

**A**

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Protein <sup>a</sup>	Modified peptide <sup>b</sup>	Position <sup>c</sup>	Obs. <sup>d</sup>	Calc. <sup>e</sup>	dM <sup>f</sup>	dM <sup>g</sup>	sequence found with modifications	HC
	ITPNLAEFA <sub>58</sub> (+156)FSLYR	50 - 63	899.48439	899.48425	-9.9	2	-	
A1AT	A <sub>360</sub> (+138)VLTIDEK	360 - 367	512.8005	513.8065	-1.006	-4.7	+	

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a. Type of modification.

b. Site of modified peptide.

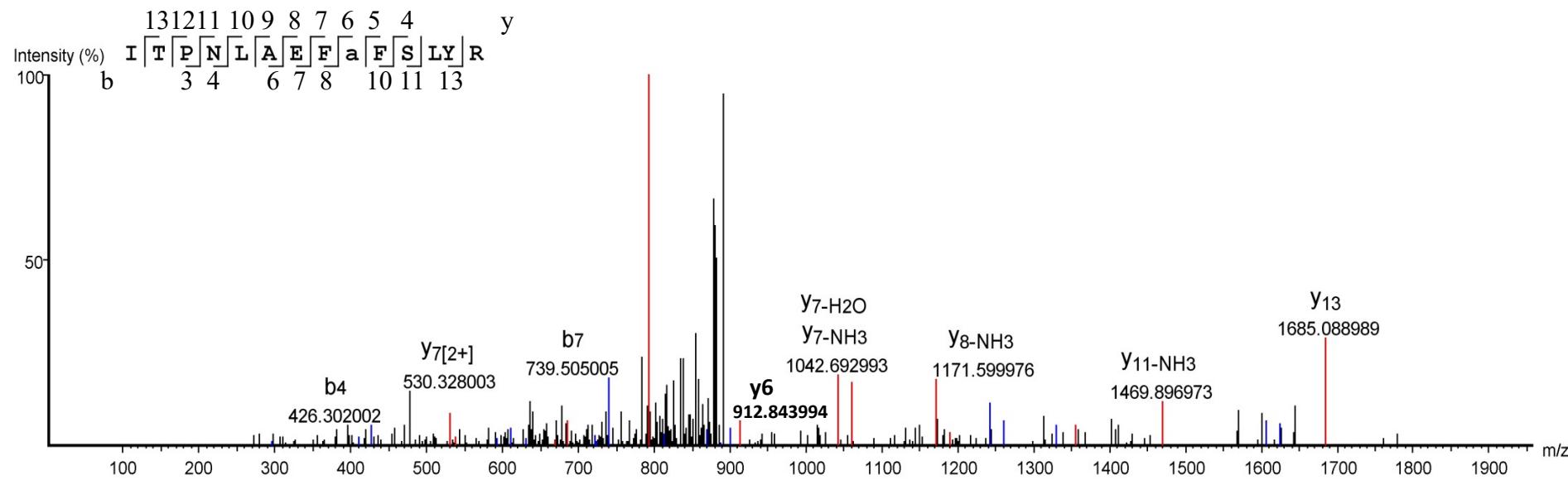
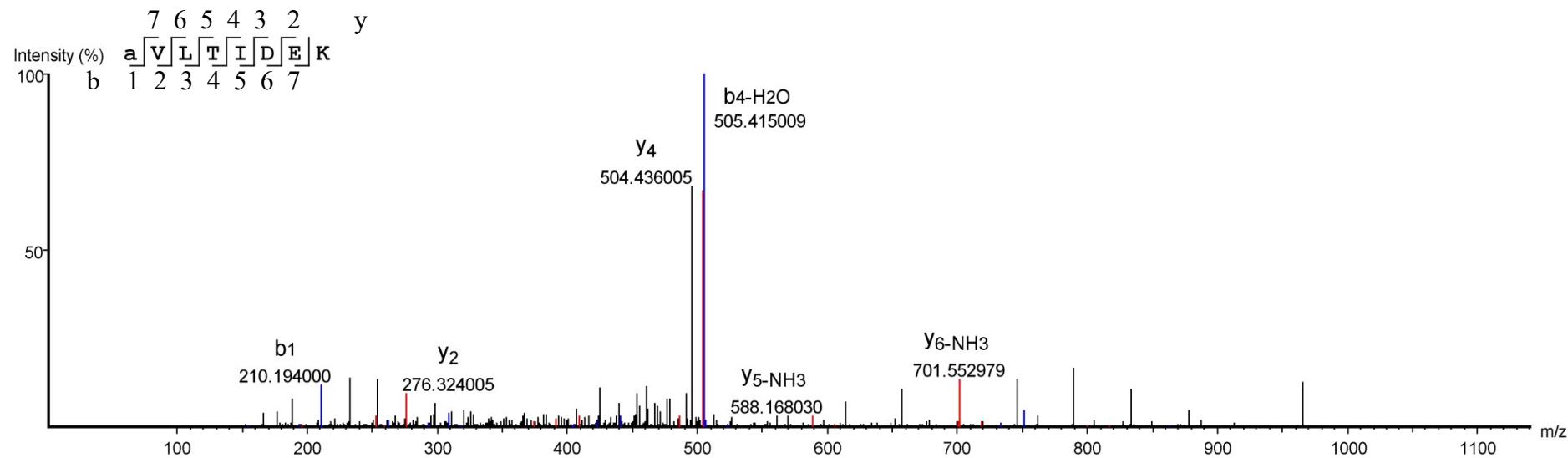
c. Amino acid positions of the first and the last residues in accession number (P01009).

d. Obs.: observed m/z of the modified peptides.

e. Calc.: calculated m/z of the modified peptides.

f. dM (Obs.-Calc.): mass accuracy.

g. Modified peptide mass accuracy (ppm).

**B****pSS****HC**

C

## Serum

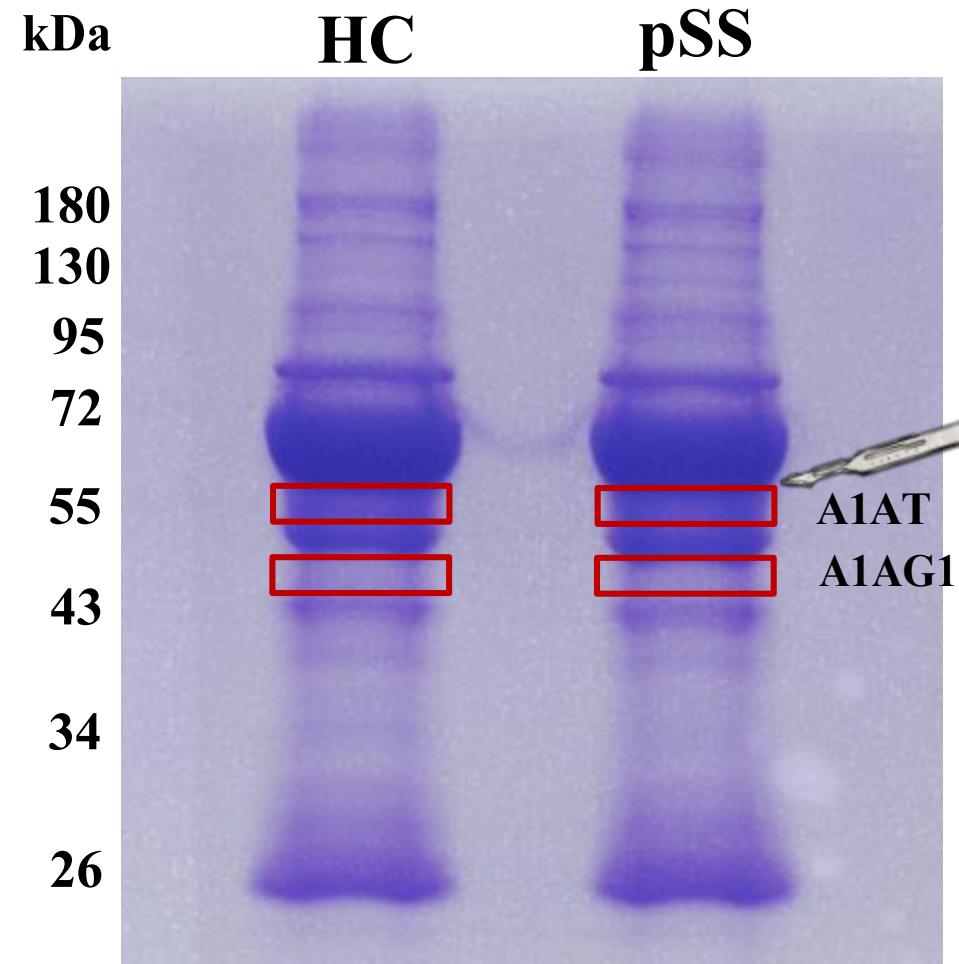


Figure S1. Identification of 4-hydroxy-2-nonenal (HNE) modifications of A1AT (**A**). A representative MS/MS spectrum of the peptide sequence of <sup>50</sup>-ITPNLAEFAFSLYR-<sup>63</sup> and the modified peptide bearing the HNE modification at alanine 58 in primary Sjögren's syndrome (pSS) (**B**, upper panel). The MS/MS spectrum of <sup>360</sup>-AVLTIDEK-<sup>367</sup> and the modified peptide bearing the HNE modification at alanine 360 in healthy controls (HCs) (**B**, bottom panel). The gel was rapidly stained with Coomassie brilliant blue, and gel bands were cut into slices according to the molecular weight of A1AG1 (48 kDa) and A1AT (55 kDa), respectively (**C**).