## **Supporting Information**

**Figure S-1.** Spectral complexity contained within a single 2D-LC fraction and demonstration of the enhanced dynamic range obtained with quadrupole marching. a) Broadband MS illustrating two putative forms from deconvolution of low-resolution MS. b) High-resolution broadband MS with no isotopic clusters identified by THRASH. c) Concatenated spectra from "quadrupole marching" windows (6 windows taken) yielding four molecular species, all of which were identified by MS/MS analysis.

**Figure S-2.** Calculating QWASI ratios of alleles from a single human being using Top Down LC-MS. a) Broadband FTMS data summed across entire region of elution of the Charcot-Leyden crystal protein. b) Expanded views of the visible charge states of Charcot-Leyden from a). c) The equation used to determine the expression ratios of both alleles of a heterozygote.







