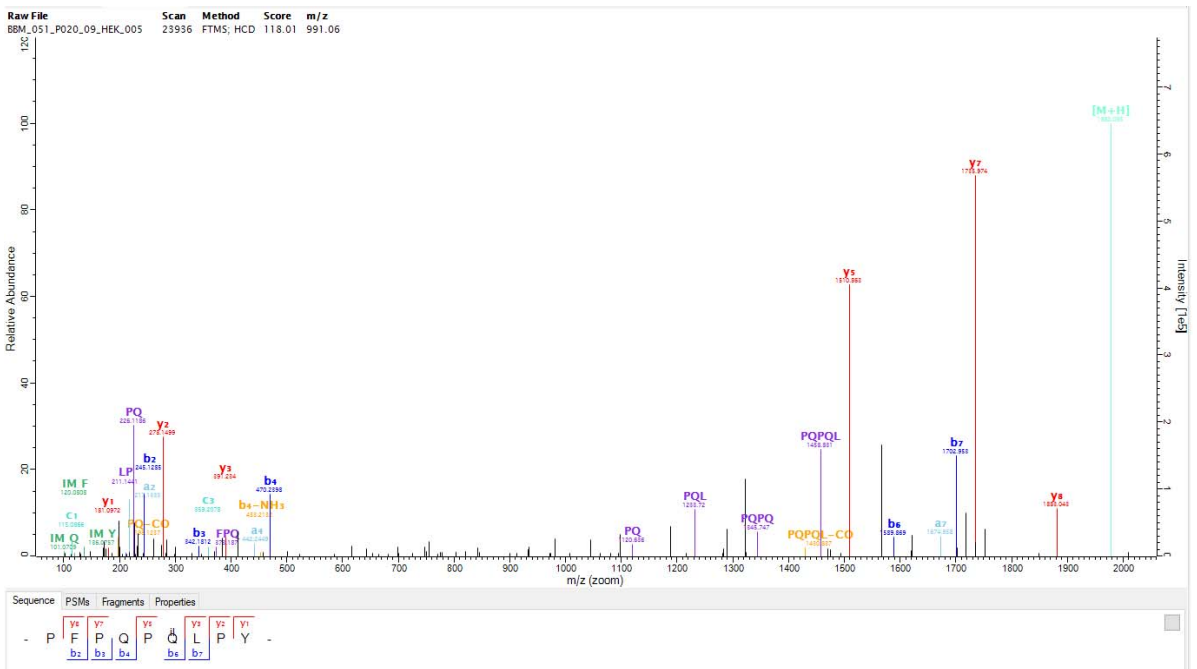
b2 b3 b4 b5 b6 b7

Raw File Scan Method Score m/z  
BSM\_051\_P020\_09\_HEK\_006 30125 FTMS; HCD 164.89 1286.19

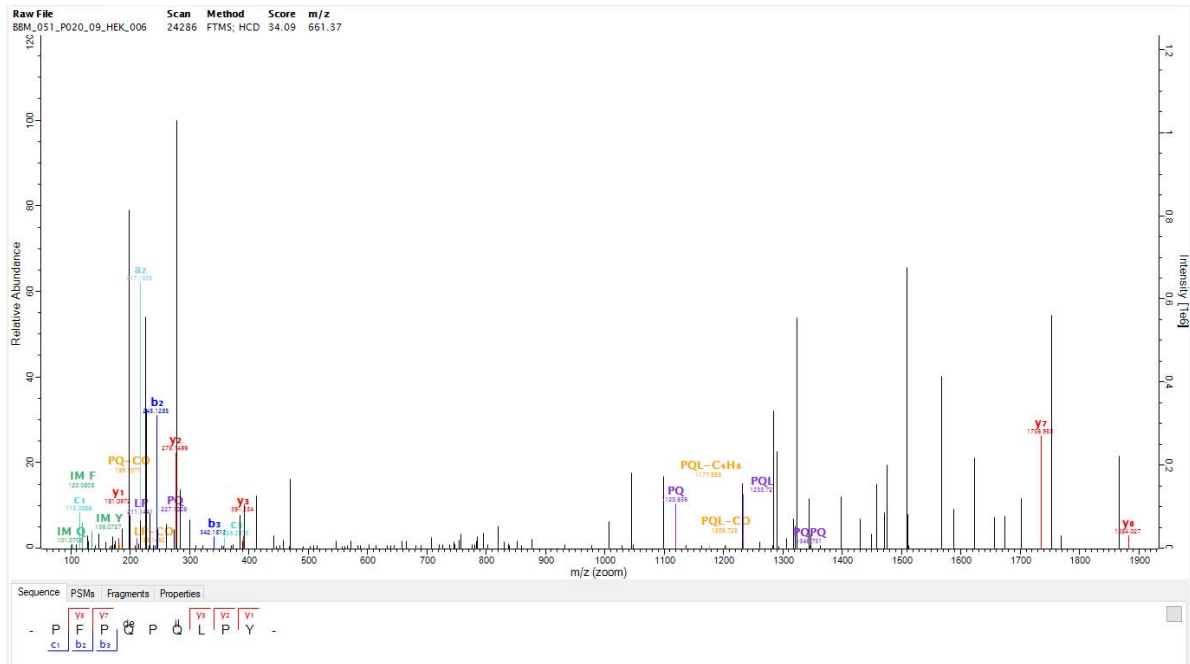
W



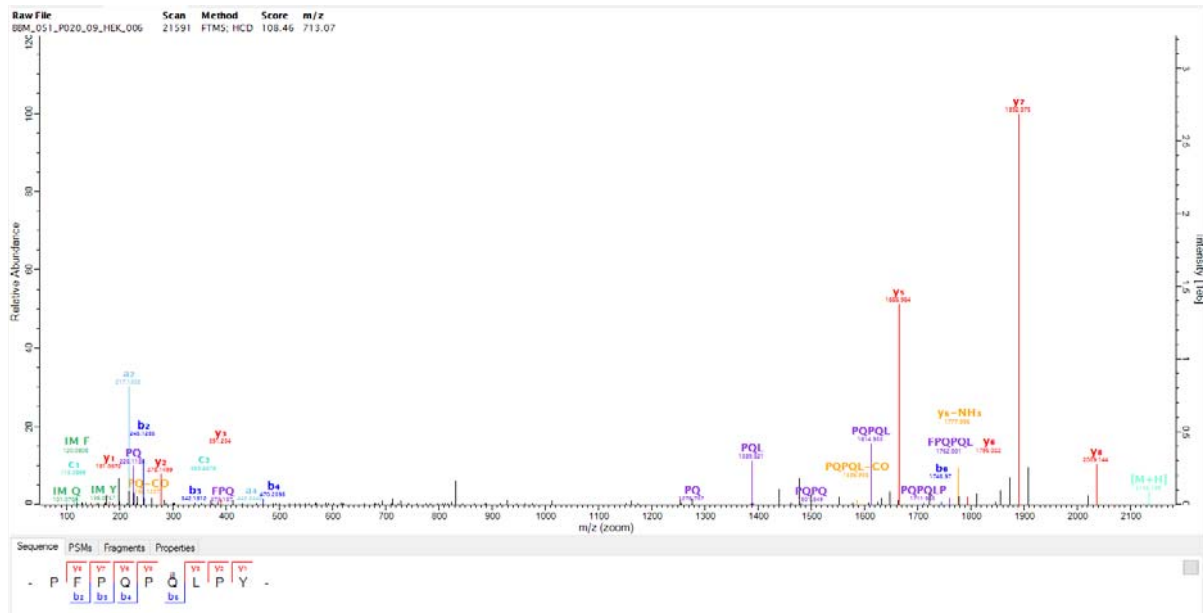
X



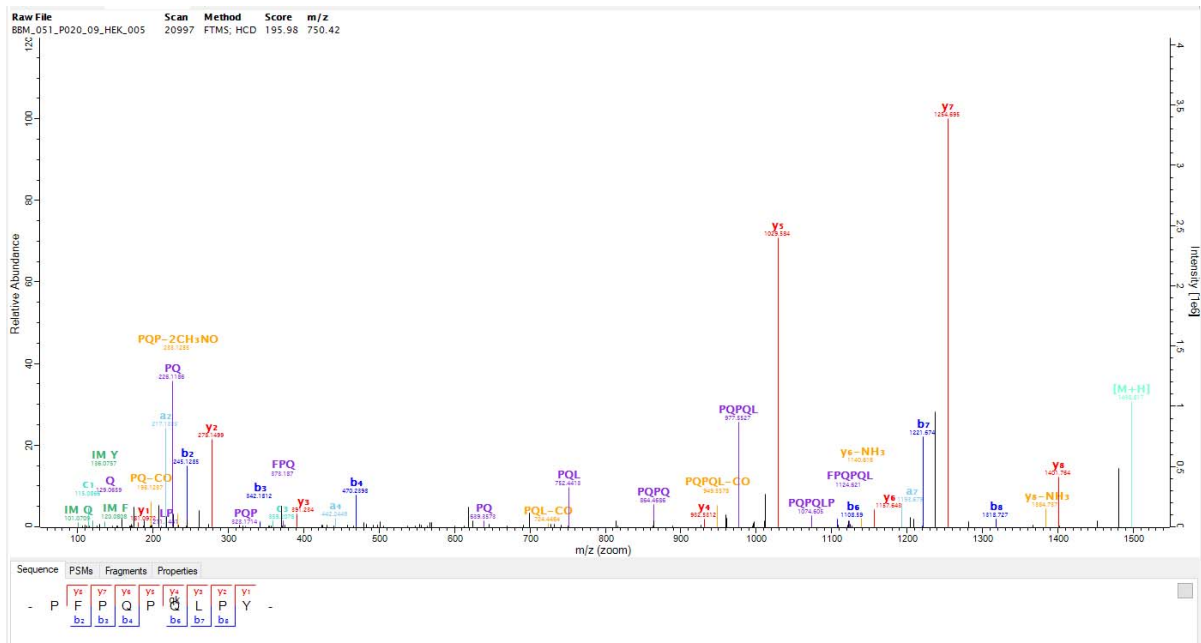
Y



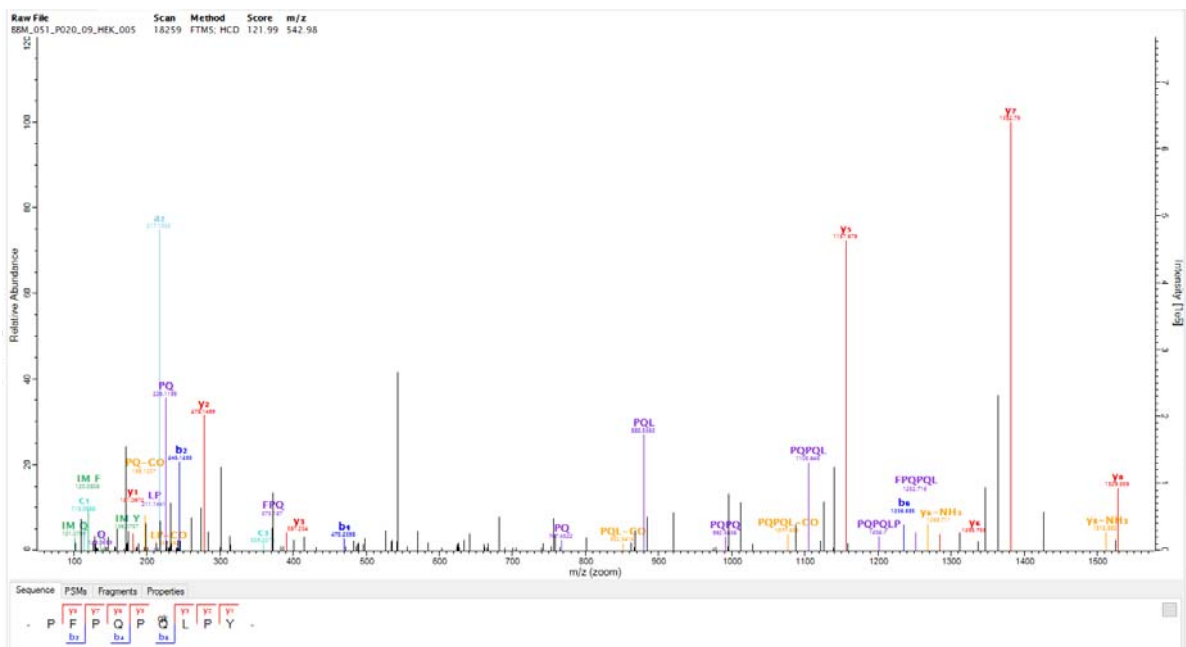
Z



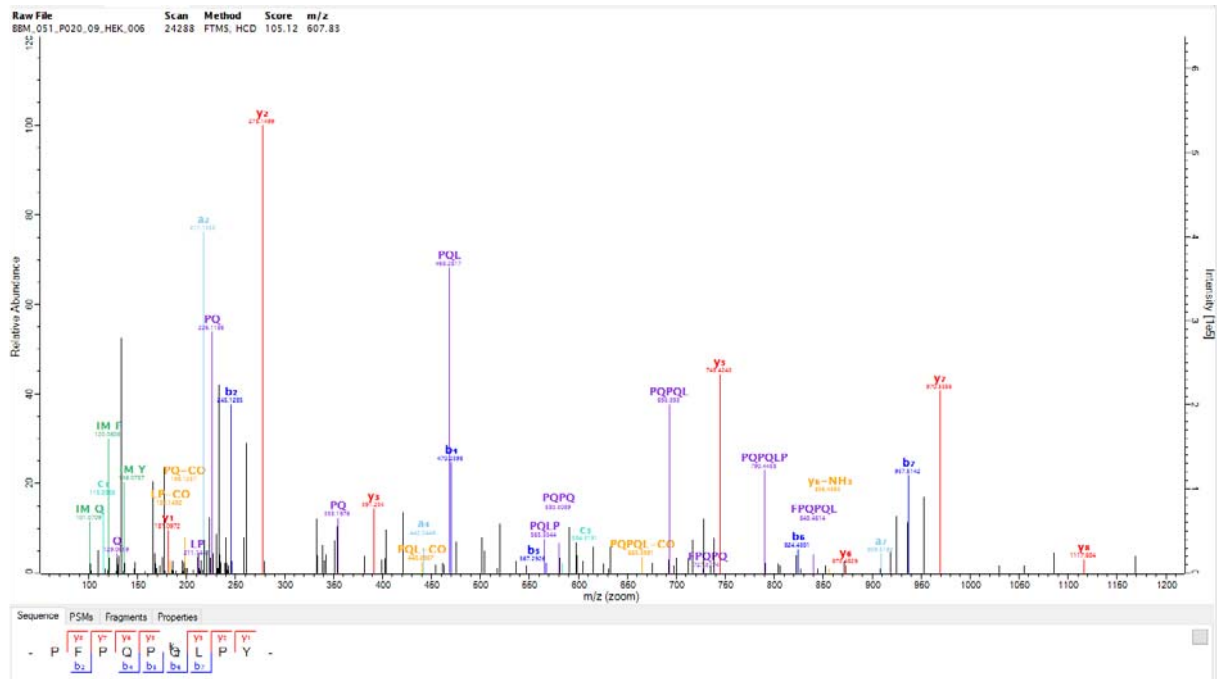
AA



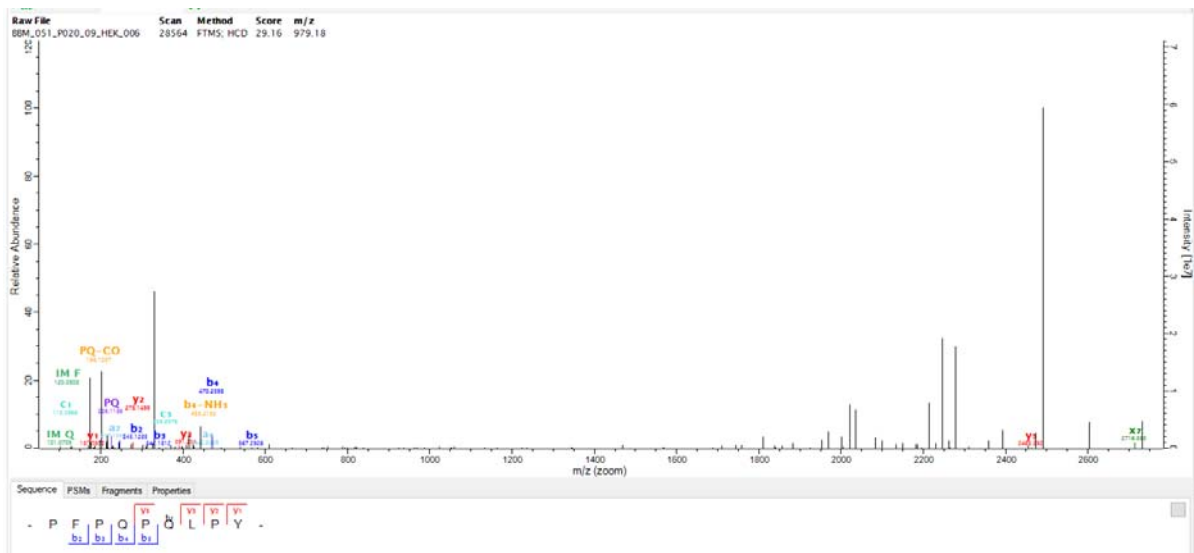
AB



AC



AD



Raw File: BBM\_051\_P070\_09\_MEK\_005 Scan Method Score m/z 35536 FTMS; HCD 93.2 937.51

Sequence: PSMs Fragments Properties

Y1 Y2 Y3 Y4 Y5 Y6 Y7 Y8 Y9 Y10 Y11 Y12 Y13 Y14 Y15 Y16 Y17 Y18 Y19 Y20 Y21 Y22 Y23 Y24 Y25 Y26 Y27 Y28 Y29 Y30 Y31 Y32 Y33 Y34 Y35 Y36 Y37 Y38 Y39 Y40 Y41 Y42 Y43 Y44 Y45 Y46 Y47 Y48 Y49 Y50 Y51 Y52 Y53 Y54 Y55 Y56 Y57 Y58 Y59 Y6



Raw File Scan Method Score m/z

BBM\_051\_P020\_09-HEK-005 24334 FTMS: HCD 151.51 873.98

Relative Abundance

m/z (zoom)

Intensity [a.u.]

Sequence PSMs Fragments Properties

- P Y8 Y7 Y4 Y3 Y2 Y1 -

c1 b2 b4 b6 b7 b8

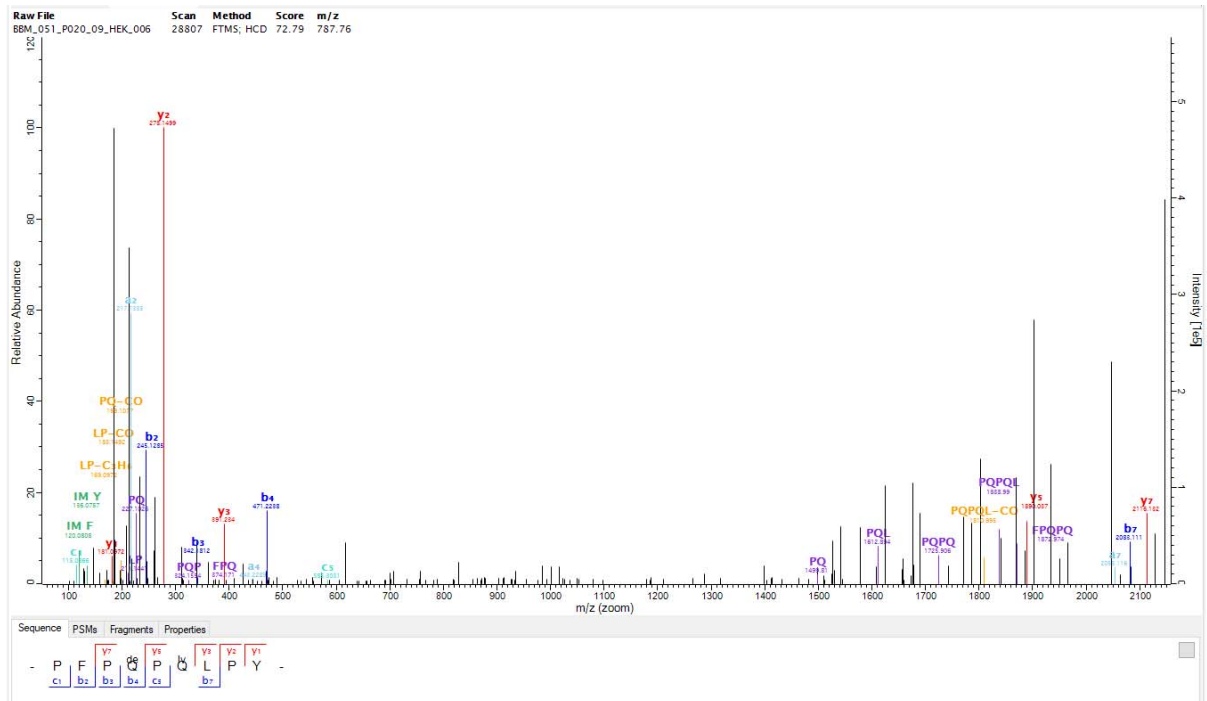
Raw File  
 BSM\_051\_P020\_09\_HEK\_005 Scan Method Score m/z  
 28857 FTMS; HCD 104.75 787.43

Sequence PSMs Fragments Properties

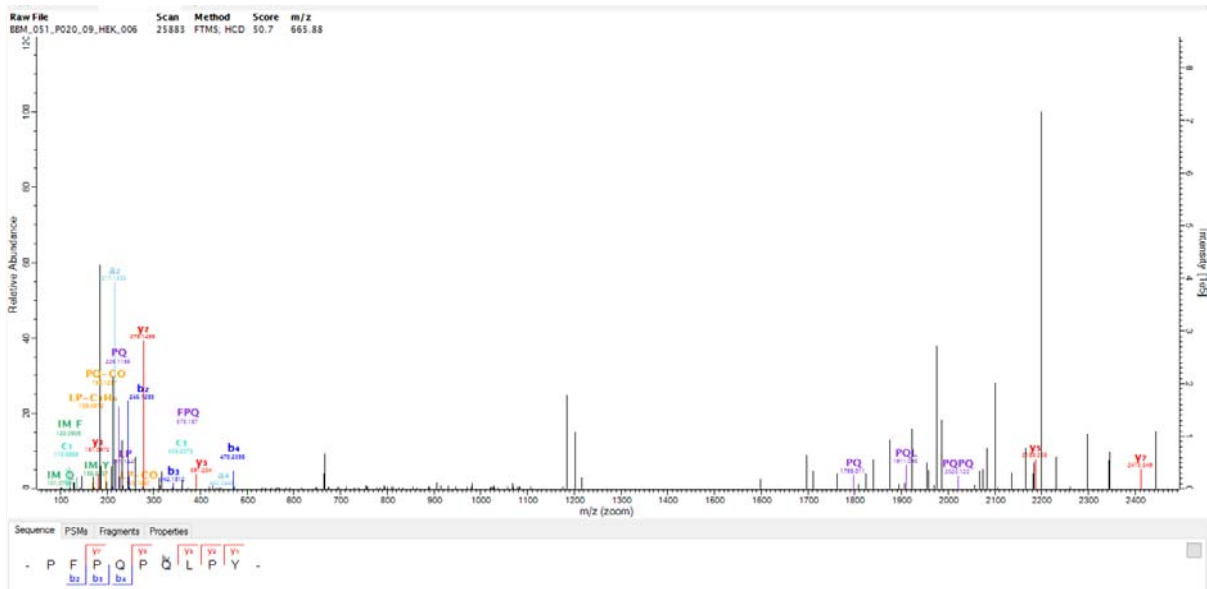
- P F P Q P Q L P Y -

Y2+ b2 Y- b3 Y3 b4 Y1 b5

AI



AJ



Supplemental Figure S3. **MS/MS spectra of isopeptides between human tissue transglutaminase (TG2) and PepQ (PFPQPQLPY) or its deamidated form PepE.** The fragments are marked in different colors as follows: y-fragments in red; b-fragments in blue; a- and c-fragments in turquoise; internal fragments in violet or green (single amino acids); fragments with losses of NH<sub>3</sub> or CO marked in orange. For every identified isopeptide a single MS/MS spectrum with annotated fragments is shown:

A, QEYVLTQQGFIYQGSAKFIK/PepE  
B, FLKNAGR/PepQ  
C, FLKNAGR/PepE  
D, WKNHGCQR/PepQ  
E, AIKEGDLSTK/PepQ  
F, AIKEGDLSTK/PepE  
G, SLIVGLKISTK/PepQ  
H, ISTKSVGR/PepQ  
I, ISTKSVGRDER/PepQ  
J, EDITHTYKYPEGSSEER/PepQ  
K, EDITHTYKYPEGSSEER/PepE  
L, DEREDITHTYKYPEGSSEER/PepQ  
M, LEAKEETGMAMR/PepQ  
N, LEAKEETGMAMR/PepE  
O, ANHLNKLEAKEETGMAMR/PepQ  
P, ANHLNKLEAKEETGMAMR/Pep  
Q, ANHLNK/PepQ  
R, ANHLNK/PepE  
S, SVPLCILYEKYR/PepQ  
T, DCLTESNLIKVR/PepQ  
U, YRDCLTESNLIKVR/PepQ  
V, DLYLENPEIKIR/PepQ  
W, DLYLENPEIKIR/PepE  
X, ILGEPKQK/PepQ  
Y, ILGEPKQK/PepE  
Z, ILGEPKQKR/PepQ  
AA, QKR/PepQ  
AB, QKRK/PepQ  
AC, K/PepQ  
AD, TVEIPDPVEAGEEVKVR/PepQ  
AE, MDLLPLHMGHLKLVVNFESDKLK/PepQ  
AF, AVKGFR/PepQ  
AG, AVKGFR/PepE  
AH, LVVNFESDKLK/PepQ  
AI, LVVNFESDKLK/PepE  
AJ, LVVNFESDKLKAVK/PepQ.