

Supplementary Figure 1: Tandem mass spectrum of HyD-Crys peptide 15-31 with trioxidation on C18.

The tandem mass spectrum of +2 charged trioxidized H γ D-Crys peptide 15-31 (m/z=1047.4512) is plotted. The calculated monoisotopic mass of this peptide is 2093.8946 Da which is within 2 ppm of the calculated monoisotopic mass of 2093.8982 Da. b- and y- ions are labeled and b- or y- ions with trioxidation are marked with asterisk which clearly indicates trioxidation occurs on C18.





The selected ion chromatograms of the +2 charged H γ D-Crys peptide 99-114 with (m/z=990.39) or without trioxidation on C108 (m/z=994.91) are plotted. The level of C108 trioxidation was very low.

No figure was made for C78 oxidation. C78 is in a very short tryptic peptide 77-79 which was hard to detect in our experiments. C78 oxidation was detected in the peptide 77-88 which has a missed cleavage site. Very often, trypsin will not cleave near a modification site. Therefore, relative intensity of the oxidized peptide, 77-88, compared to the

unmodified peptide is not a true representation of the amount of oxidized C78. Therefore, it was not possible to reliably compare the relative levels of C78 and C18 trioxidation.



Supplementary Figure 3: Selected Ion Chromatograms of HyD-Crys peptide 15-31 in trypsin digested UV-irradiated samples.

 $H\gamma D$ -Crys was irradiated for 60 minutes and the supernatant and pellet were separated as described in the method. Proteins from the supernatant and pellet were digested by trypsin and analyzed by LC-MS/MS. The selected ion chromatograms of the +2 charged $H\gamma D$ -Crys peptide 15-31 (m/z=1047.45, peak B) with and without trioxidation (m/z=1051.97, peak A) are plotted. The results indicate that the level of trioxidation of C18 was much higher in the pellet than in the supernatant.



Supplementary Figure 4: Turbidity Curves of WT $H\gamma$ D-Crys Removing the UVR Source at Various Times.

HγD-Crys was irradiated for the indicated time period and then the UV lamp was covered up while turbidity measurements continued. Samples contained protein at 1 mg/ml in reaction buffer, and light scattering was monitored at 600 nm as a function of UVR exposure time. 60 minutes (blue diamonds), 45 minutes (red squares), 30 minutes (green triangles), 15 minutes (purple X's), 5 minutes (turquoise hashes).