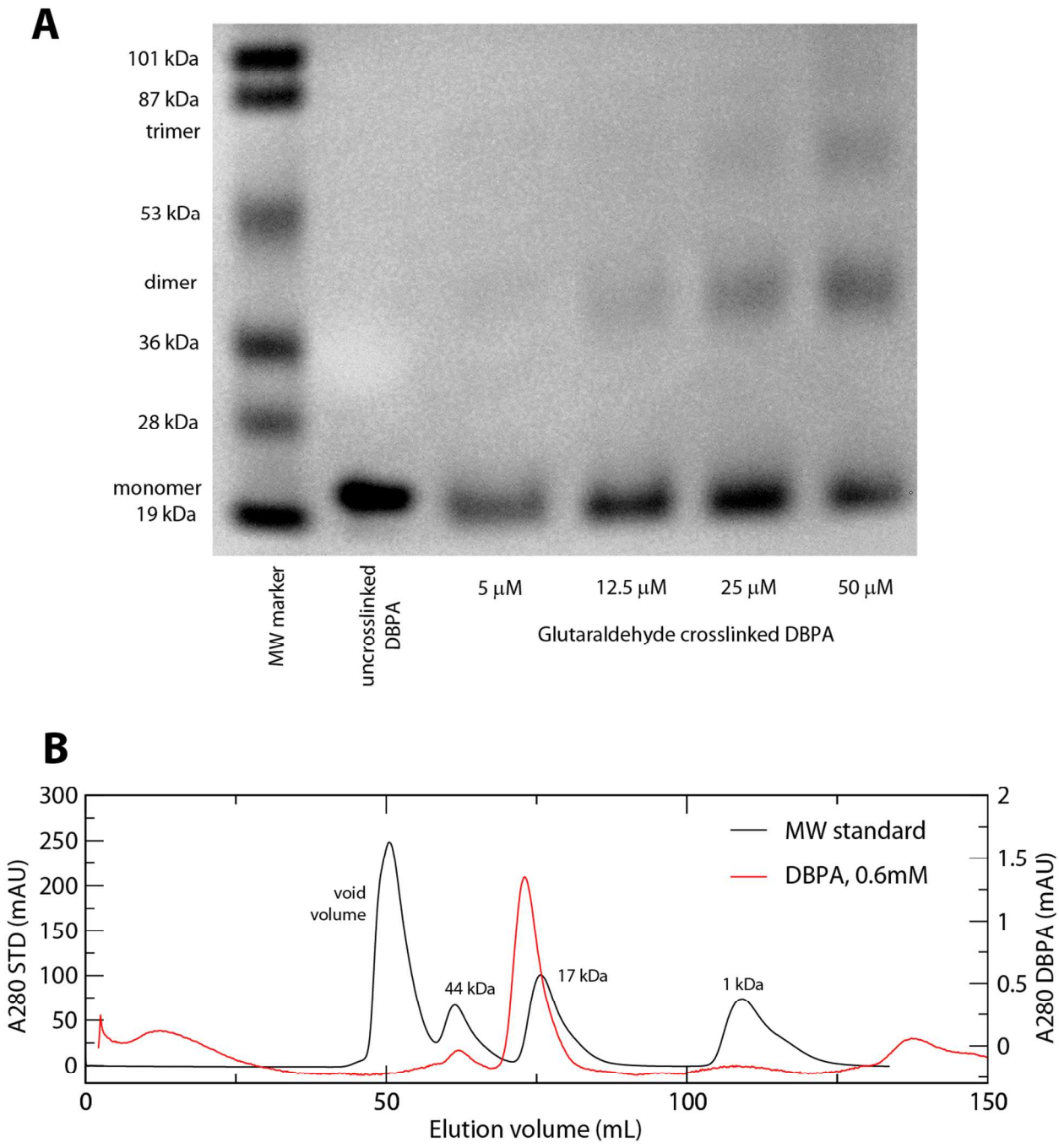

Figure S1. A) SDS-PAGE analysis of glutaraldehyde-crosslinked DBPA. Lane 1: MW marker; lane 2: uncrosslinked DBPA; lane 3 to 6: DBPA cross linked at increasing protein concentrations. B) size exclusion chromatogram of 0.6 mM DBPA (red) and molecular weight standards (black). Please consult the experimental methods section for experimental methods on cross linking and SEC chromatography.

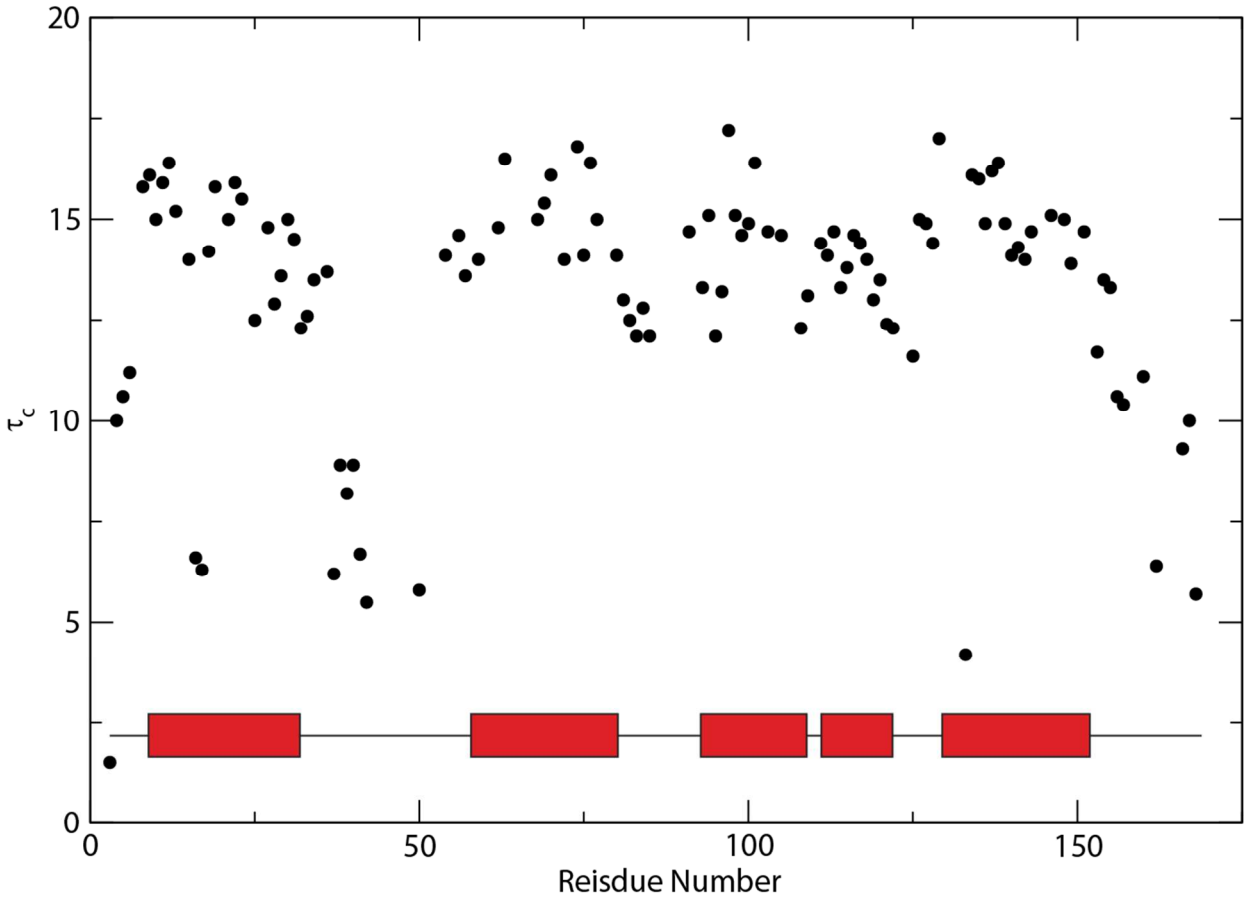

Figure S2. Rotational correlation time of DBPA in the absence of GAGs. Secondary structure of the protein is shown schematically with red rectangles representing alpha helical regions and straight line representing unstructured regions. Note the flexibility in the linker between helices one and two as well at the C-terminus.

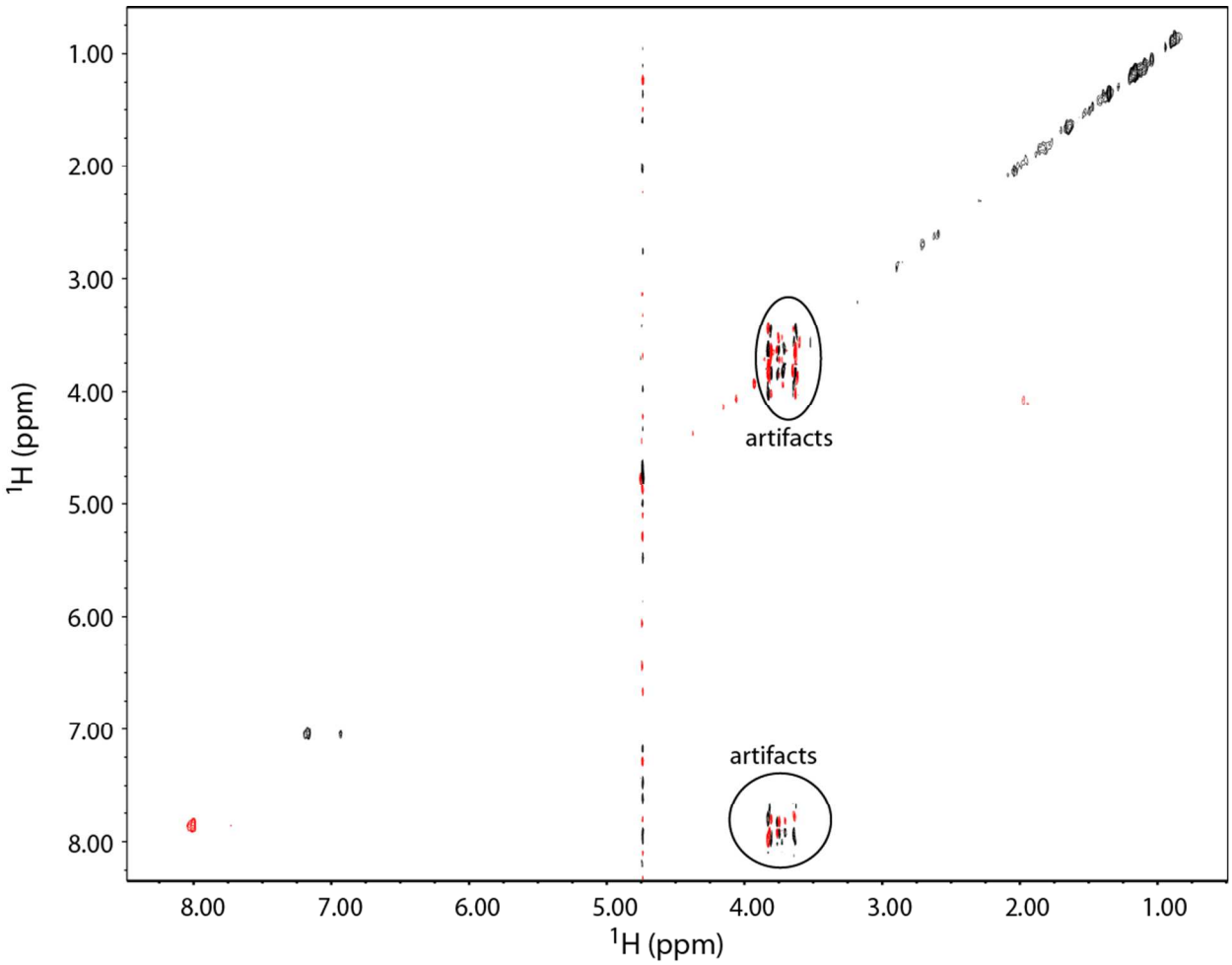

Figure S3. 2D-^{13}C, ^{15}N-filtered, ^{13}C-edited NOESY of a sample containing 0.5 mM ^{13}C, ^{15}N-labeled DBPA and 0.5 mM unlabeled DBPA.

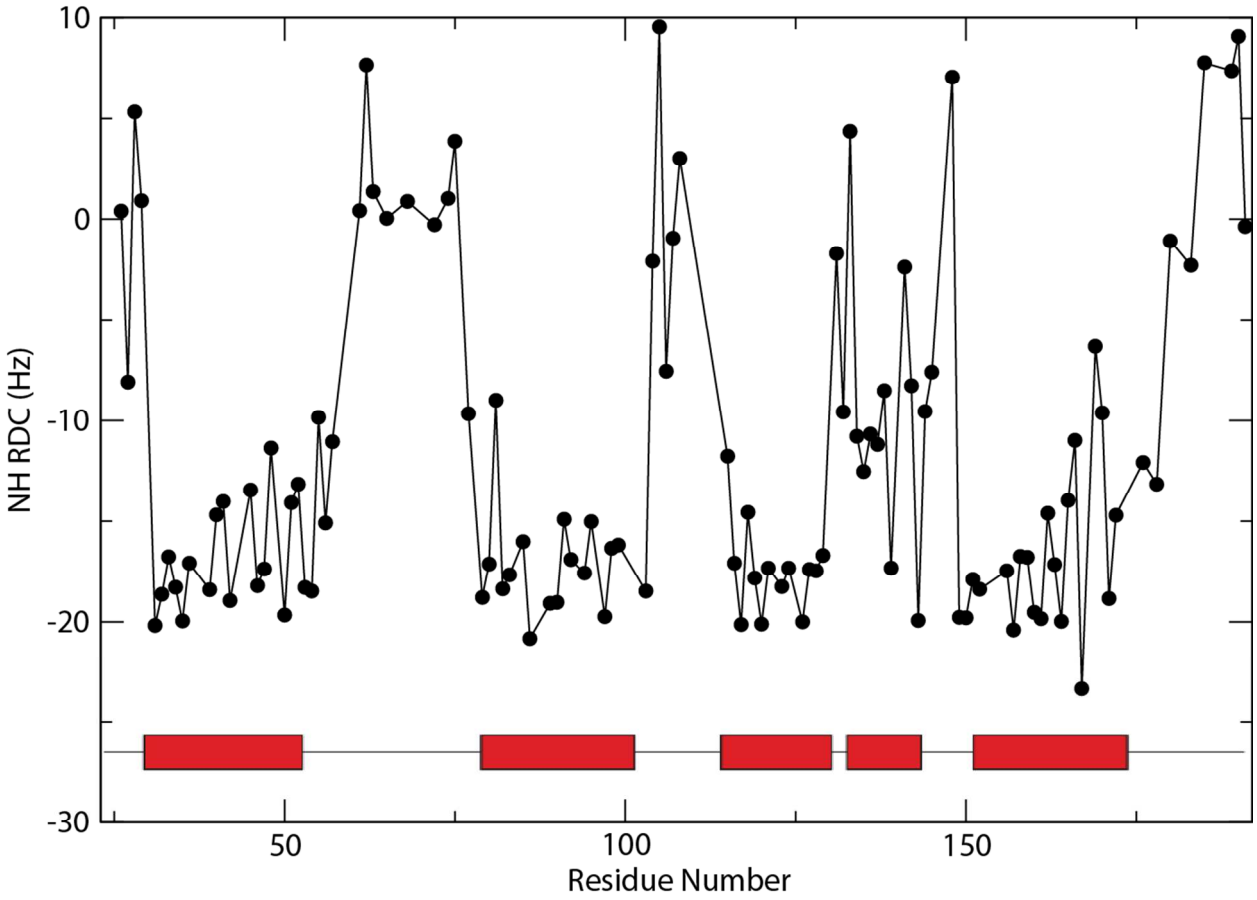

Figure S4. Backbone HN RDCs of DBPA aligned in 7% neutral polyacrylamide gel. Secondary structure of the protein is shown schematically with red rectangles representing α helical regions and straight line representing unstructured regions.

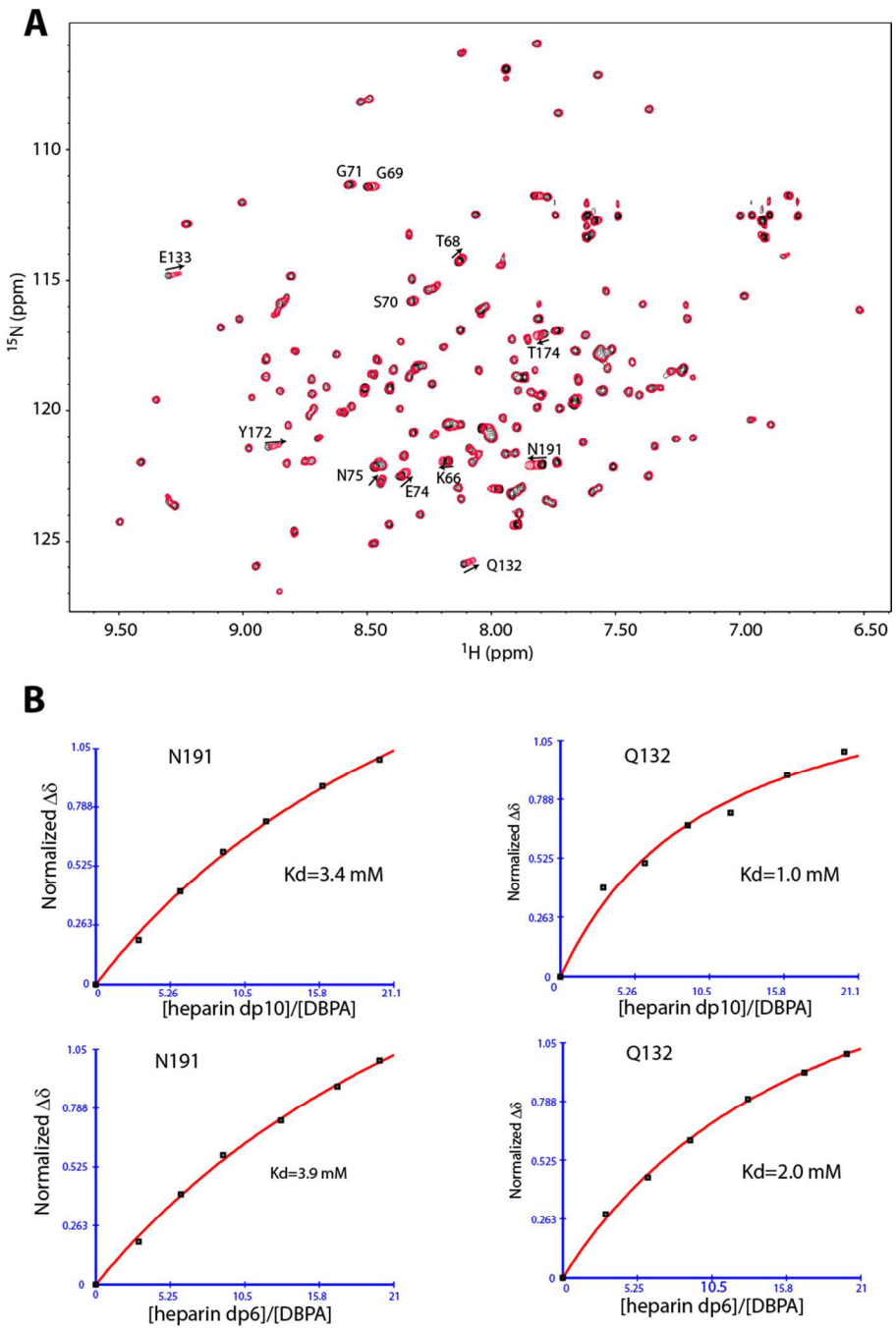

Figure S5. A) ^{15}N-HSQCs of DBPA-heparin dp6 titration. Black contours represent HSQC of DBPA without heparin dp6. Each successive red contour represents heparin dp6 concentrations of 0.3, 0.6, 0.9, 1.2, 1.6 and 2.0 mM, respectively. The concentration of DBPA is 01 mM. B) Fittings of Kds for DBPA-heparin interactions using chemical shift changes observed in N191 and Q132. Details of the fitting are described in the Experimental Procedures section. Briefly, normalized chemical shift changes for each residue are plotted against the ratio of ligand and protein concentrations. The resulting curve is fitted using the software xcrvfit according to the equation

$\Delta\delta = \Delta\delta_{max} \frac{(Lt+Pt+Kd)-\sqrt{(Lt+Pt+Kd)^2-4LtPt}}{2Pt}$, where Lt=total ligand concentration, Pt=total protein concentration, Kd=dissociation constant and $\Delta\delta_{max}$=maximum chemical shift change possible for the residue.

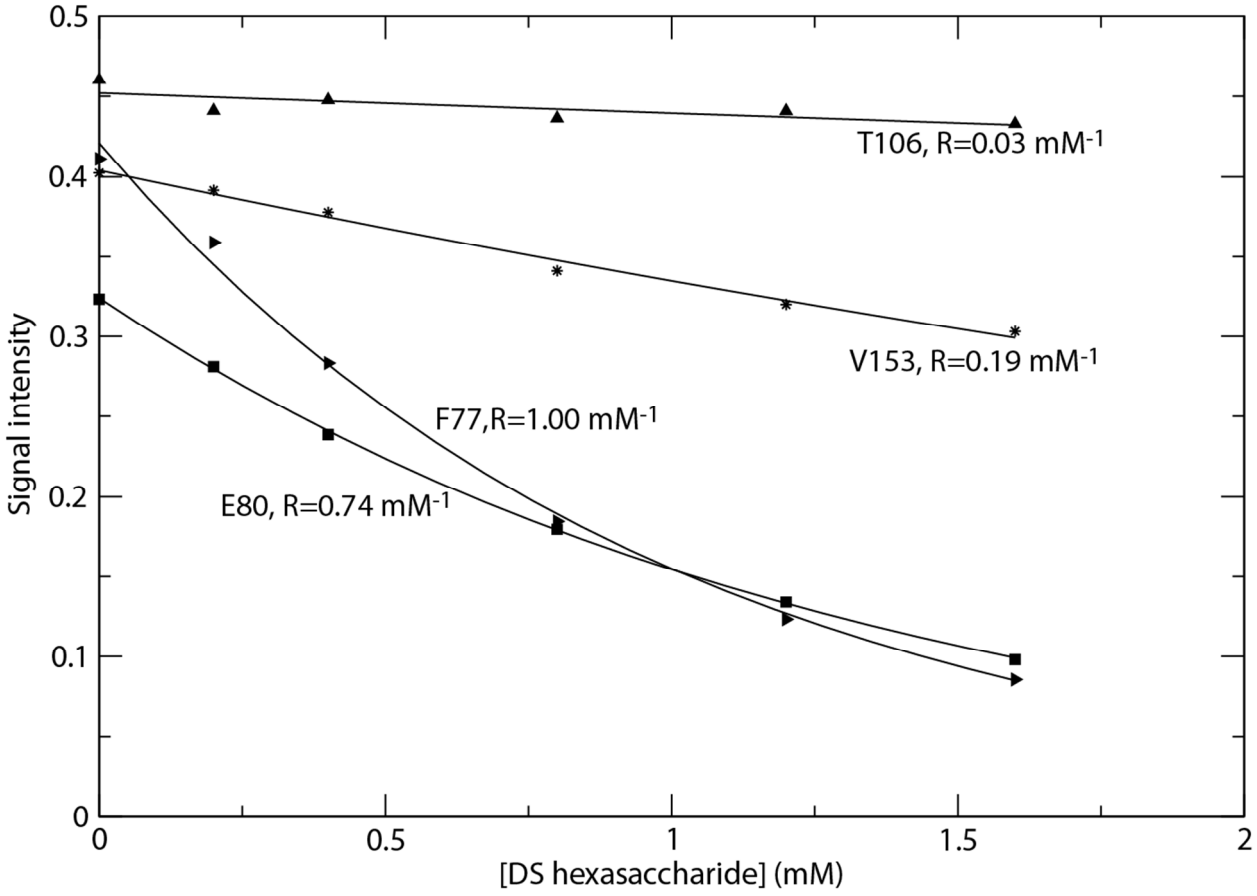

Figure S6. Fitting of signal decay observed during DS hexasaccharide titration of DBPA. The residues are chosen to illustrate the range of variation in the decay rate.

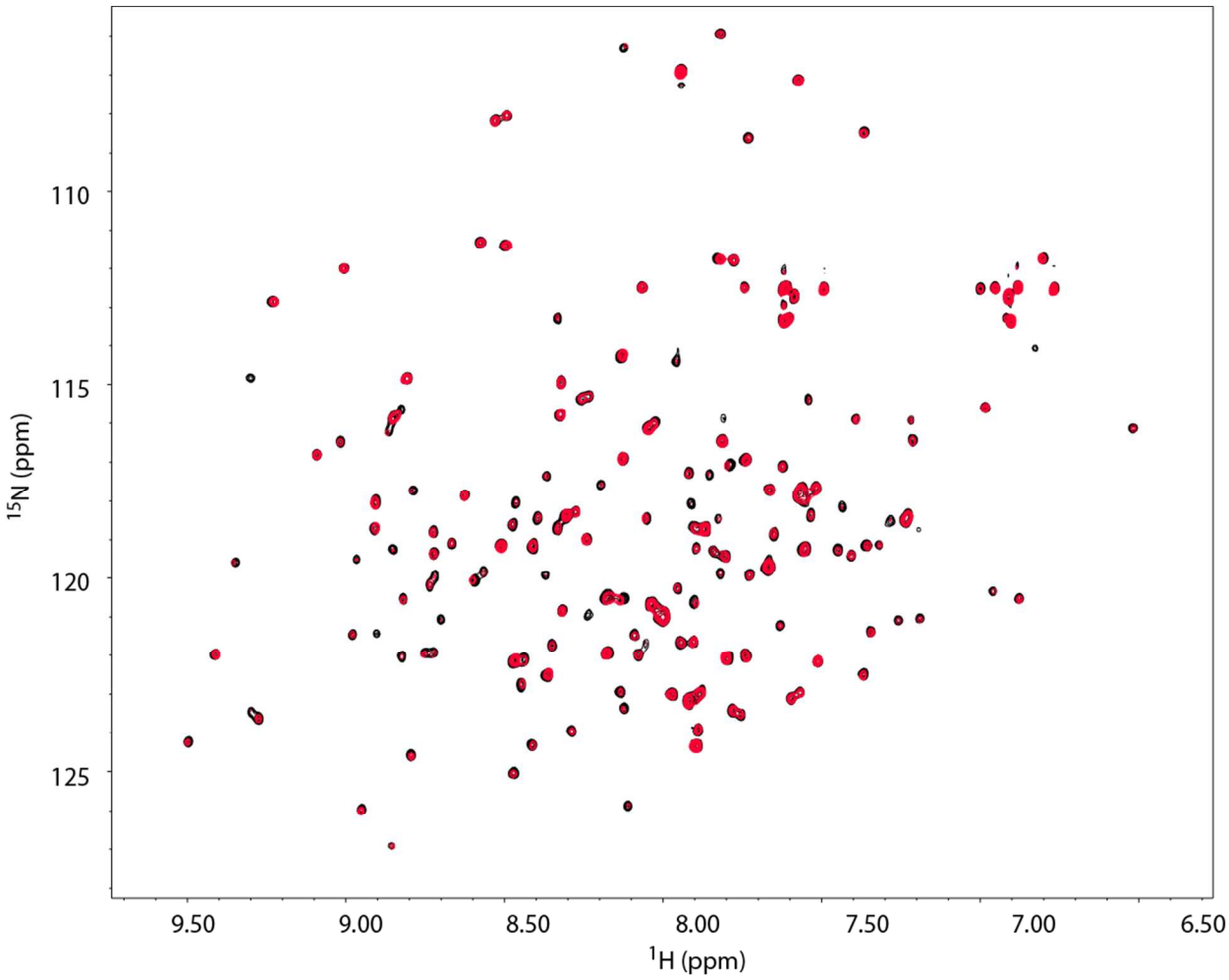

Figure S7. ^{15}N-HSQCs of DBPA in the presence (red contours) and absernce (black contours) of six molar equivalence of DS dp14.