

Supporting information for:

**A streamlined data analysis pipeline for the identification of sites of
citrullination**

Aaron J. Maurais^{‡2}, Ari J. Salinger^{‡1,2}, Micaela Tobin¹, Scott A. Shaffer^{1,3}, Eranthie
Weerapana^{*2}, Paul R. Thompson^{*1}

¹Department of Biochemistry and Molecular Pharmacology, University of Massachusetts
Medical School, LRB 826, 364 Plantation Street, Worcester, MA 01605, USA.

²Department of Chemistry, Boston College, Chestnut Hill, MA 02467, USA.

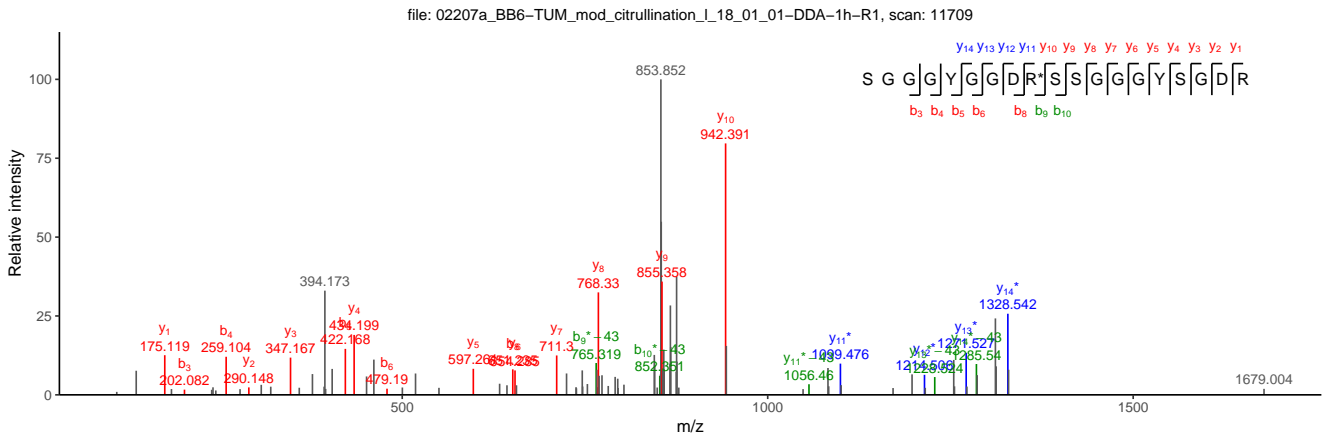
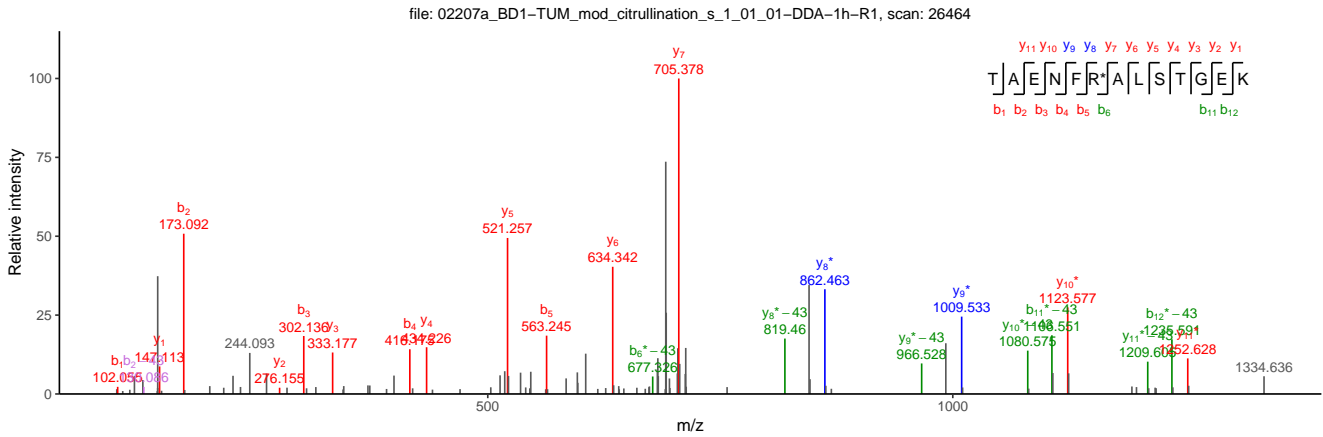
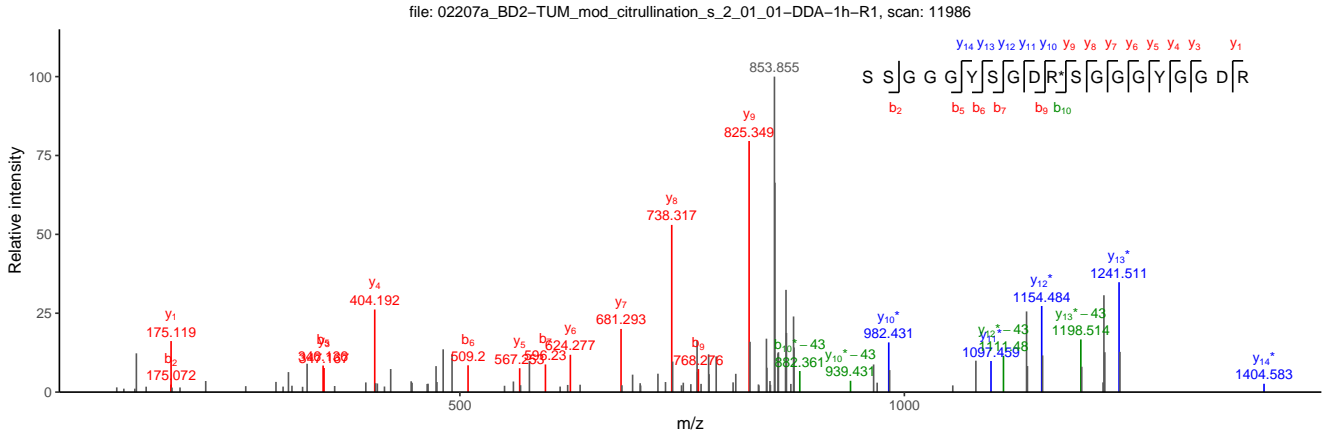
³Mass Spectrometry Facility, University of Massachusetts Medical School, Shrewsbury,
MA 01545, USA.

[‡]These authors contributed equally to this work

^{*}Corresponding authors; paul.thompson@umassmed.edu, eranthie@bc.edu

Ground truth: true, ionFinder classification: true

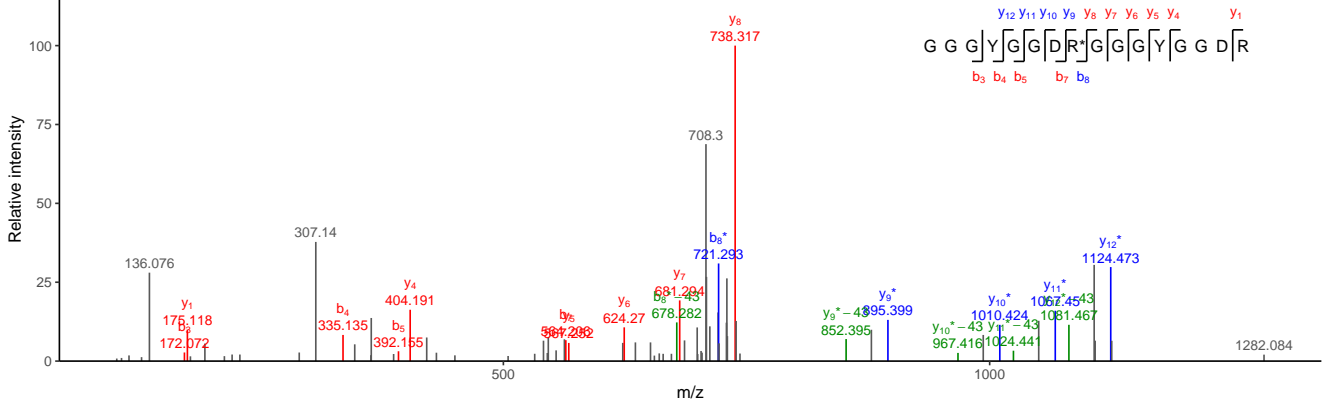
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



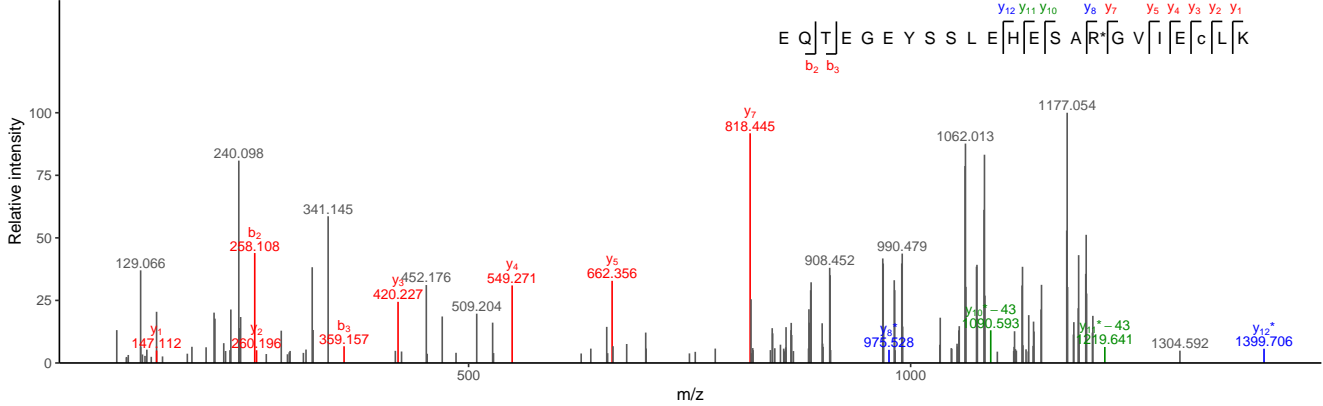
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

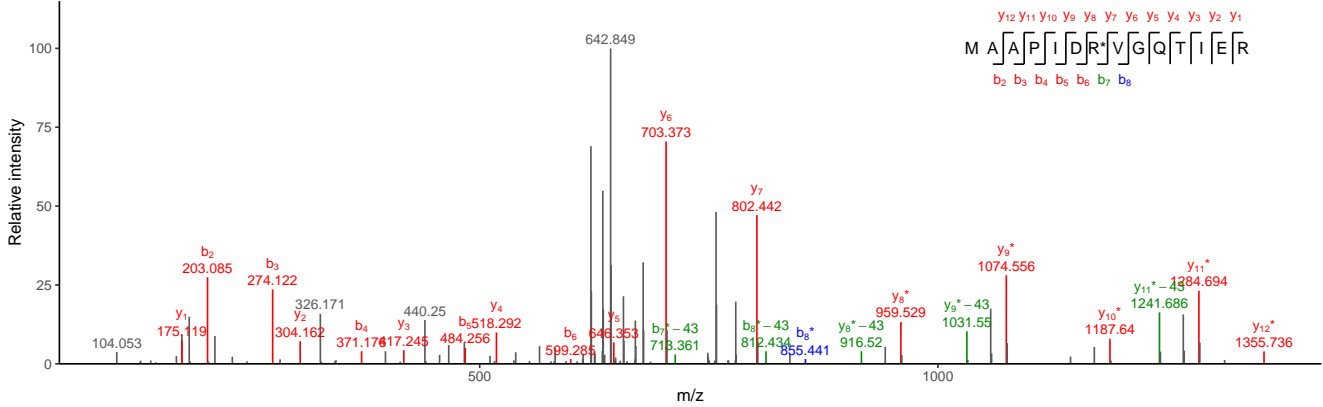
file: 02207a_BB1-TUM_mod_citrullination_I_13_01_01-DDA-1h-R1, scan: 12990



file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 38470



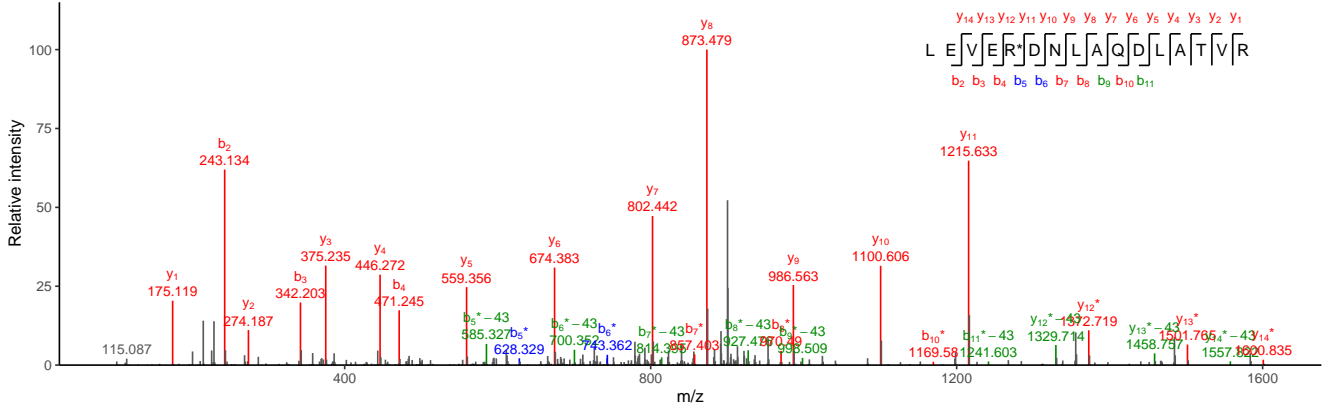
file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 35805



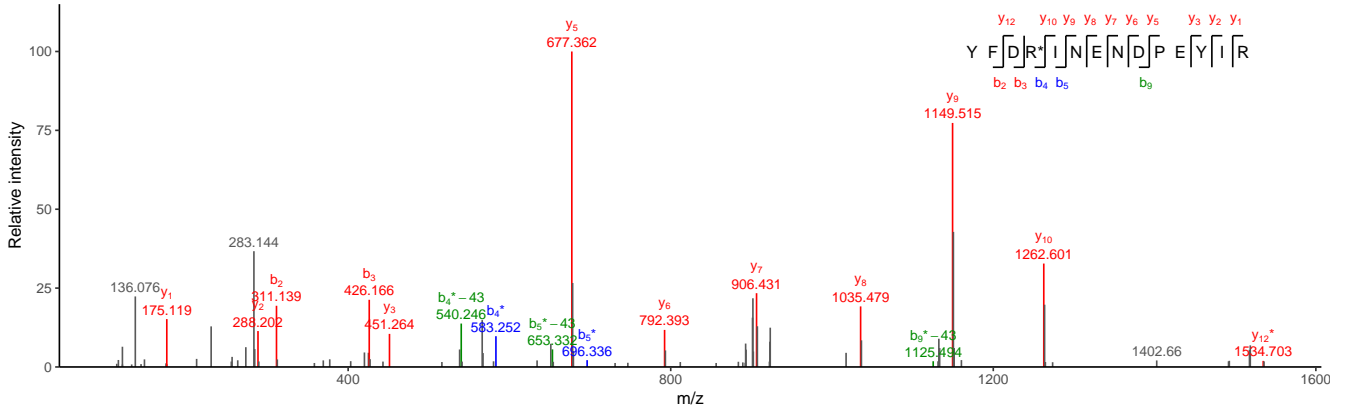
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

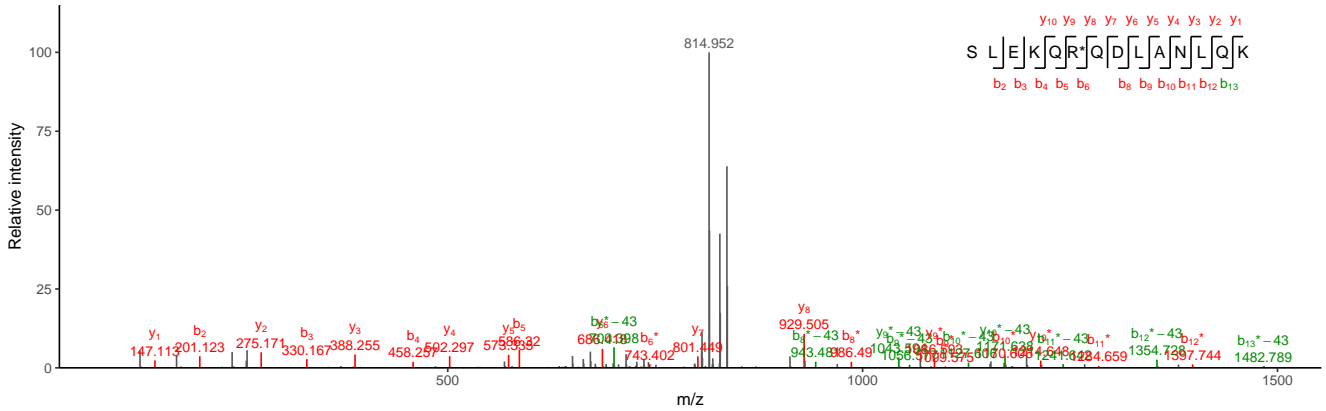
file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 42781



file: 02207a_BA9-TUM_mod_citrullination_L_9_01_01-DDA-1h-R1, scan: 34873



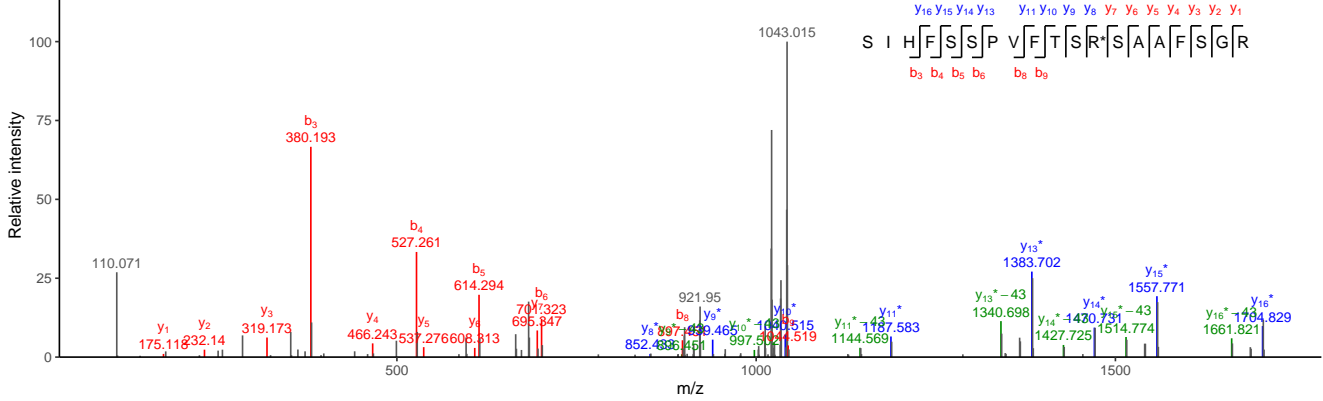
file: 02207a_BA9-TUM_mod_citrullination_L_9_01_01-DDA-1h-R1, scan: 24354



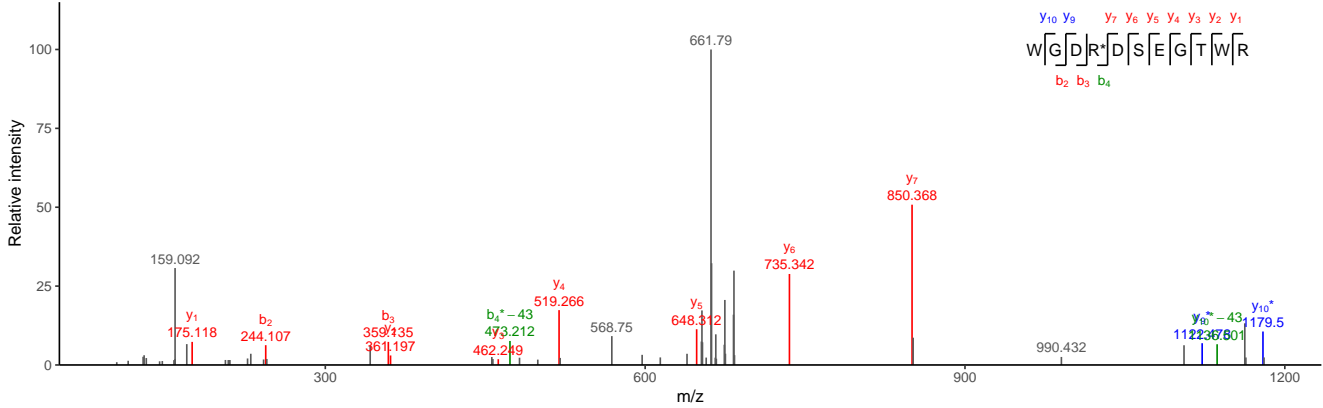
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

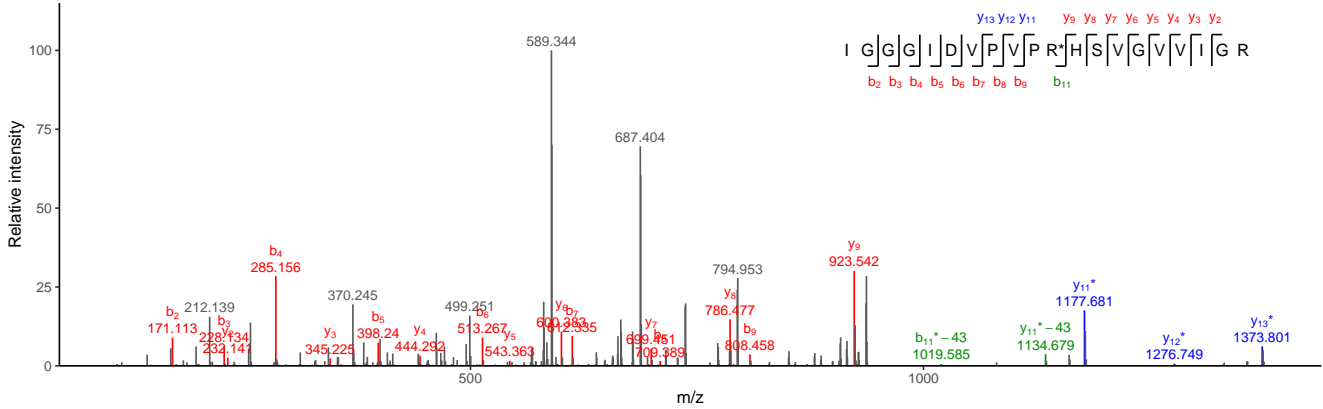
file: 02207a_BB6-TUM_mod_citrullination_I_18_01_01-DDA-1h-R1, scan: 45213



file: 02207a_BA4-TUM_mod_citrullination_I_4_01_01-DDA-1h-R1, scan: 28509



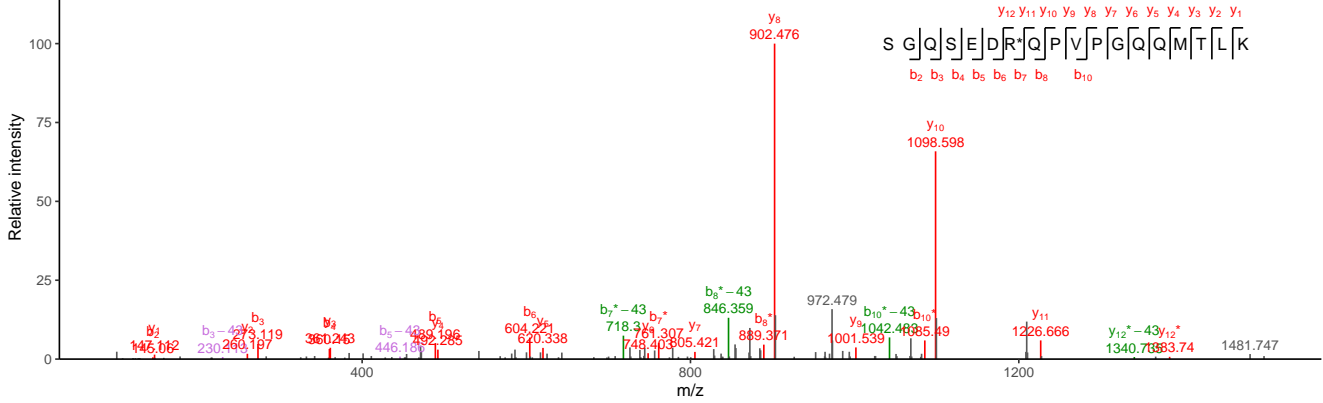
file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 40271



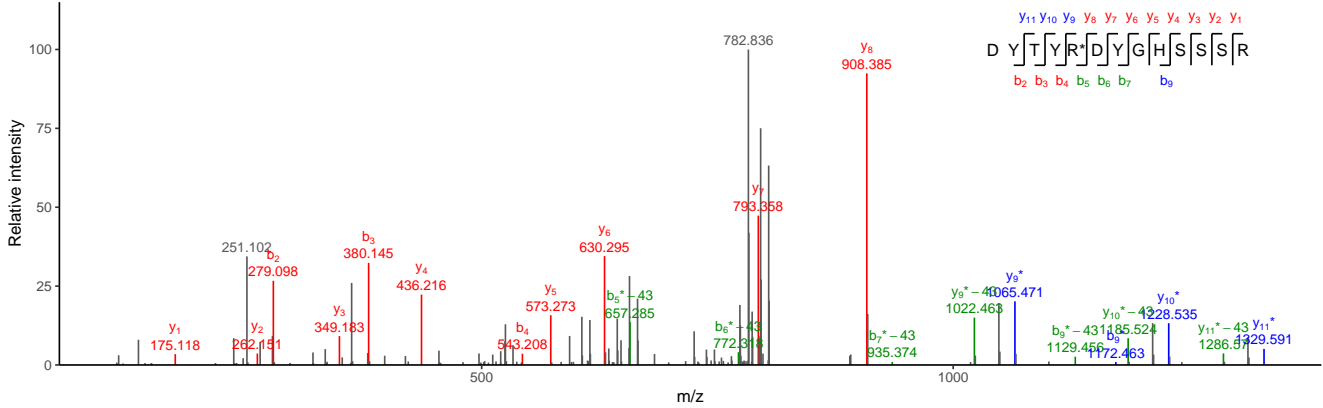
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

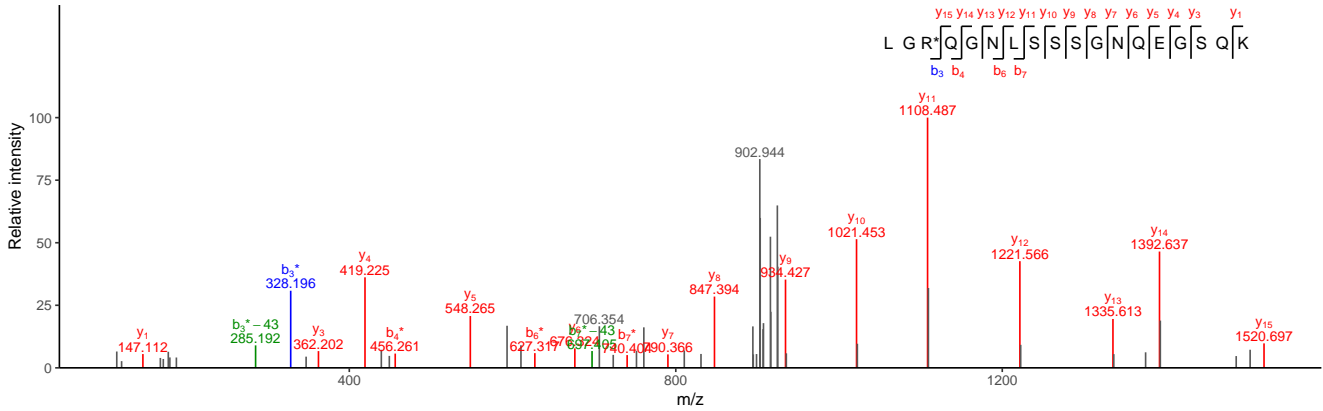
file: 02207a_BB4-TUM_mod_citrullination_I_16_01_01-DDA-1h-R1, scan: 24935



file: 02207a_BA8-TUM_mod_citrullination_I_8_01_01-DDA-1h-R1, scan: 17957



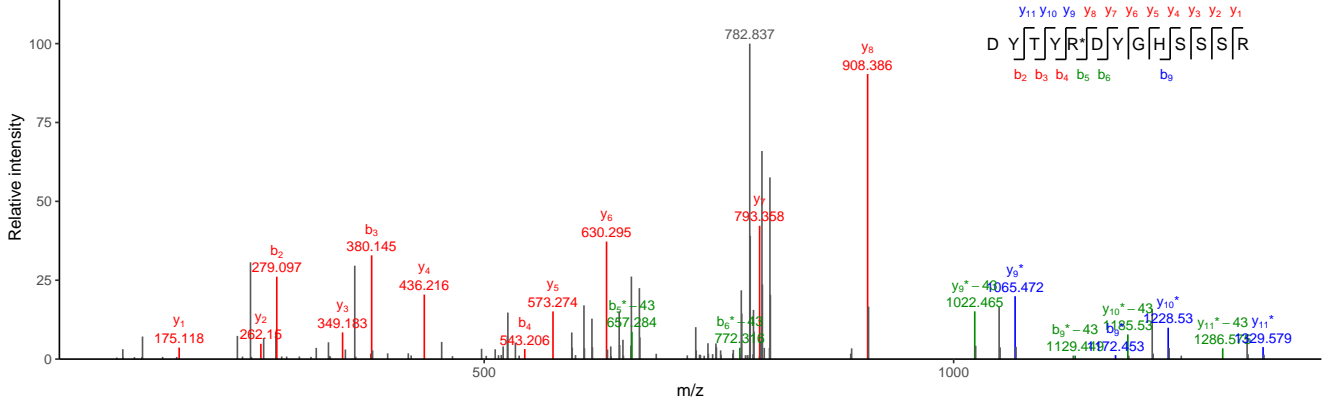
file: 02207a_BB5-TUM_mod_citrullination_I_17_01_01-DDA-1h-R1, scan: 10841



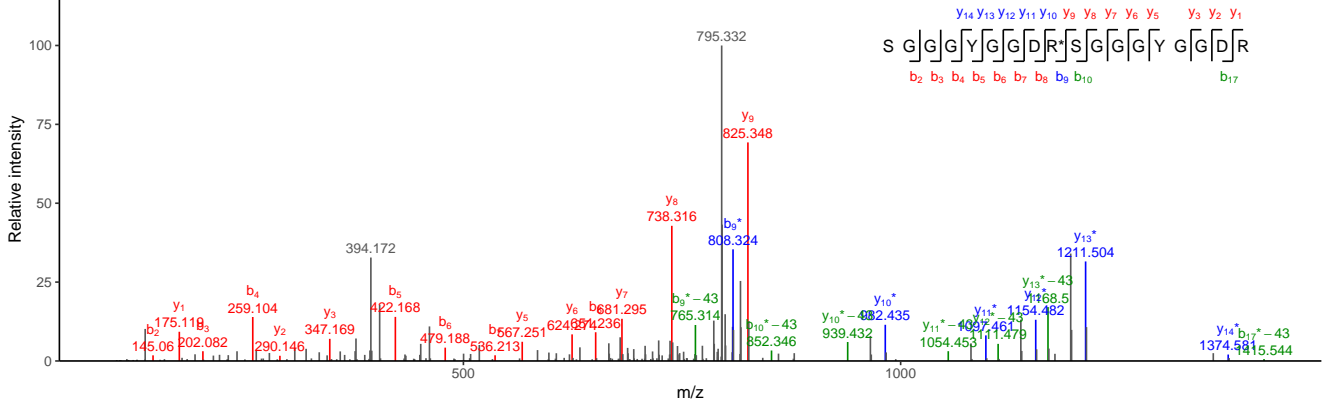
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

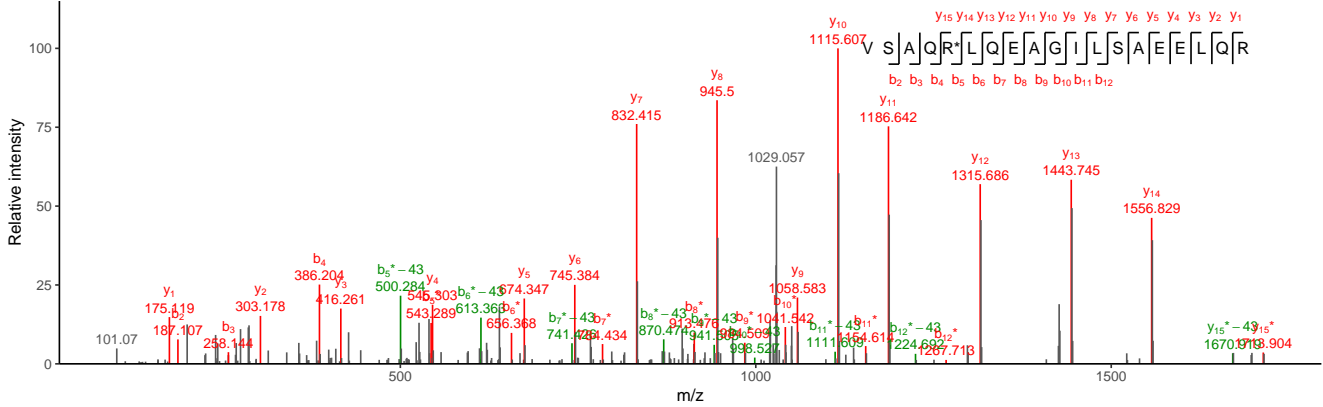
file: 02207a_BA8-TUM_mod_citrullination_I_8_01_01-DDA-1h-R1, scan: 18102



file: 02207a_BB4-TUM_mod_citrullination_I_16_01_01-DDA-1h-R1, scan: 11694

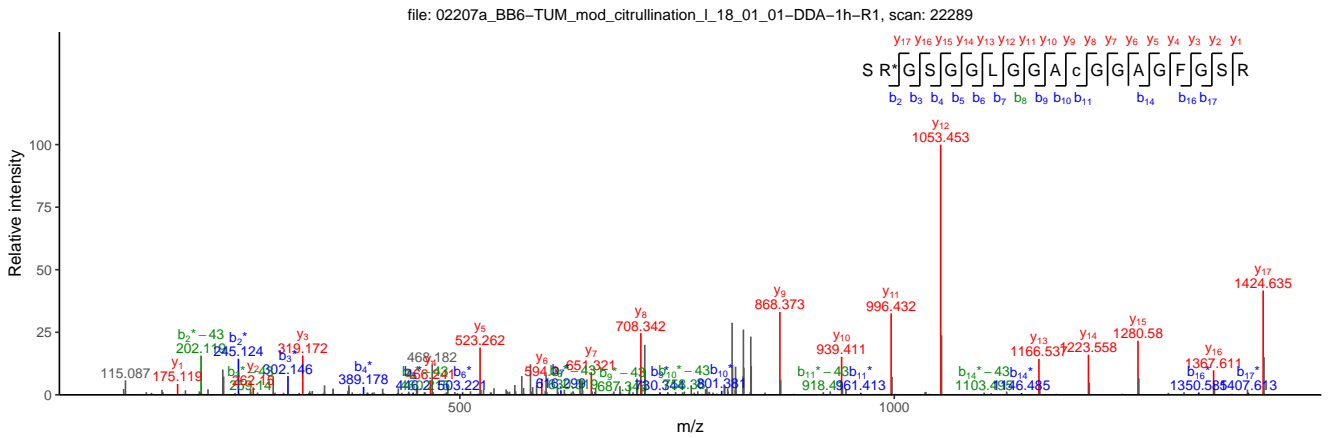
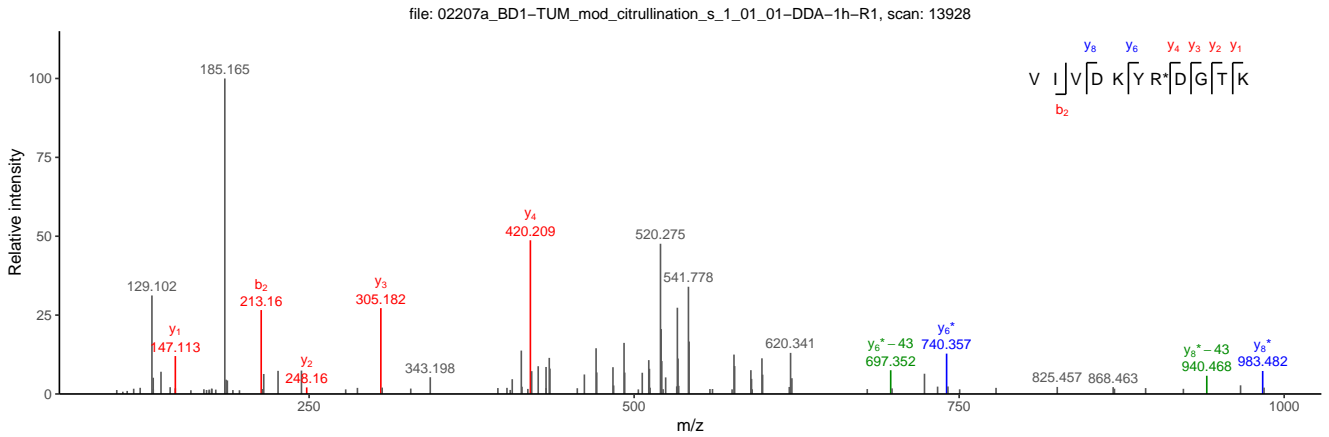
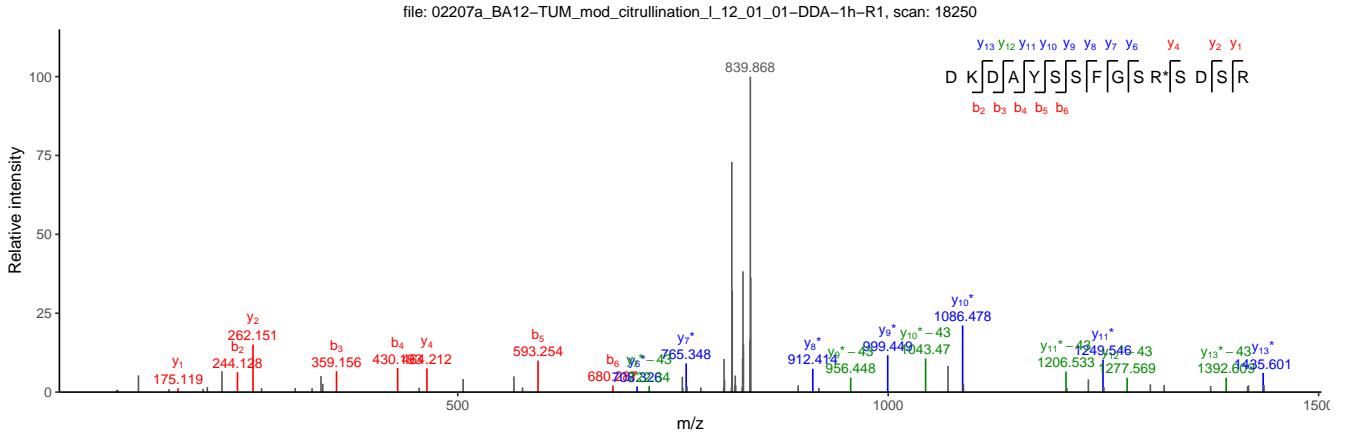


file: 02207a_BB6-TUM_mod_citrullination_I_18_01_01-DDA-1h-R1, scan: 43341



Ground truth: true, ionFinder classification: true

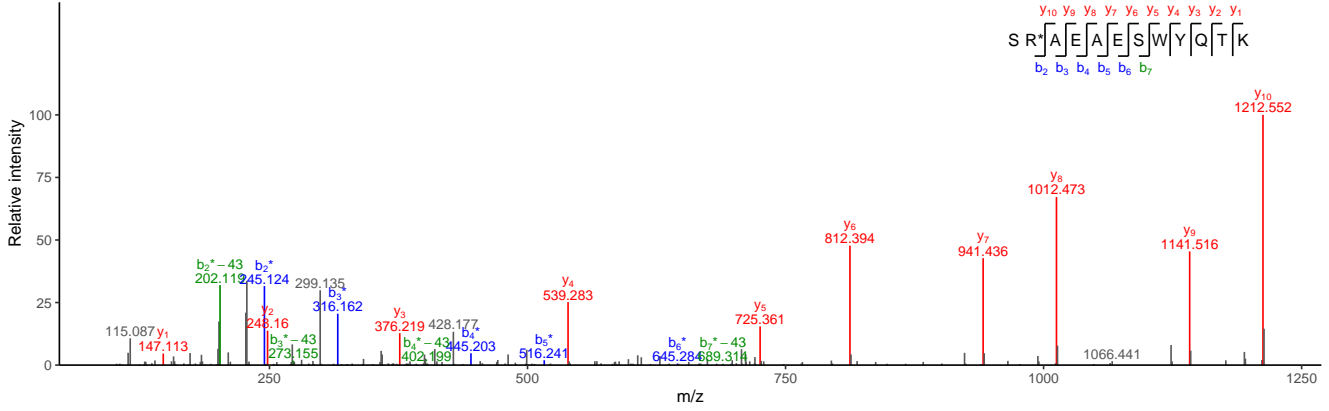
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



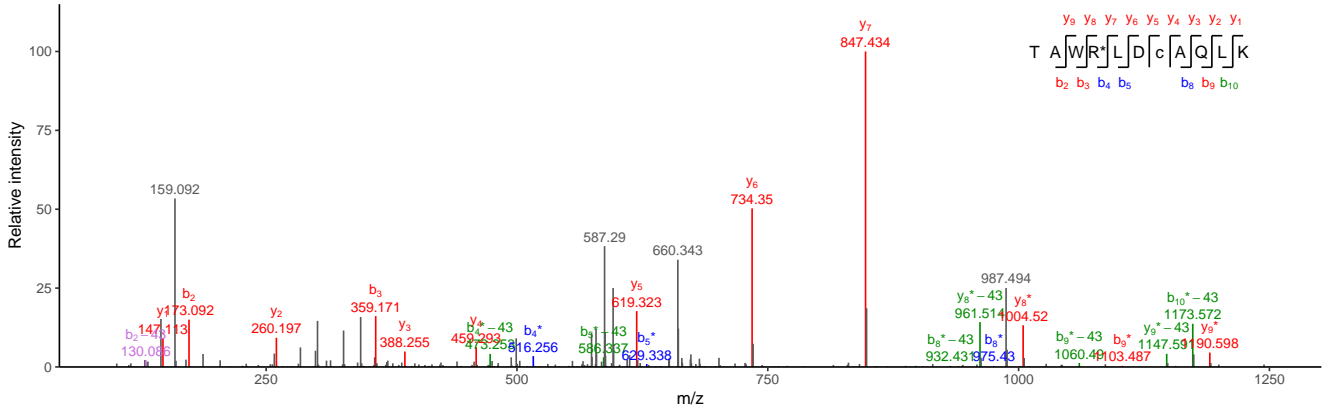
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

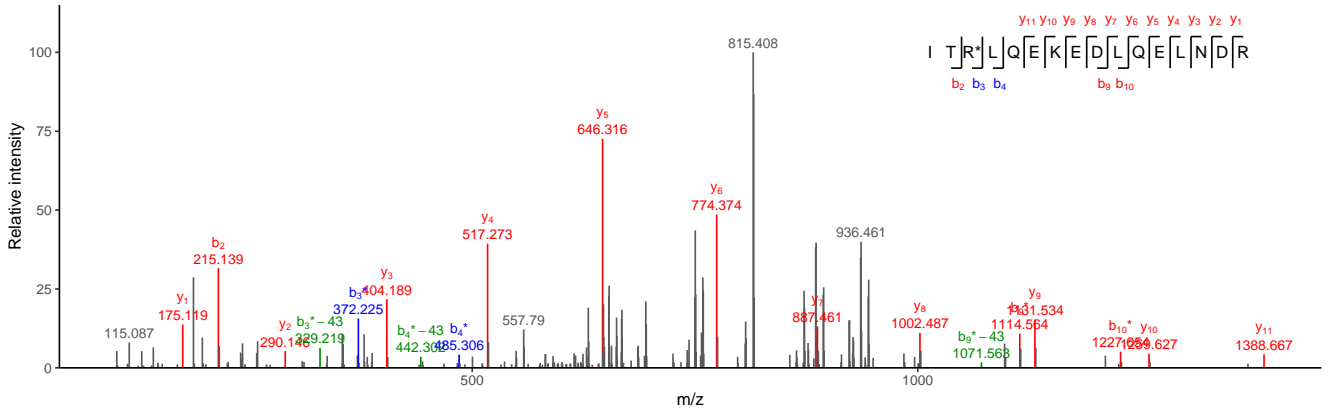
file: 02207a_BA5-TUM_mod_citrullination_L_5_01_01-DDA-1h-R1, scan: 25825



file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 35879



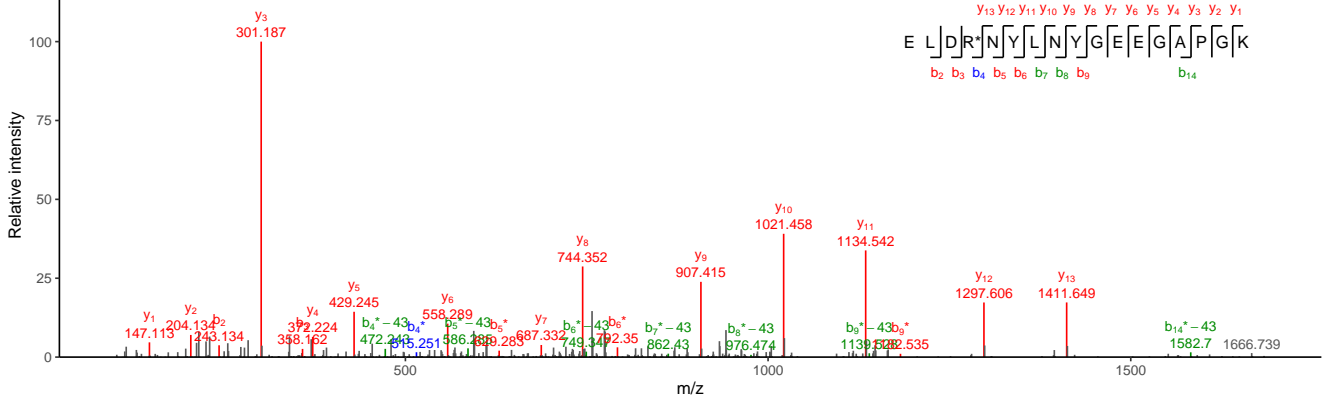
file: 02207a_BB2-TUM_mod_citrullination_L_14_01_01-DDA-1h-R1, scan: 28335



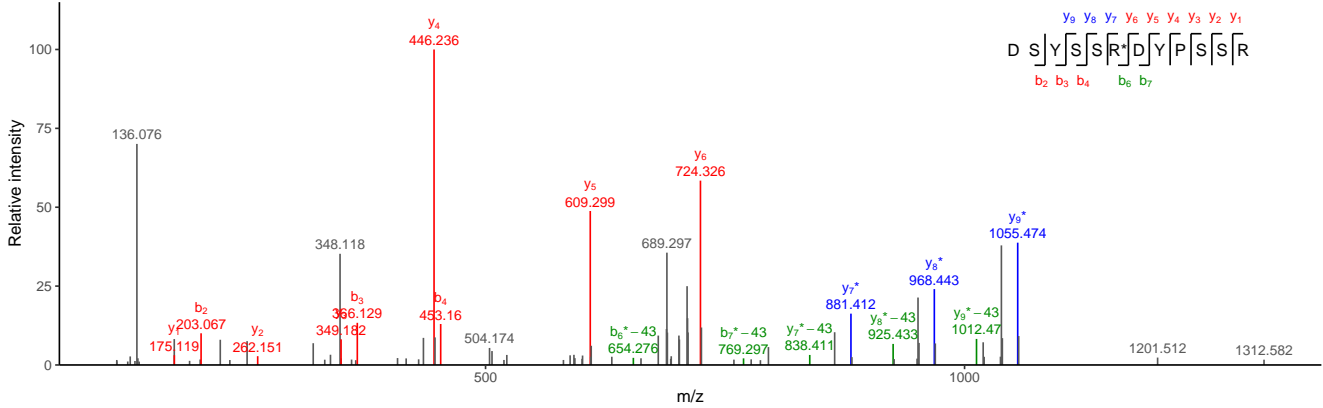
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

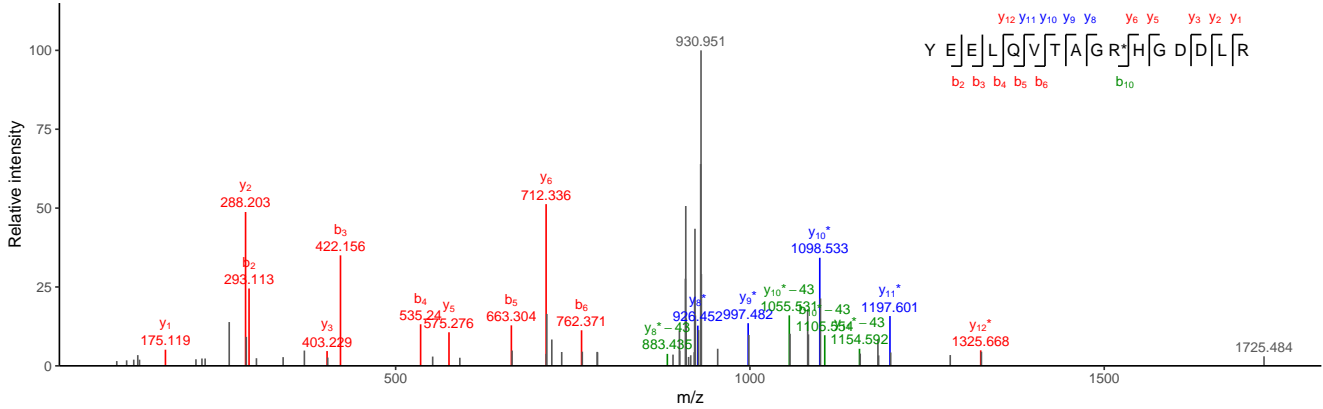
file: 02207a_BB2-TUM_mod_citrullination_L14_01_01-DDA-1h-R1, scan: 31930



file: 02207a_BD1-TUM_mod_citrullination_s1_01_01-DDA-1h-R1, scan: 14464



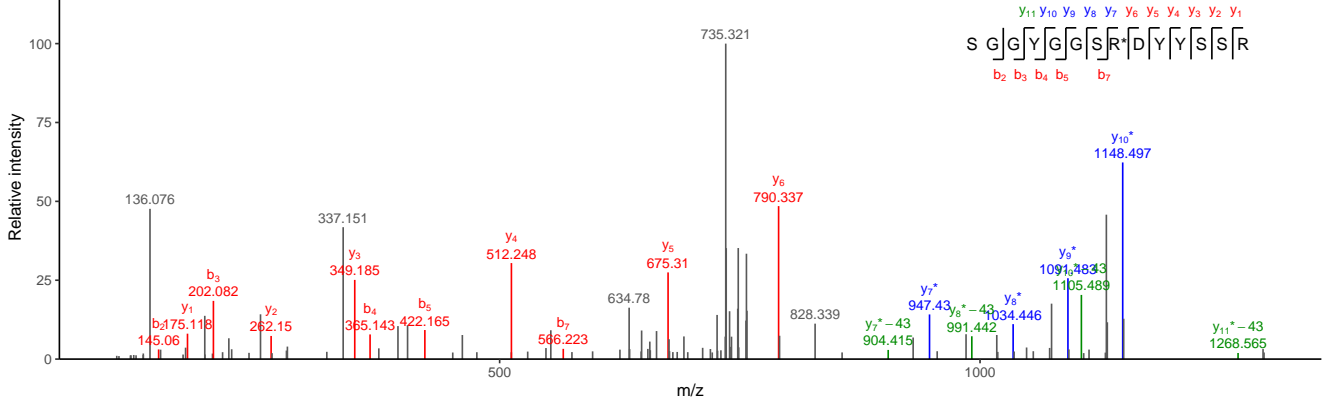
file: 02207a_BD1-TUM_mod_citrullination_s1_01_01-DDA-1h-R1, scan: 25997



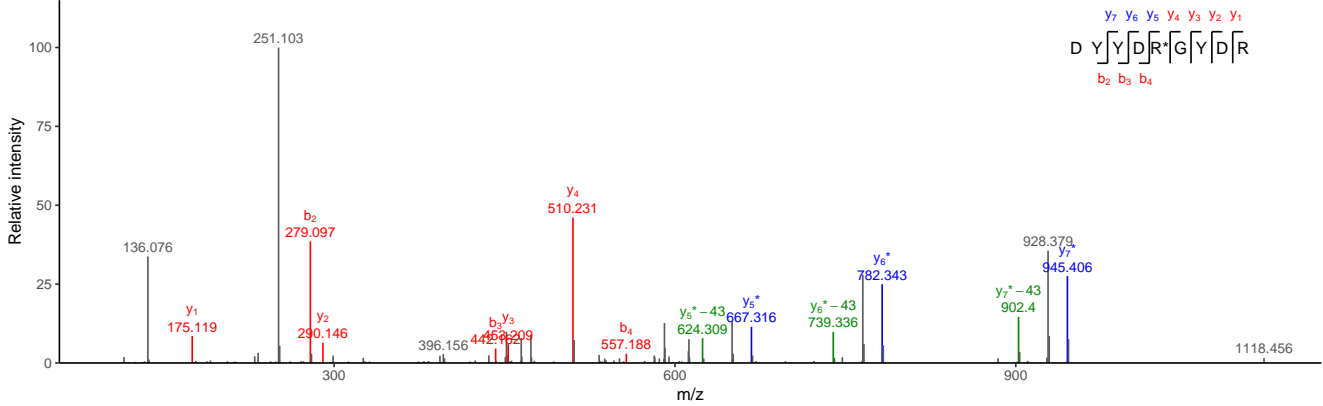
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

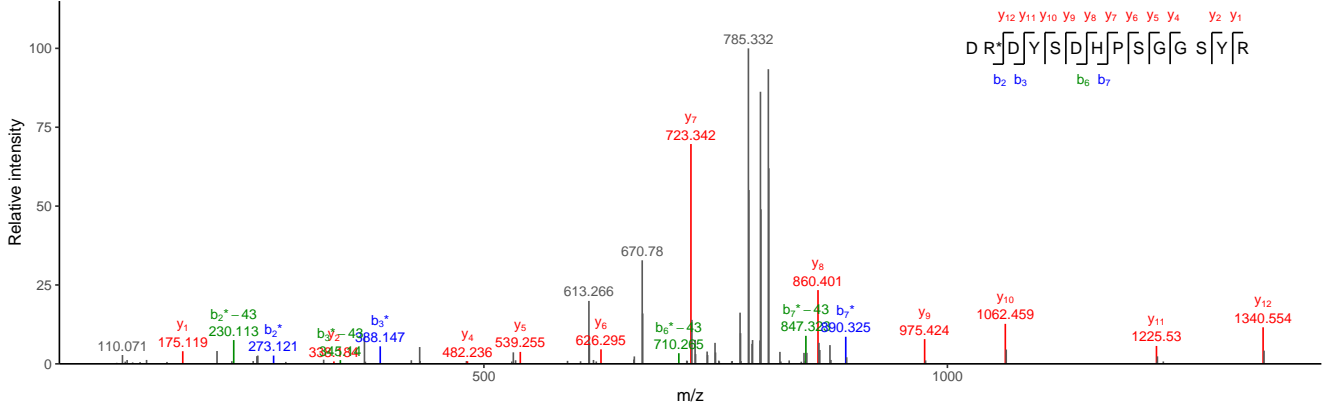
file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 17850



file: 02207a_BA2-TUM_mod_citrullination_L_2_01_01-DDA-1h-R1, scan: 21011



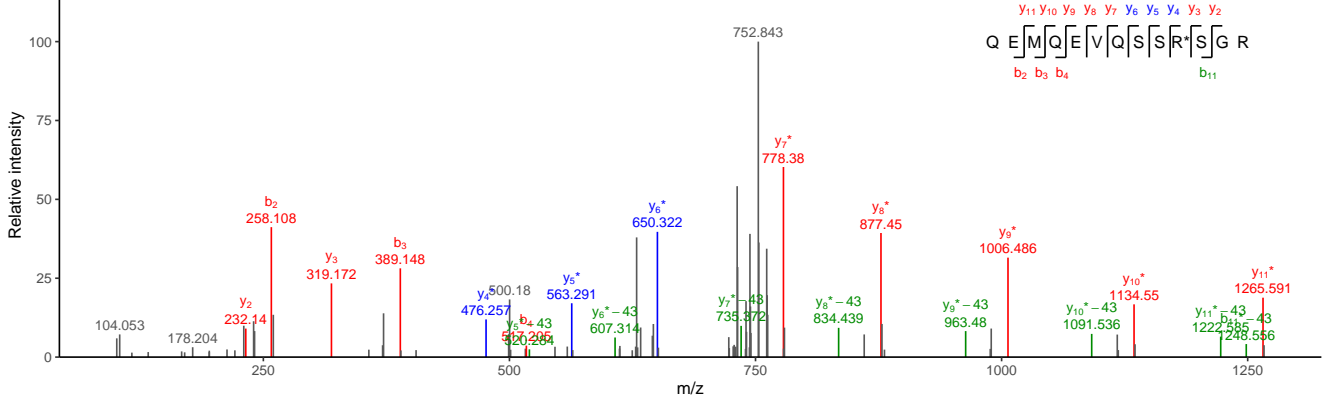
file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 10900



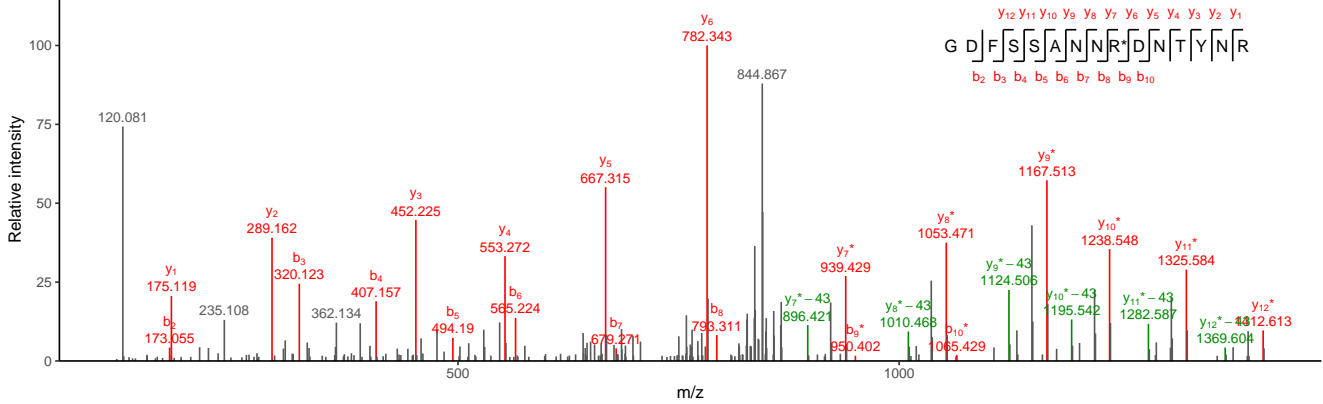
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

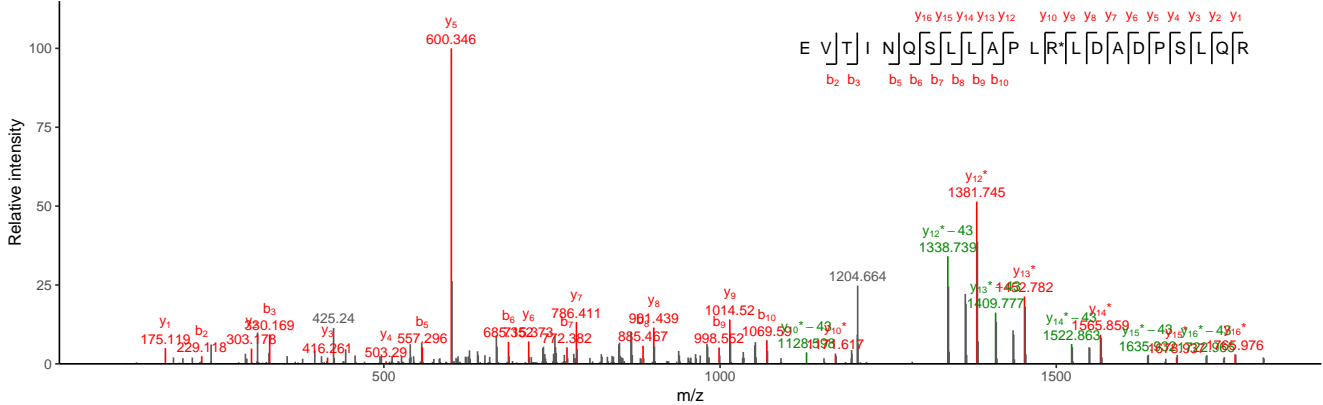
file: 02207a_BA7-TUM_mod_citrullination_L_7_01_01-DDA-1h-R1, scan: 10670



file: 02207a_BA10-TUM_mod_citrullination_L_10_01_01-DDA-1h-R1, scan: 15436



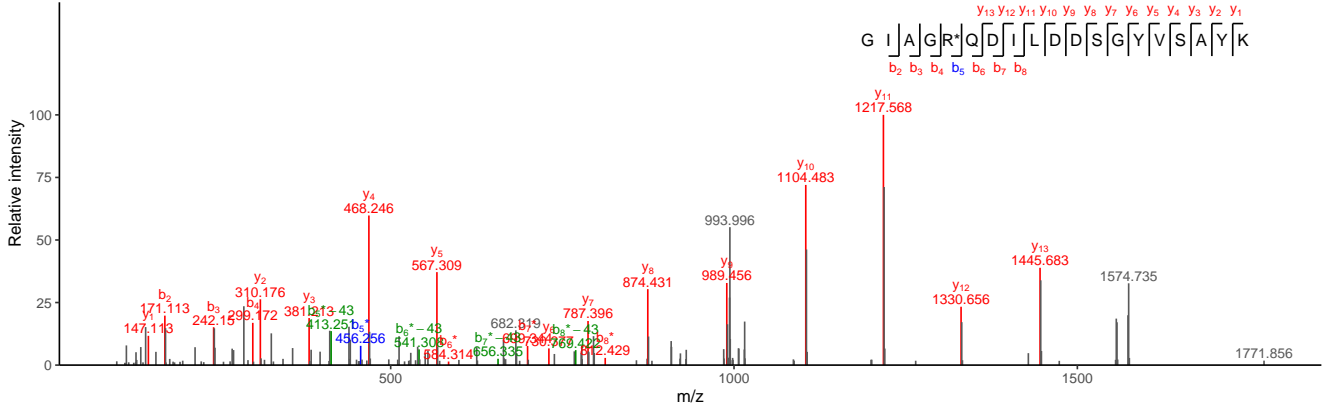
file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 54701



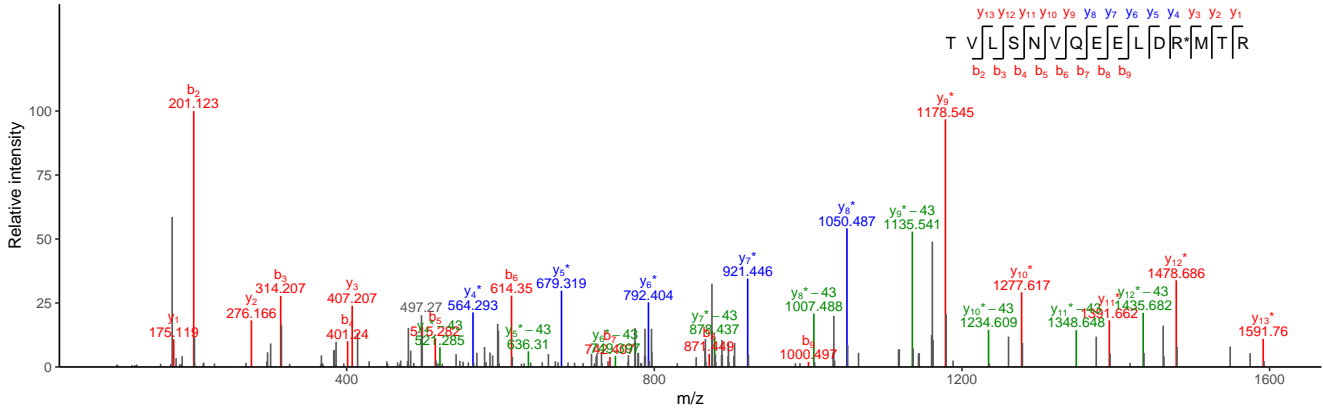
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

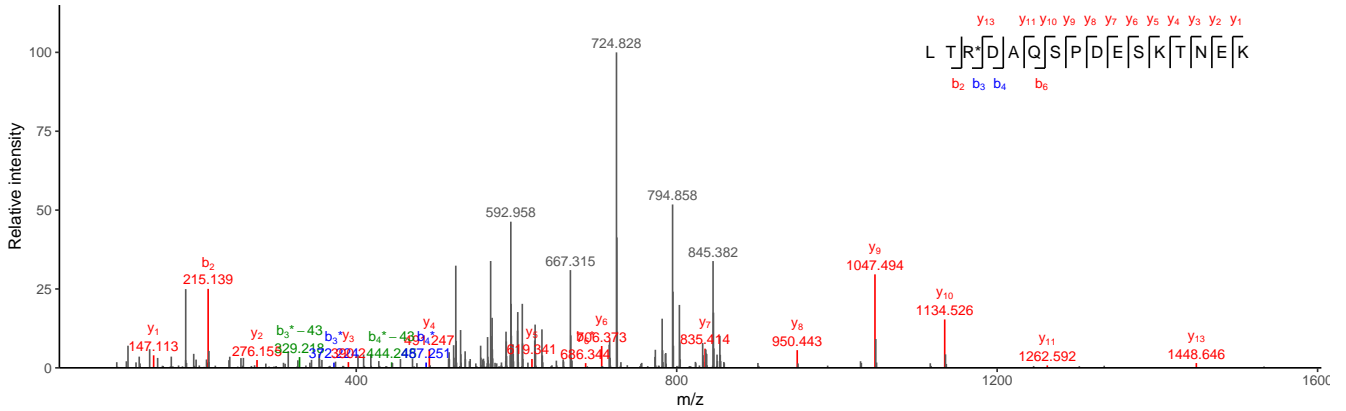
file: 02207a_BB6-TUM_mod_citrullination_I_18_01_01-DDA-1h-R1, scan: 38875



file: 02207a_BA12-TUM_mod_citrullination_I_12_01_01-DDA-1h-R1, scan: 53722



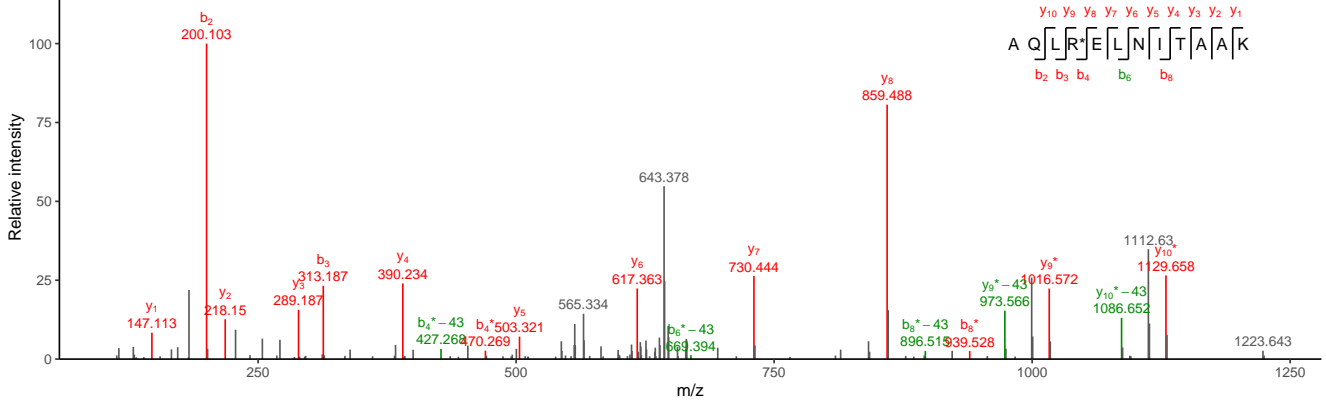
file: 02207a_BB1-TUM_mod_citrullination_I_13_01_01-DDA-1h-R1, scan: 6579



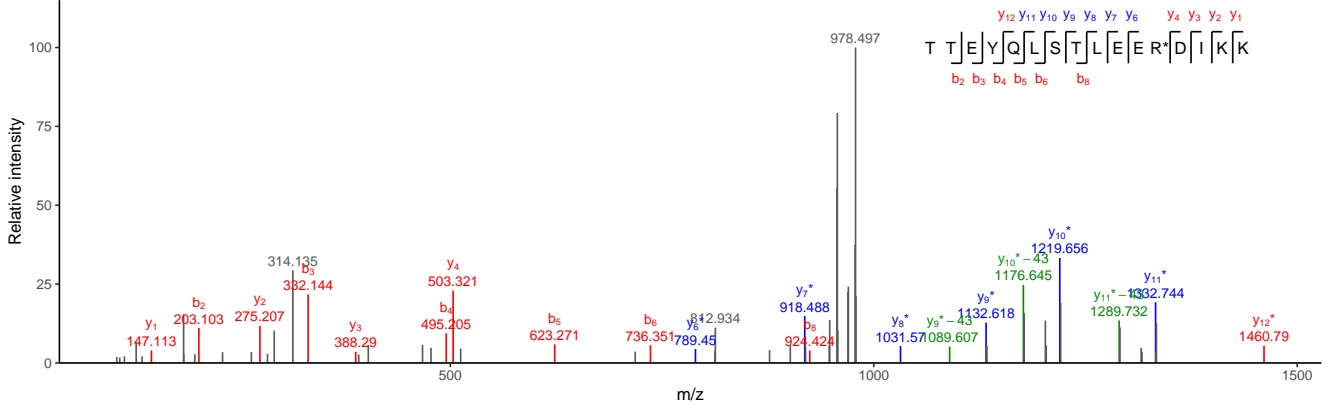
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

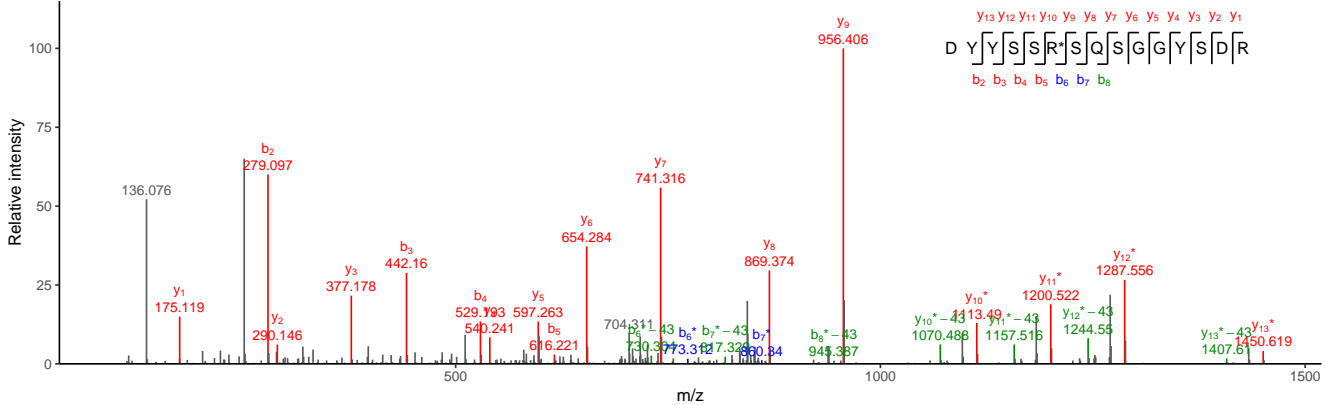
file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 32606



file: 02207a_BB1-TUM_mod_citrullination_l_13_01_01-DDA-1h-R1, scan: 30672



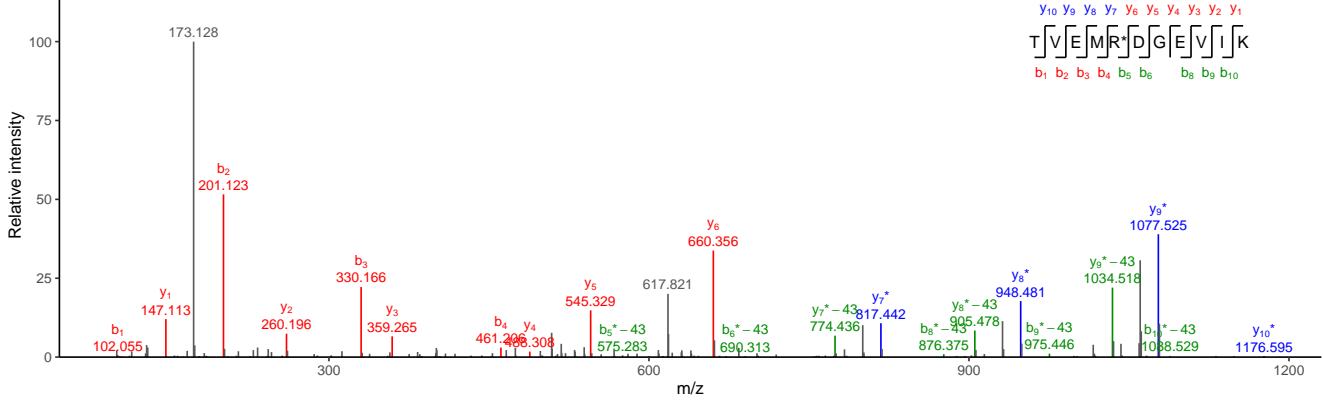
file: 02207a_BB1-TUM_mod_citrullination_l_13_01_01-DDA-1h-R1, scan: 17335



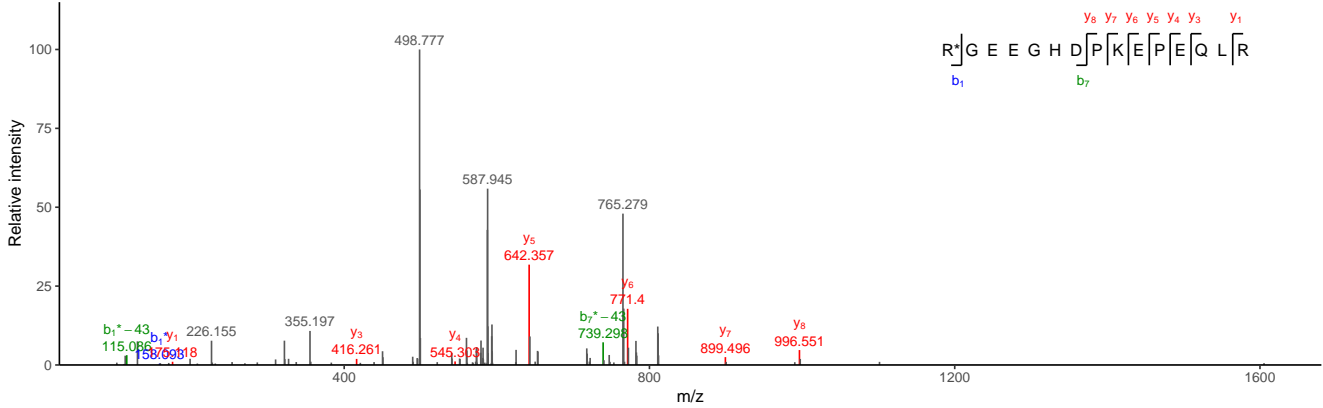
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

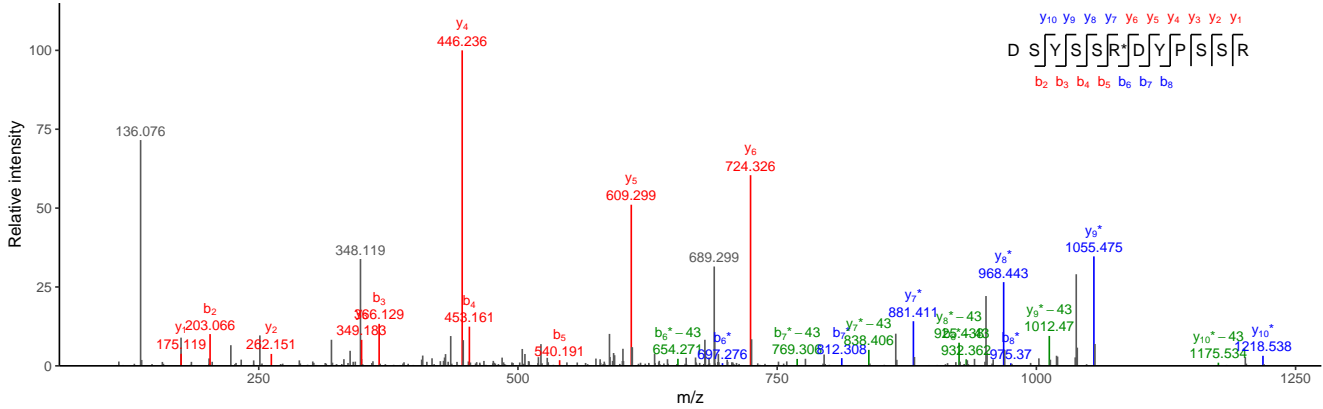
file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 24613



file: 02207a_BB2-TUM_mod_citrullination_l_14_01_01-DDA-1h-R1, scan: 11178



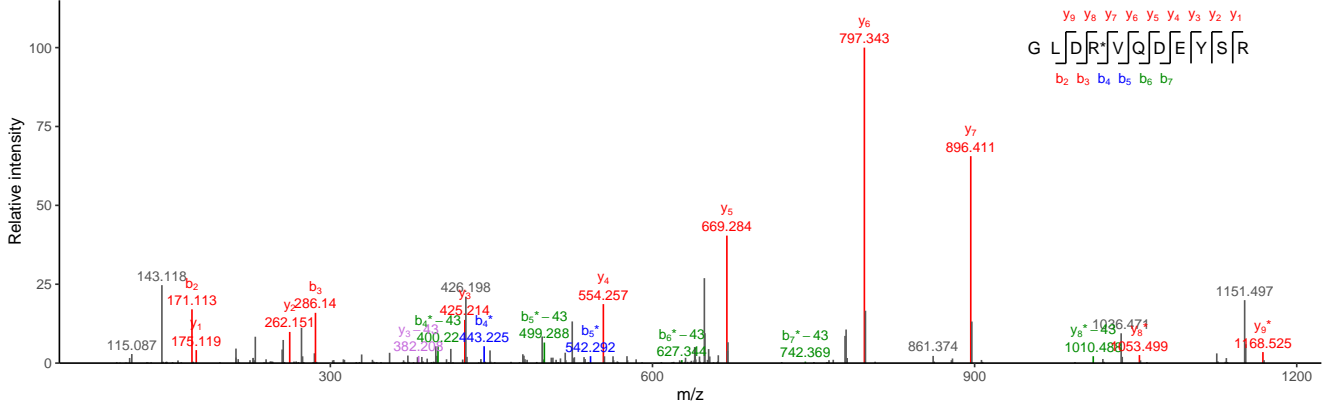
file: 02207a_BA6-TUM_mod_citrullination_l_6_01_01-DDA-1h-R1, scan: 14852



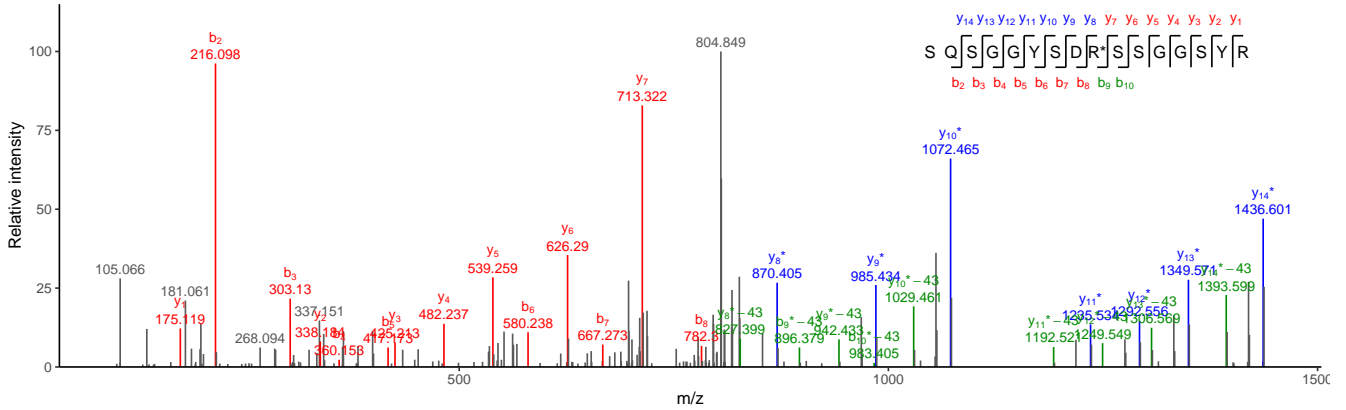
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

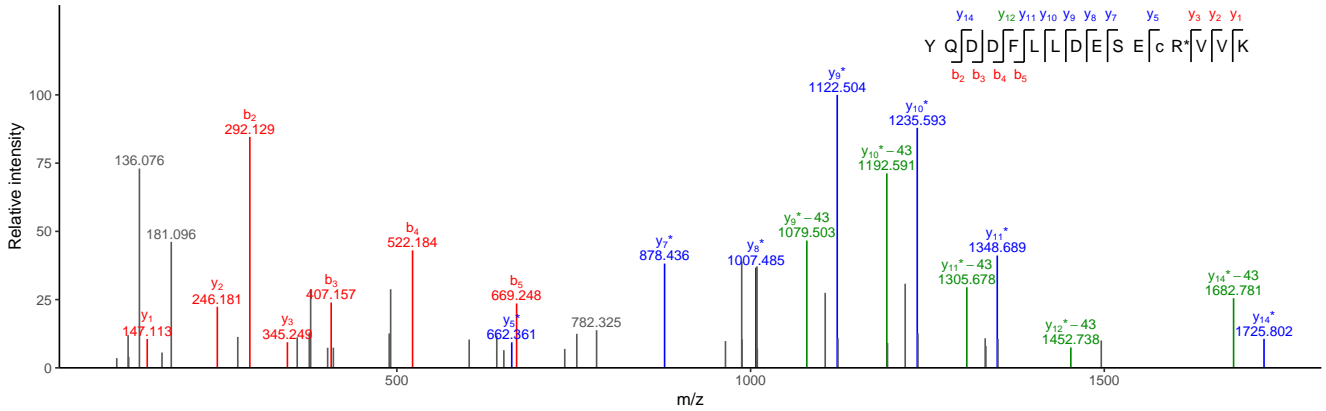
file: 02207a_BA4-TUM_mod_citrullination_L_4_01_01-DDA-1h-R1, scan: 22441



file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 10179



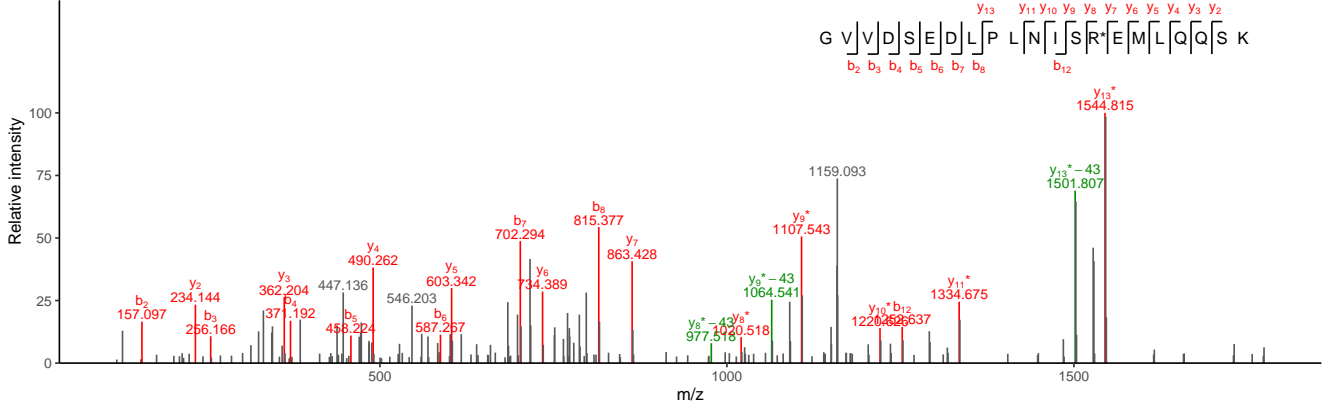
file: 02207a_BB2-TUM_mod_citrullination_L_14_01_01-DDA-1h-R1, scan: 41854



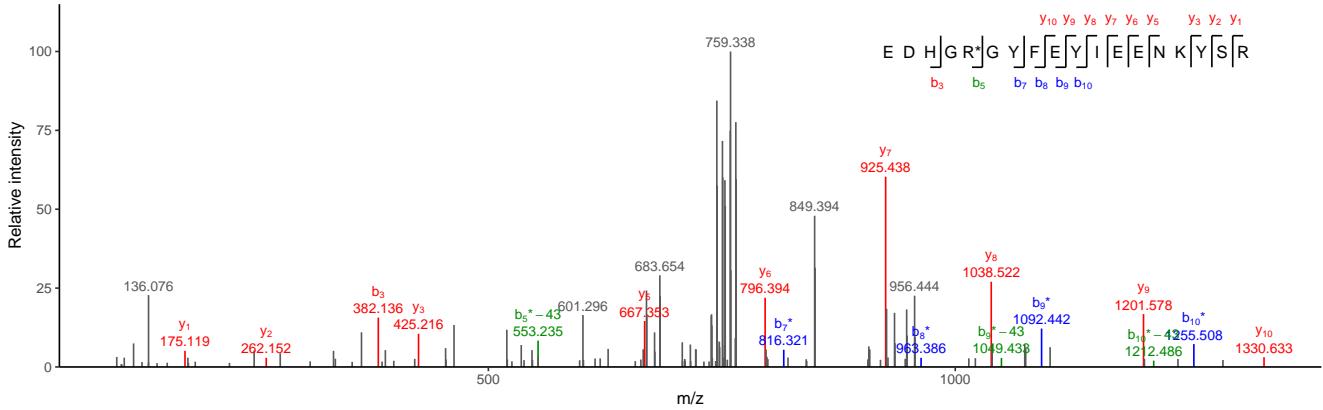
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

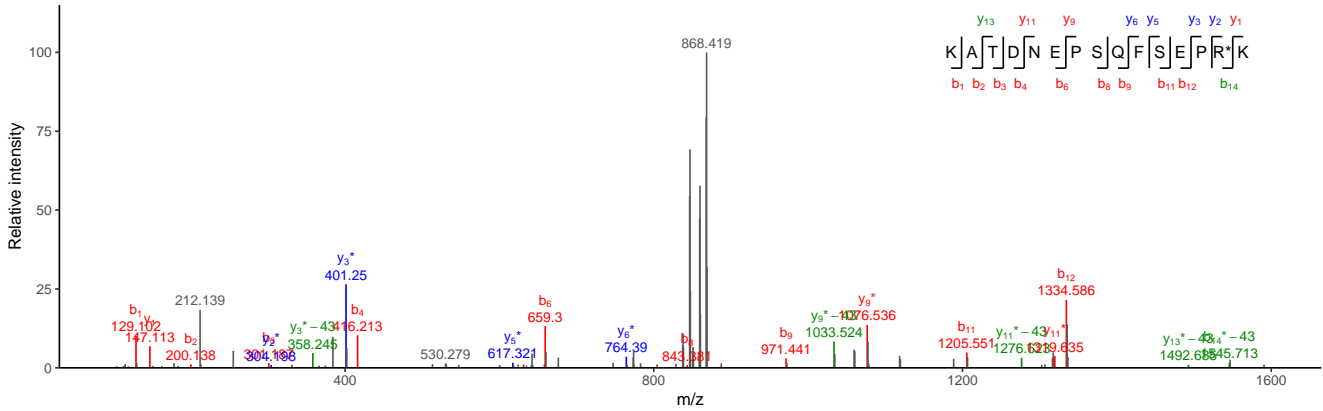
file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 51608



file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 32954

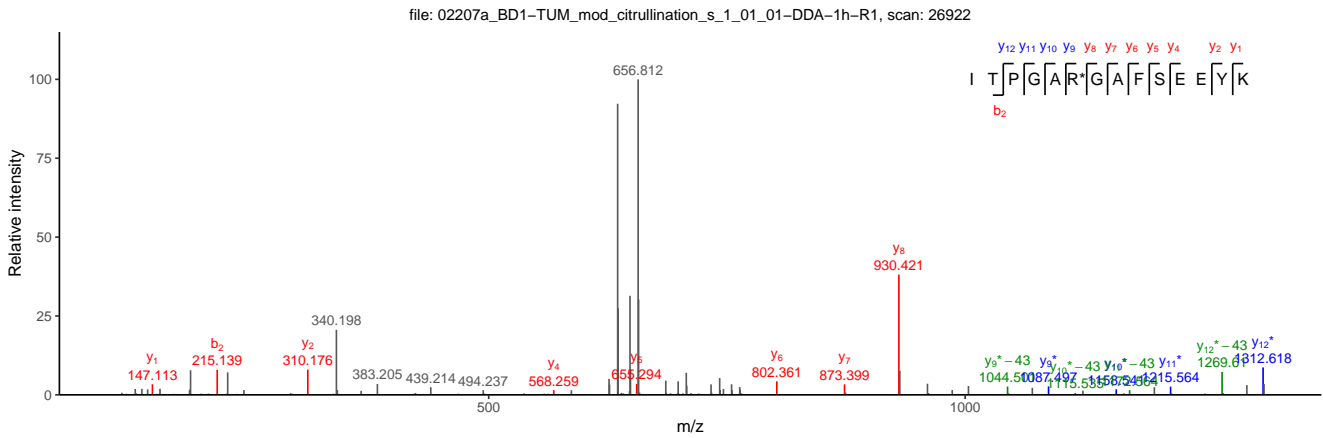
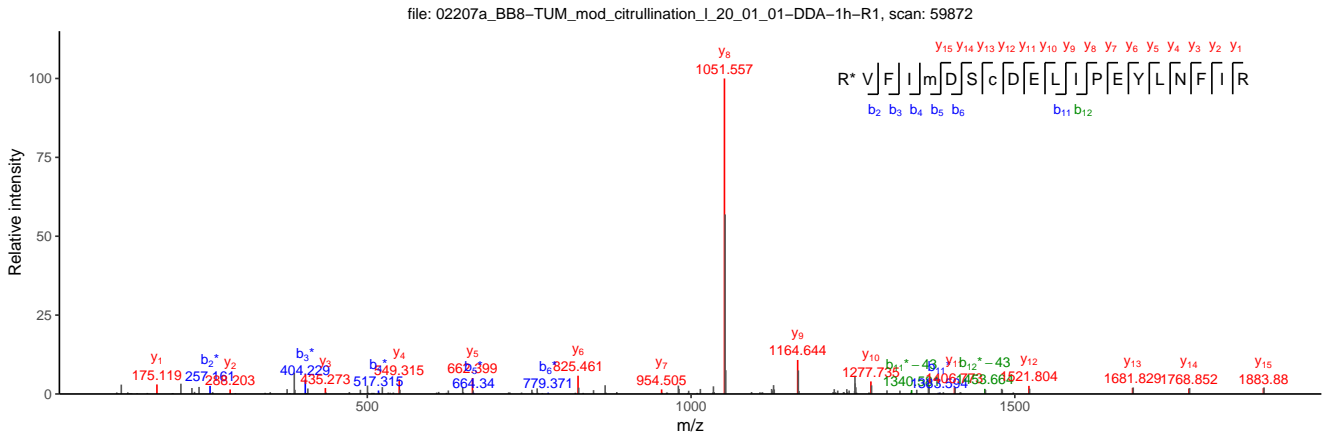
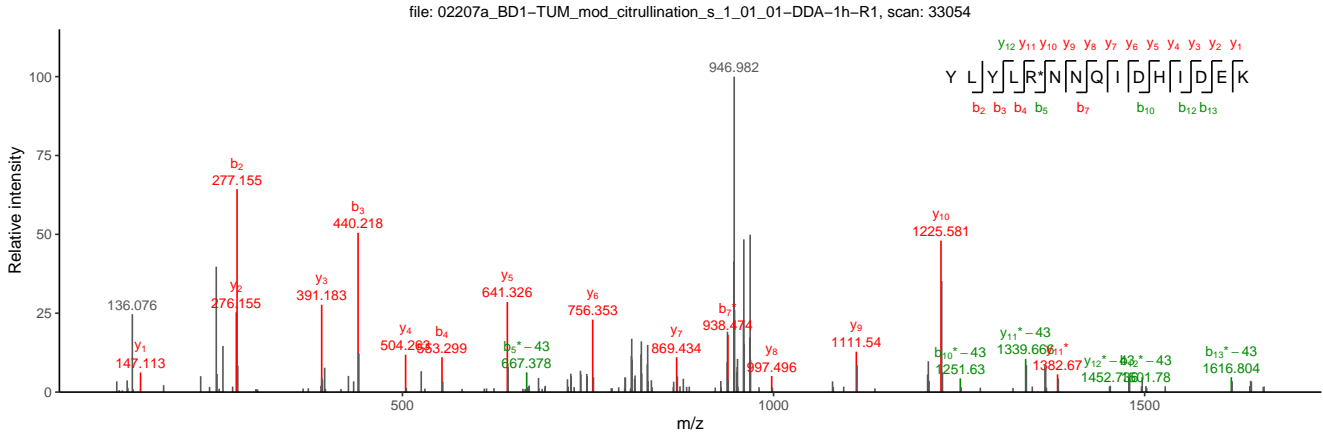


file: 02207a_BB1-TUM_mod_citrullination_l_13_01_01-DDA-1h-R1, scan: 14689



Ground truth: true, ionFinder classification: true

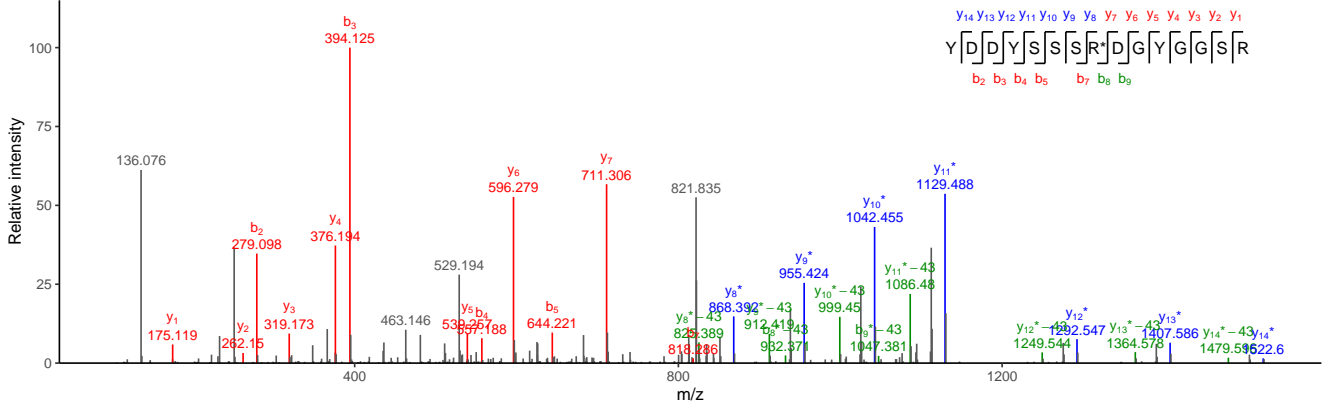
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



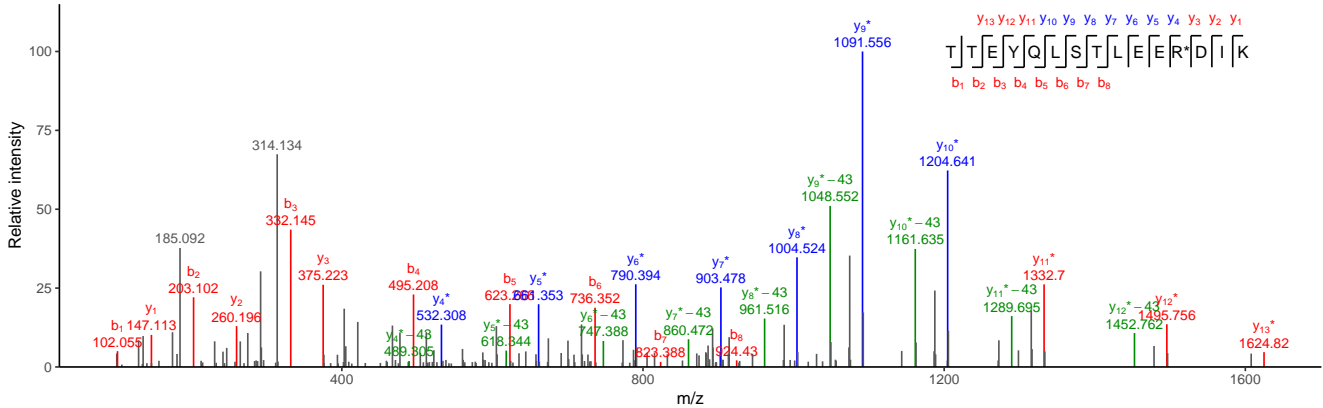
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

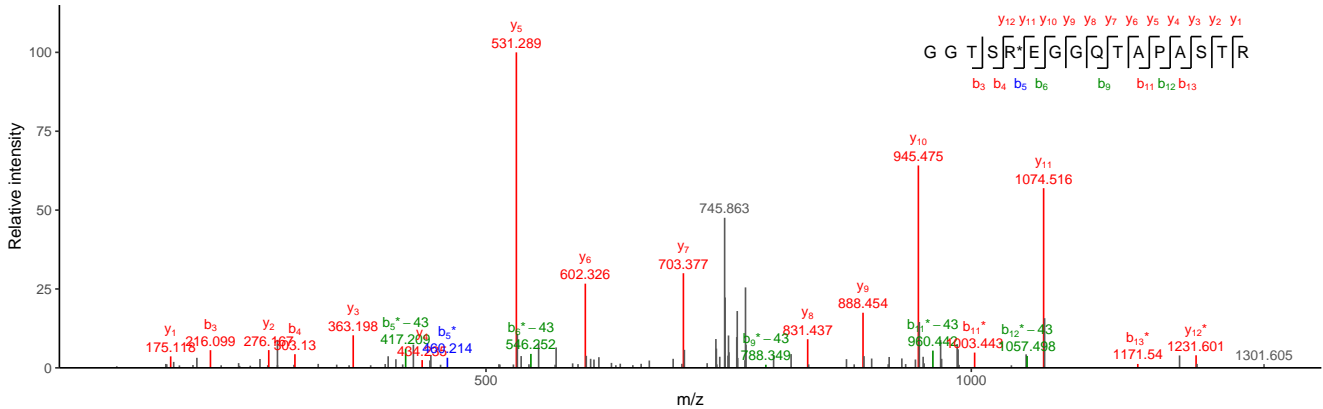
file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 16409



file: 02207a_BA11-TUM_mod_citrullination_l_11_01_01-DDA-1h-R1, scan: 35499



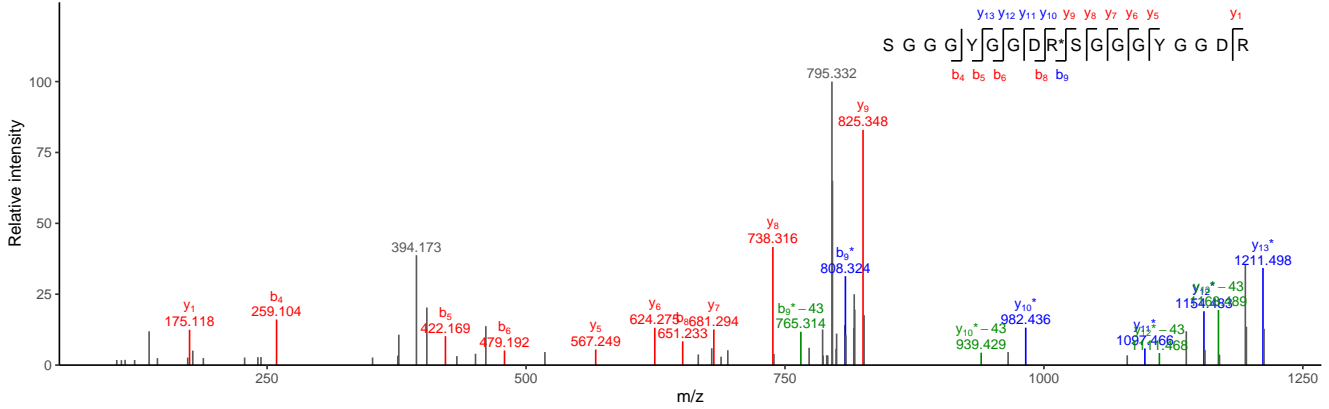
file: 02207a_BA12-TUM_mod_citrullination_l_12_01_01-DDA-1h-R1, scan: 5823



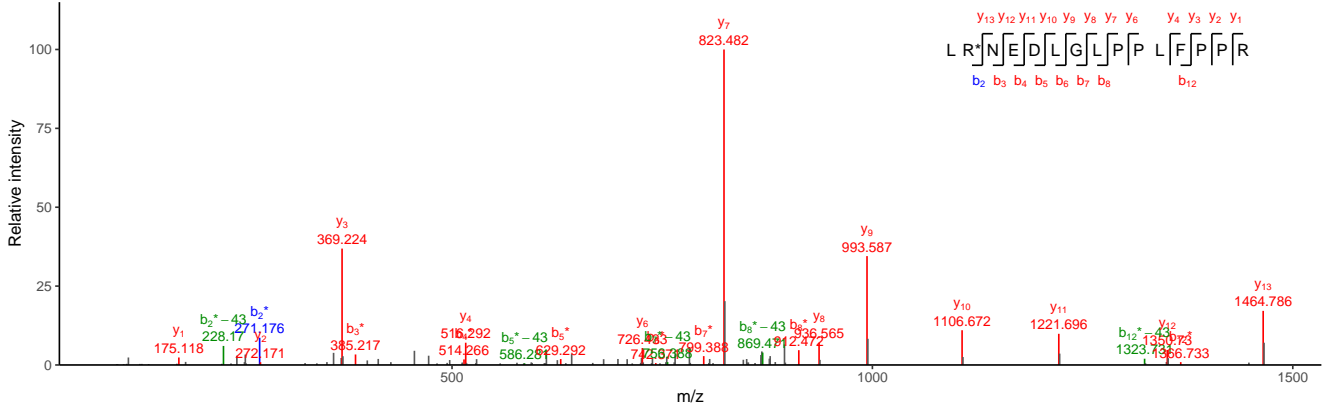
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

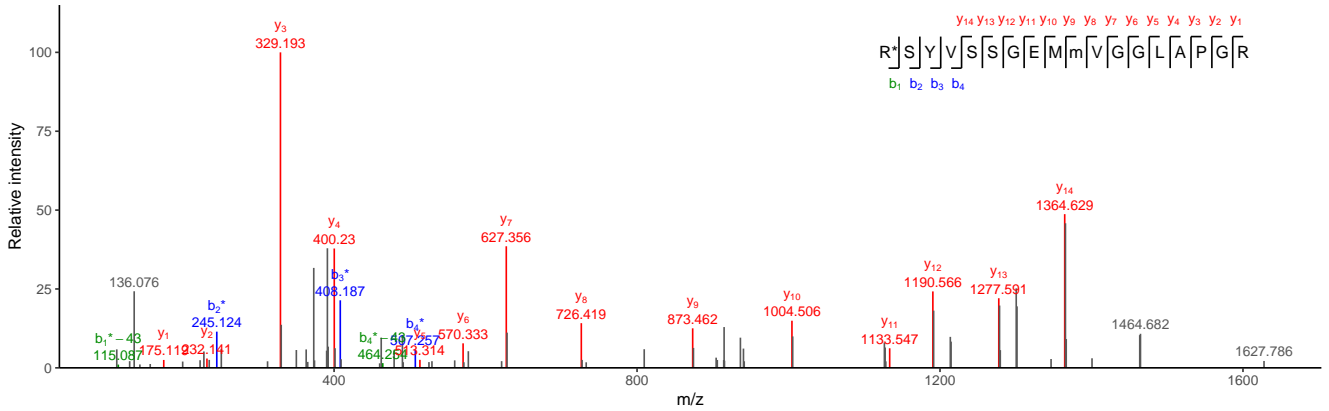
file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 12449



file: 02207a_BA11-TUM_mod_citrullination_L_11_01_01-DDA-1h-R1, scan: 52932



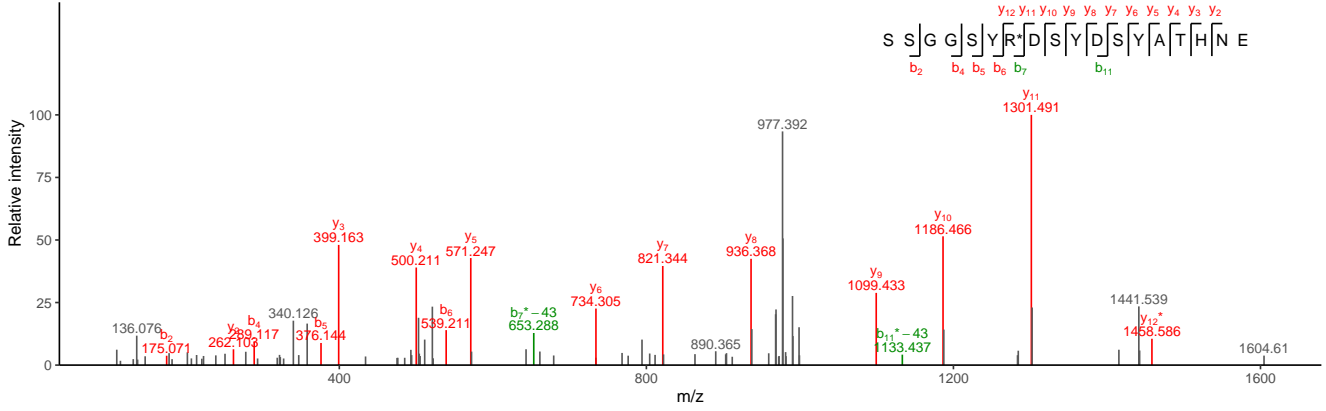
file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 33252



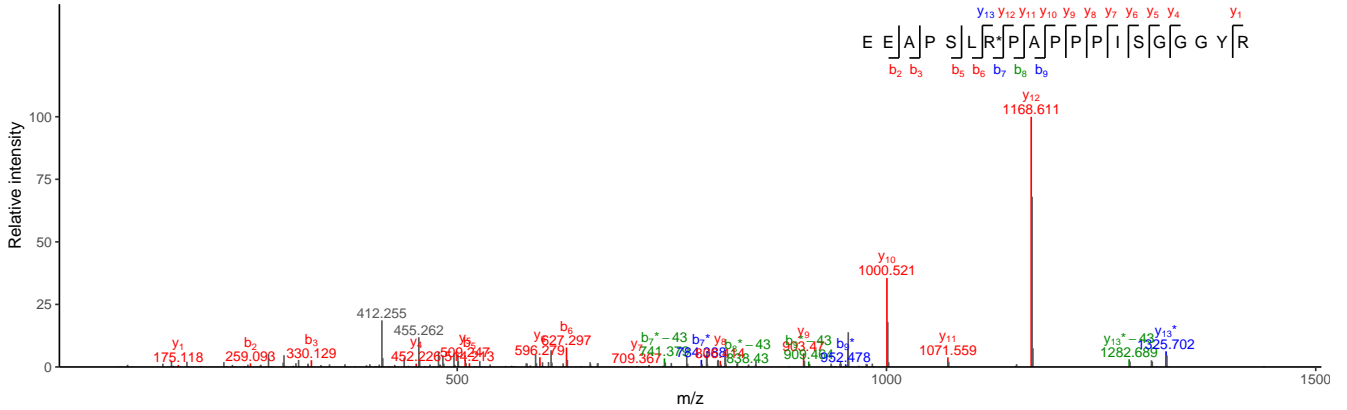
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

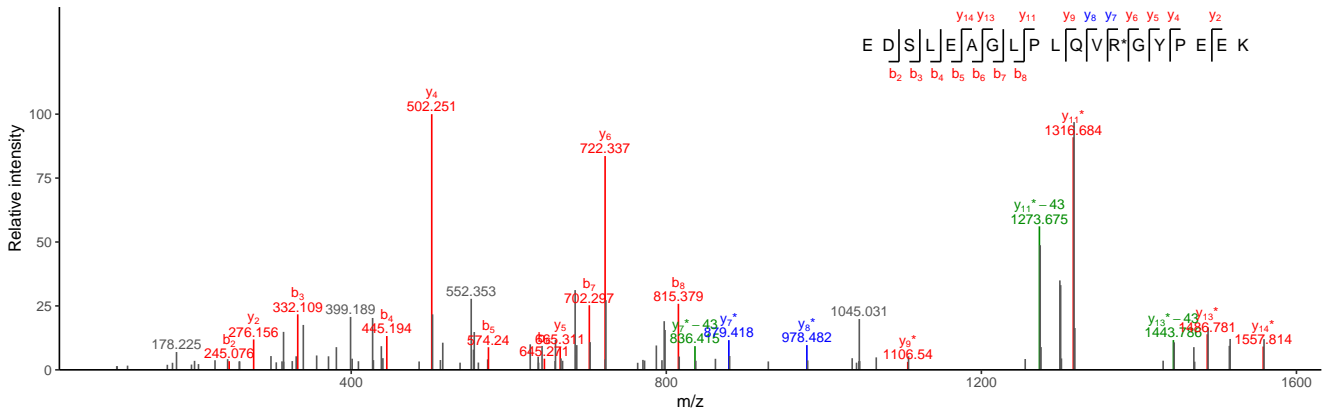
file: 02207a_BB10-TUM_mod_citrullination_L_22_01_01-DDA-1h-R1, scan: 21466



file: 02207a_BB6-TUM_mod_citrullination_L_18_01_01-DDA-1h-R1, scan: 33391



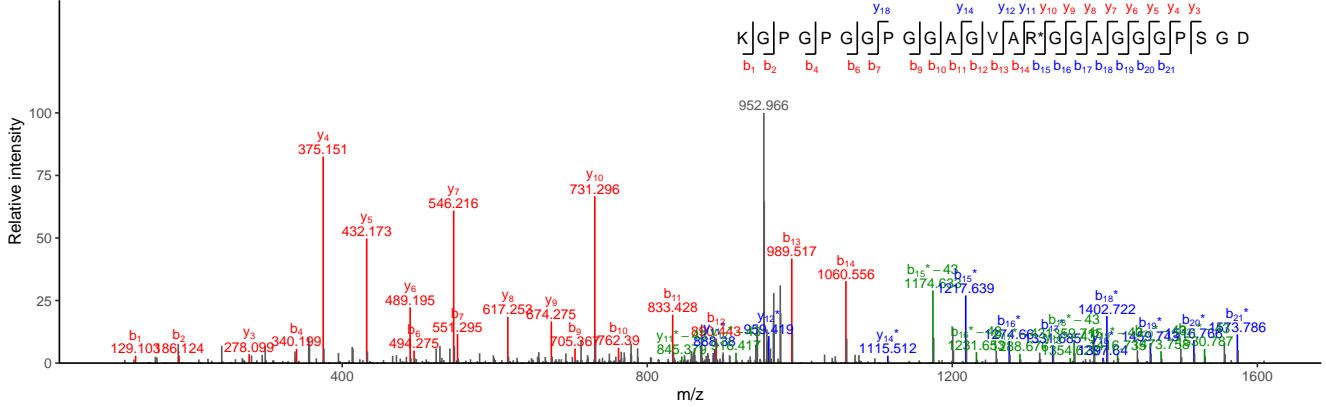
file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 43378



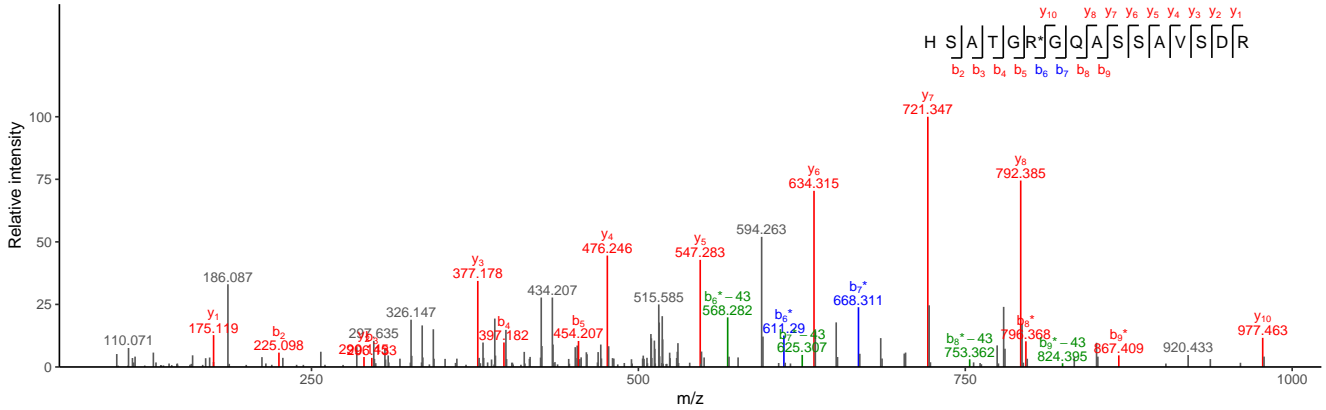
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

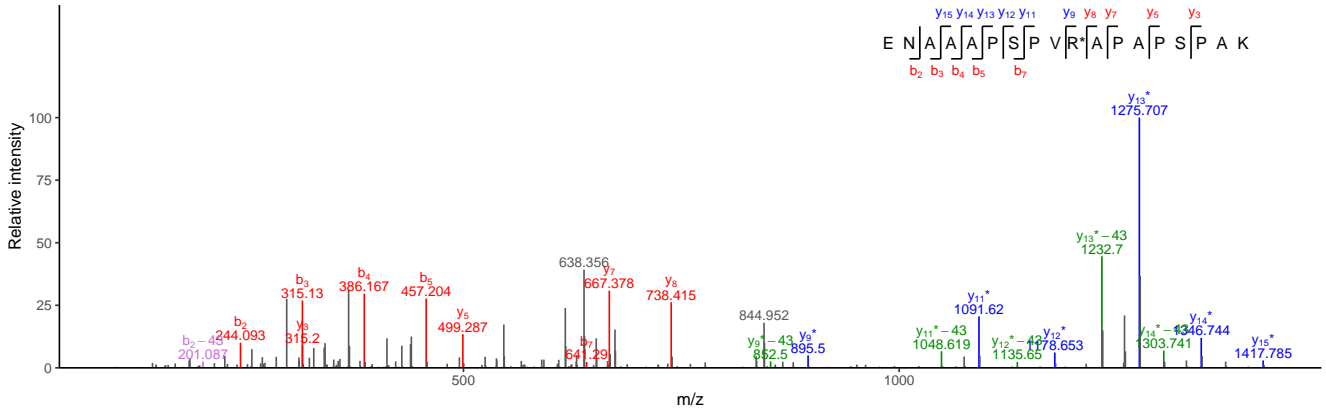
file: 02207a_BB10-TUM_mod_citrullination_I_22_01_01-DDA-1h-R1, scan: 13576



file: 02207a_BA12-TUM_mod_citrullination_I_12_01_01-DDA-1h-R1, scan: 6812



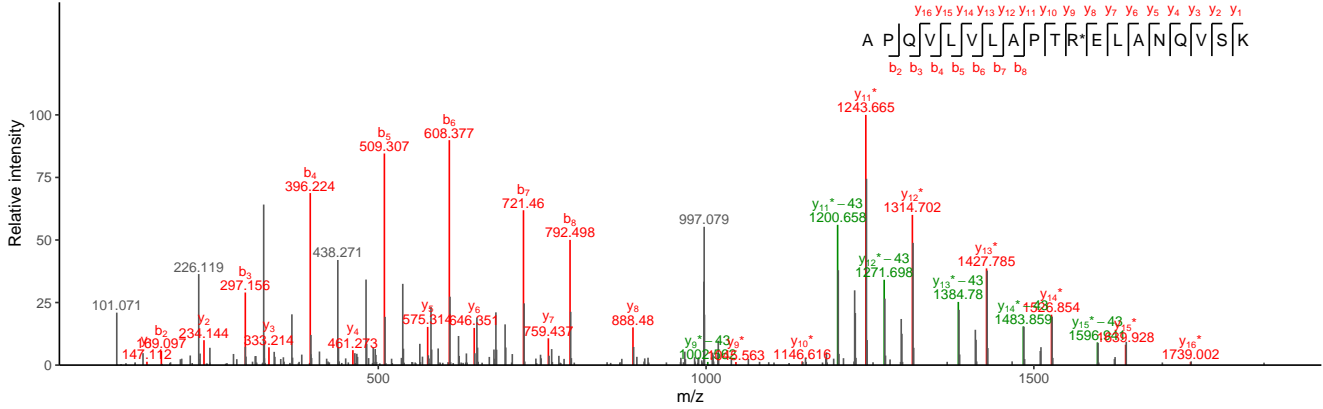
file: 02207a_BB8-TUM_mod_citrullination_I_20_01_01-DDA-1h-R1, scan: 21552



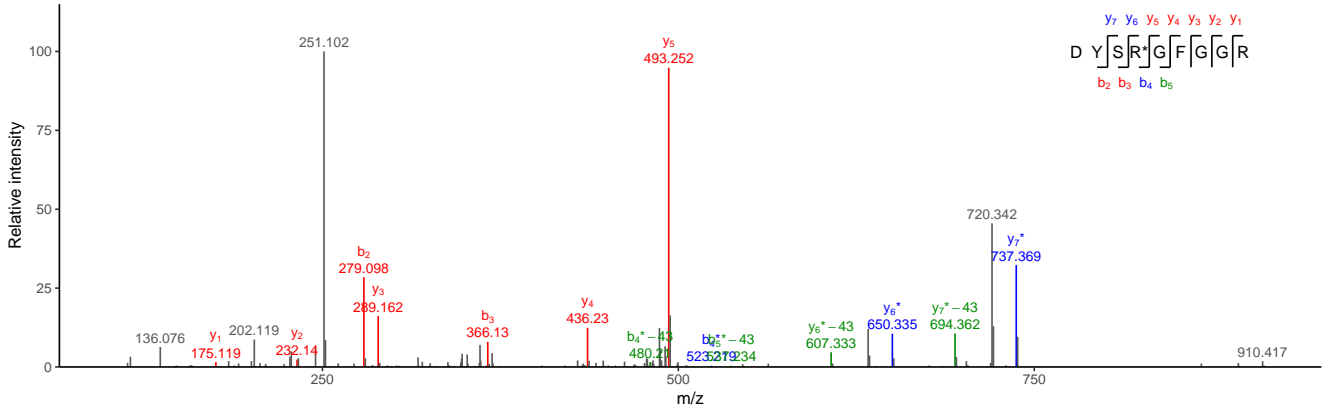
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

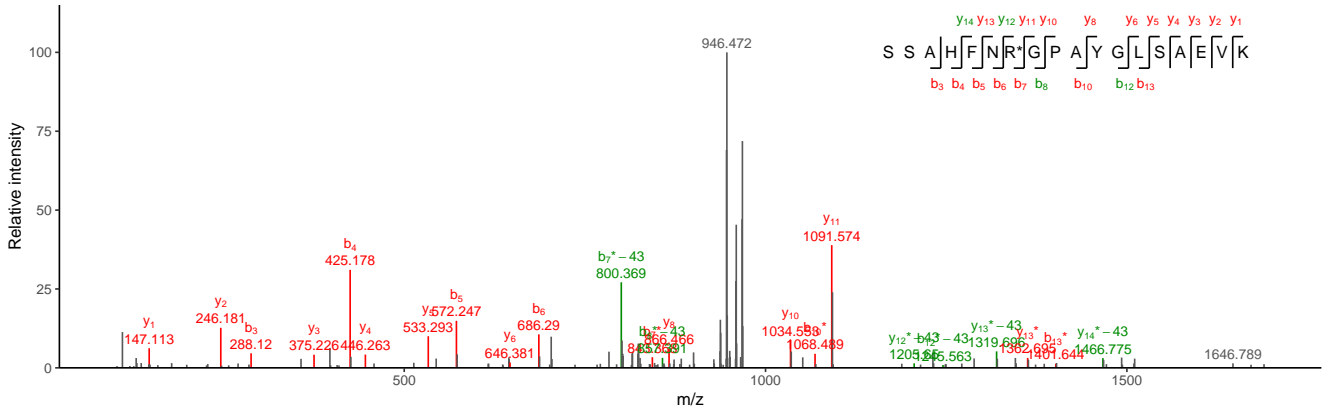
file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 47052



file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 19235

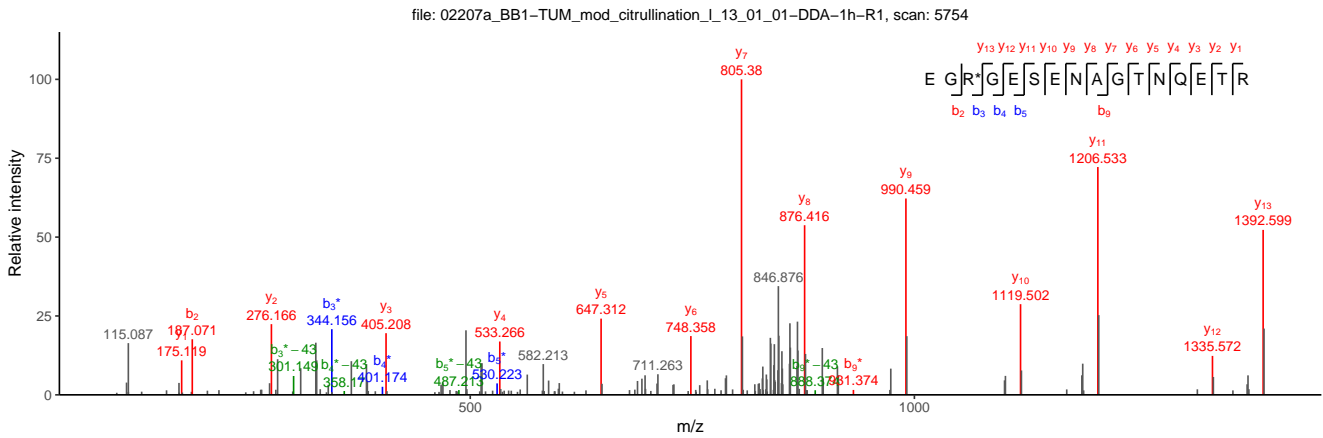
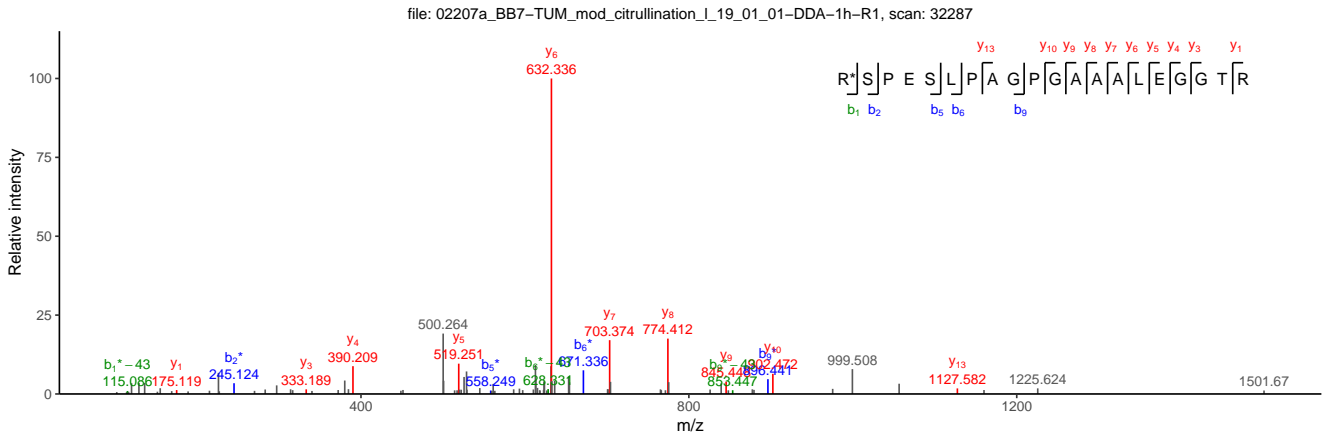
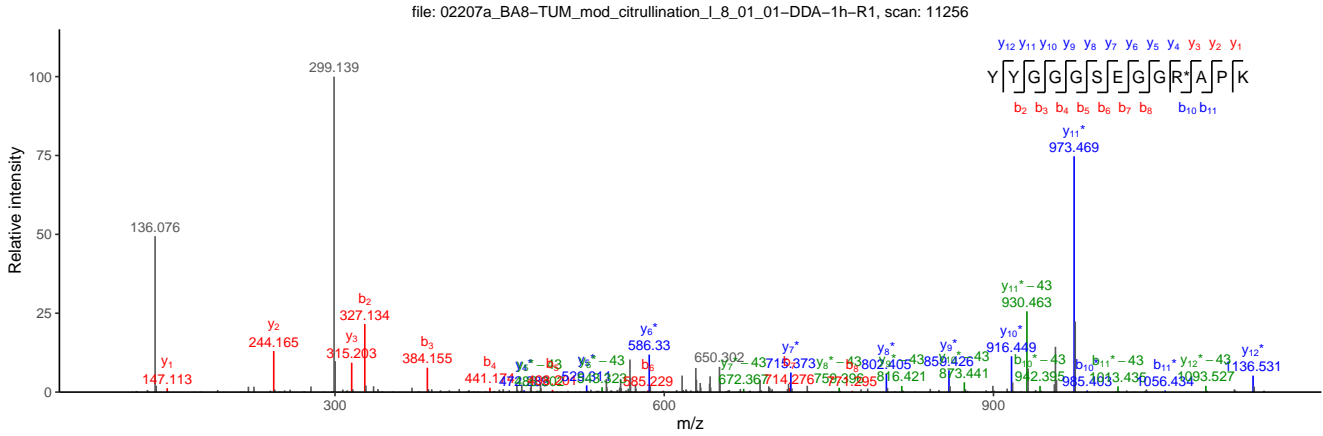


file: 02207a_BB5-TUM_mod_citrullination_L_17_01_01-DDA-1h-R1, scan: 33818



Ground truth: true, ionFinder classification: true

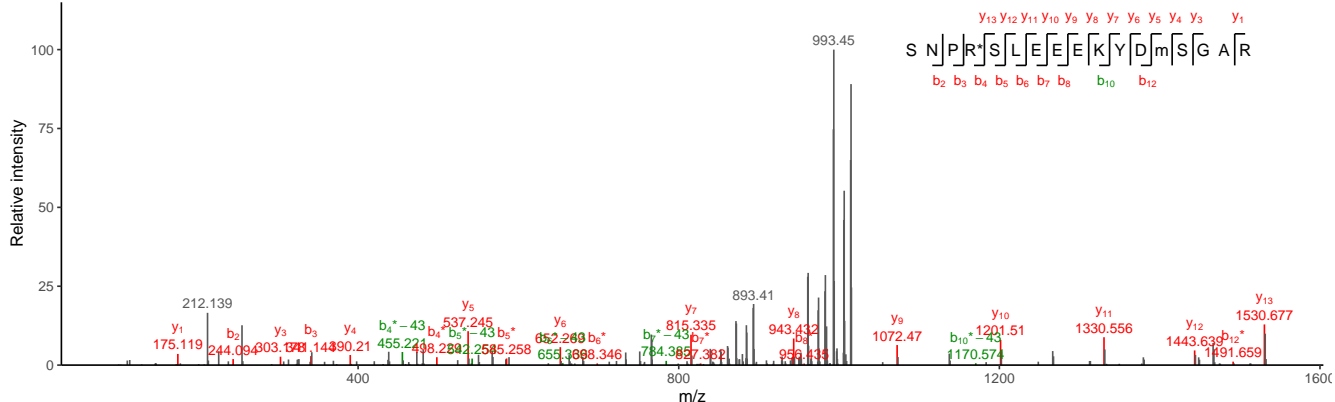
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



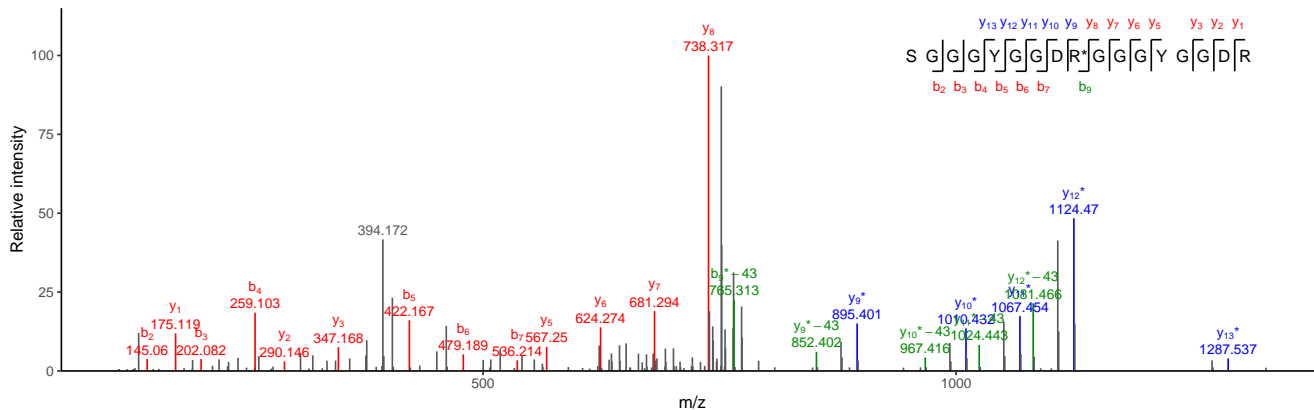
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

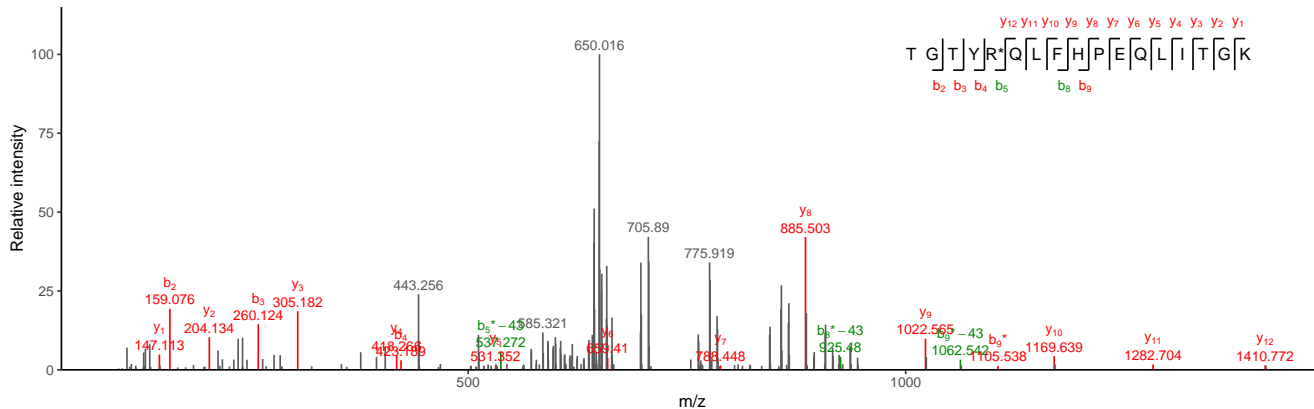
file: 02207a_BB3-TUM_mod_citrullination_I_15_01_01-DDA-1h-R1, scan: 23417



file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 12023



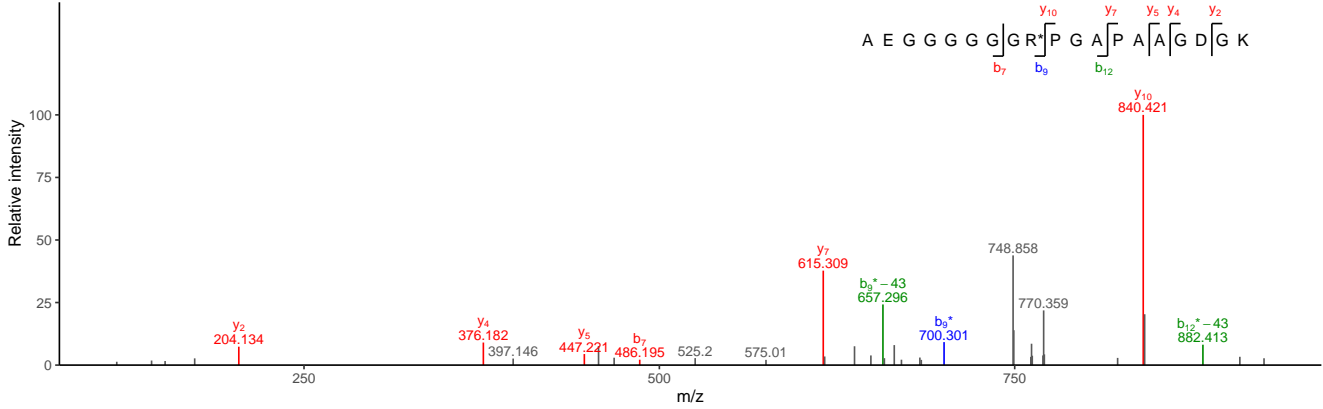
file: 02207a_BB3-TUM_mod_citrullination_I_15_01_01-DDA-1h-R1, scan: 42389



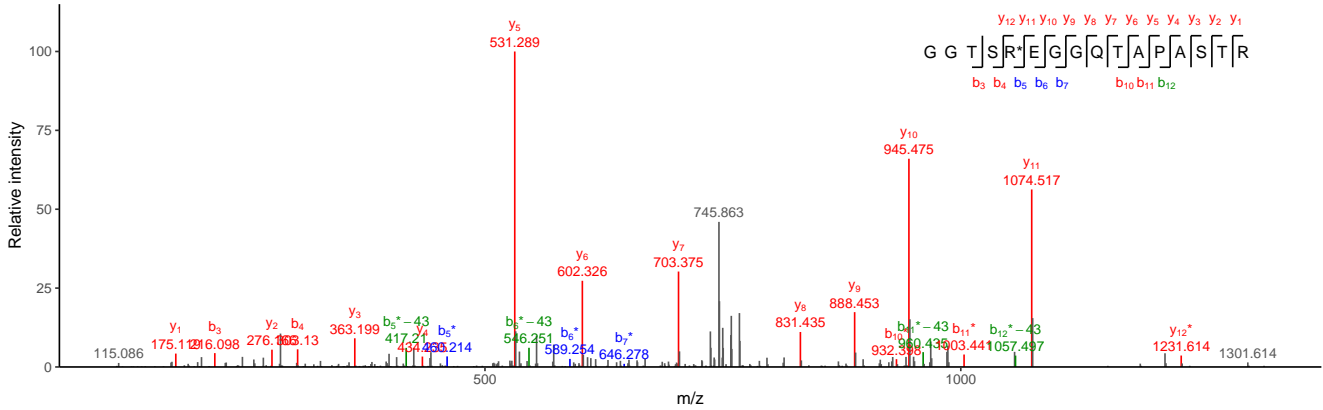
Ground truth: true, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

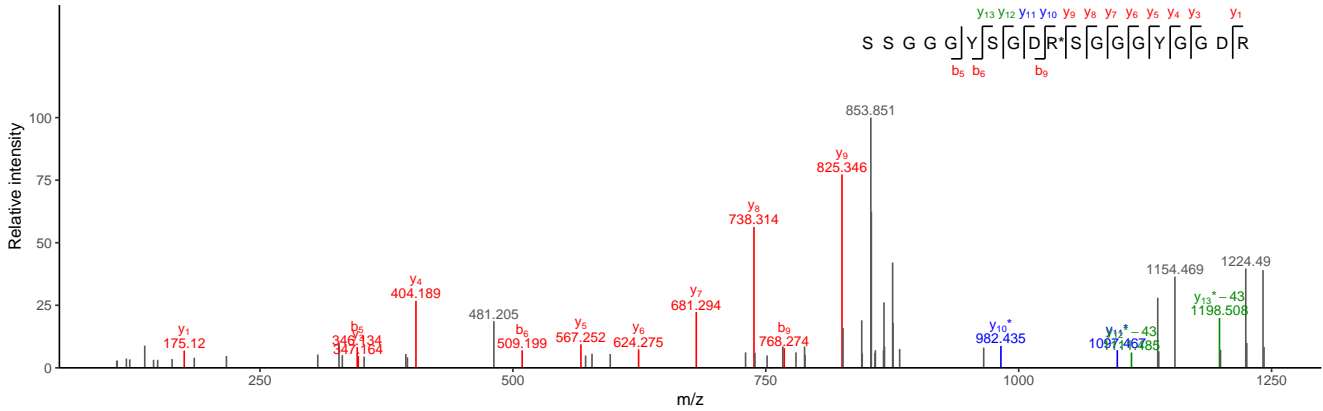
file: 02207a_BB5-TUM_mod_citrullination_L17_01_01-DDA-1h-R1, scan: 7190



file: 02207a_BA12-TUM_mod_citrullination_L12_01_01-DDA-1h-R1, scan: 5517



file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 12143



Ground truth: true, ionFinder classification: true

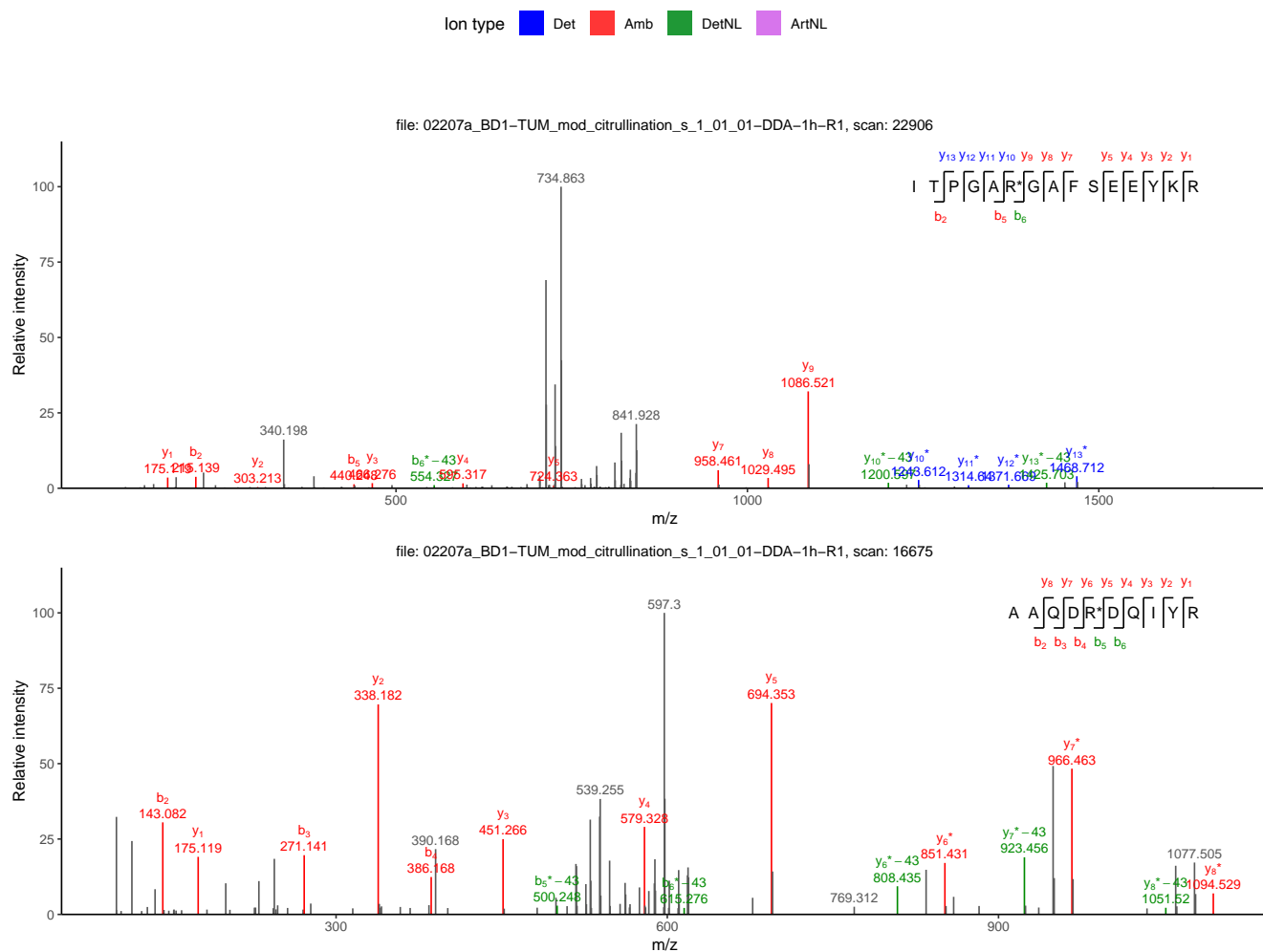
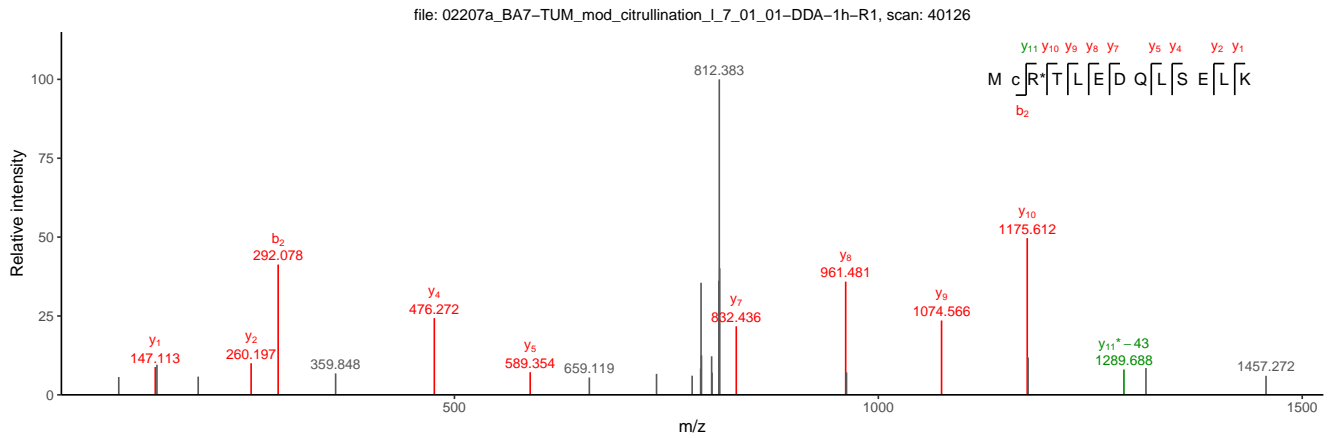
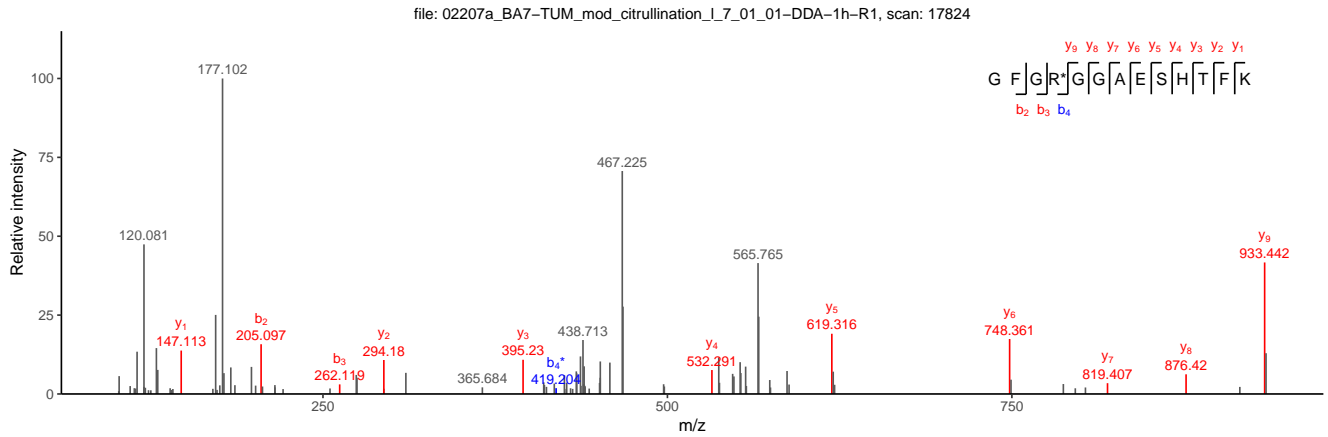
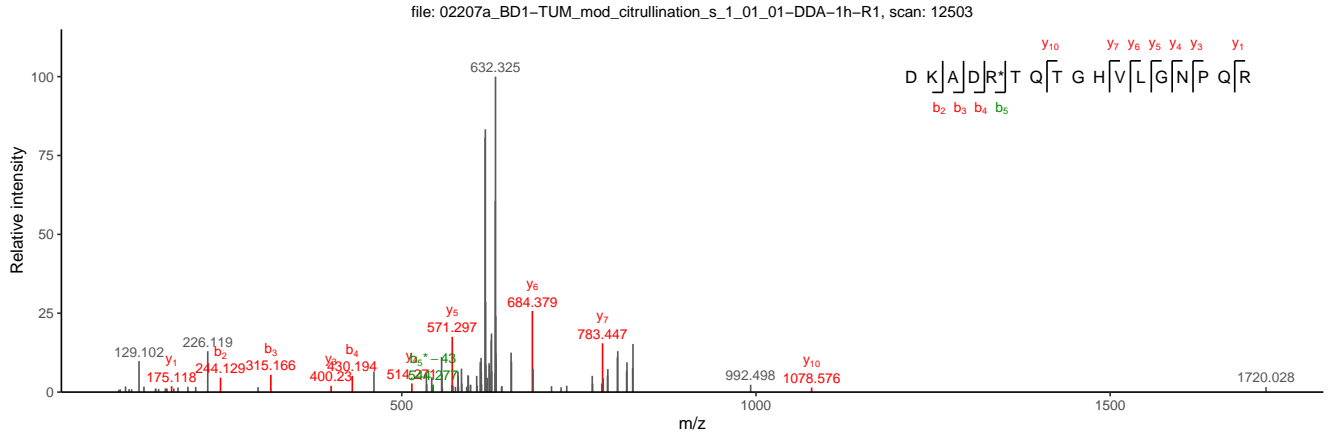


Figure S1. MS² spectra from 77 PSMs of synthetic spectra which were identified as *true* by ionFinder and classified as *true* in the ground truth dataset.

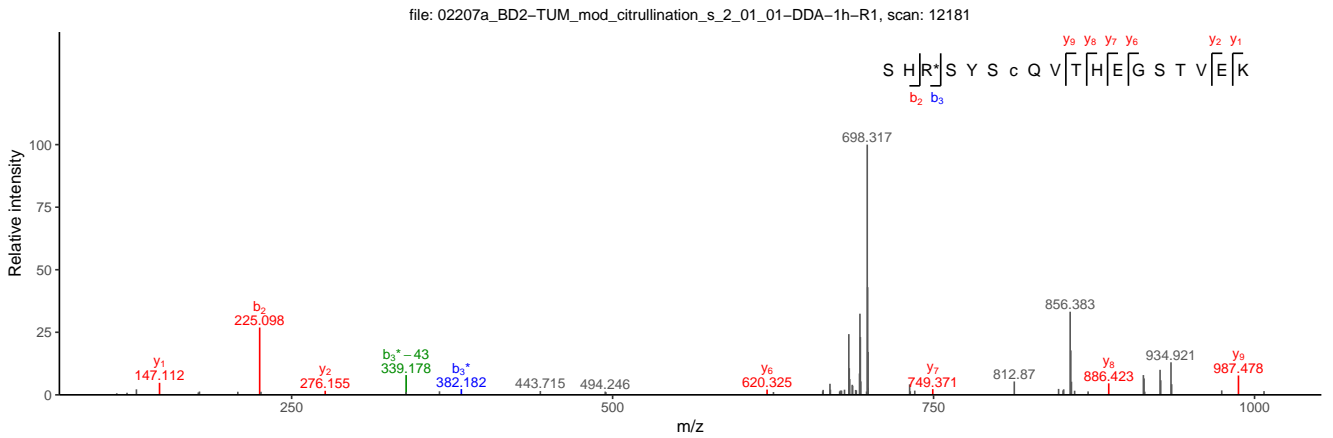
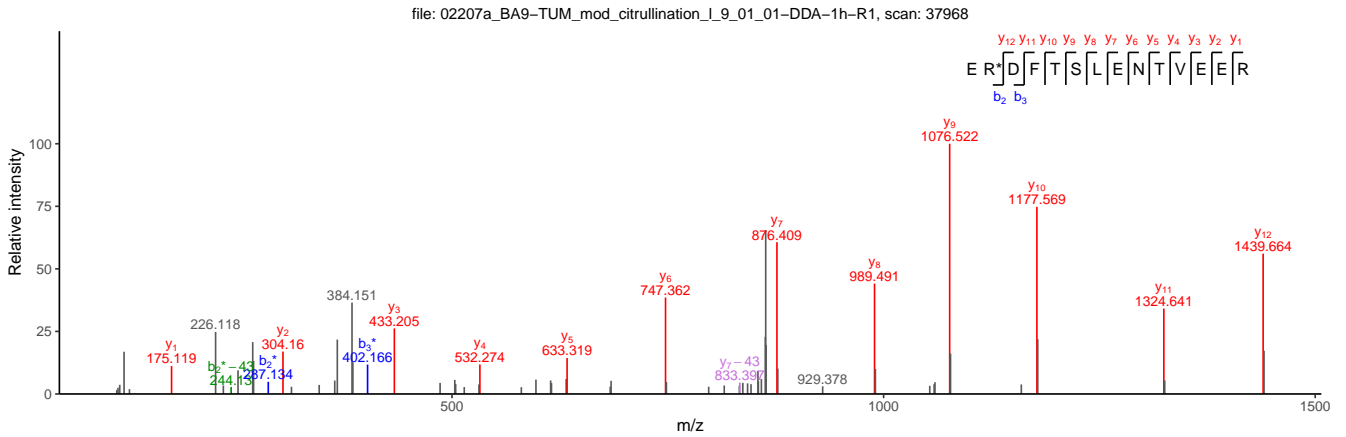
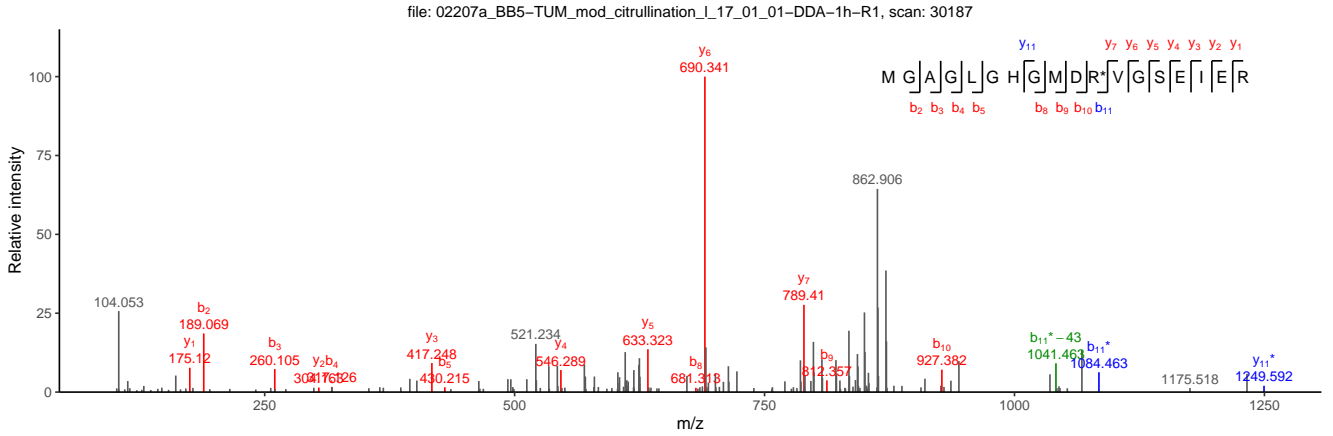
Ground truth: true, ionFinder classification: likely

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



Ground truth: true, ionFinder classification: likely

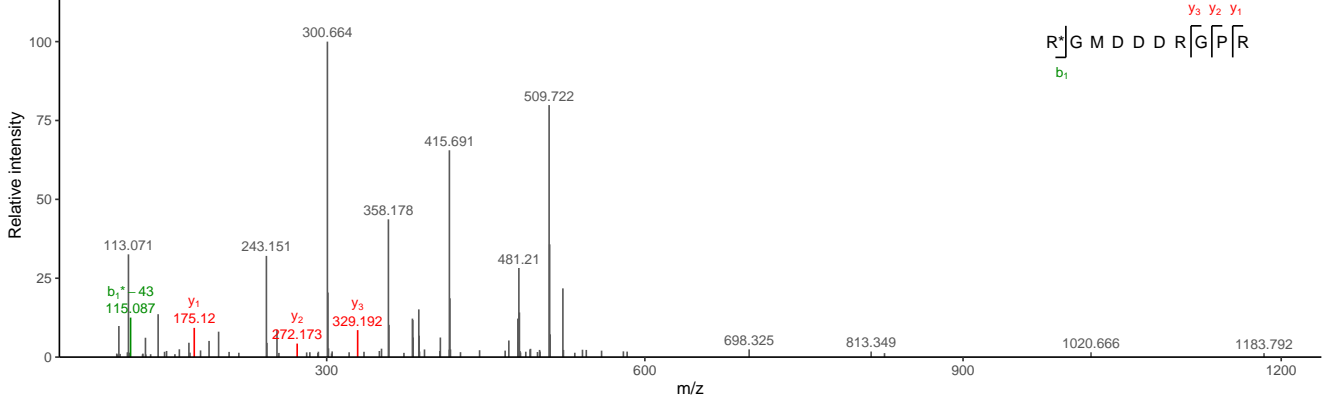
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



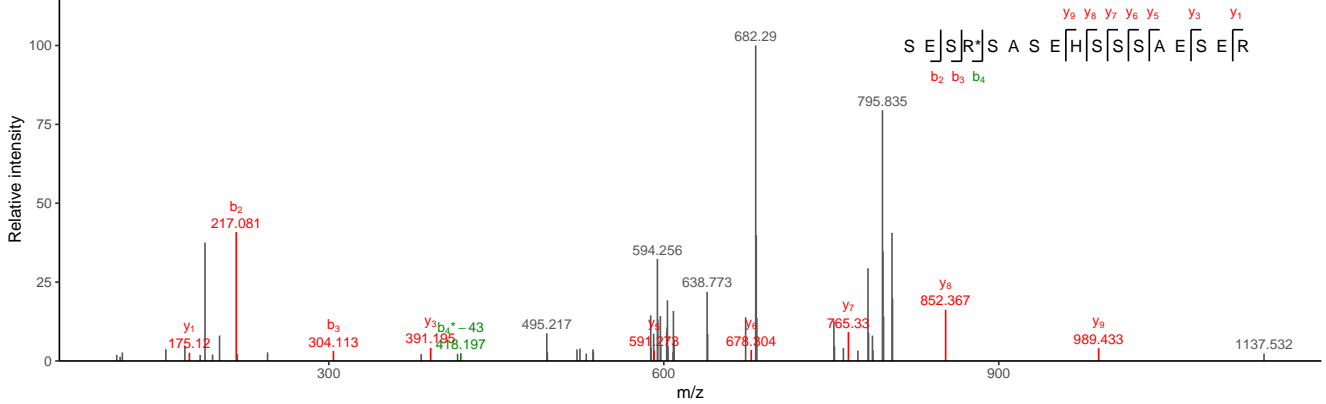
Ground truth: true, ionFinder classification: likely

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

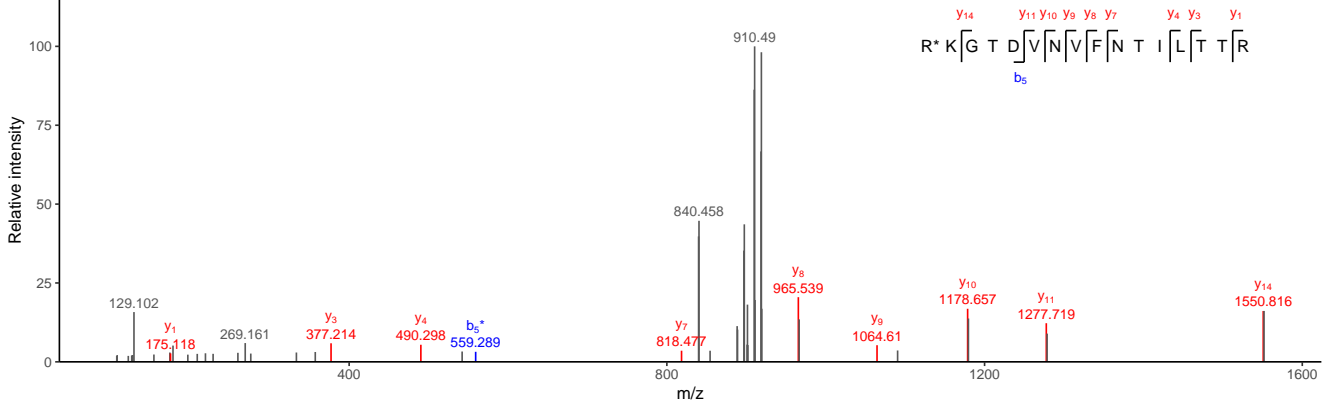
file: 02207a_BA2-TUM_mod_citrullination_I_2_01_01-DDA-1h-R1, scan: 6275



file: 02207a_BB3-TUM_mod_citrullination_I_15_01_01-DDA-1h-R1, scan: 3948

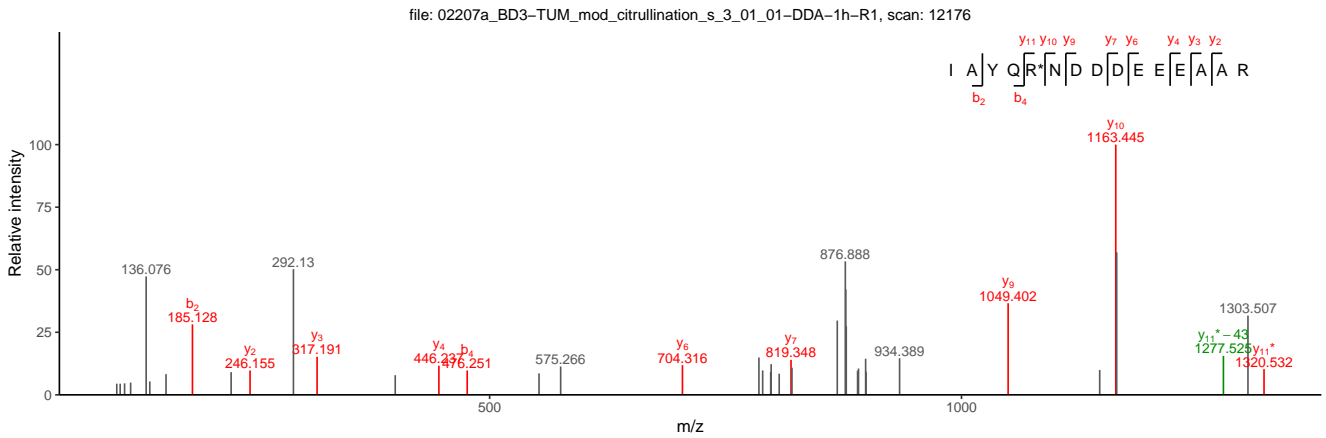
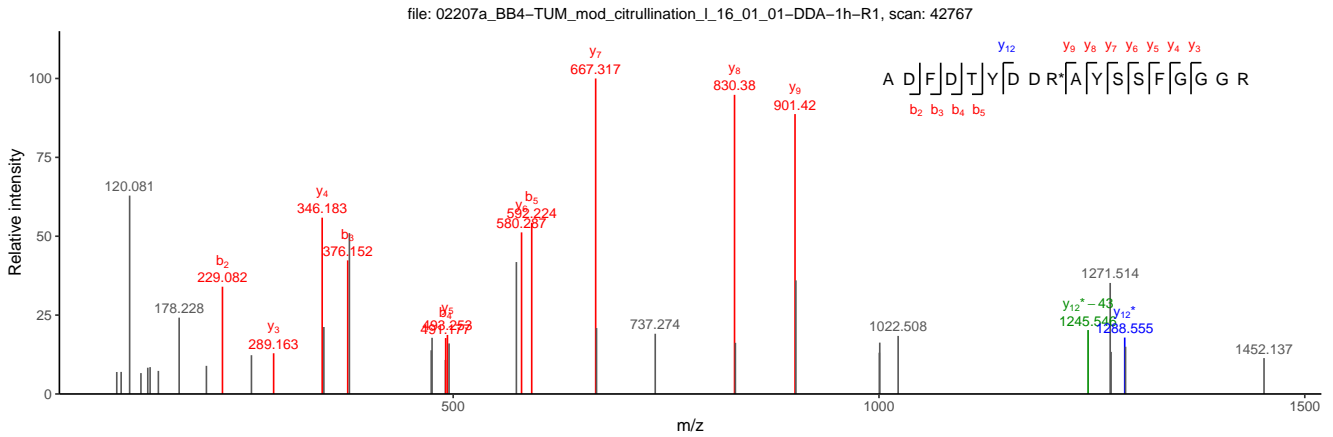
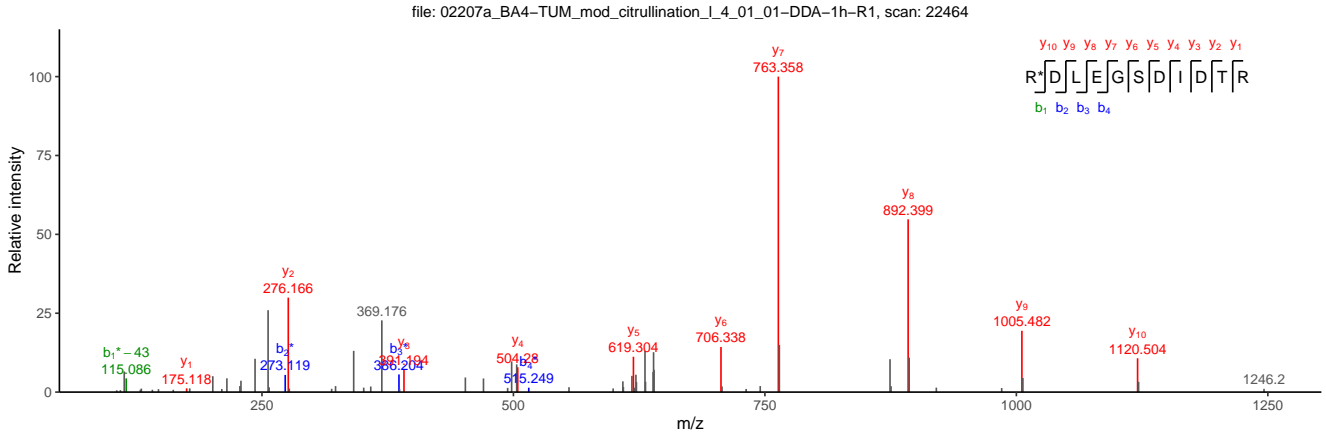


file: 02207a_BB1-TUM_mod_citrullination_I_13_01_01-DDA-1h-R1, scan: 43883



Ground truth: true, ionFinder classification: likely

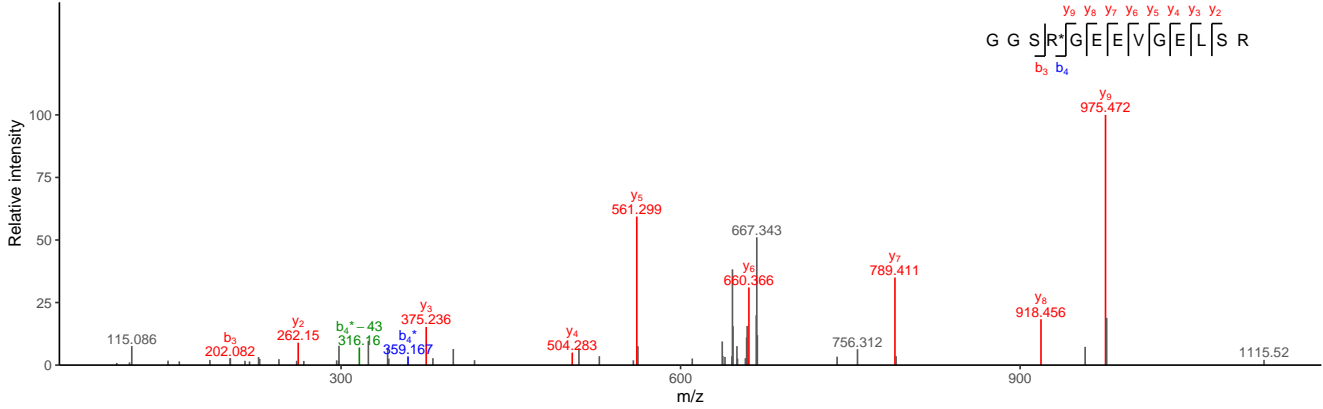
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



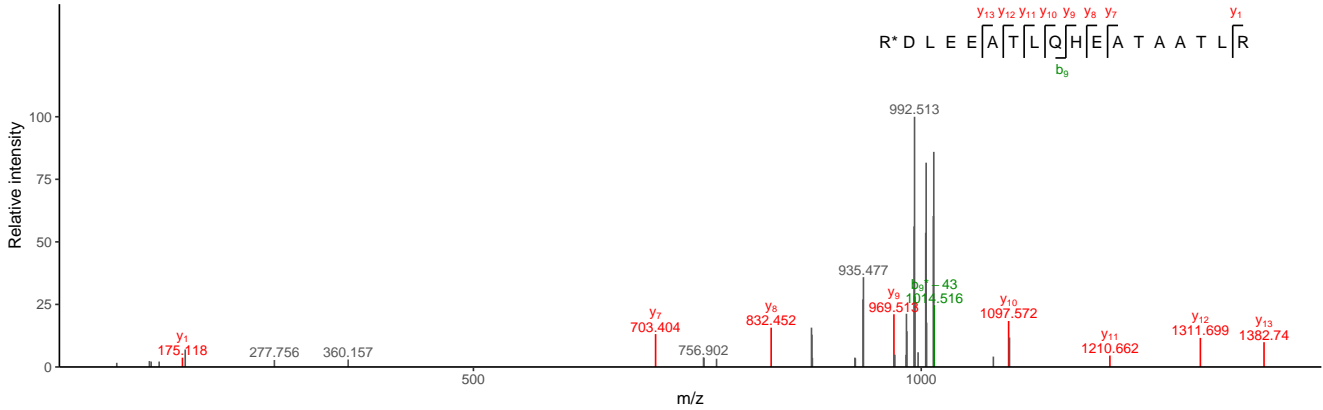
Ground truth: true, ionFinder classification: likely

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

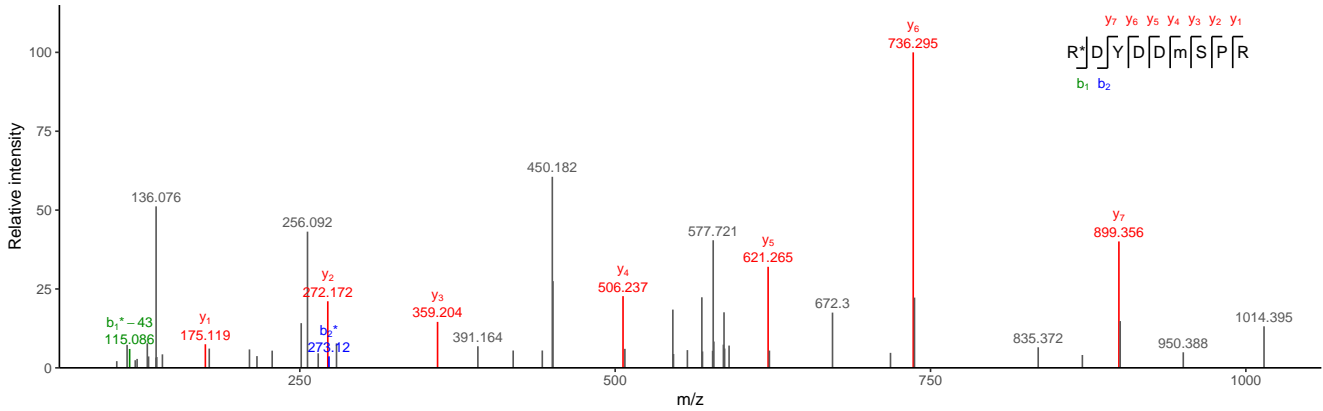
file: 02207a_BA7-TUM_mod_citrullination_L7_01_01-DDA-1h-R1, scan: 20171



file: 02207a_BB5-TUM_mod_citrullination_L17_01_01-DDA-1h-R1, scan: 33016



file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 8427



Ground truth: true, ionFinder classification: likely

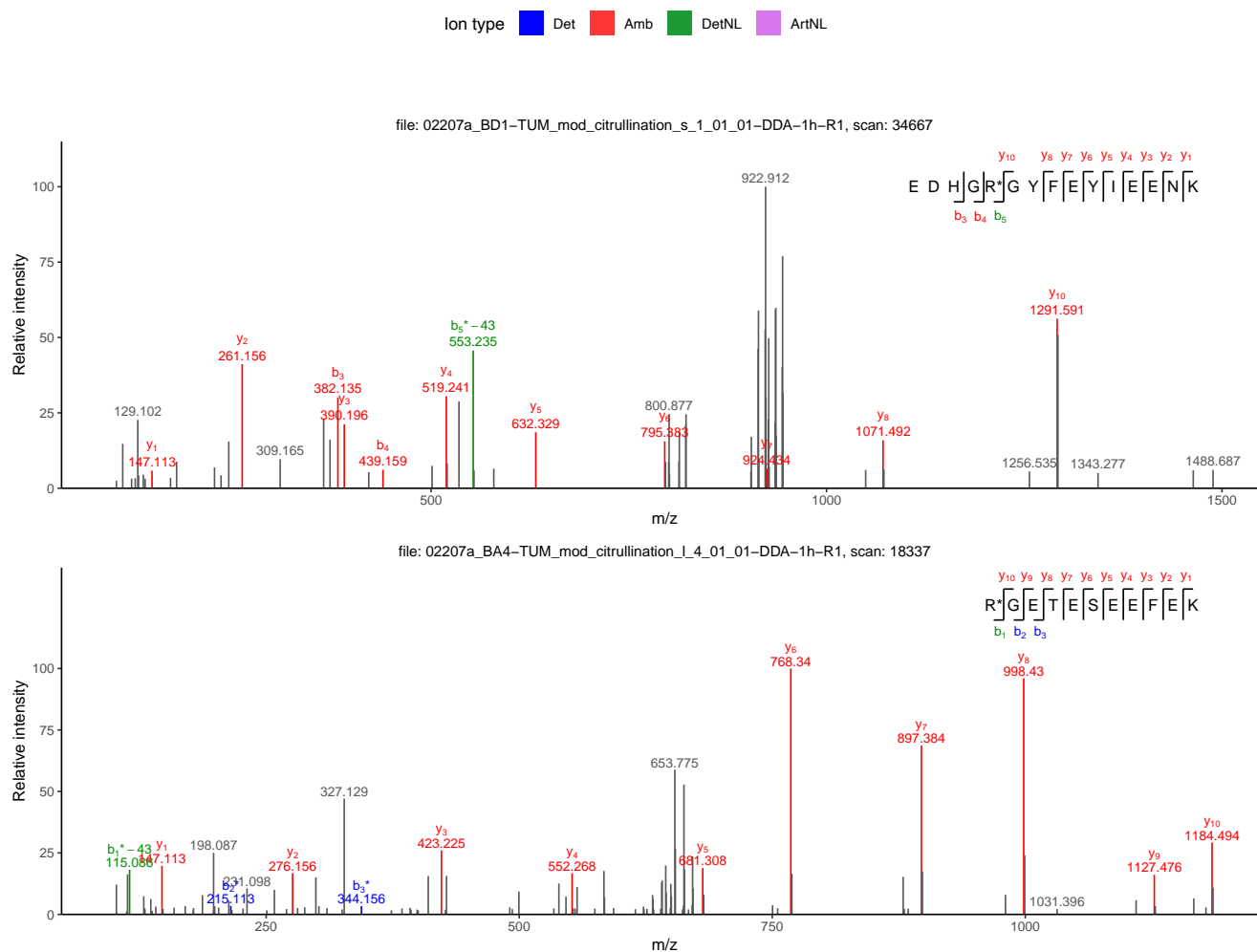
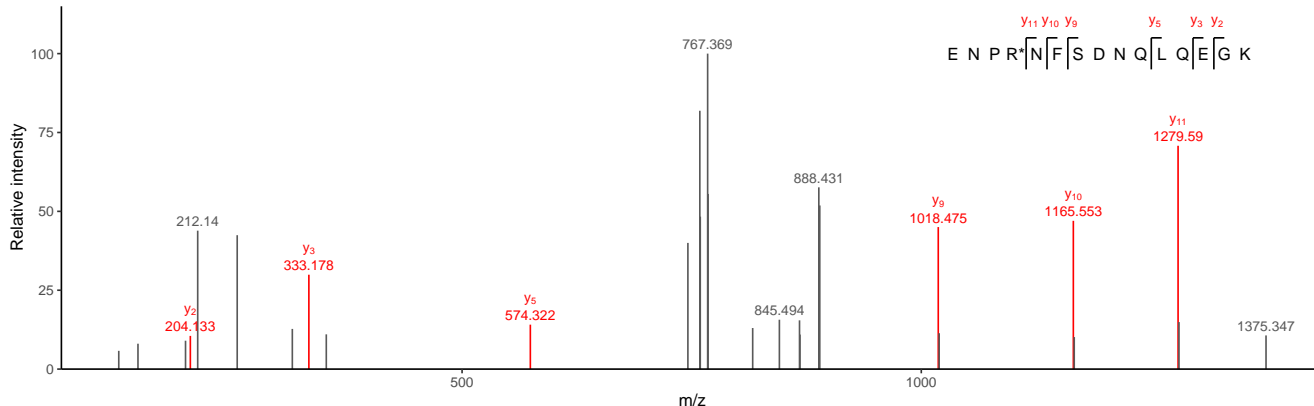


Figure S2. MS² spectra from 17 PSMs of synthetic spectra which were identified as *likely* by ionFinder and classified as *true* in the ground truth dataset.

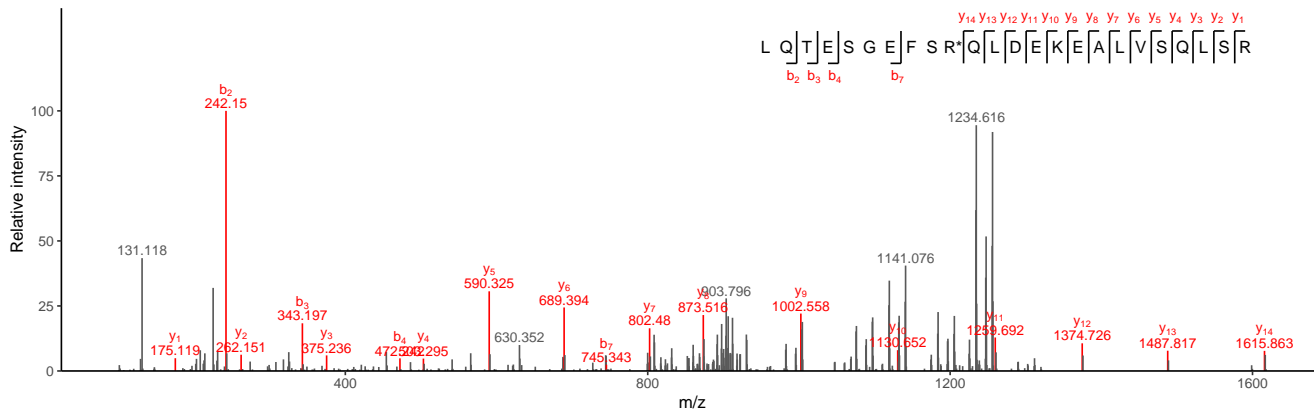
Ground truth: true, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

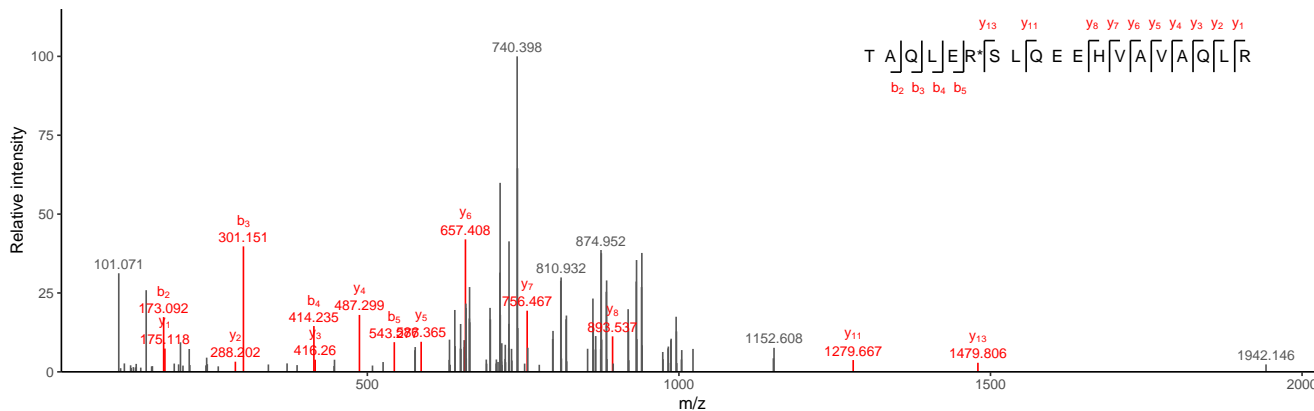
file: 02207a_BD1-TUM_mod_citrullination_s_1_01_01-DDA-1h-R1, scan: 21467



file: 02207a_BD2-TUM_mod_citrullination_s_2_01_01-DDA-1h-R1, scan: 42615



file: 02207a_BB6-TUM_mod_citrullination_L_18_01_01-DDA-1h-R1, scan: 36953



Ground truth: true, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

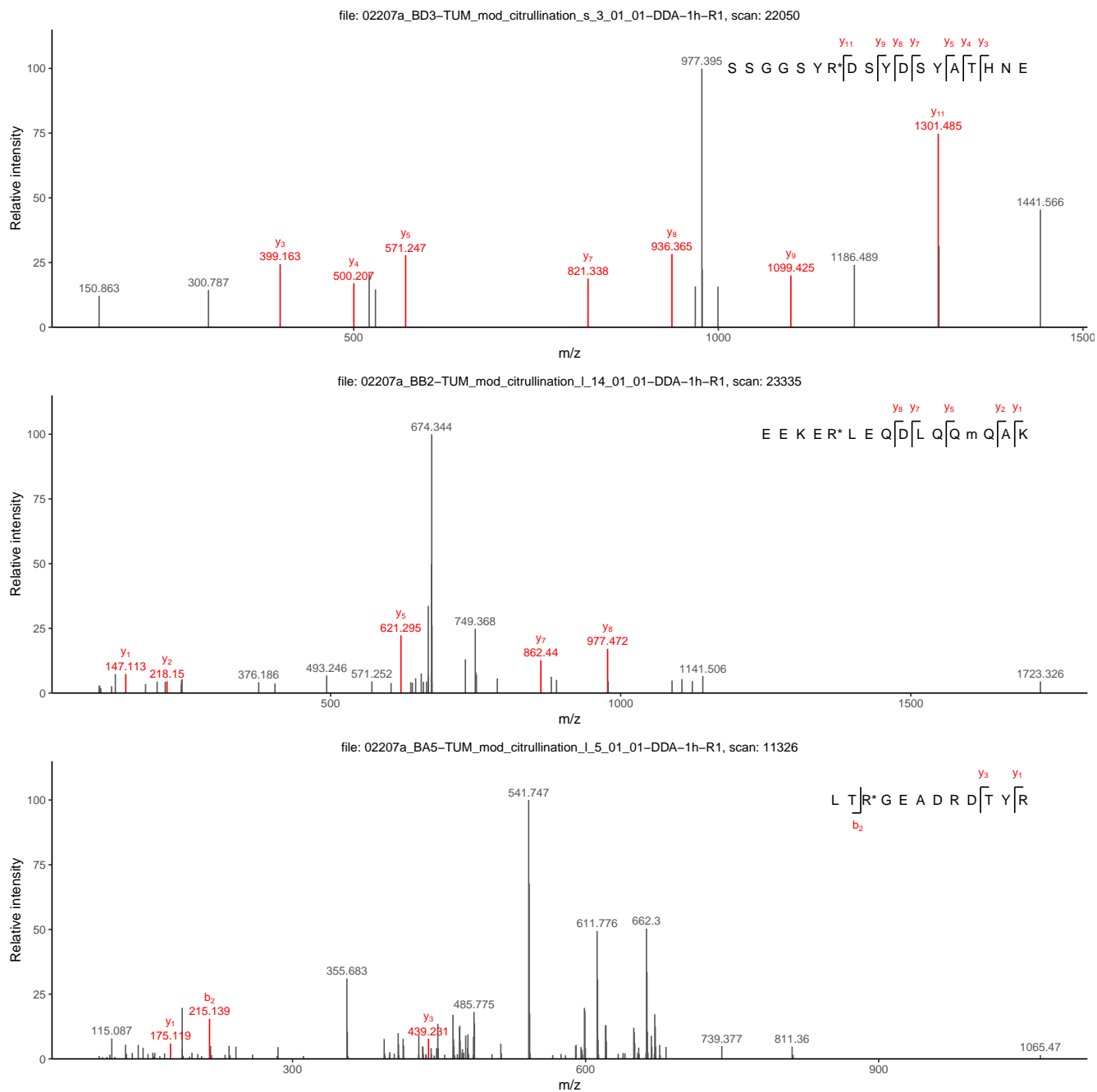
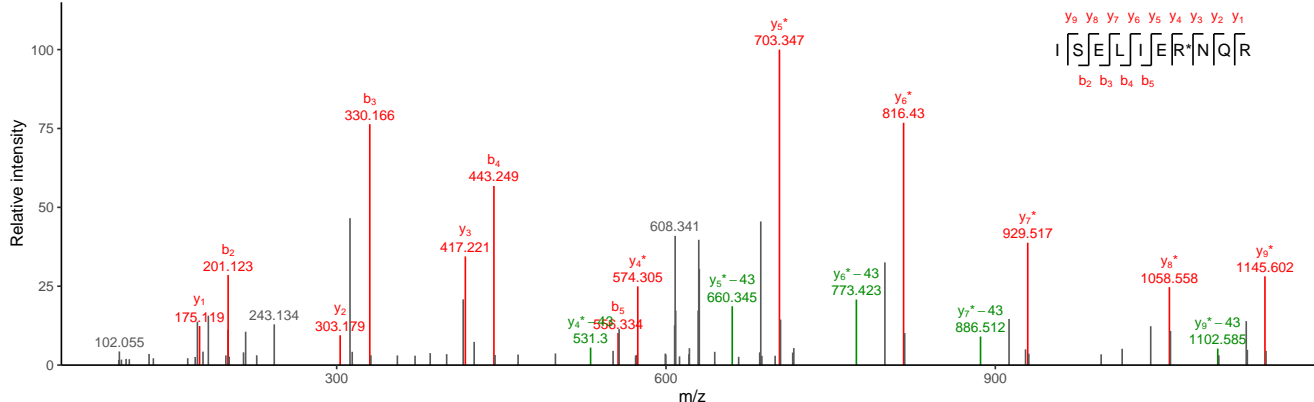


Figure S3. MS² spectra from 6 PSMs of synthetic spectra which were identified as *ambiguous* by ionFinder and classified as *true* in the ground truth dataset.

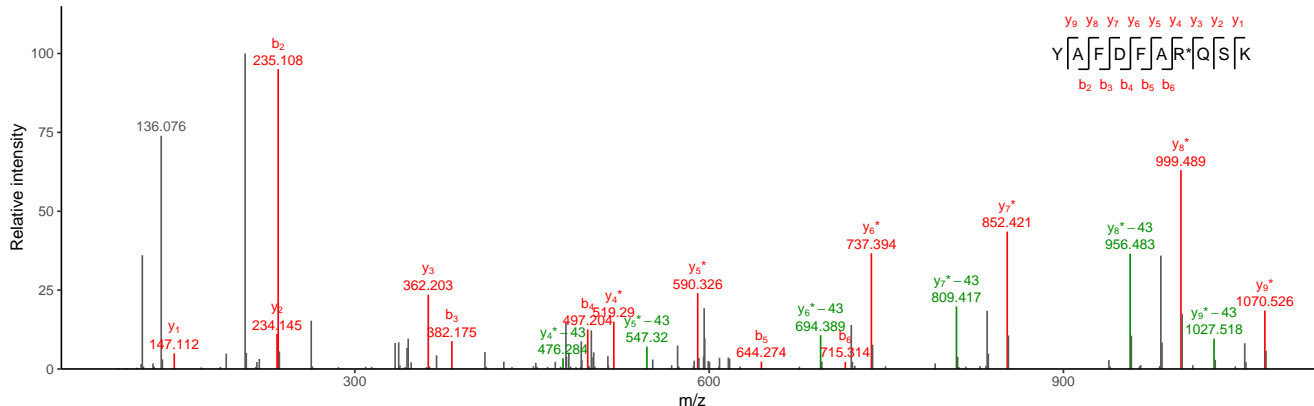
Ground truth: false, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

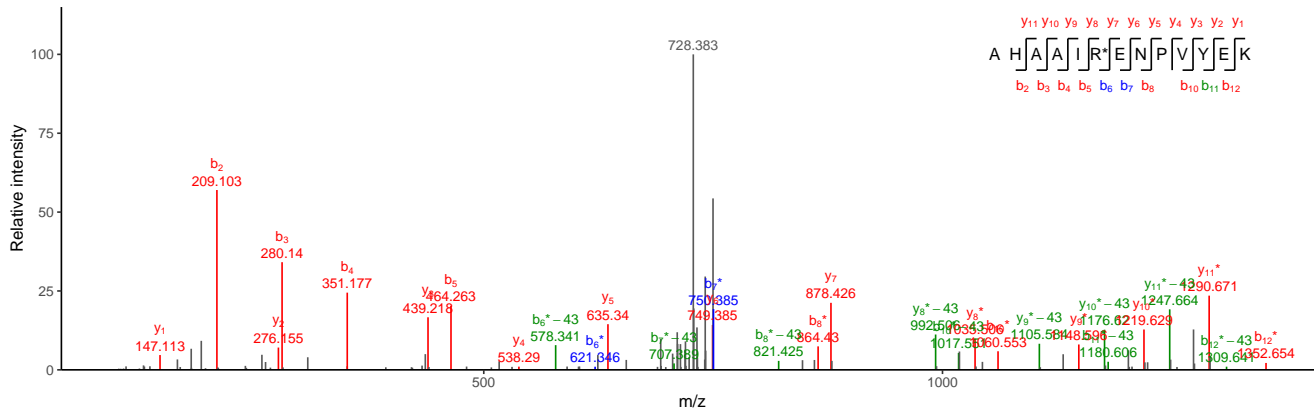
file: 02207a_BA6-TUM_mod_citrullination_L_6_01_01-DDA-1h-R1, scan: 23976



file: 02207a_BA3-TUM_mod_citrullination_L_3_01_01-DDA-1h-R1, scan: 35297



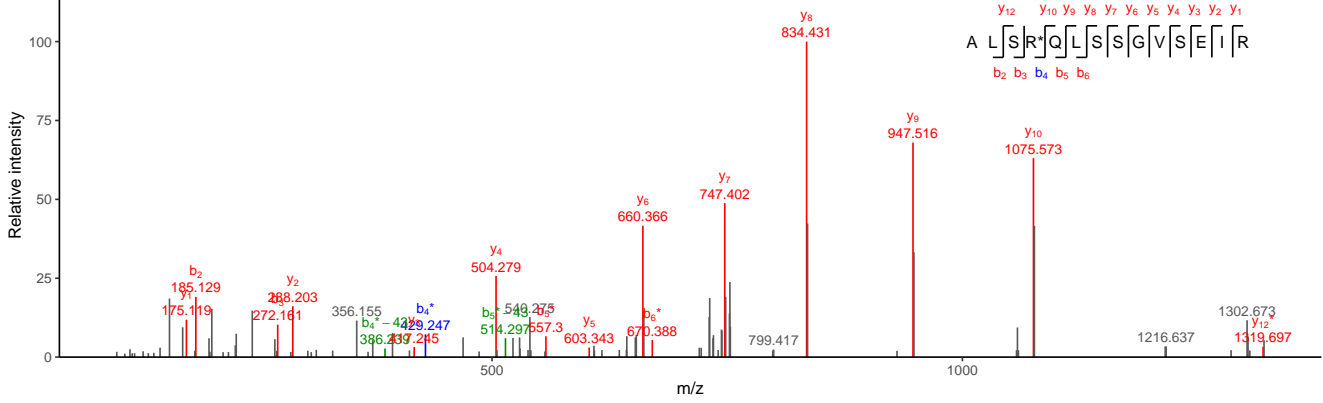
file: 02207a_BA7-TUM_mod_citrullination_L_7_01_01-DDA-1h-R1, scan: 18332



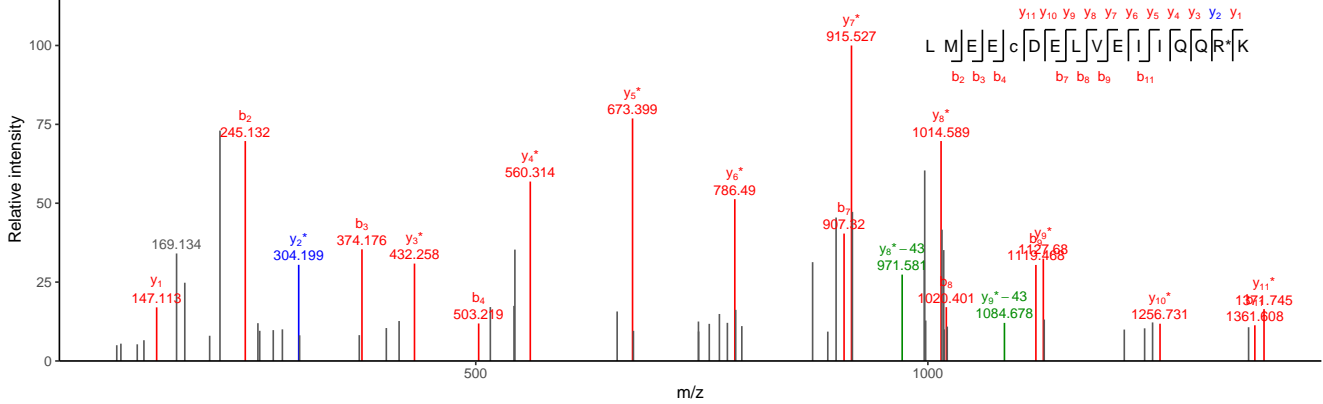
Ground truth: false, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

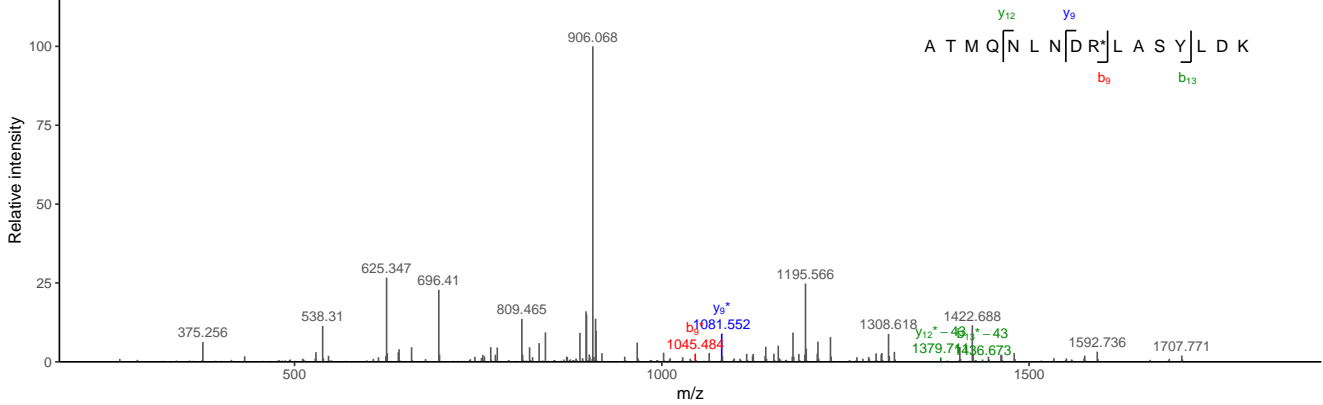
file: 02207a_BA9-TUM_mod_citrullination_I_9_01_01-DDA-1h-R1, scan: 30063



file: 02207a_BB2-TUM_mod_citrullination_I_14_01_01-DDA-1h-R1, scan: 47188



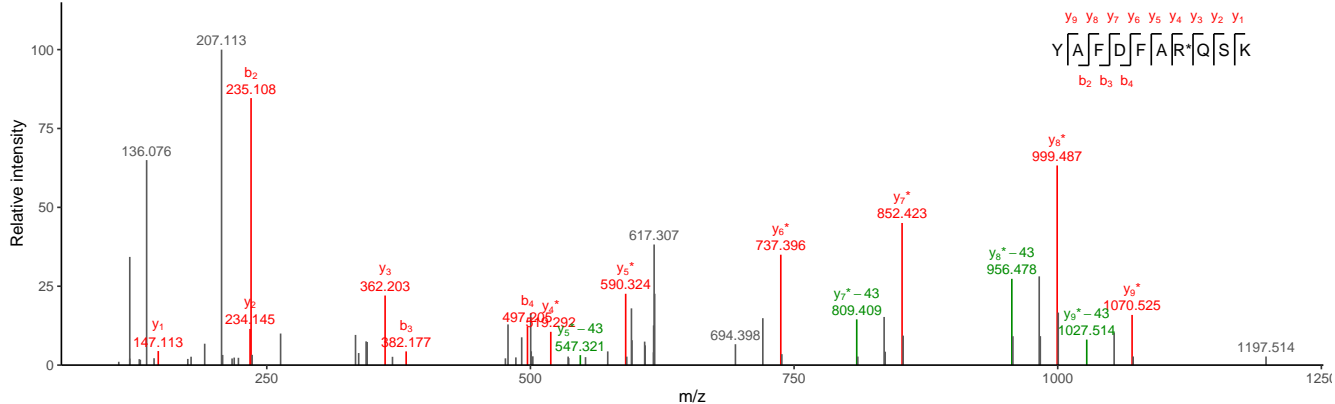
file: 02207a_BB2-TUM_mod_citrullination_I_14_01_01-DDA-1h-R1, scan: 46872



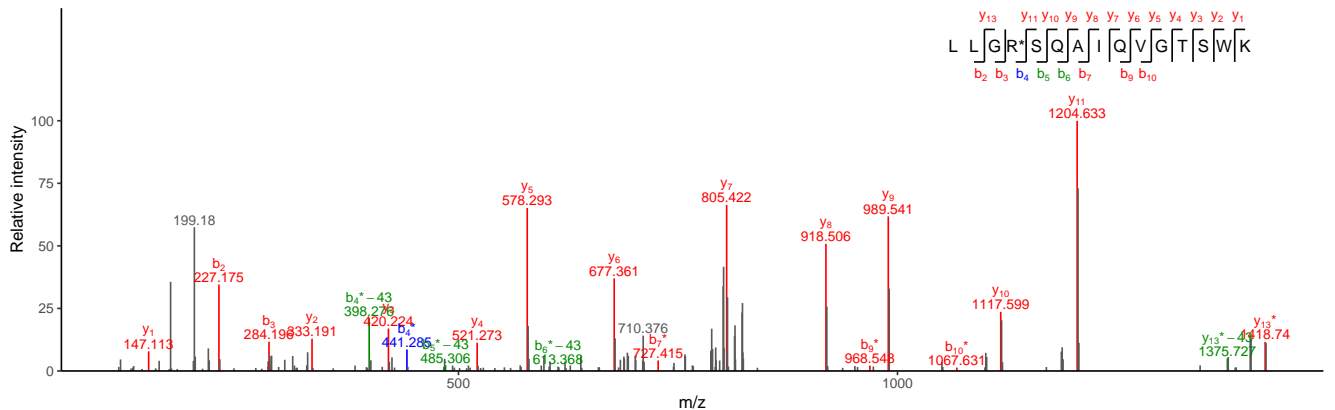
Ground truth: false, ionFinder classification: true

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

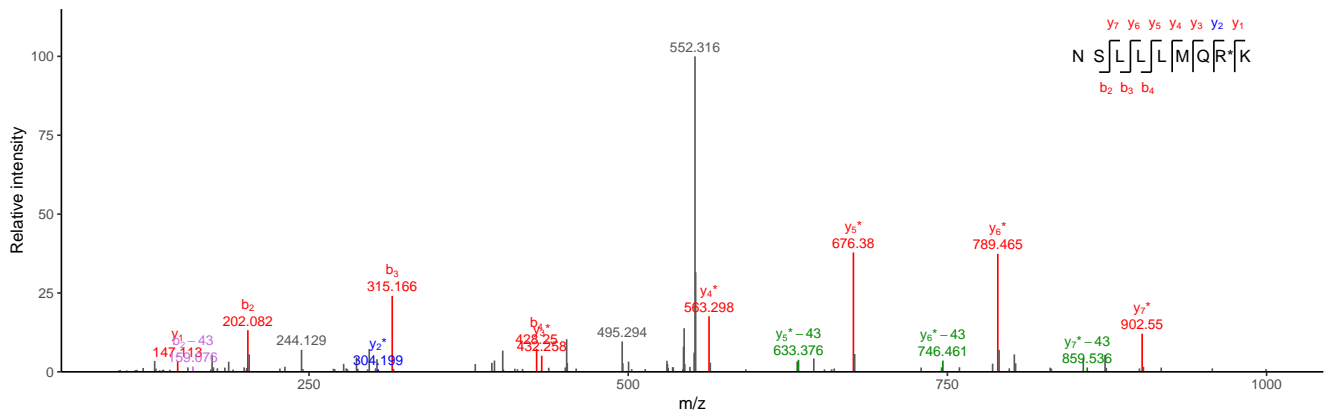
file: 02207a_BA3-TUM_mod_citrullination_L_3_01_01-DDA-1h-R1, scan: 35592



file: 02207a_BA9-TUM_mod_citrullination_L_9_01_01-DDA-1h-R1, scan: 38724



file: 02207a_BA1-TUM_mod_citrullination_L_1_01_01-DDA-1h-R1, scan: 30062



Ground truth: false, ionFinder classification: true

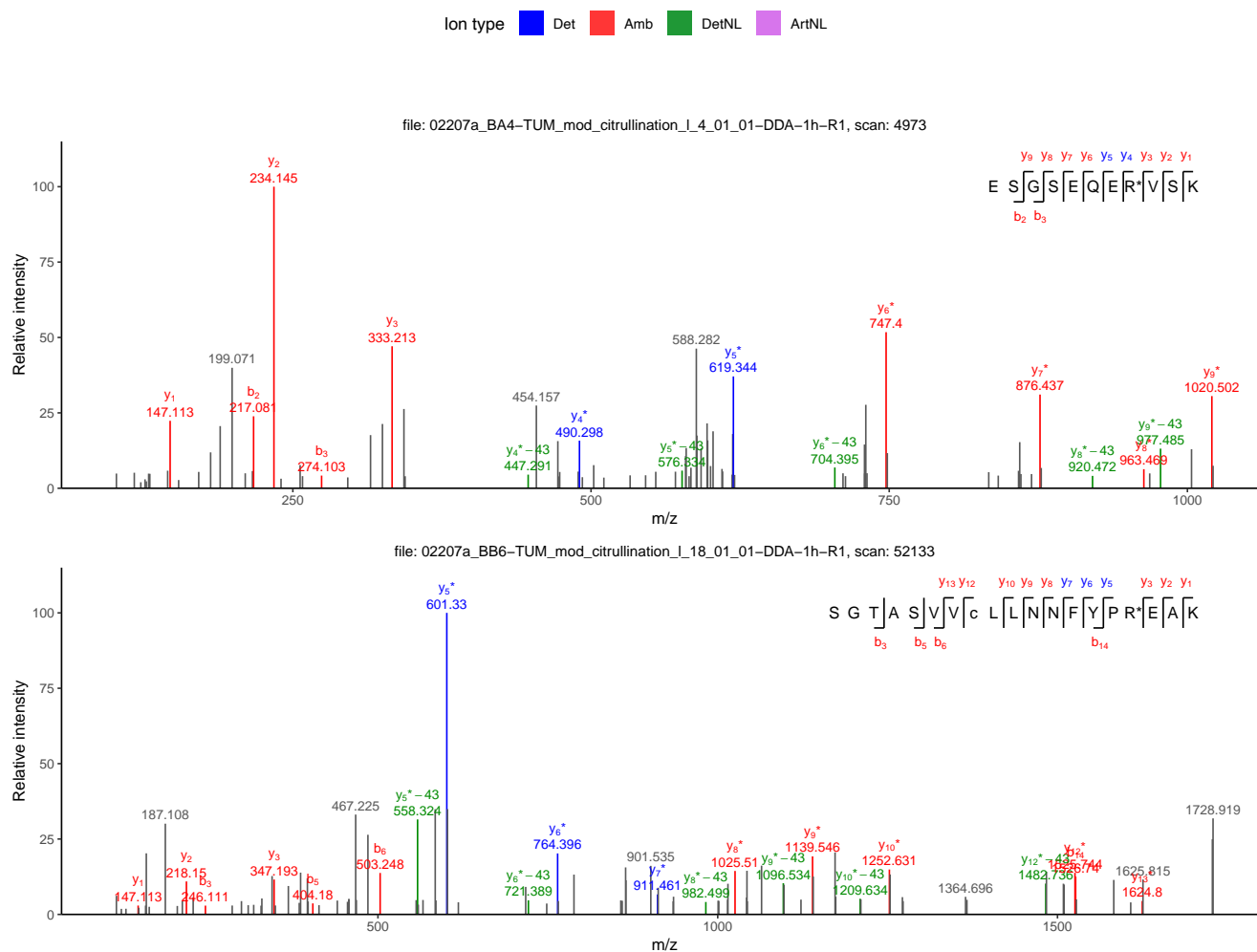
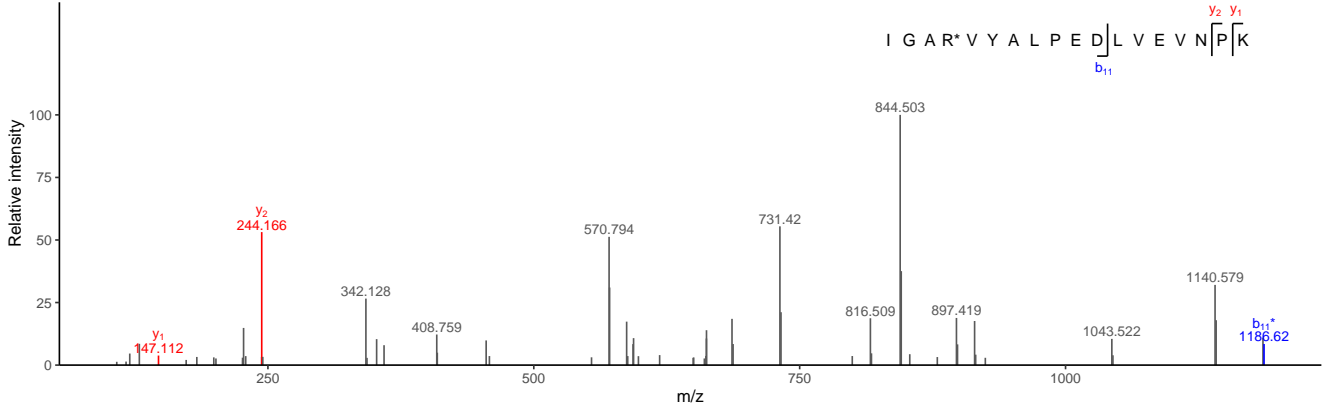


Figure S4. MS² spectra from 11 PSMs of synthetic spectra which were identified as *true* by ionFinder and classified as *false* in the ground truth dataset.

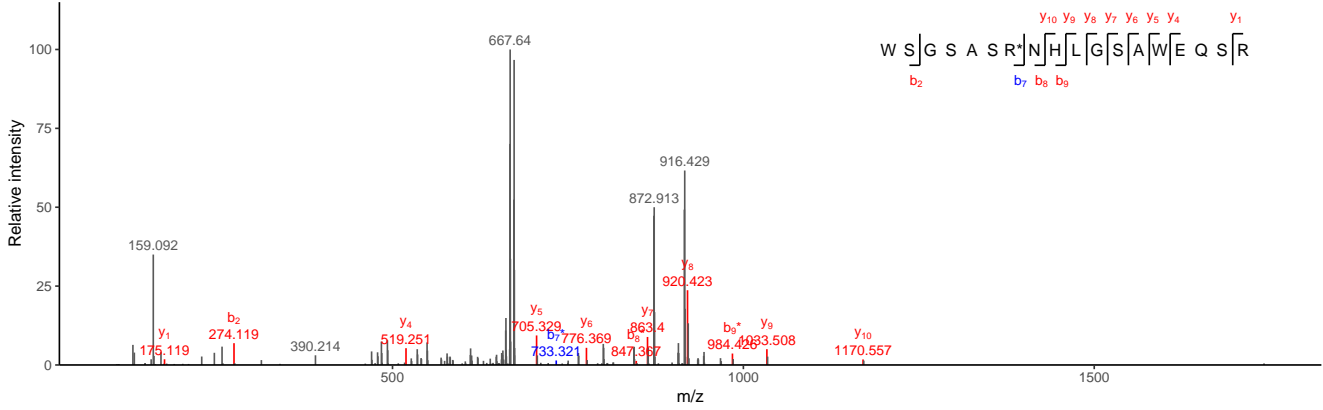
Ground truth: false, ionFinder classification: likely

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

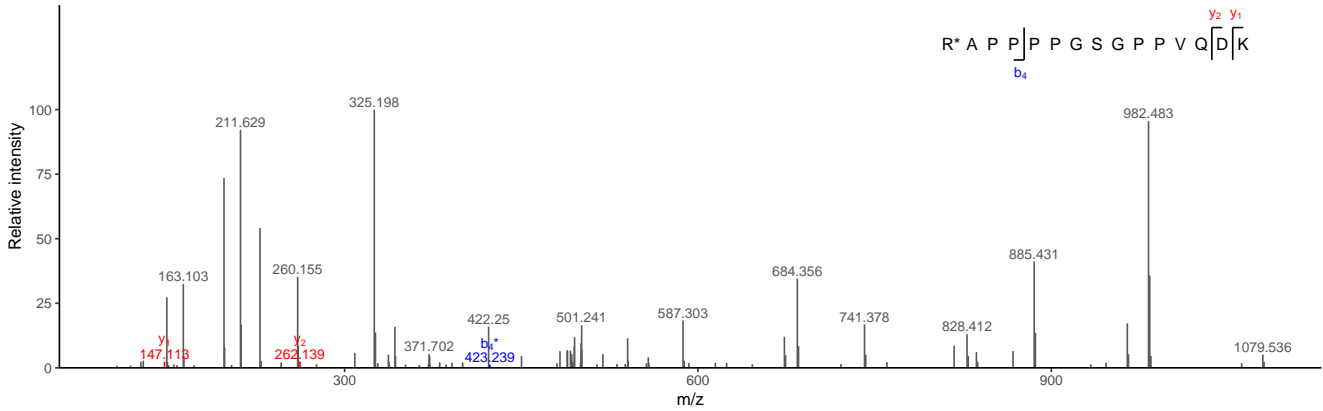
file: 02207a_BB4-TUM_mod_citrullination_I_16_01_01-DDA-1h-R1, scan: 43405



file: 02207a_BB9-TUM_mod_citrullination_I_21_01_01-DDA-1h-R1, scan: 22655



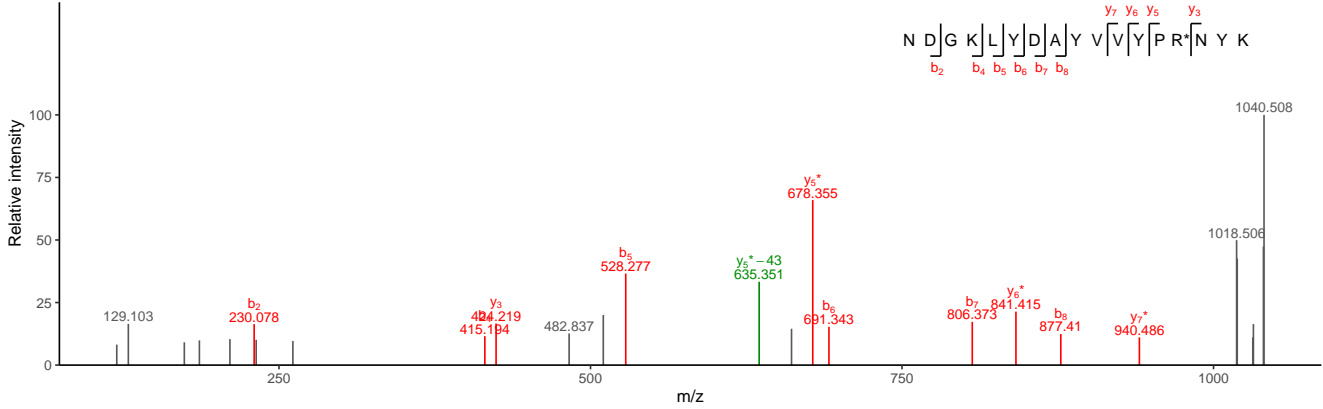
file: 02207a_BA11-TUM_mod_citrullination_I_11_01_01-DDA-1h-R1, scan: 12569



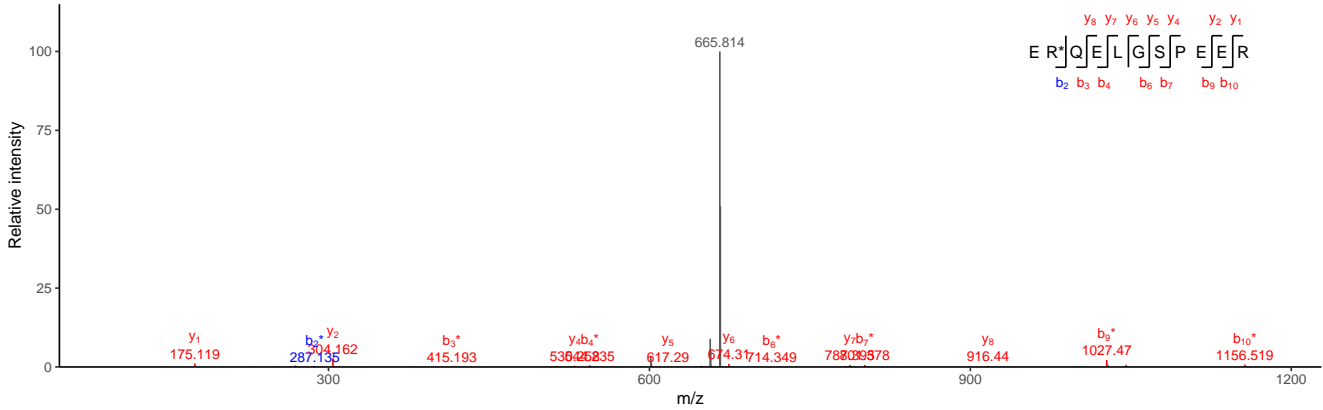
Ground truth: false, ionFinder classification: likely

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

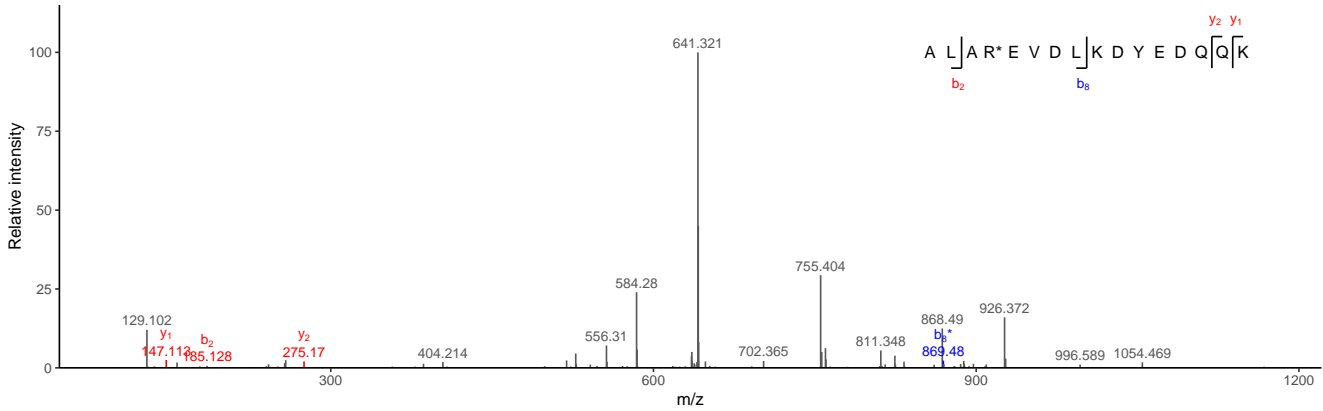
file: 02207a_BB3-TUM_mod_citrullination_L15_01_01-DDA-1h-R1, scan: 34324



file: 02207a_BA1-TUM_mod_citrullination_L1_01_01-DDA-1h-R1, scan: 11317

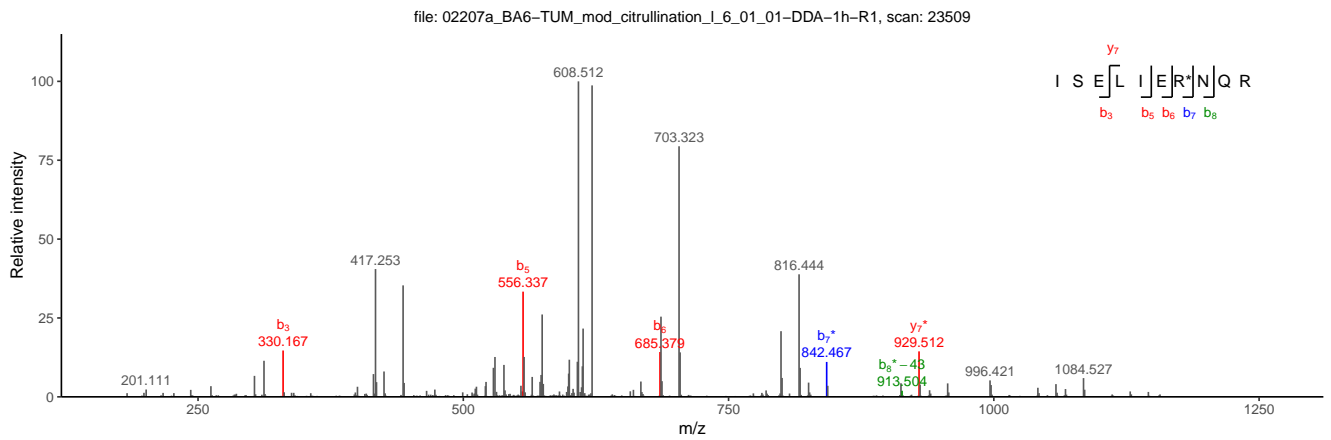
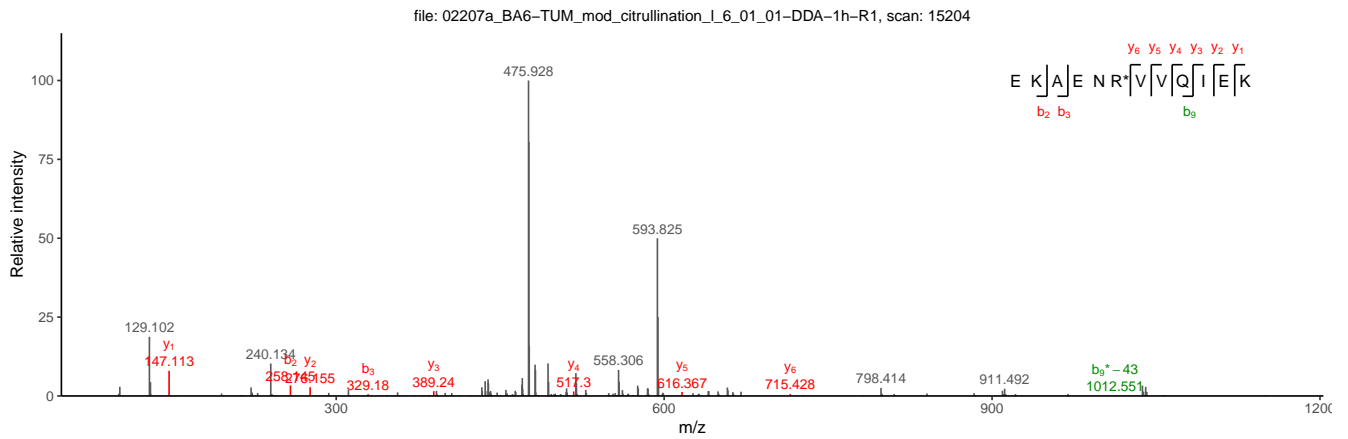
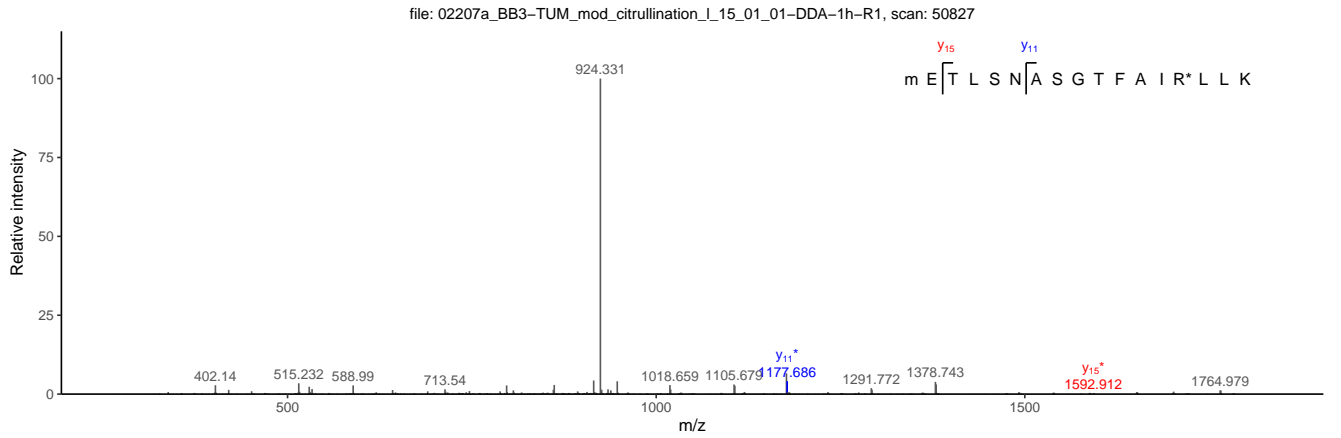


file: 02207a_BB1-TUM_mod_citrullination_L13_01_01-DDA-1h-R1, scan: 25583



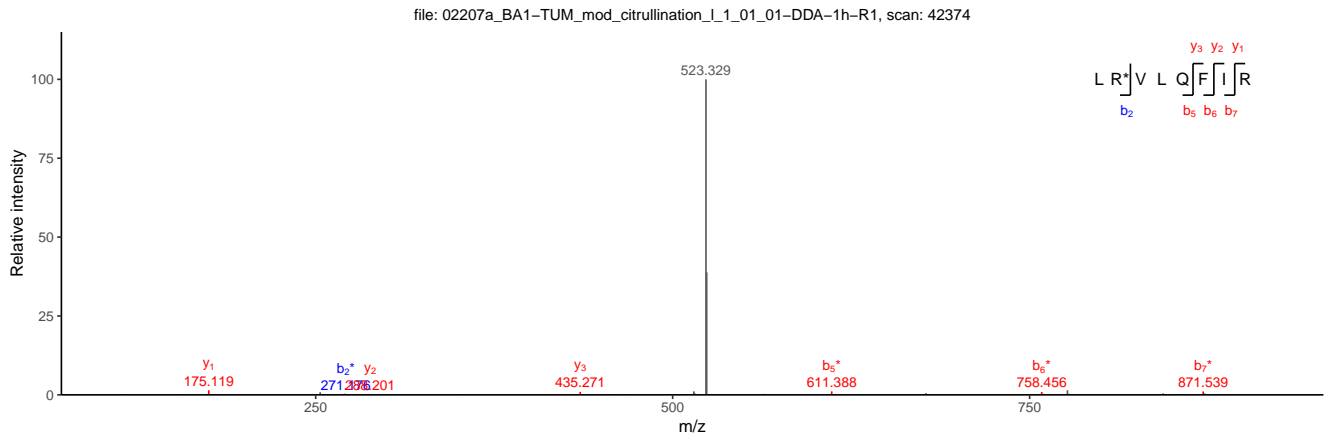
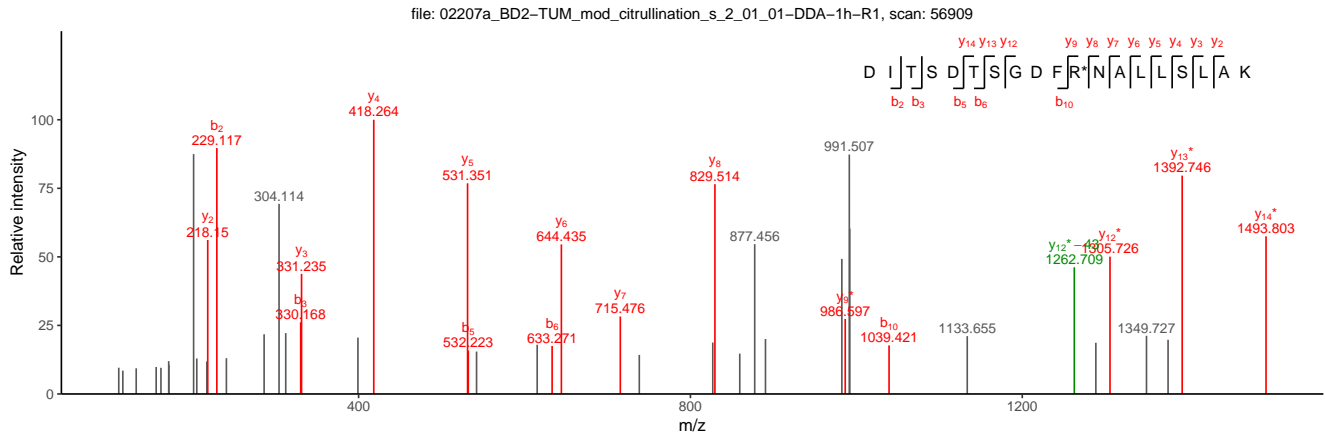
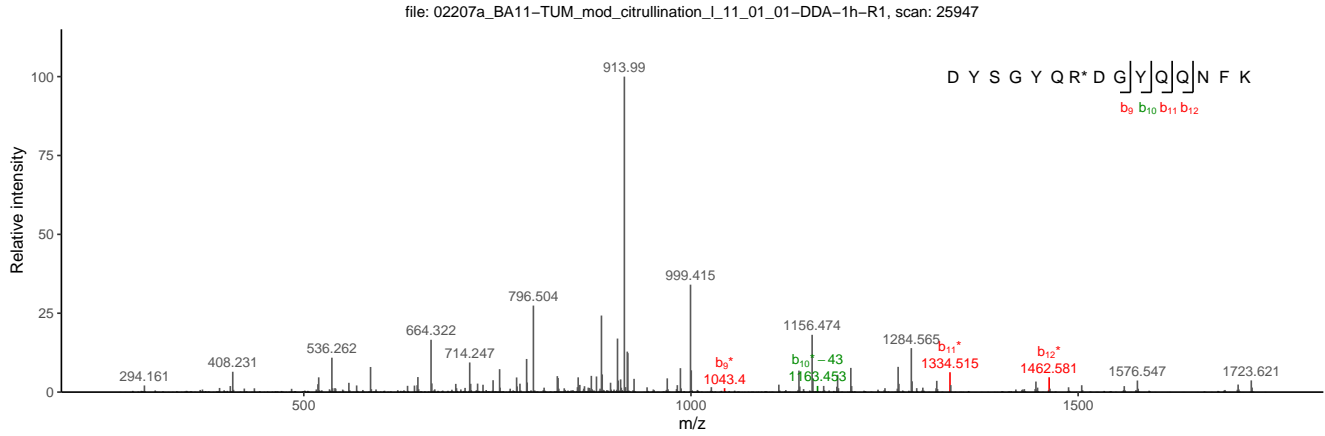
Ground truth: false, ionFinder classification: likely

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



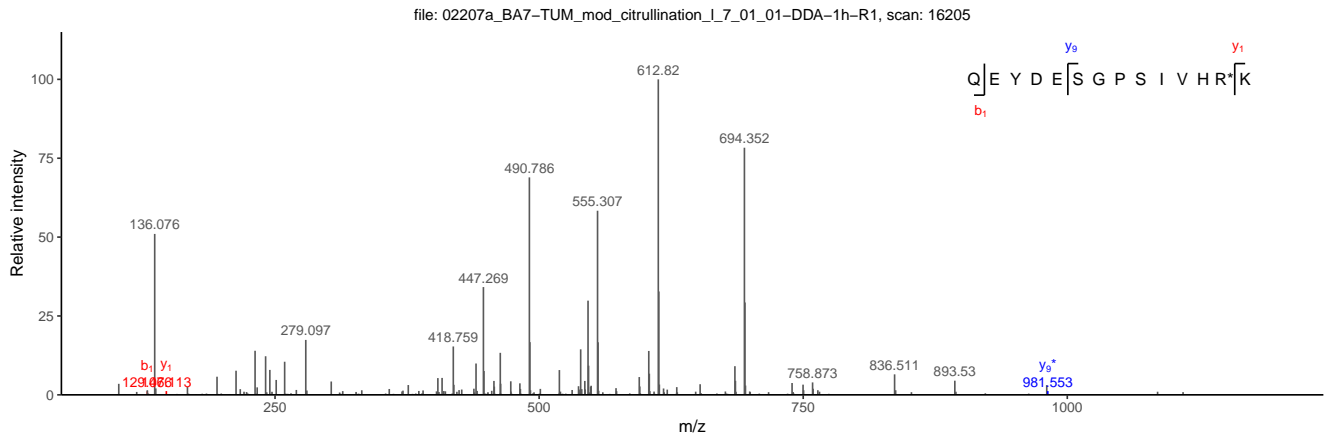
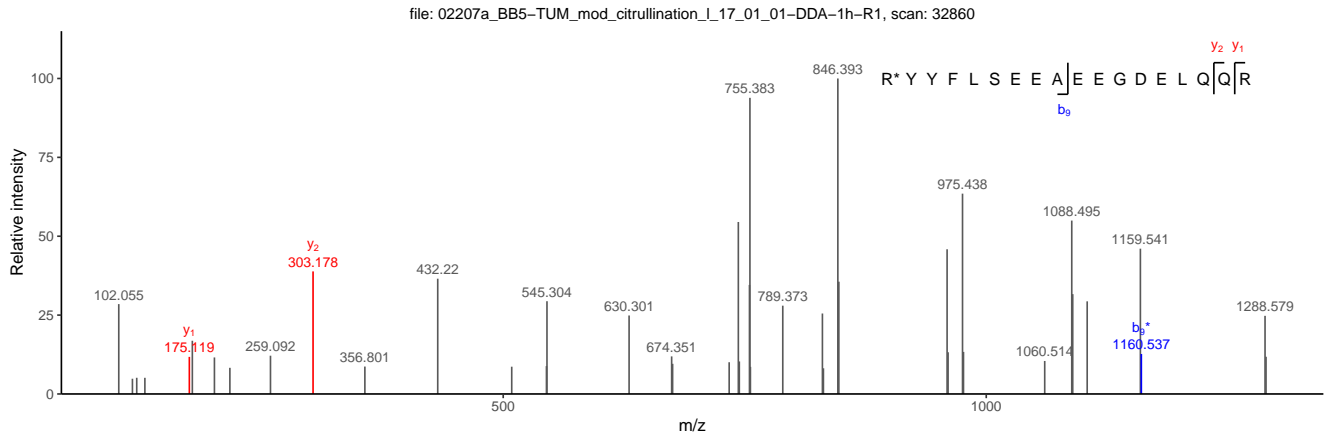
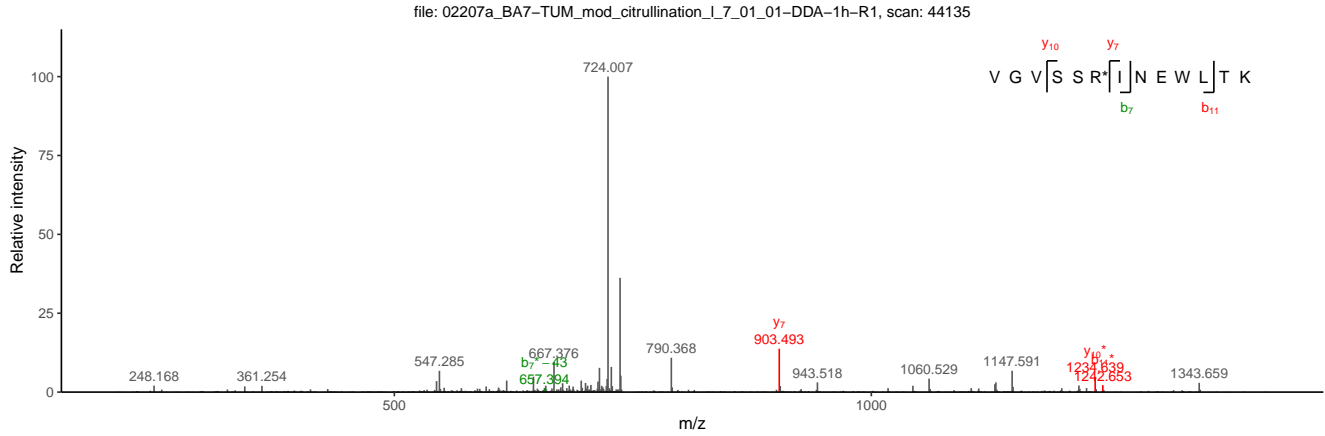
Ground truth: false, ionFinder classification: likely

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



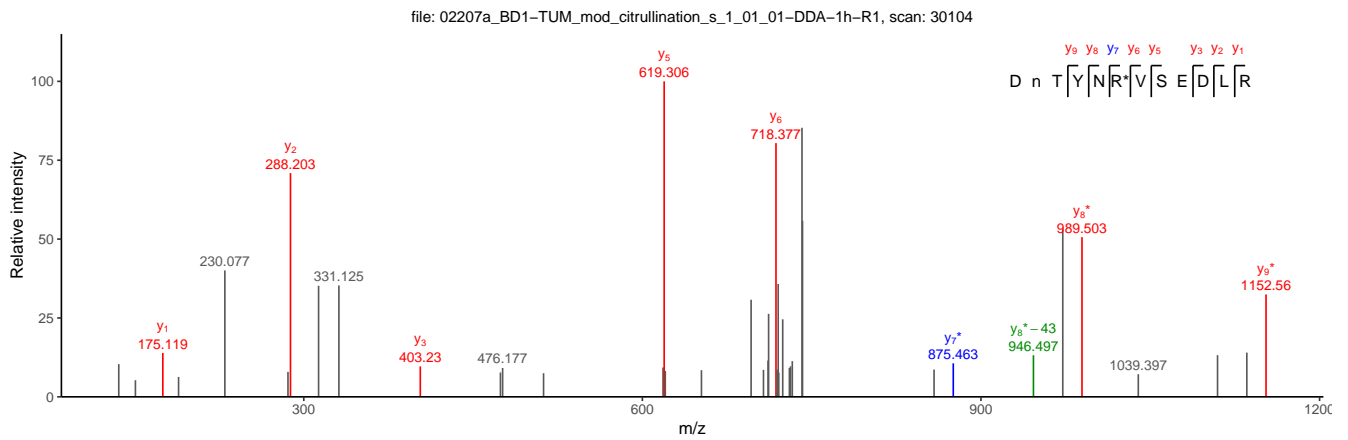
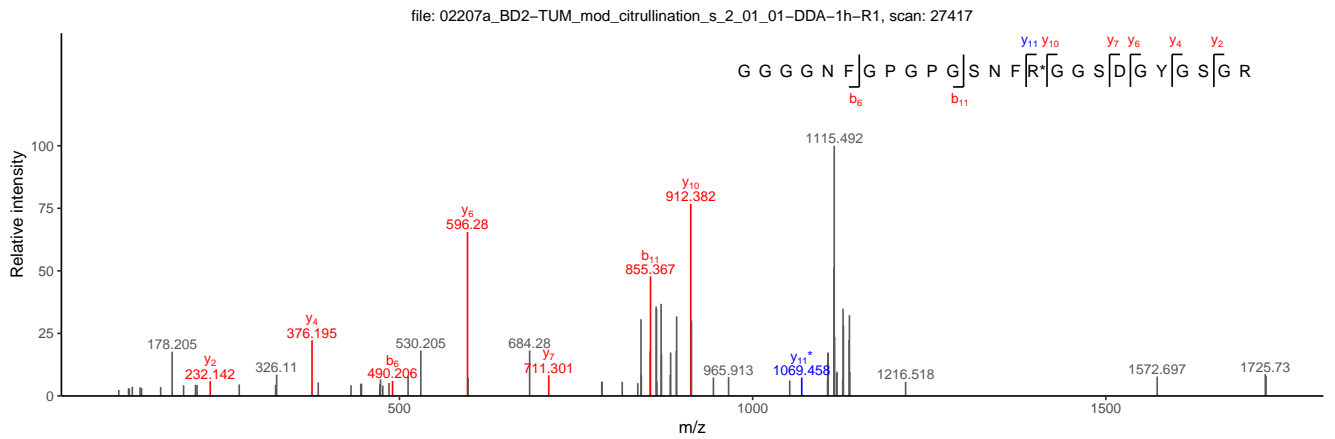
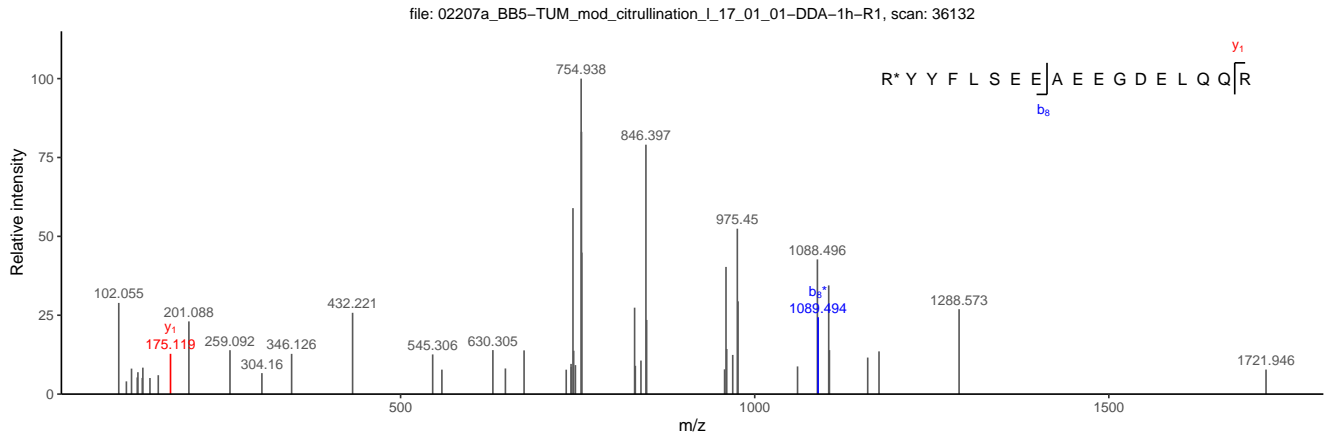
Ground truth: false, ionFinder classification: likely

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



Ground truth: false, ionFinder classification: likely

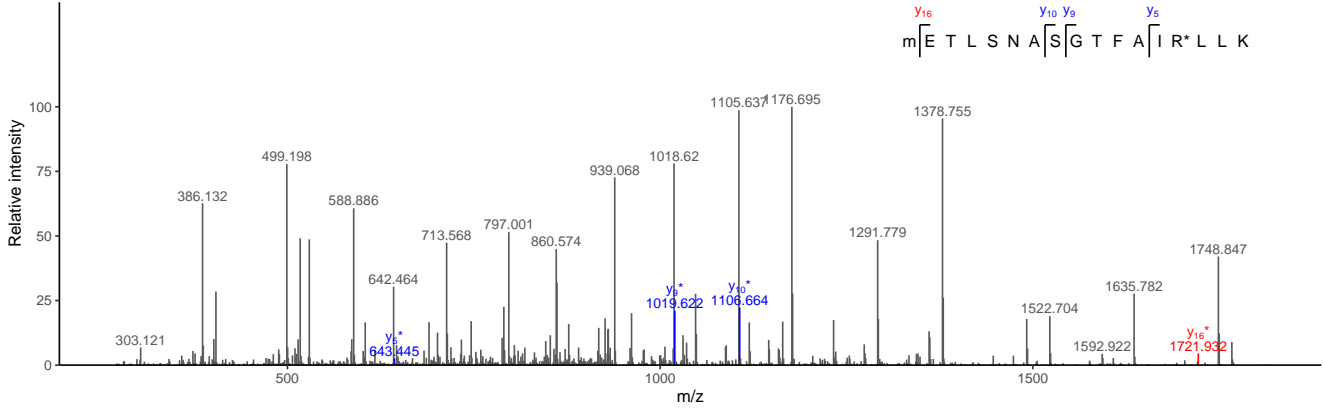
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



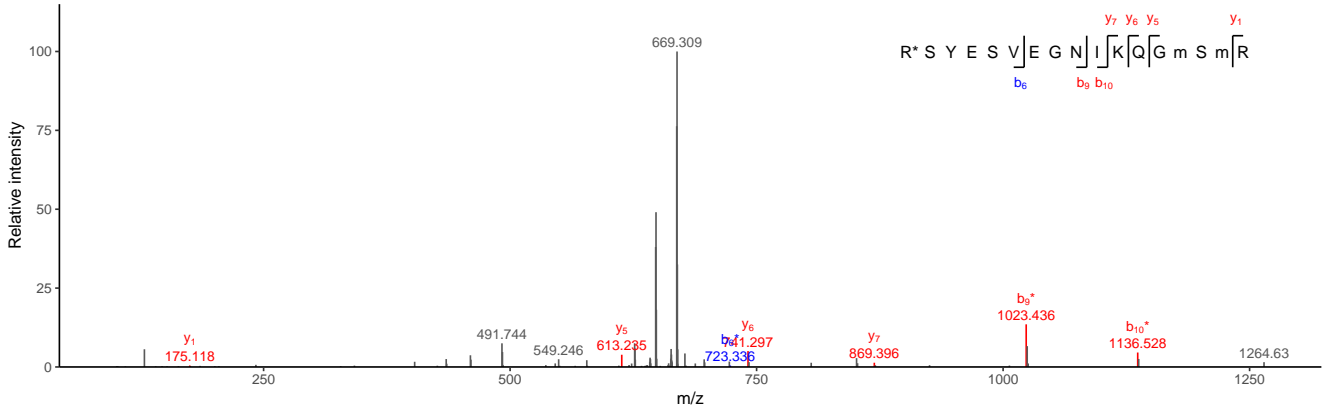
Ground truth: false, ionFinder classification: likely

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

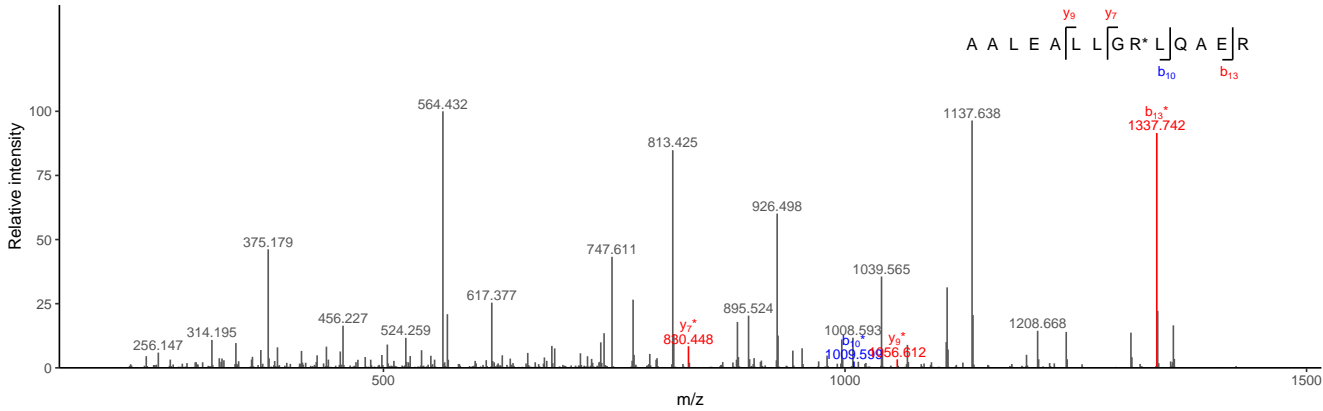
file: 02207a_BB3-TUM_mod_citrullination_I_15_01_01-DDA-1h-R1, scan: 57647



file: 02207a_BB2-TUM_mod_citrullination_I_14_01_01-DDA-1h-R1, scan: 19361



file: 02207a_BA8-TUM_mod_citrullination_I_8_01_01-DDA-1h-R1, scan: 51189



Ground truth: false, ionFinder classification: likely

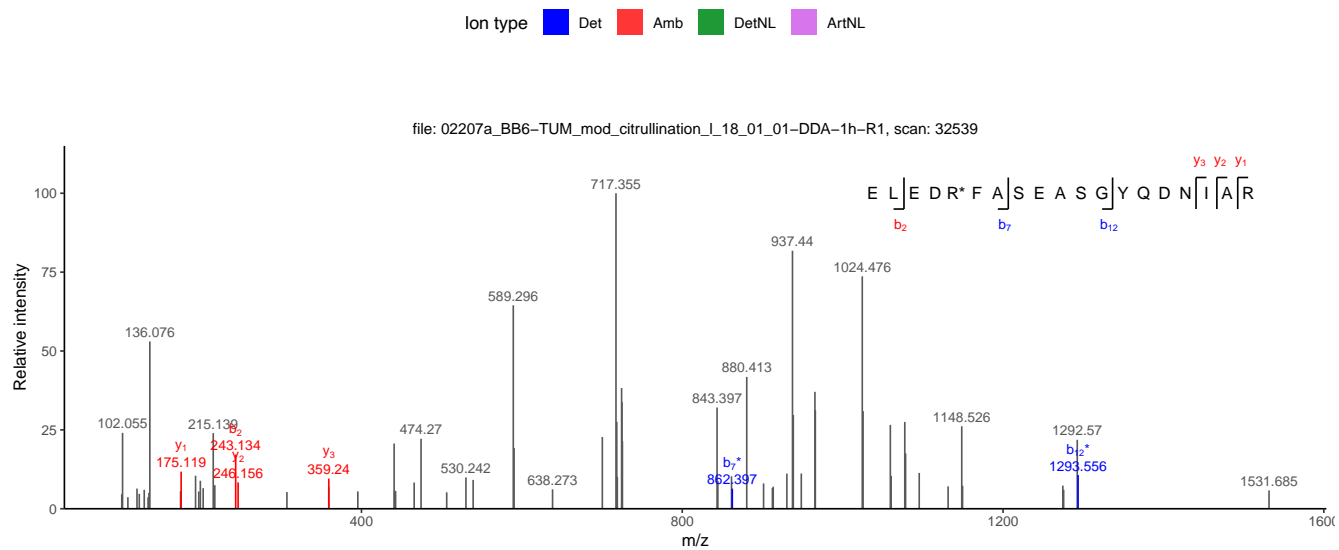
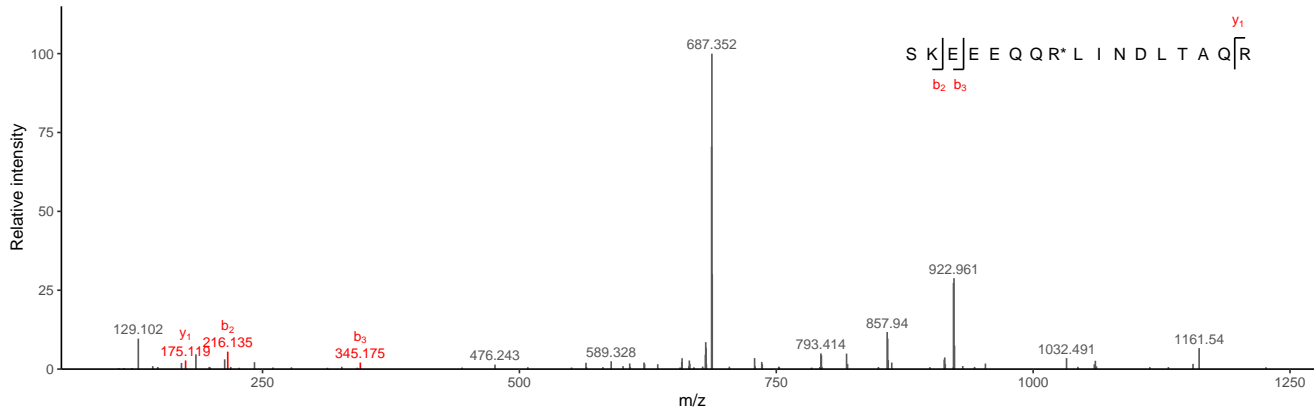


Figure S5. MS² spectra from 22 PSMs of synthetic spectra which were identified as *likely* by ionFinder and classified as *false* in the ground truth dataset.

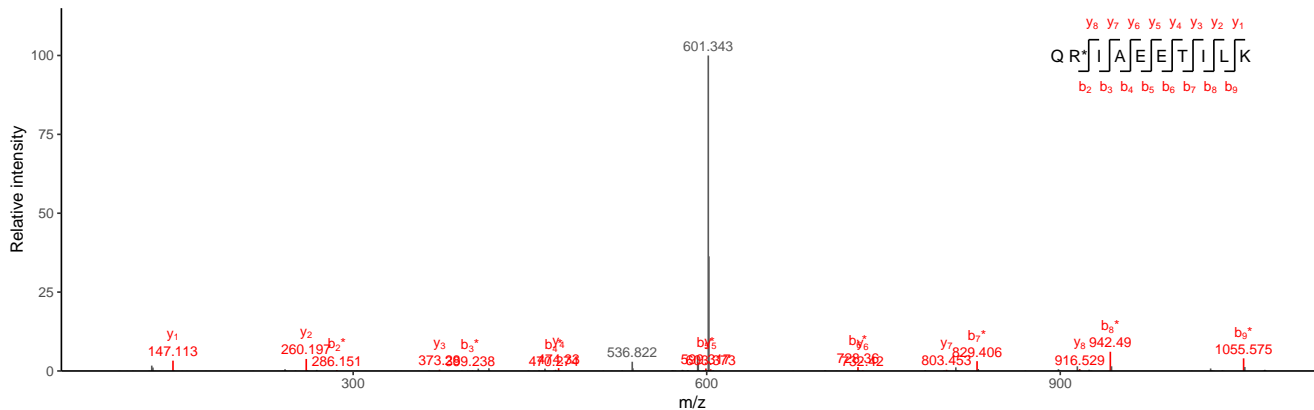
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

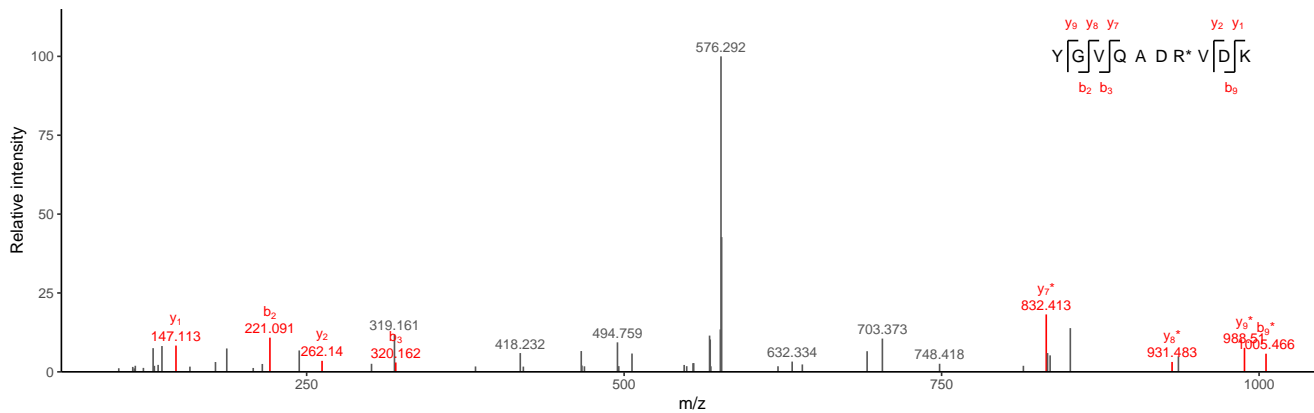
file: 02207a_BB3-TUM_mod_citrullination_I_15_01_01-DDA-1h-R1, scan: 30437



file: 02207a_BA2-TUM_mod_citrullination_I_2_01_01-DDA-1h-R1, scan: 25999

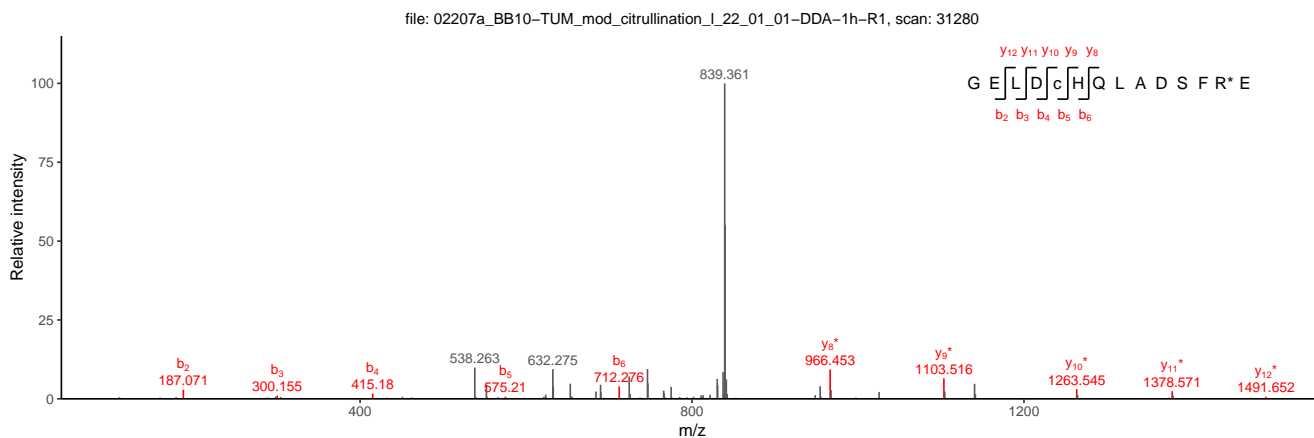
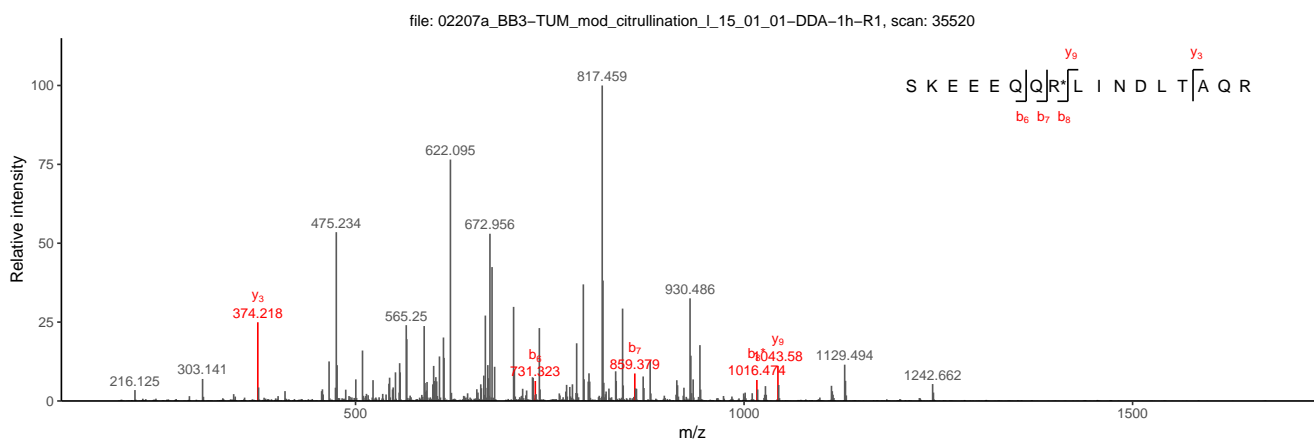
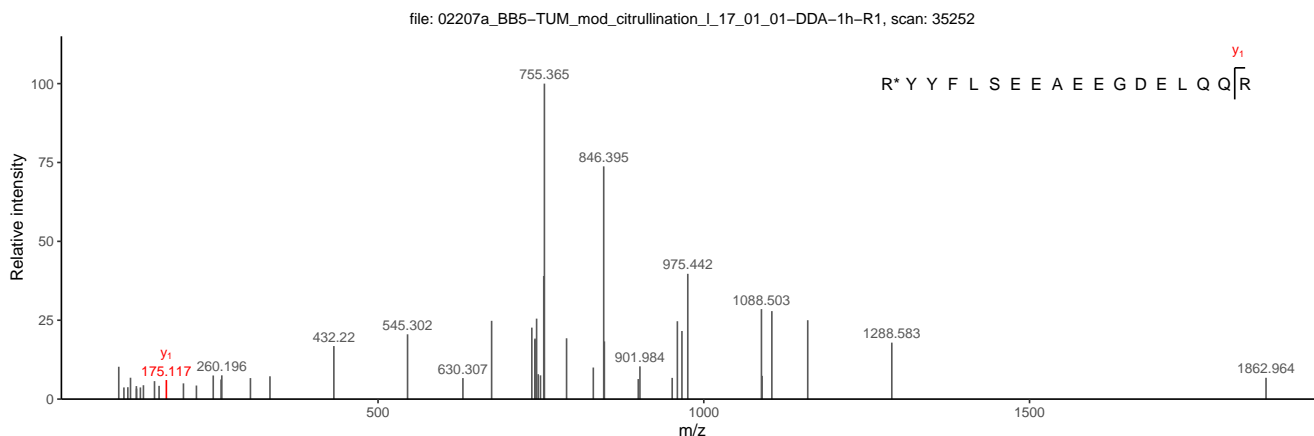


file: 02207a_BA3-TUM_mod_citrullination_I_3_01_01-DDA-1h-R1, scan: 14209



Ground truth: false, ionFinder classification: ambiguous

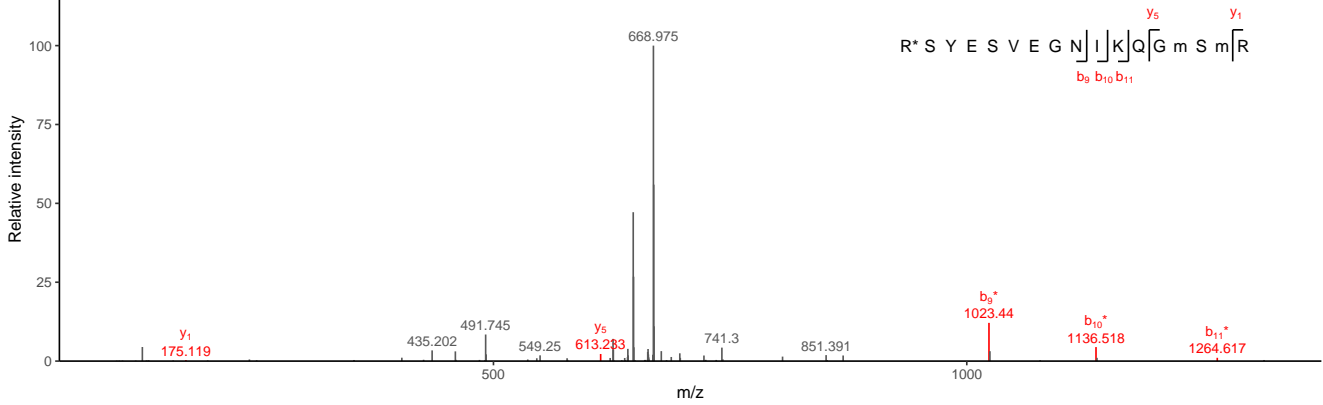
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



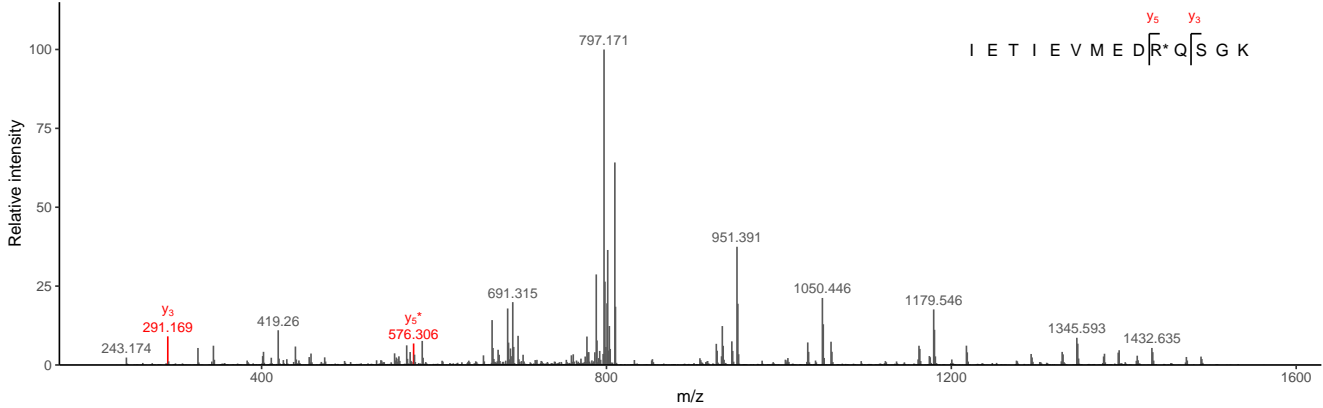
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

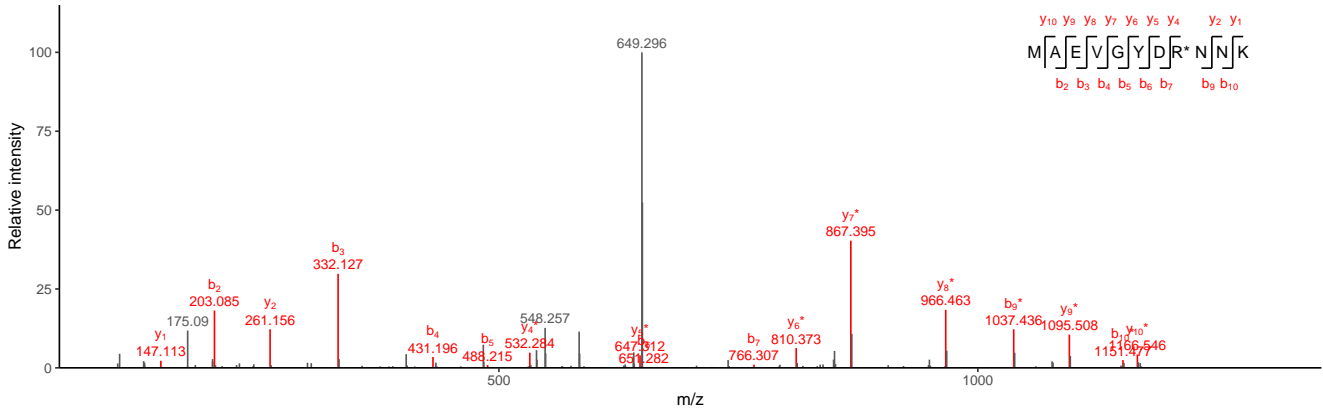
file: 02207a_BB2-TUM_mod_citrullination_I_14_01_01-DDA-1h-R1, scan: 19663



file: 02207a_BA9-TUM_mod_citrullination_I_9_01_01-DDA-1h-R1, scan: 31899

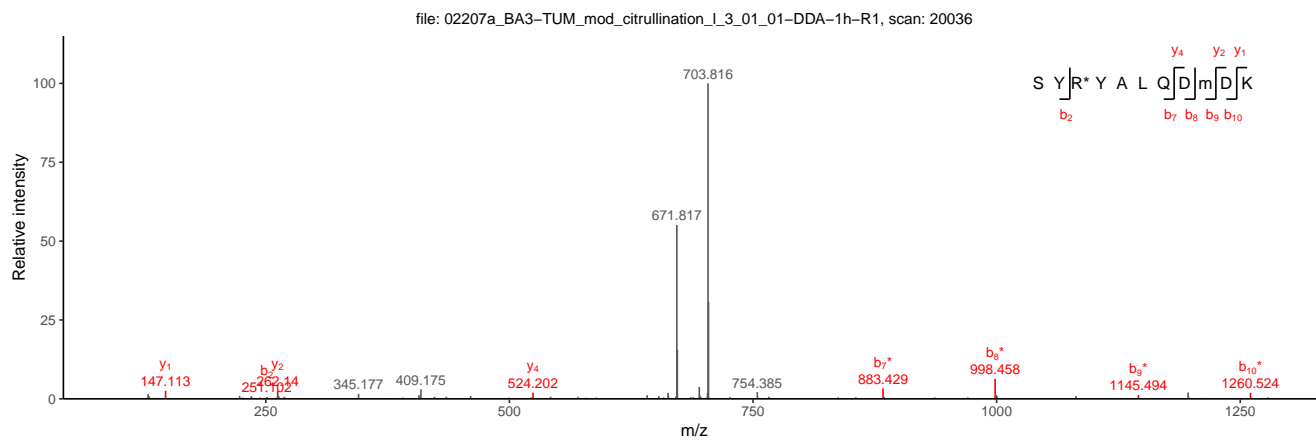
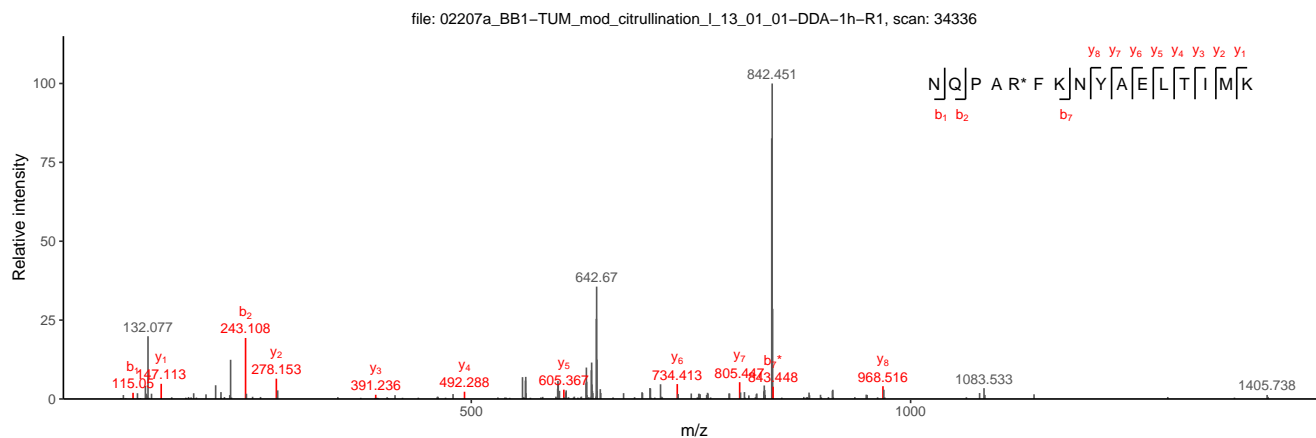
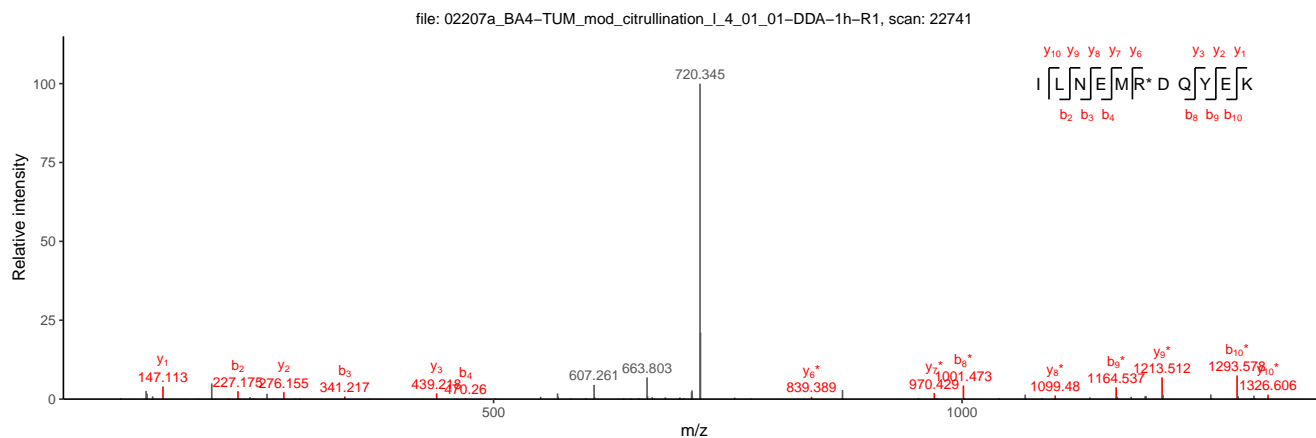


file: 02207a_BA3-TUM_mod_citrullination_I_3_01_01-DDA-1h-R1, scan: 12406



Ground truth: false, ionFinder classification: ambiguous

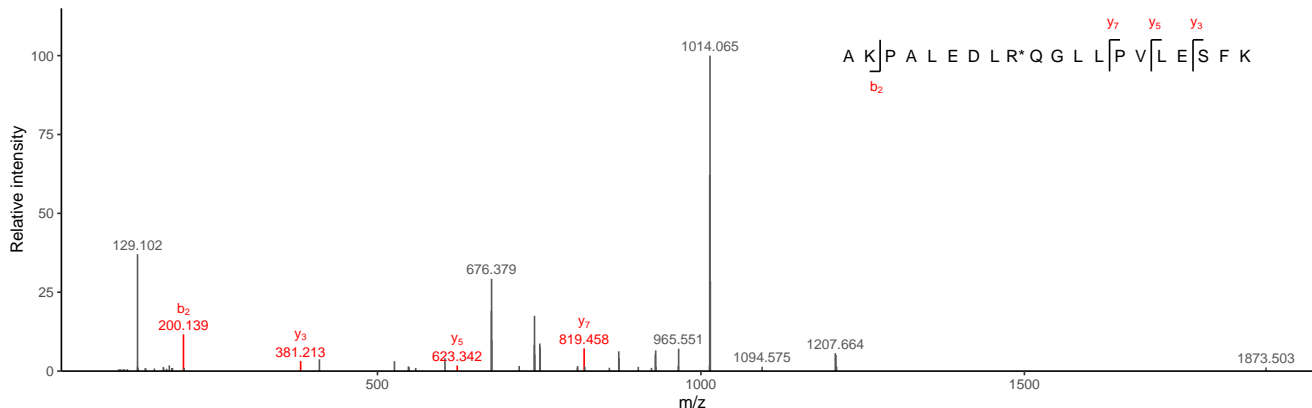
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



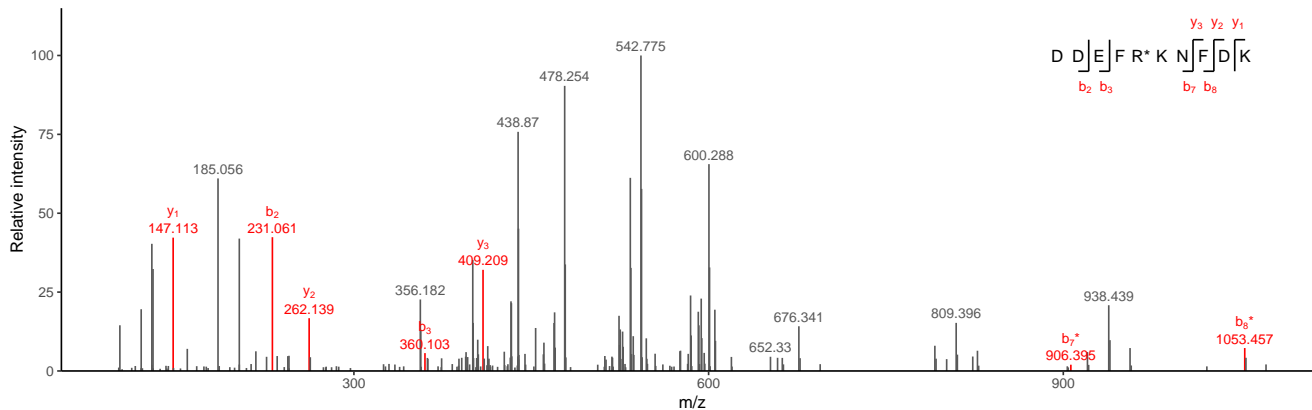
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

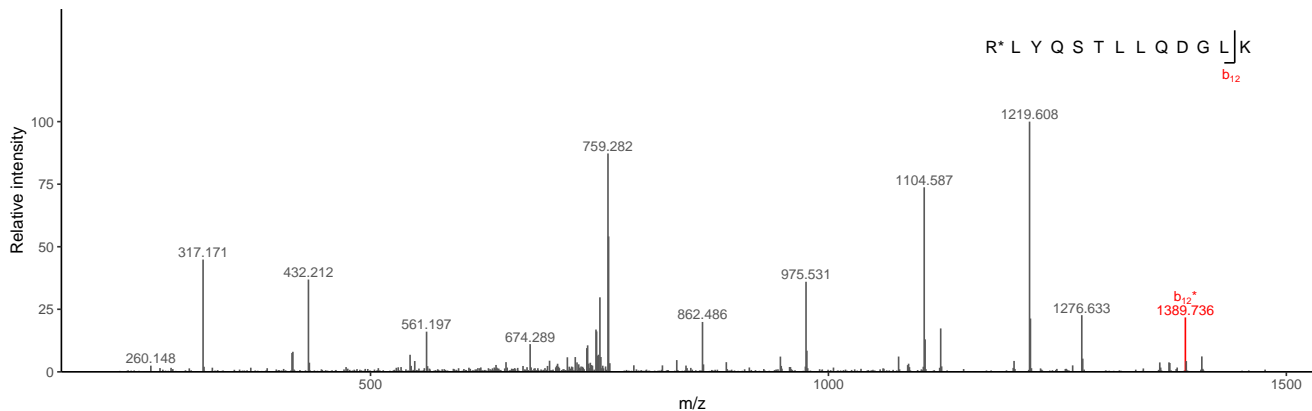
file: 02207a_BB7-TUM_mod_citrullination_L_19_01_01-DDA-1h-R1, scan: 57372



file: 02207a_BA2-TUM_mod_citrullination_L_2_01_01-DDA-1h-R1, scan: 21437



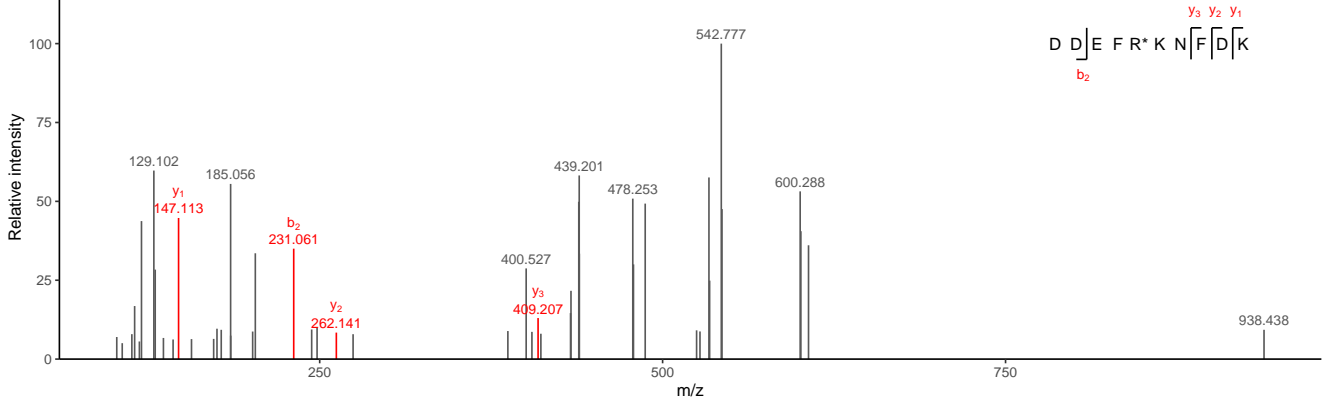
file: 02207a_BA7-TUM_mod_citrullination_L_7_01_01-DDA-1h-R1, scan: 34699



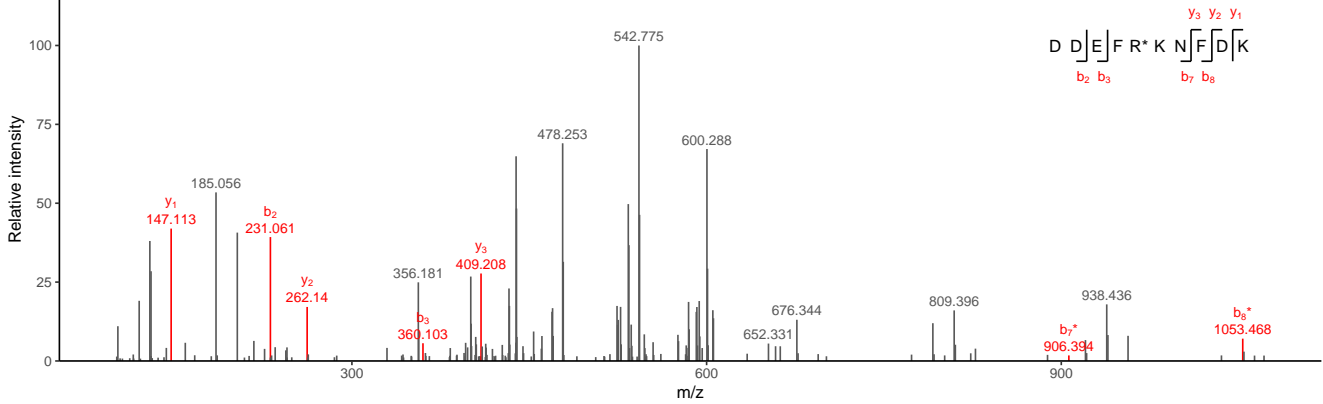
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

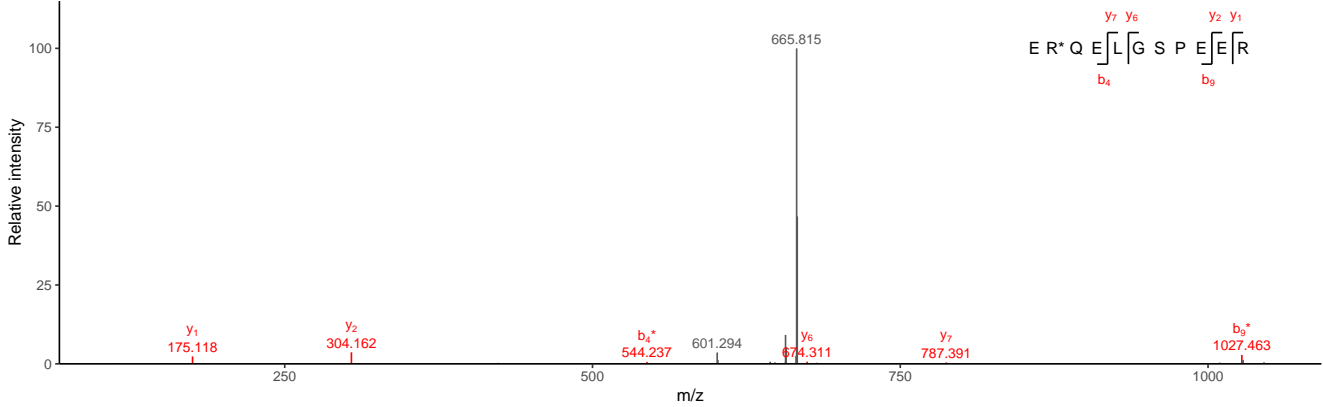
file: 02207a_BA2-TUM_mod_citrullination_L_2_01_01-DDA-1h-R1, scan: 32249



file: 02207a_BA3-TUM_mod_citrullination_L_3_01_01-DDA-1h-R1, scan: 15889

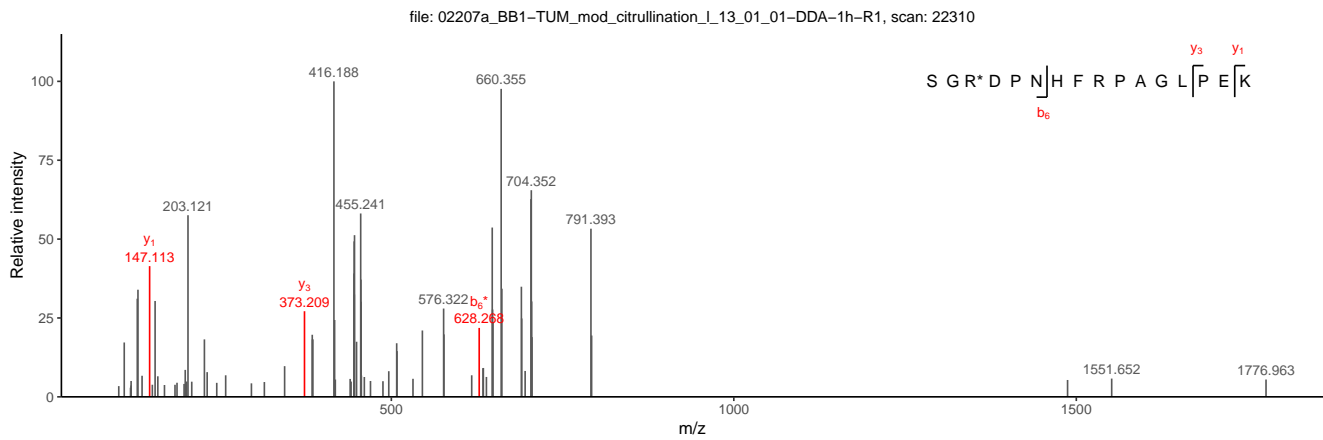
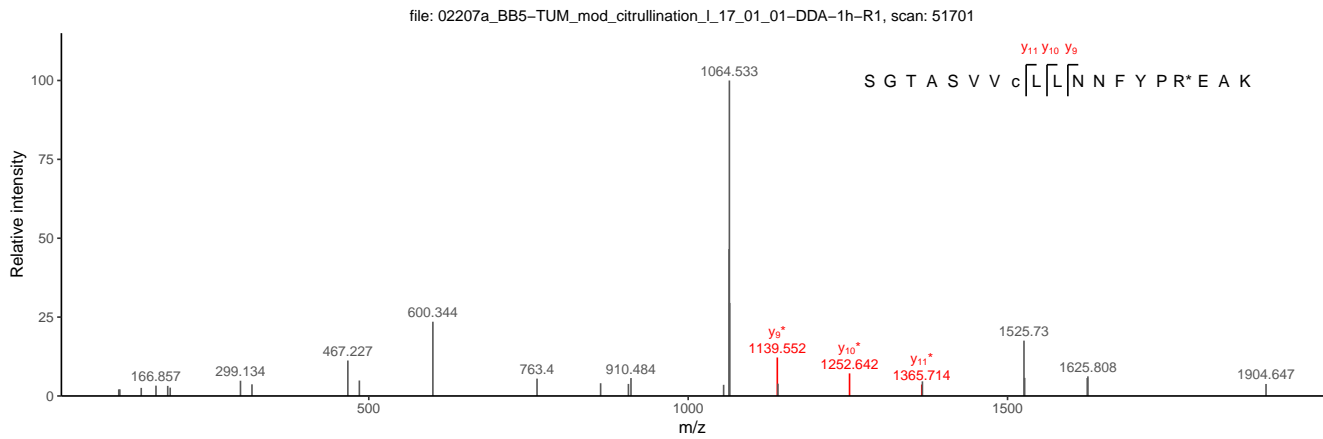
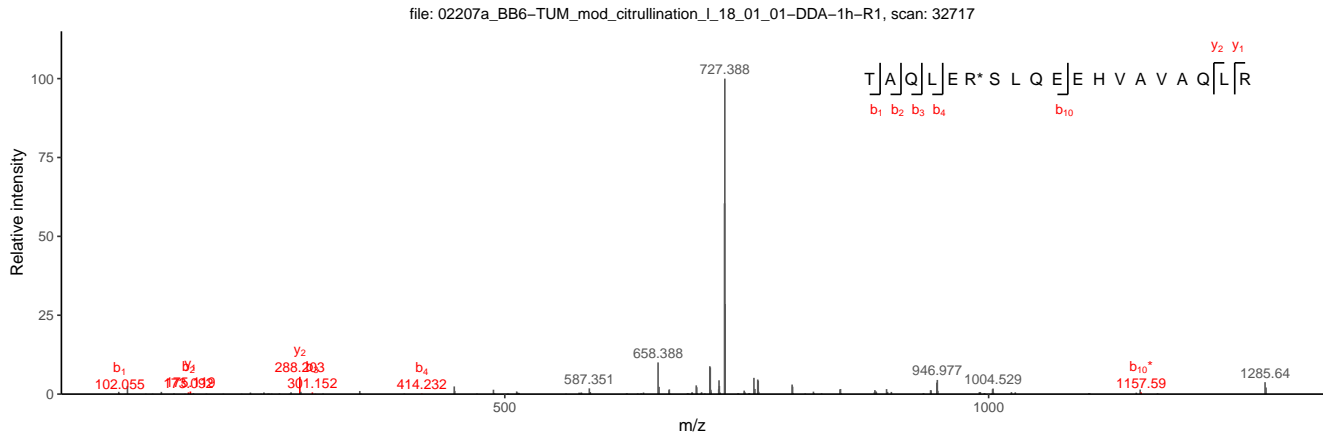


file: 02207a_BA1-TUM_mod_citrullination_L_1_01_01-DDA-1h-R1, scan: 14414



Ground truth: false, ionFinder classification: ambiguous

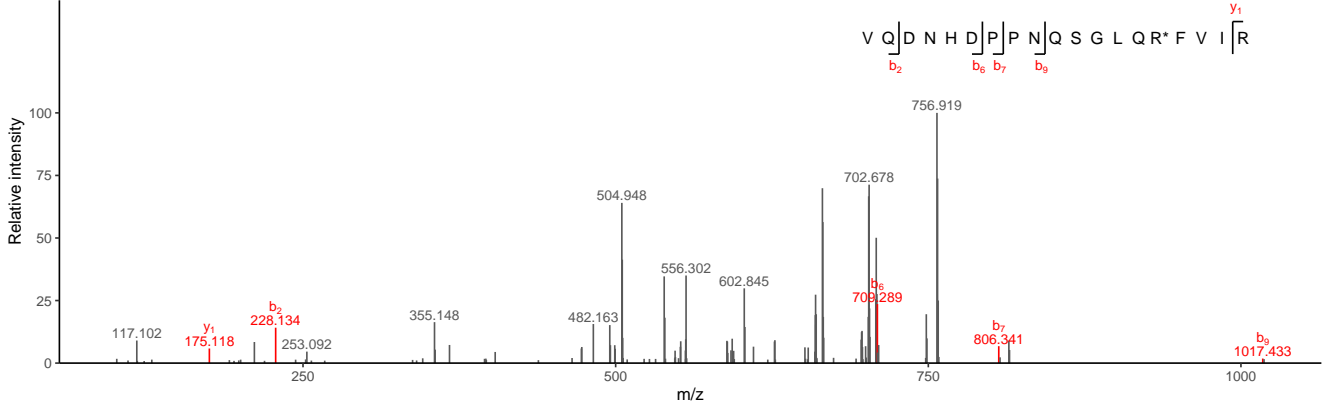
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



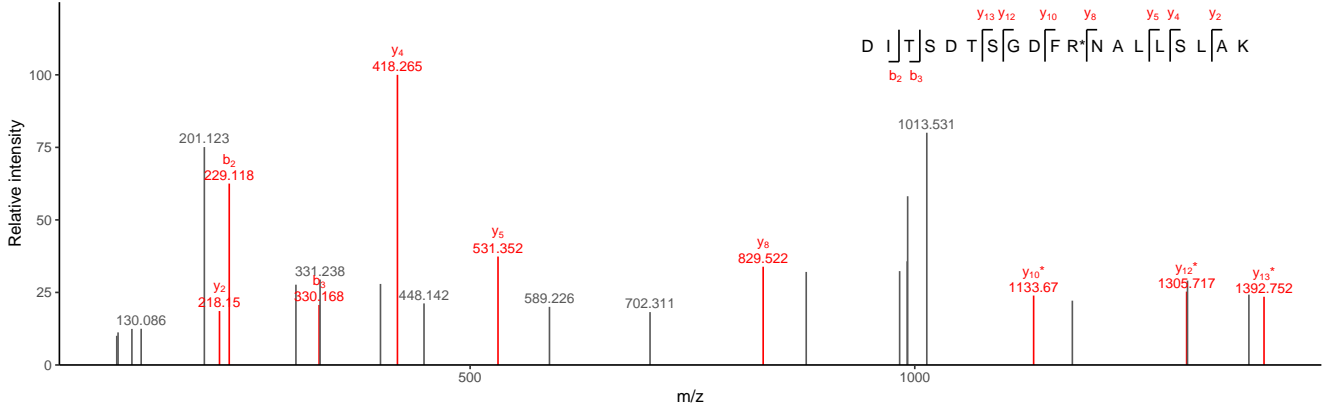
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

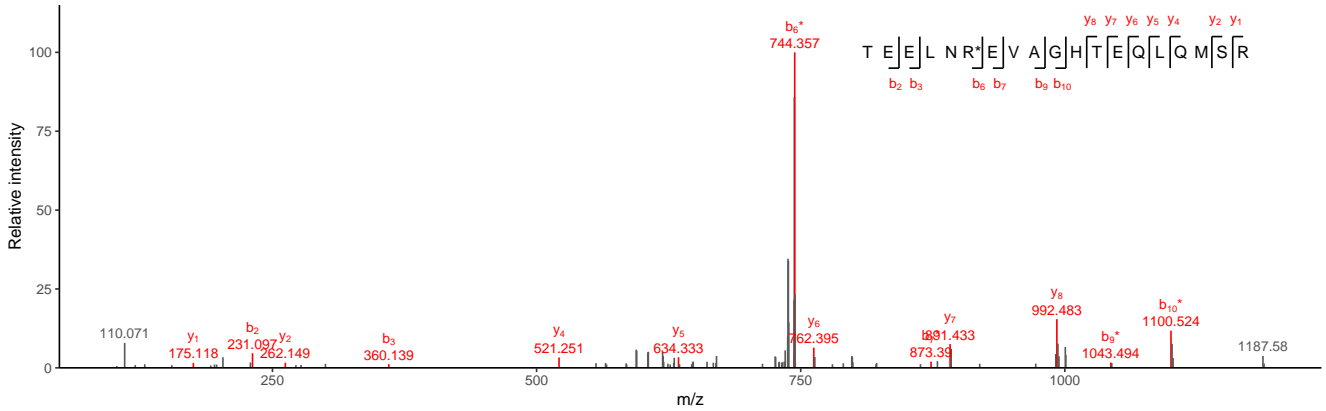
file: 02207a_BB5-TUM_mod_citrullination_I_17_01_01-DDA-1h-R1, scan: 33276



file: 02207a_BB6-TUM_mod_citrullination_I_18_01_01-DDA-1h-R1, scan: 56941

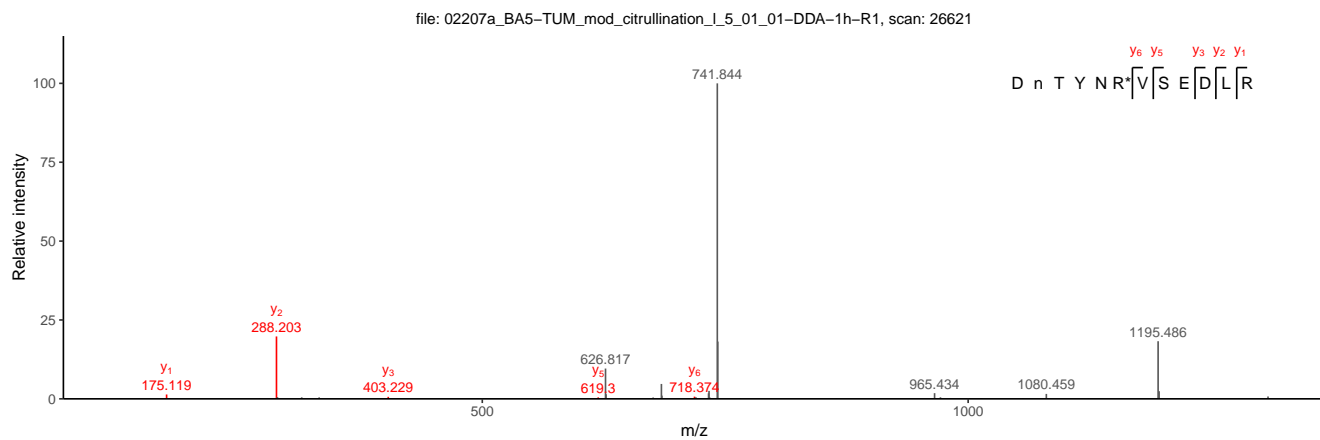
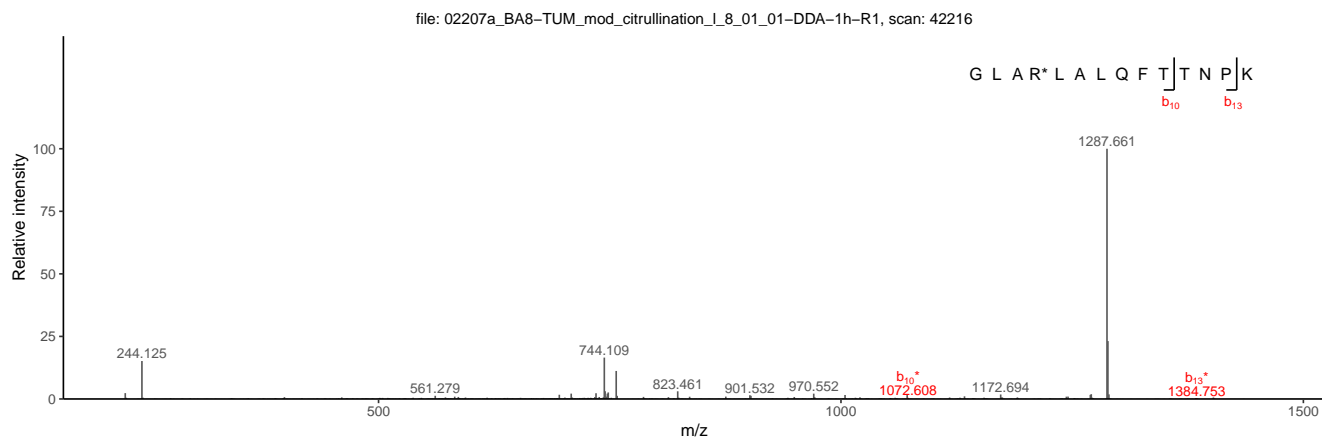
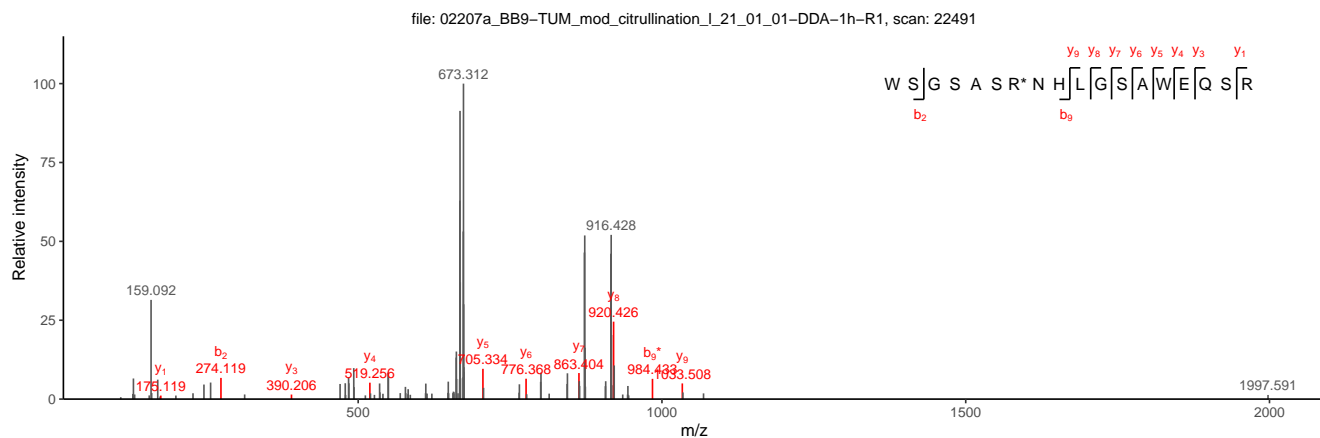


file: 02207a_BB7-TUM_mod_citrullination_I_19_01_01-DDA-1h-R1, scan: 26556



Ground truth: false, ionFinder classification: ambiguous

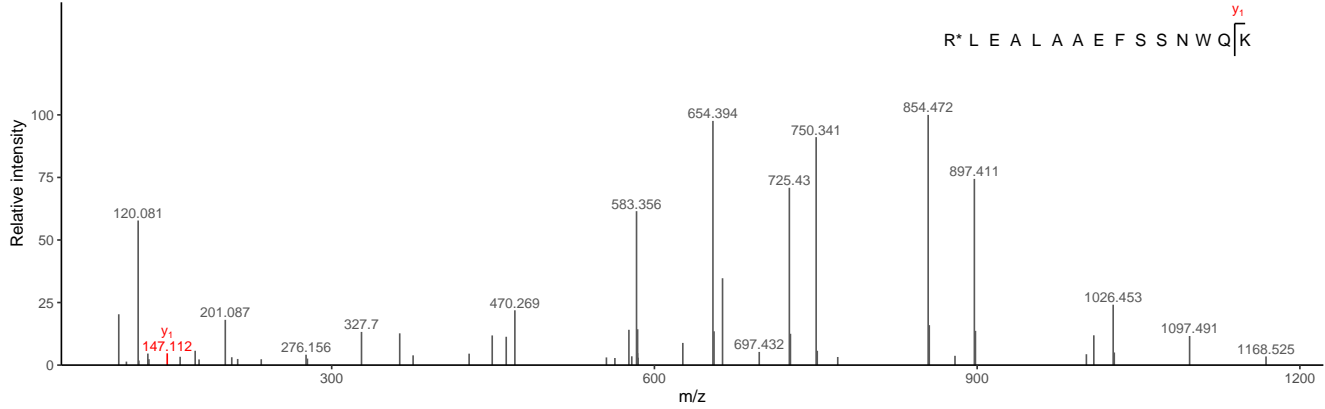
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



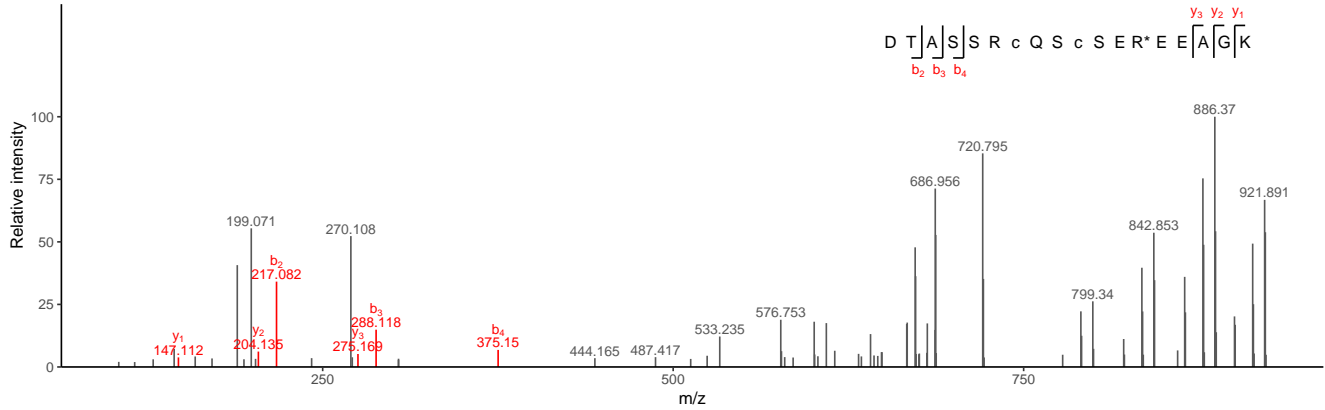
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

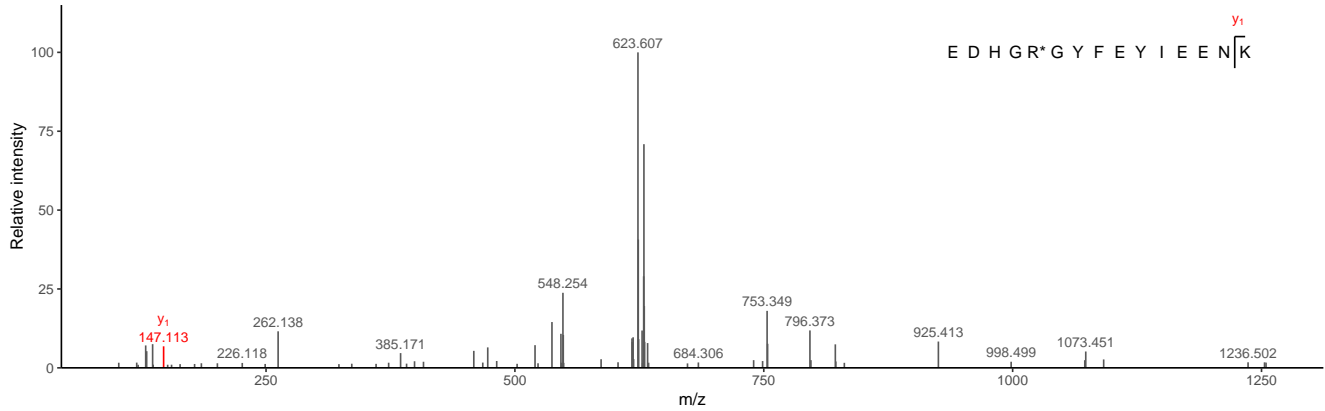
file: 02207a_BA11-TUM_mod_citrullination_L11_01_01-DDA-1h-R1, scan: 42591



file: 02207a_BB4-TUM_mod_citrullination_L16_01_01-DDA-1h-R1, scan: 4973

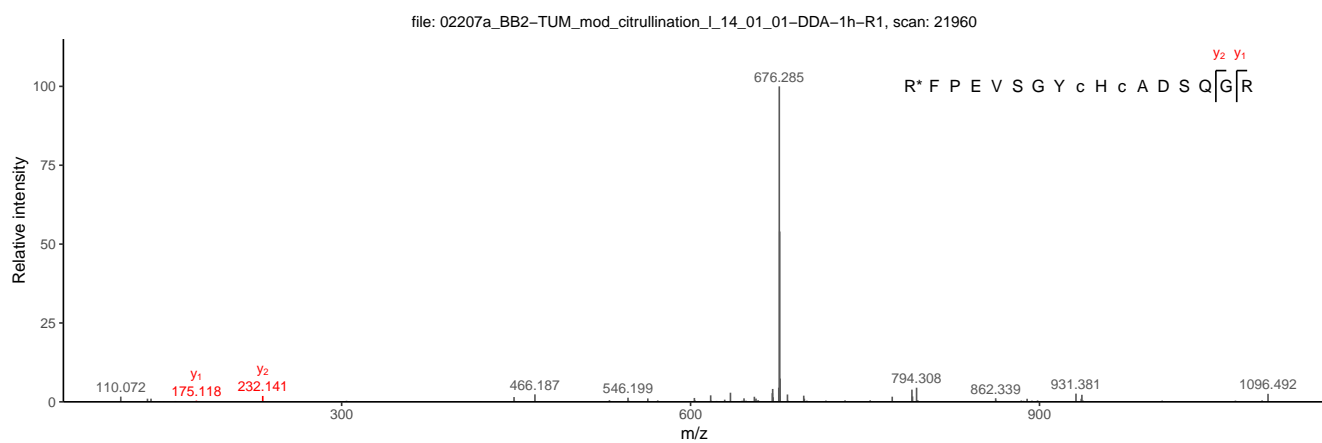
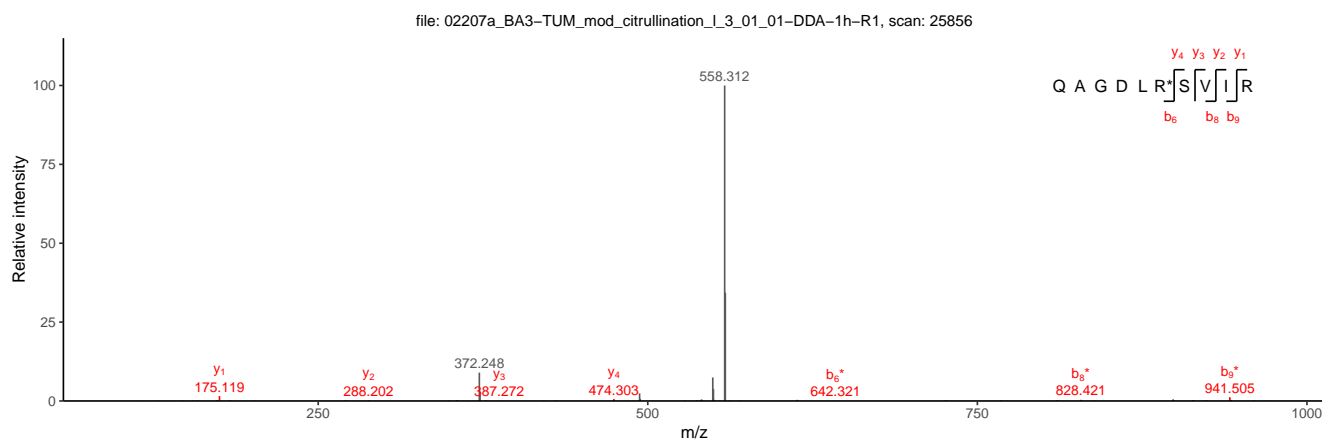
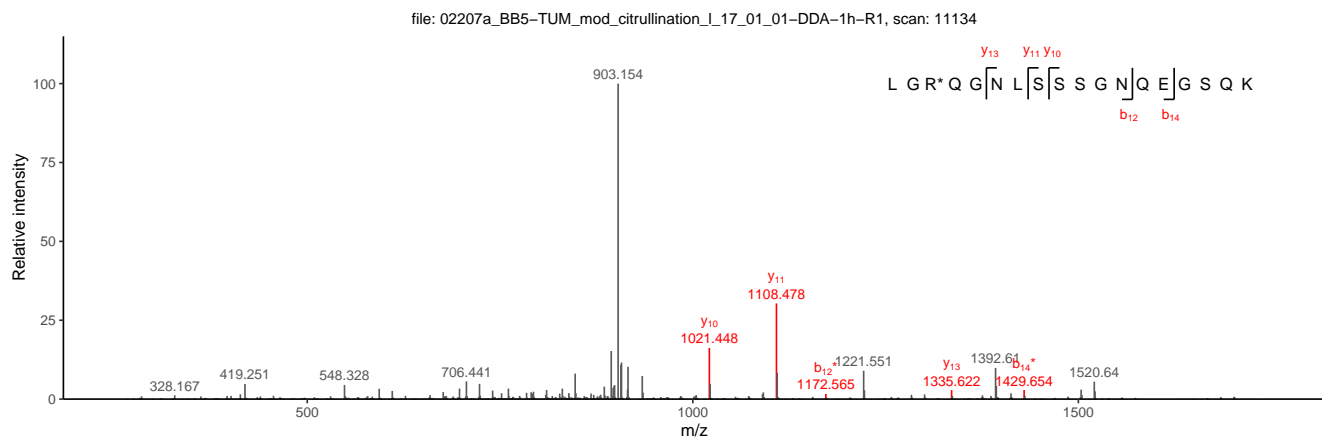


file: 02207a_BA11-TUM_mod_citrullination_L11_01_01-DDA-1h-R1, scan: 31936



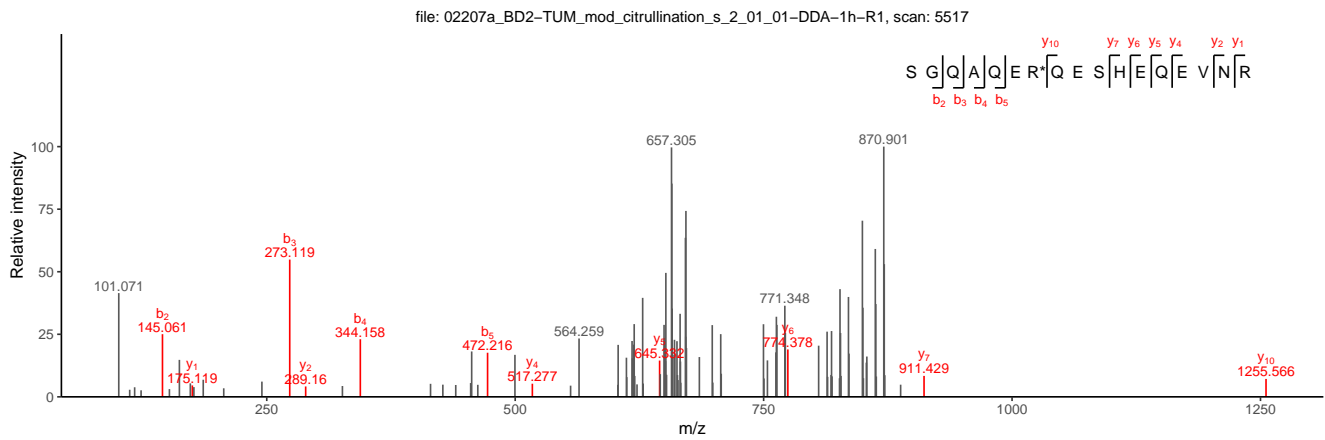
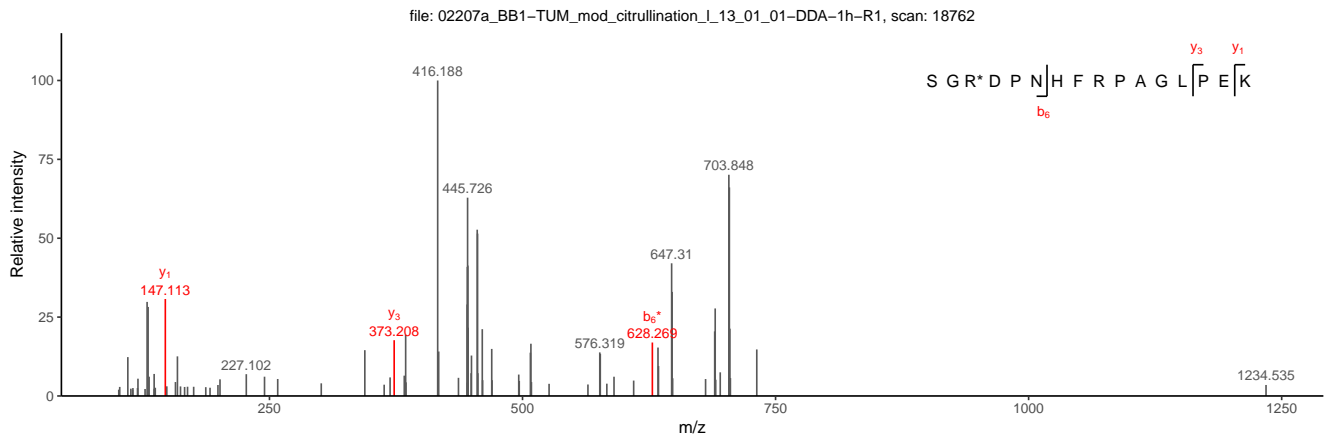
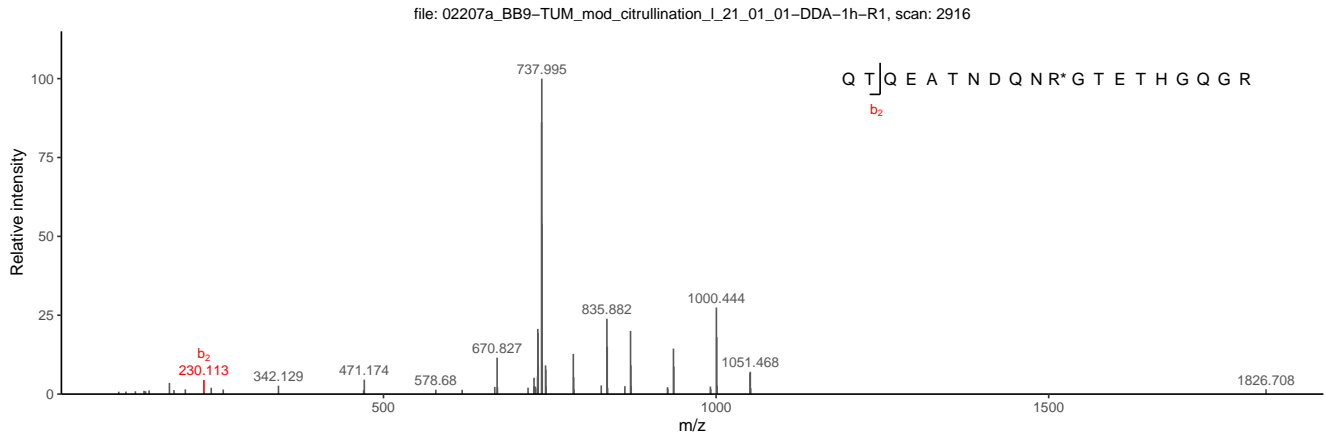
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



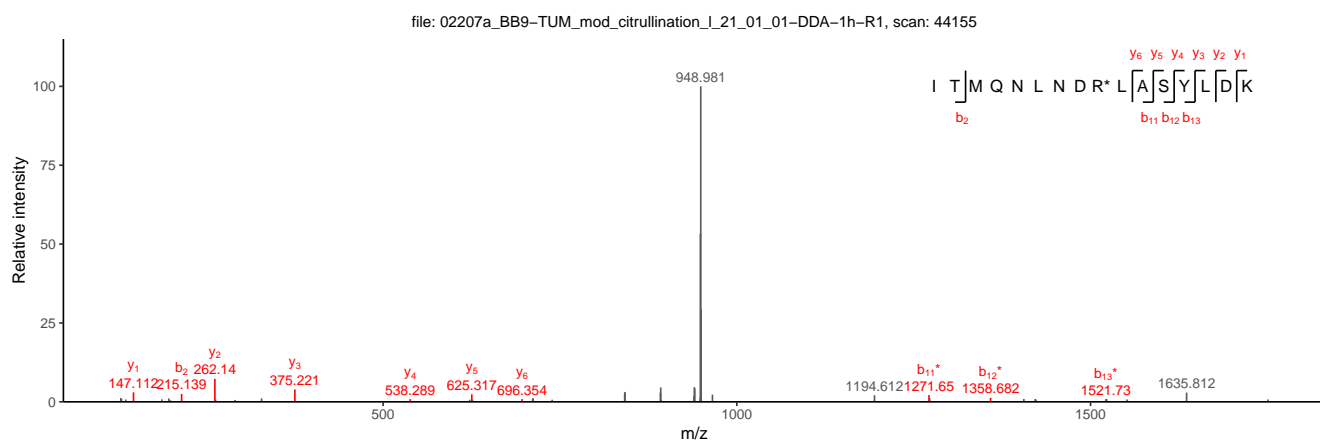
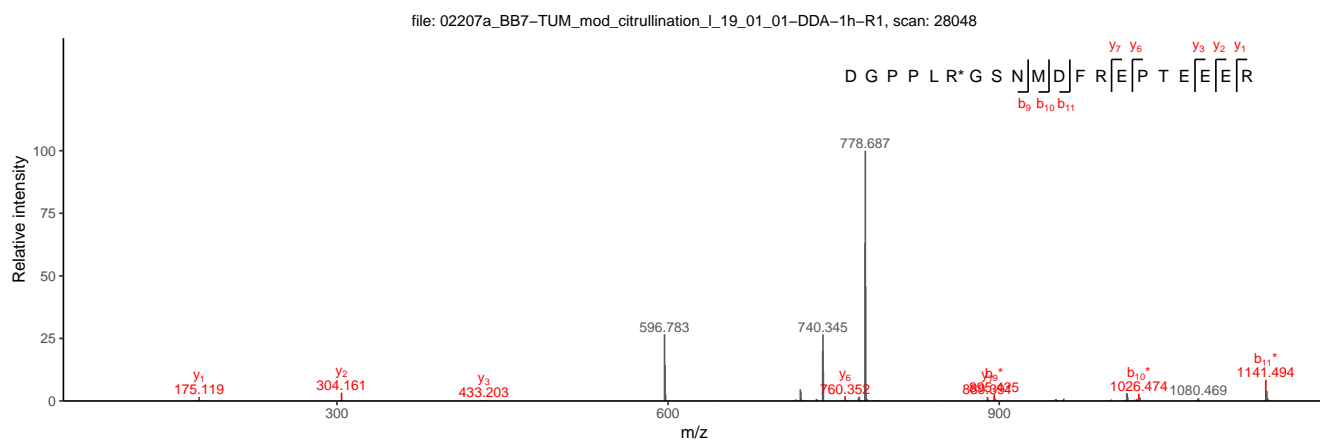
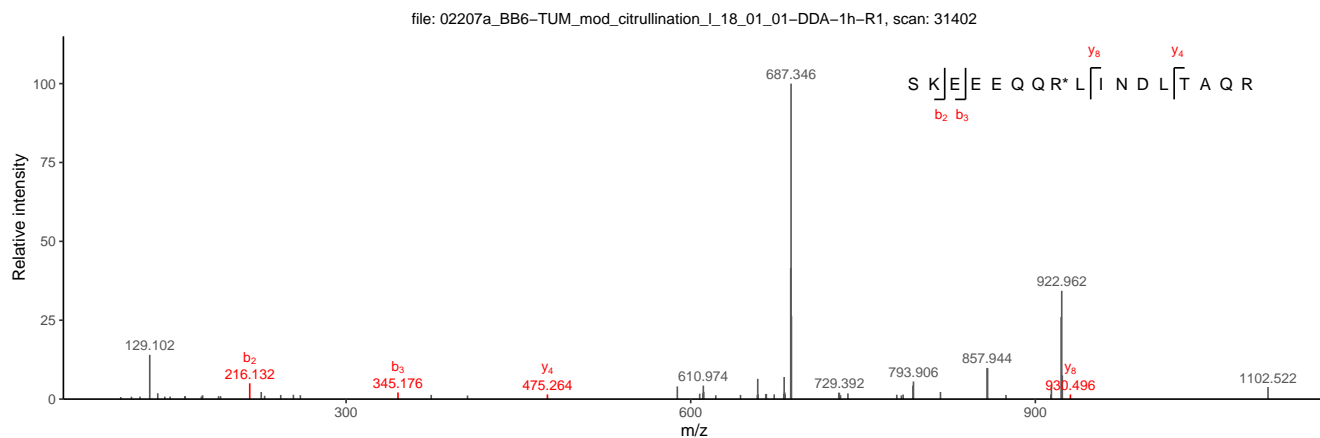
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



Ground truth: false, ionFinder classification: ambiguous

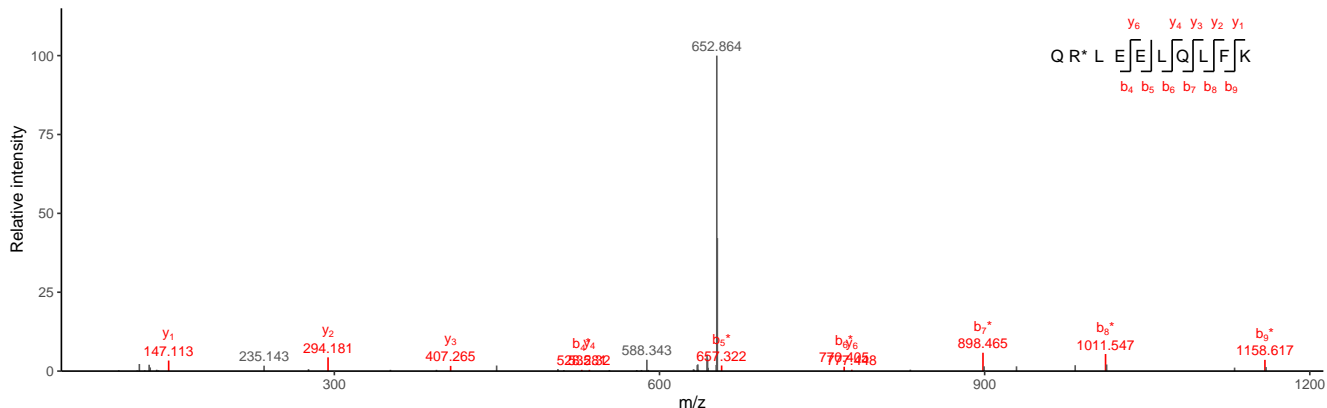
Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



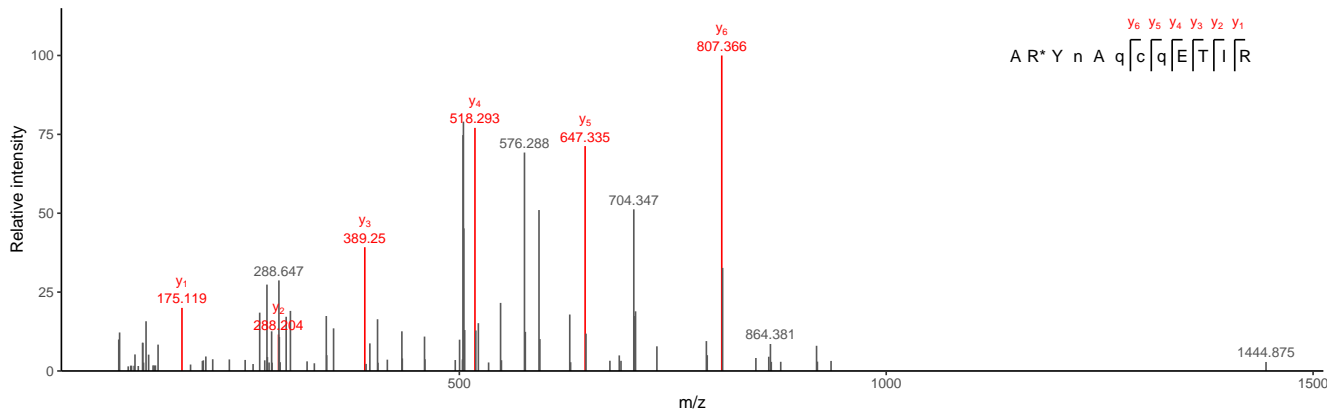
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL

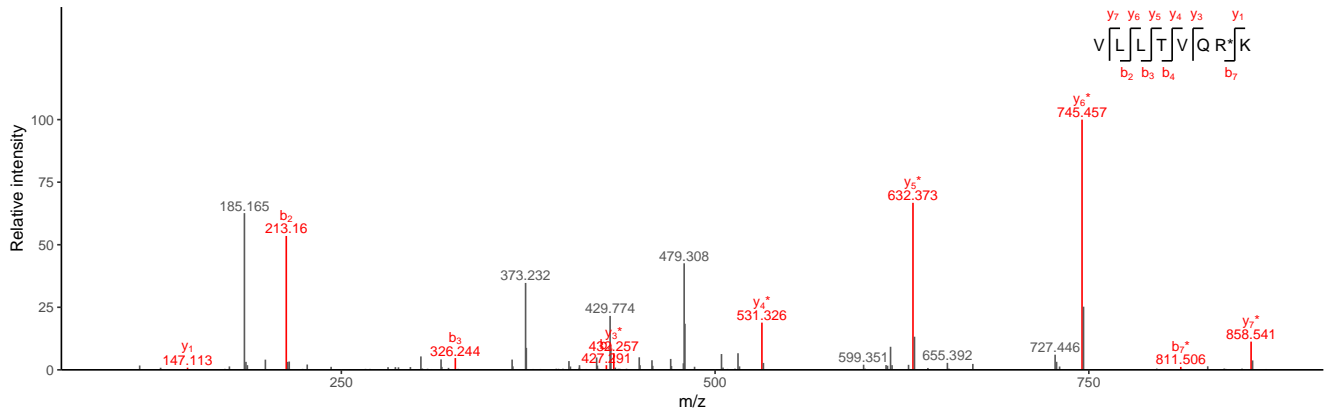
file: 02207a_BA2-TUM_mod_citrullination_L_2_01_01-DDA-1h-R1, scan: 41481



file: 02207a_BA6-TUM_mod_citrullination_L_6_01_01-DDA-1h-R1, scan: 14390

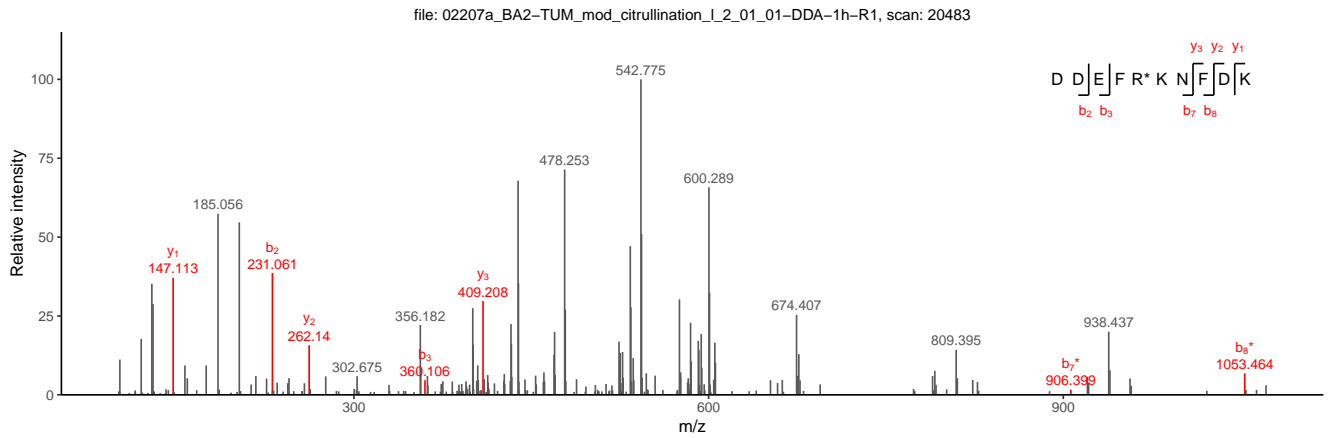
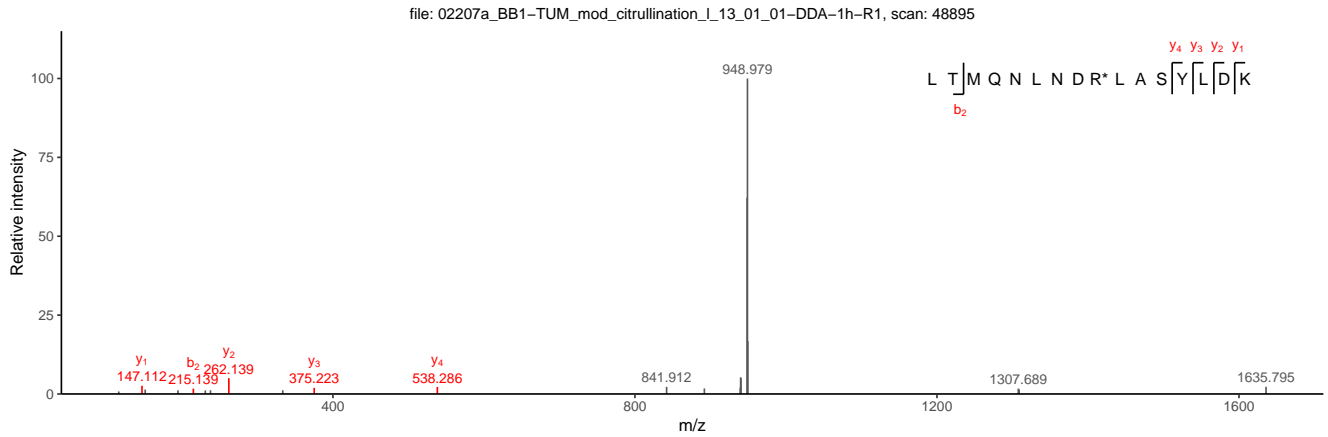
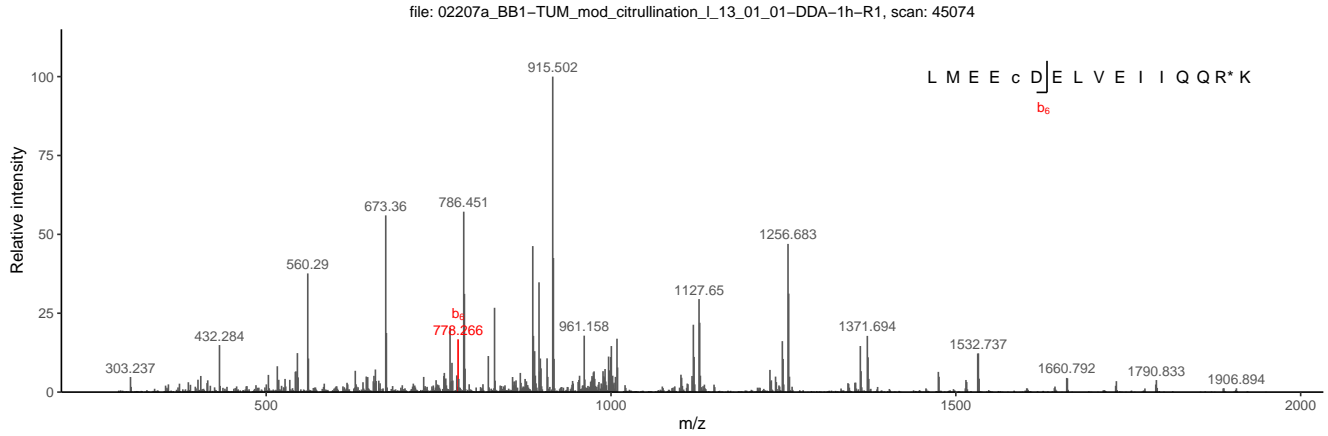


file: 02207a_BA1-TUM_mod_citrullination_L_1_01_01-DDA-1h-R1, scan: 21089



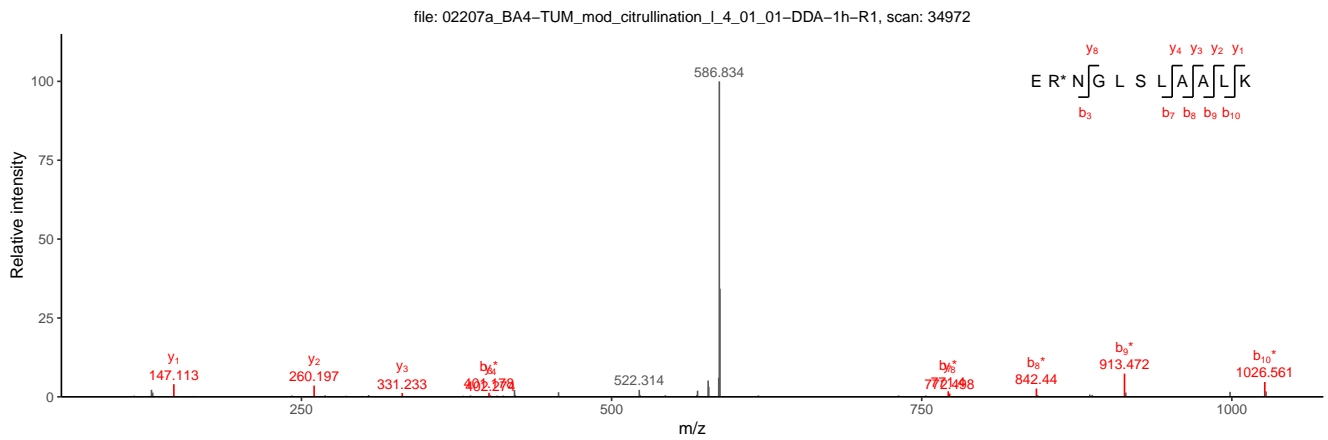
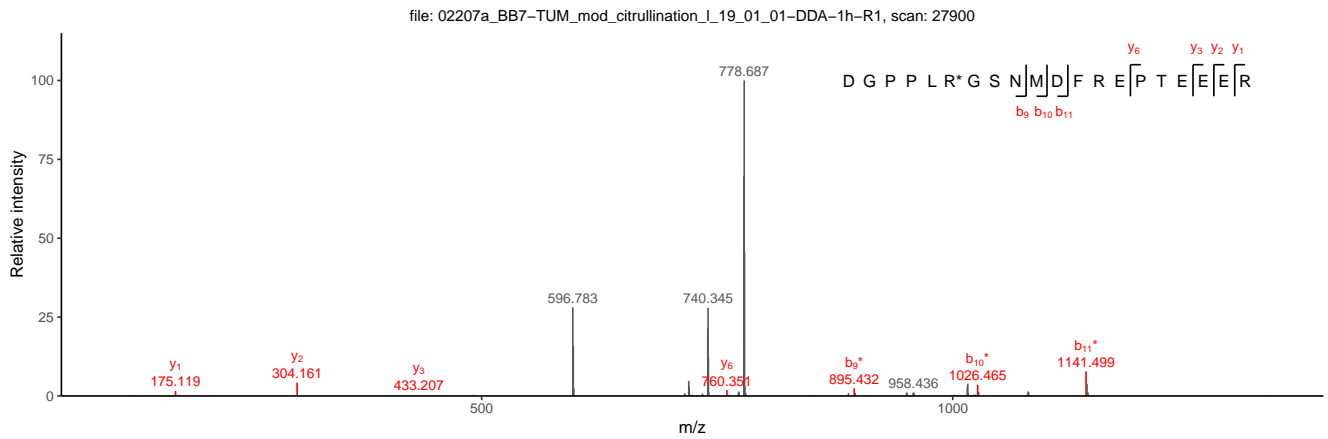
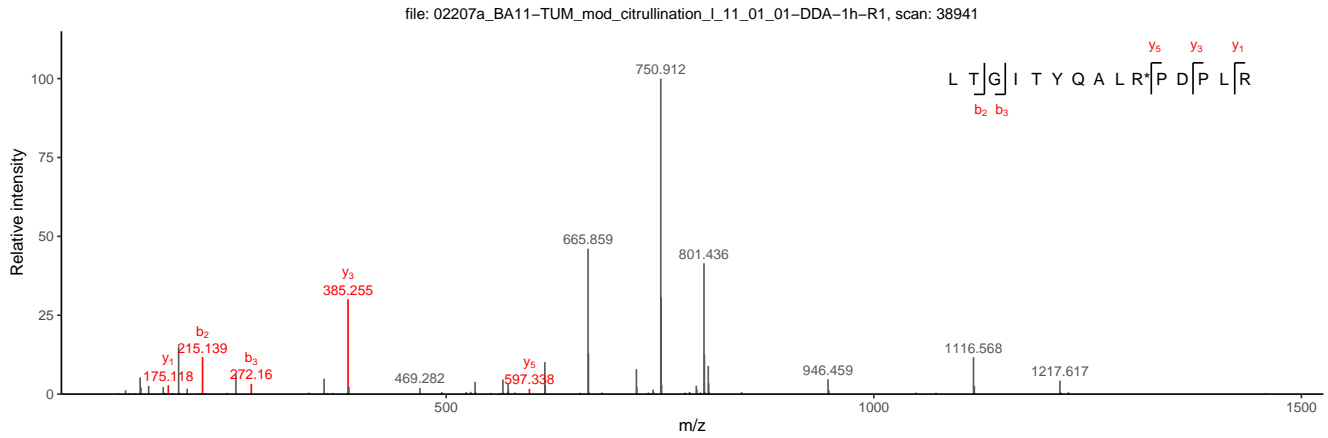
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



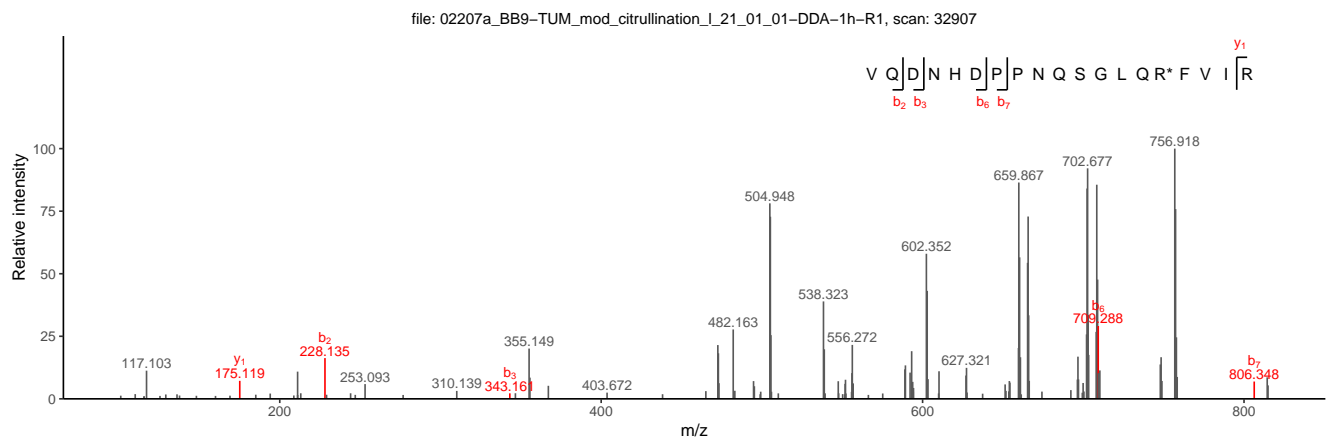
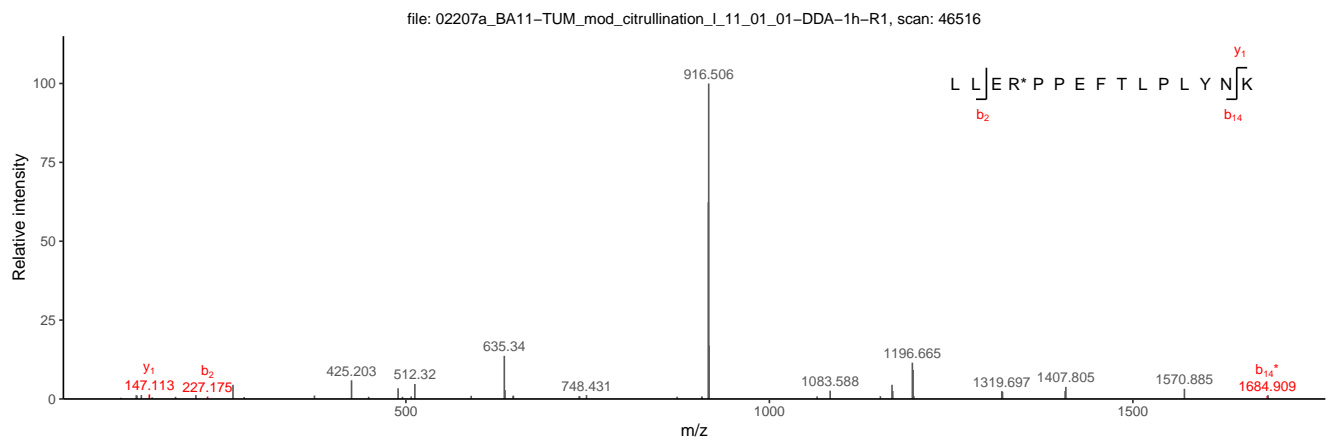
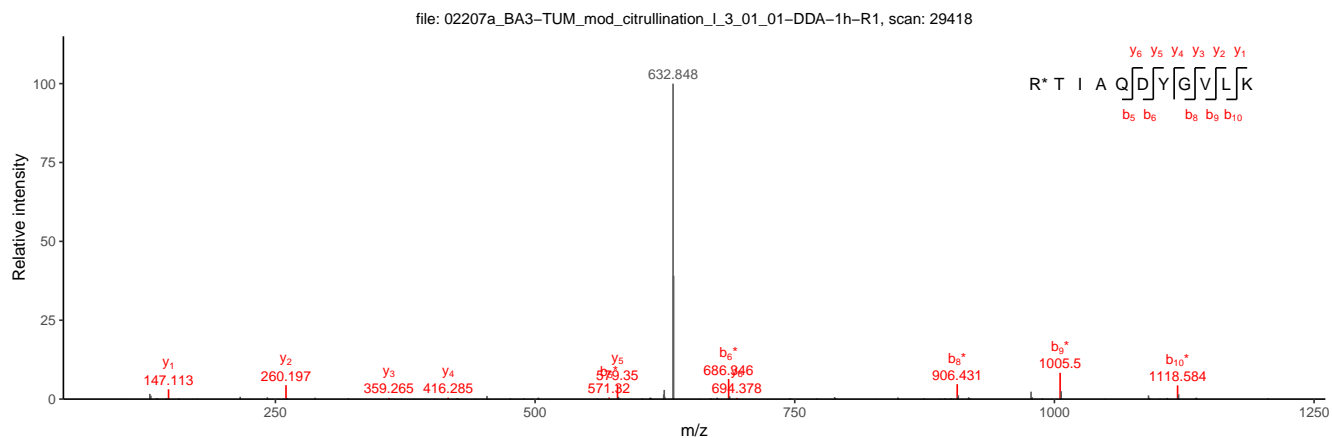
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



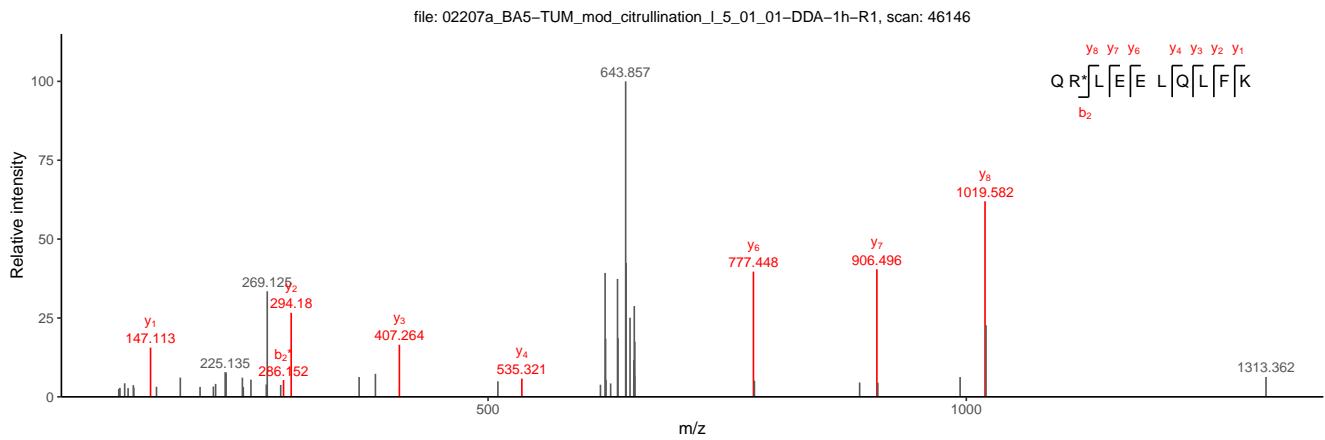
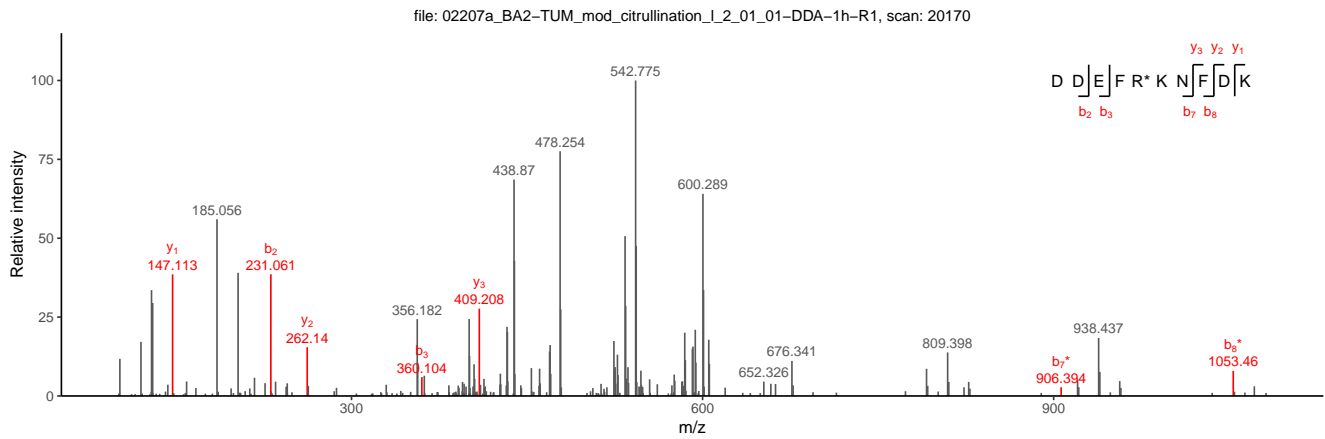
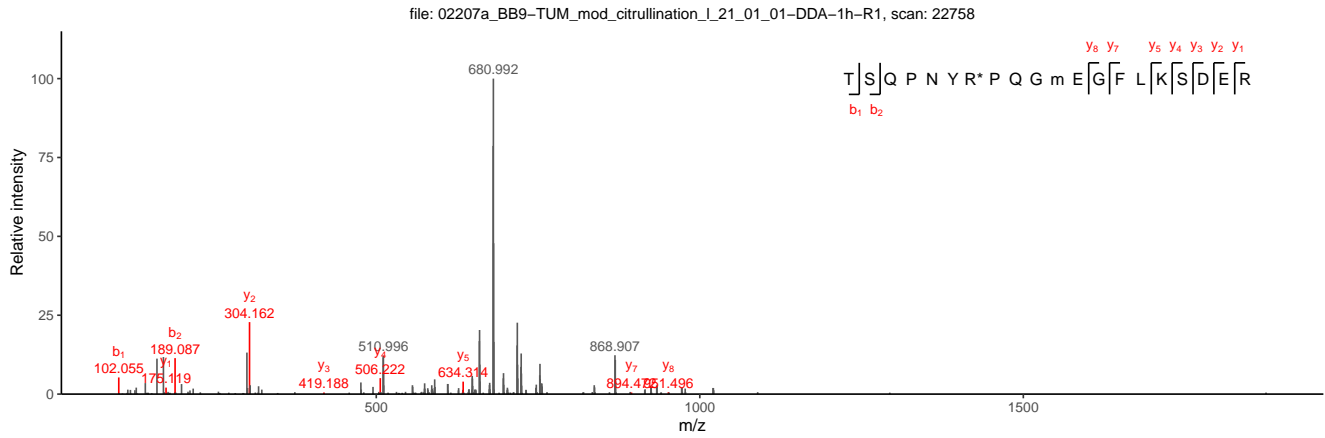
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



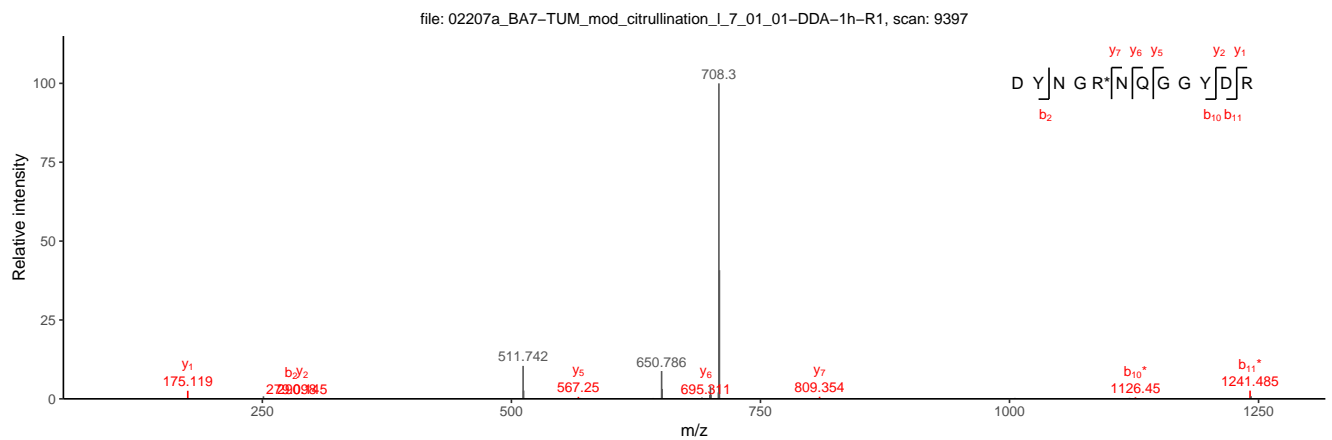
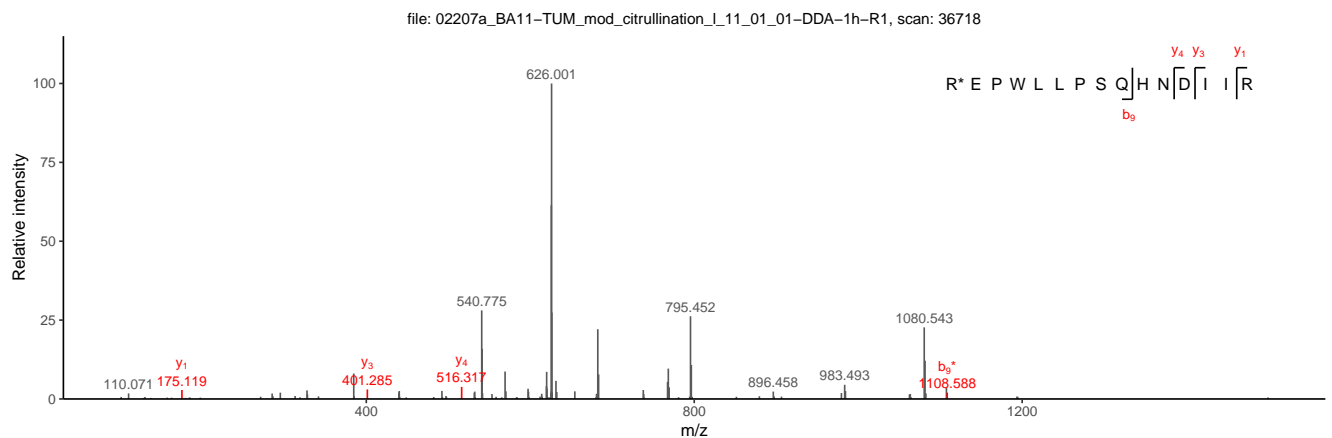
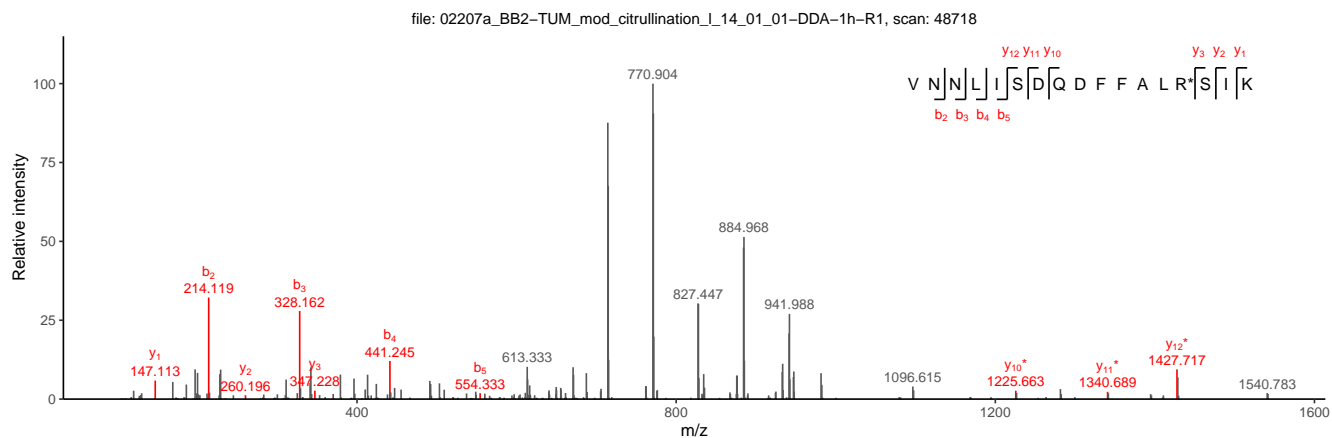
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



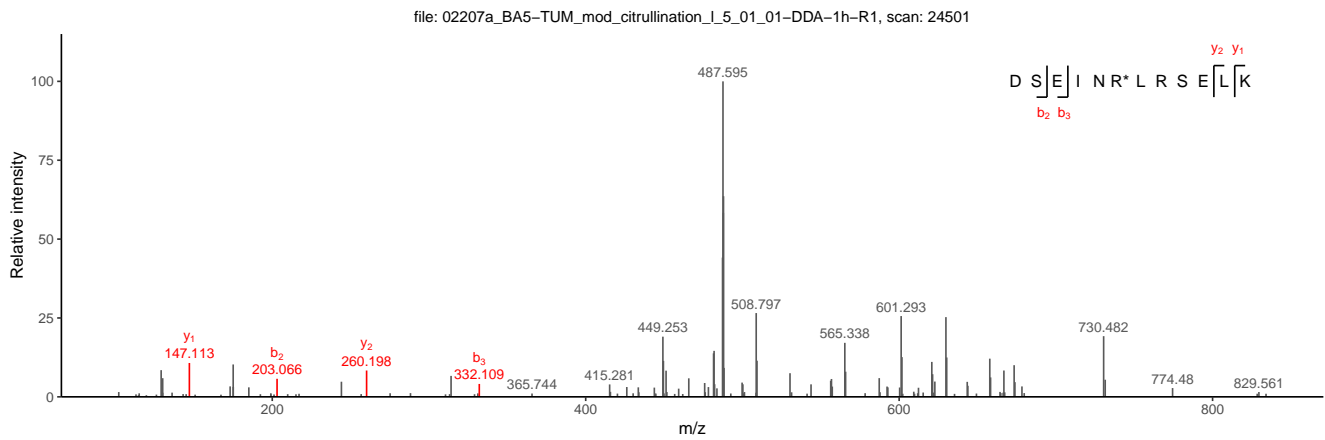
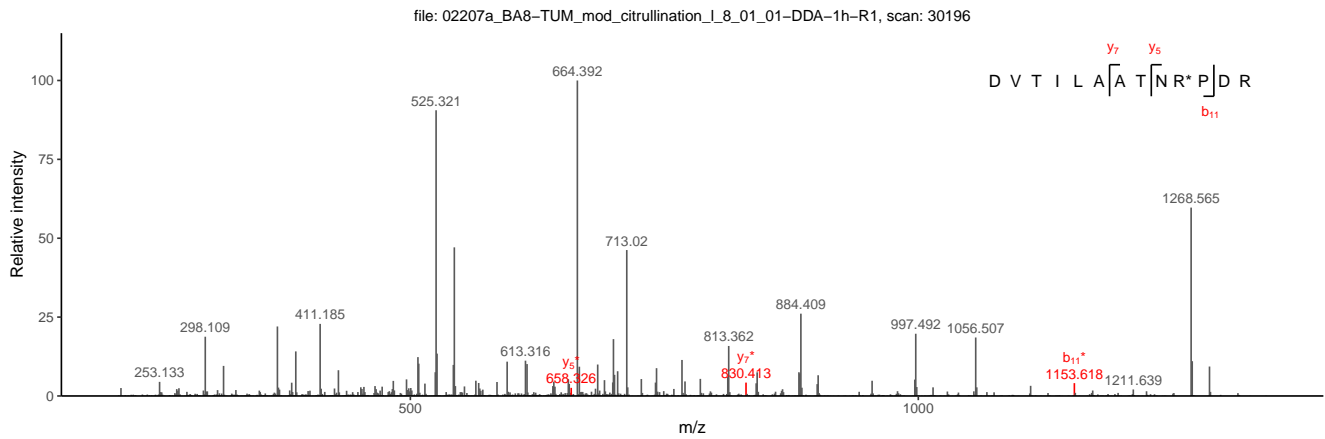
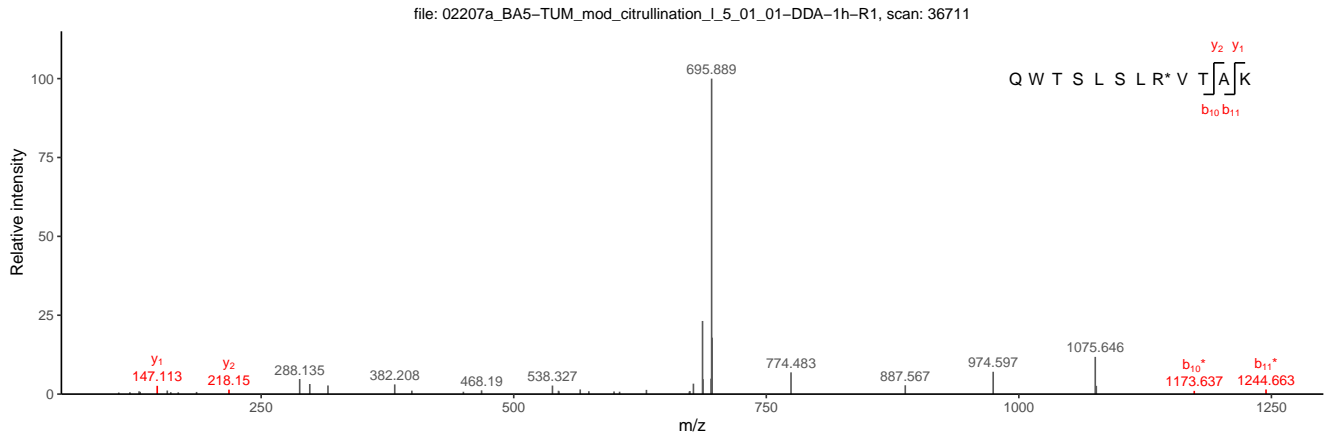
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



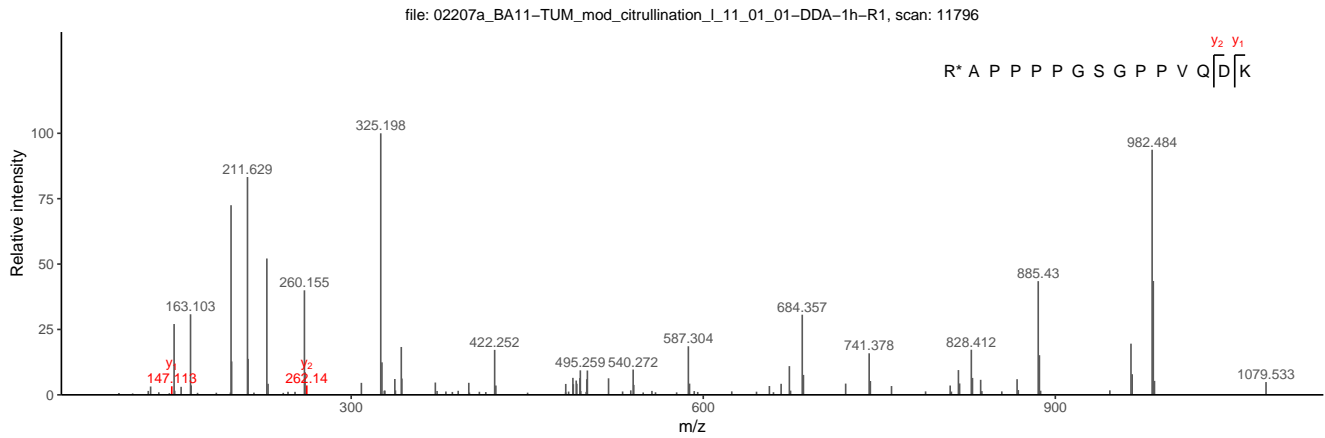
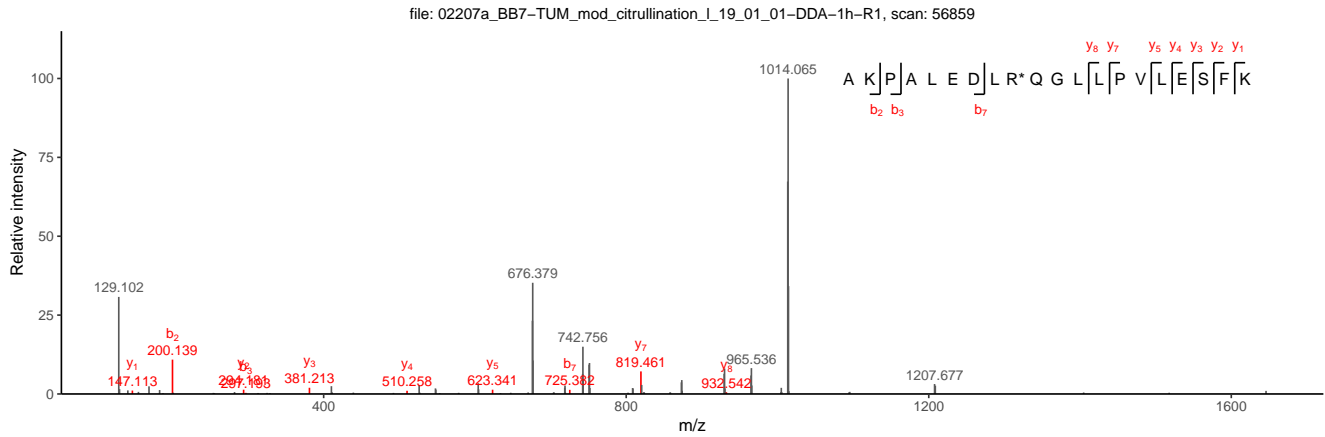
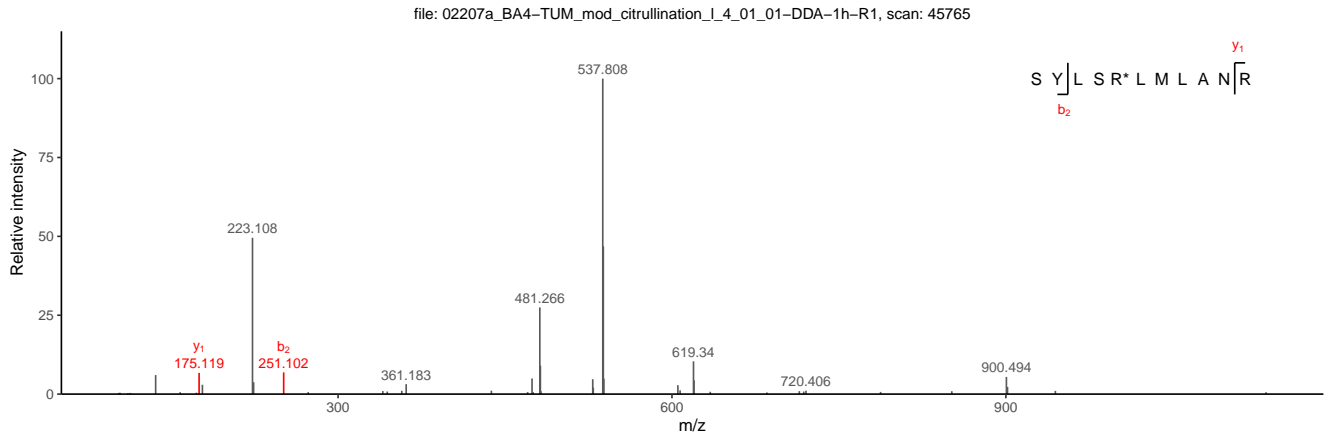
Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



Ground truth: false, ionFinder classification: ambiguous

Ion type ■ Det ■ Amb ■ DetNL ■ ArtNL



Ground truth: false, ionFinder classification: ambiguous

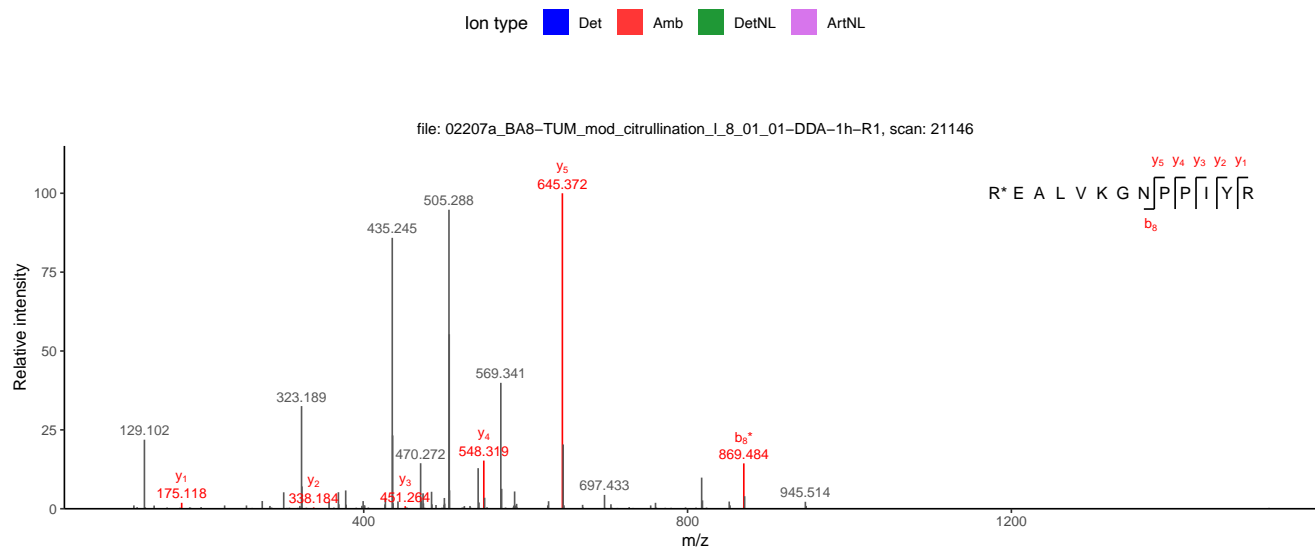


Figure S6. MS² spectra from 64 PSMs of synthetic spectra which were identified as *ambiguous* by ionFinder and classified as *false* in the ground truth dataset.

Ground truth: false, ionFinder classification: false

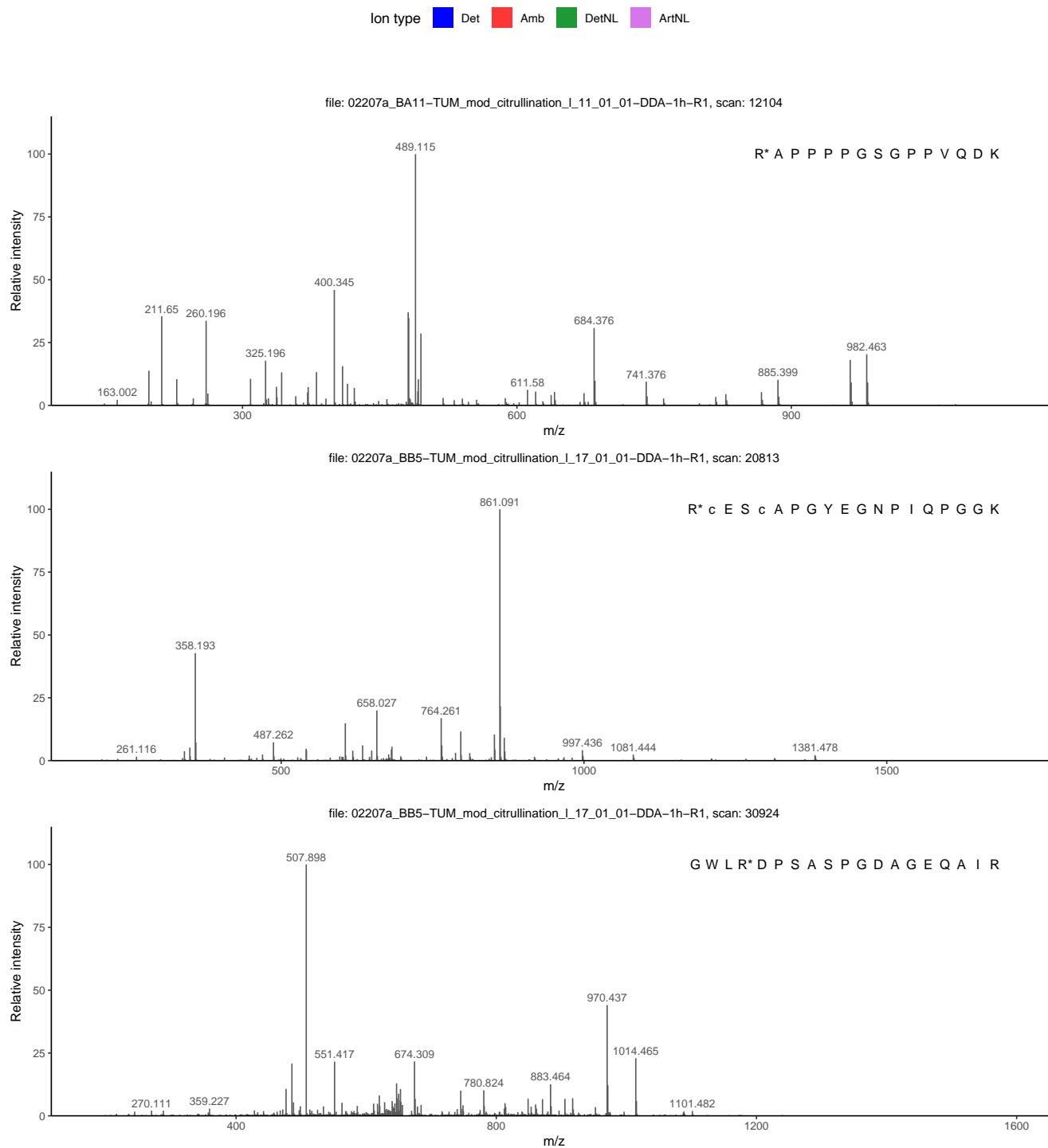


Figure S7. MS² spectra from 3 PSMs of synthetic spectra which were identified as *false* by ionFinder and classified as *false* in the ground truth dataset.

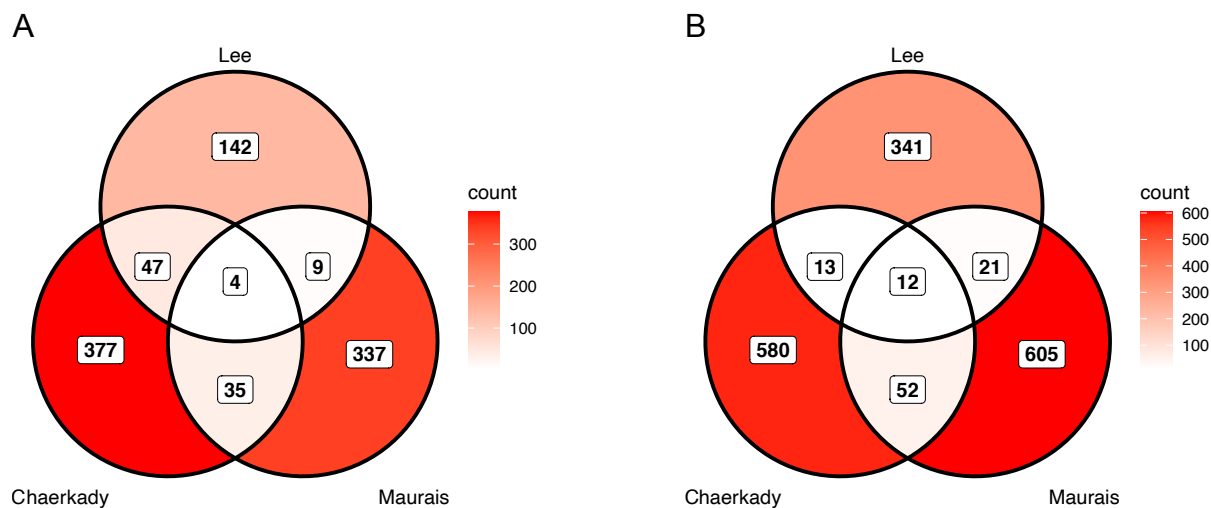


Figure S8. Number of overlapping citrulline containing (A) proteins and (B) peptides in this work, Lee *et al.*,¹ and Chaerkady *et al.*²

References

- (1) Lee, C. Y.; Wang, D.; Wilhelm, M.; Zolg, D. P.; Schmidt, T.; Schnatbaum, K.; Reimer, U.; Ponten, F.; Uhlen, M.; Hahne, H.; Kuster, B. Mining the Human Tissue Proteome for Protein Citrullination. *Mol Cell Proteomics* **2018**, *17*, 1378–1391.
- (2) Chaerkady, R.; Zhou, Y.; Delmar, J. A.; Weng, S. H. S.; Wang, J.; Awasthi, S.; Sims, D.; Bowen, M. A.; Yu, W.; Cazares, L. H.; Sims, G. P.; Hess, S. Characterization of Citrullination Sites in Neutrophils and Mast Cells Activated by Ionomycin via Integration of Mass Spectrometry and Machine Learning. *J Proteome Res* **2021**, *20*, 3150–3164.