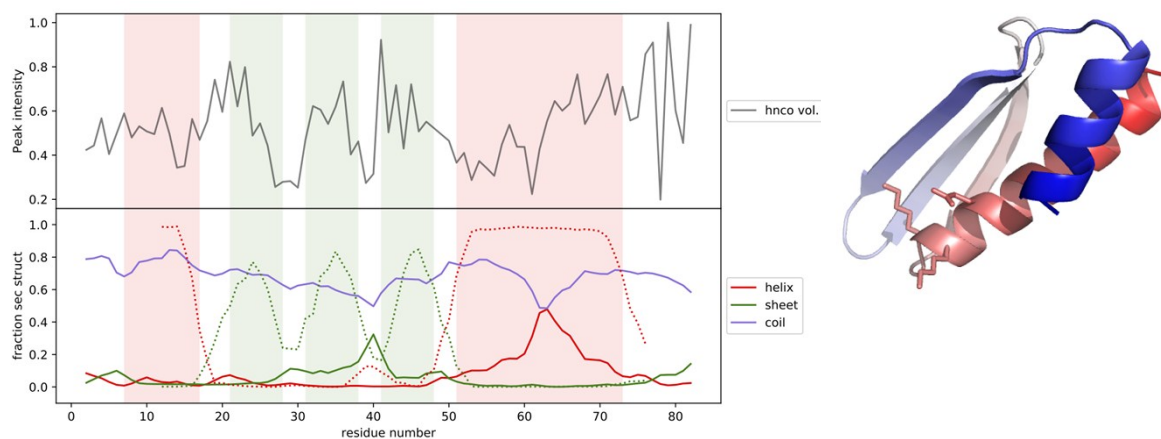### *Supplementary information*

Supplementary Figure 1

Left, d2d data and HNCO intensity for free DCL1-A at 298K. Right, structure of the folded form of DCL1-A. Helix 1 is colored blue and helix 2 is colored red. The sidechains of residues at the N terminus of helix 2 found to be pre-ordered in the free form are shown in stick representation

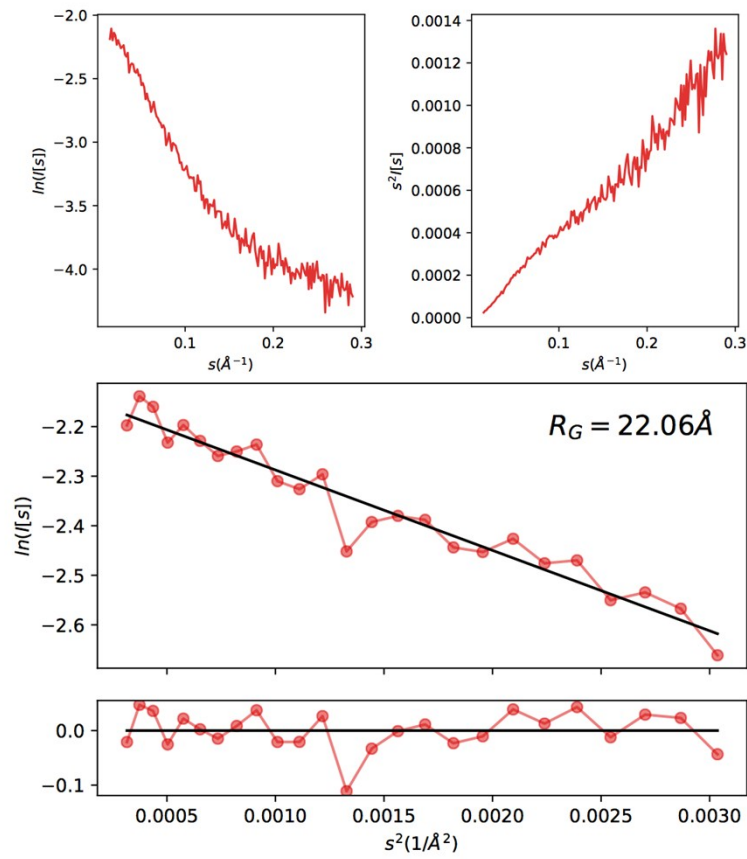Supplementary figure 1.

Supplementary Figure 2

Top, NOESY (red) and TOCSY (blue) spectra of DCL1-A at 278 K (left) and 298 K (right). Strips corresponding to residues 22-26 are shown, with horizontal lines highlighting sequential NOE peaks. HN_i-HA_{i-1} NOE peaks indicate extended structure in this region at both temperatures. Only weak intra and i-1 NOE peaks are observed. The Y23 HN-N resonances appear in a strongly overlapped region at 298K, NOE peaks corresponding to V36 and K77 appear in the same strip, centered at nearby N planes. Bottom, NOESY spectra of DCL1-A at 278 K (left) and 298 K (right). Strips corresponding to residues 60-64 are shown, with horizontal lines highlighting sequential HN-HN NOE peaks along the C-terminal helical region. No HN-HN peaks were detected at 298 K.
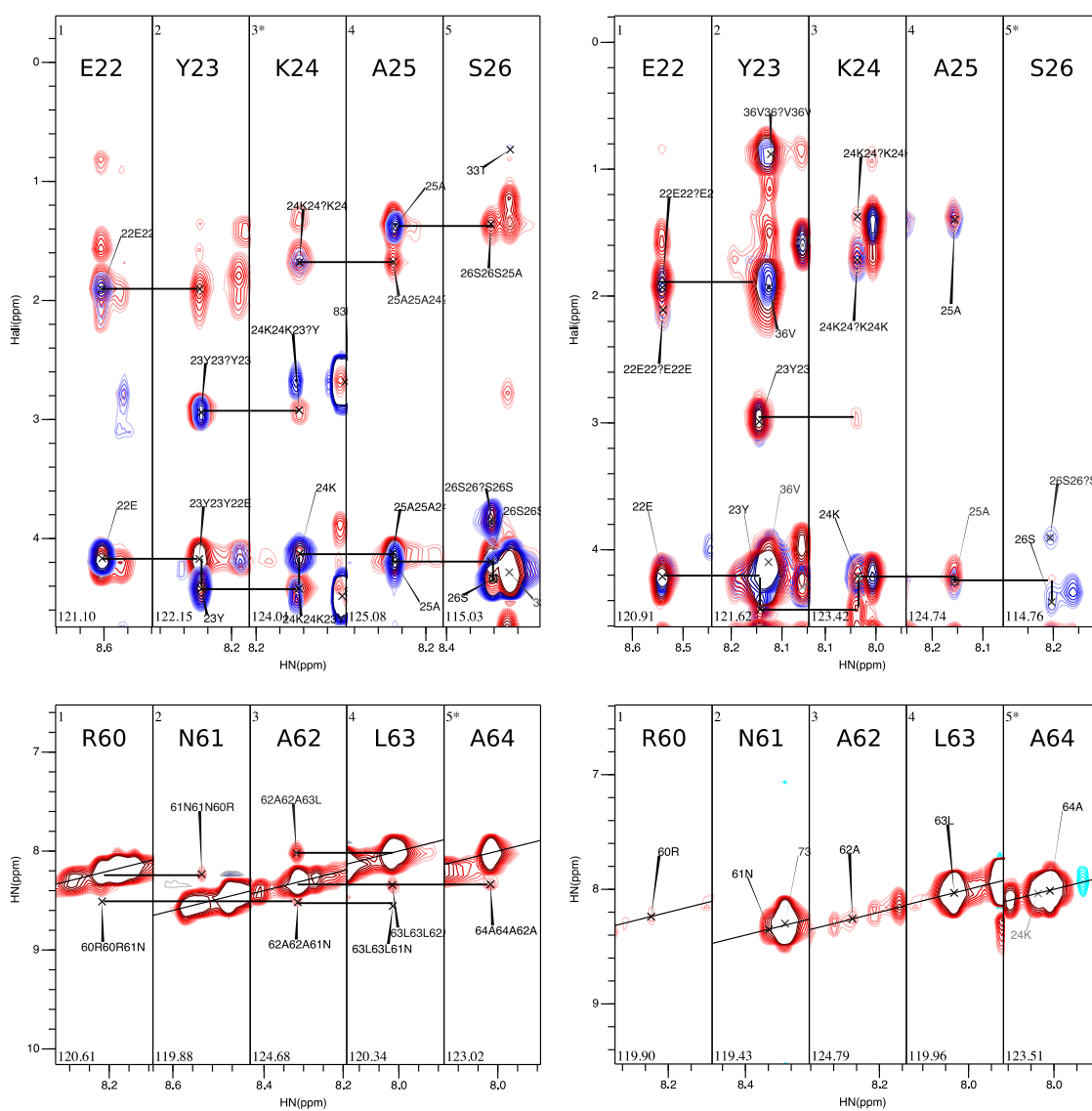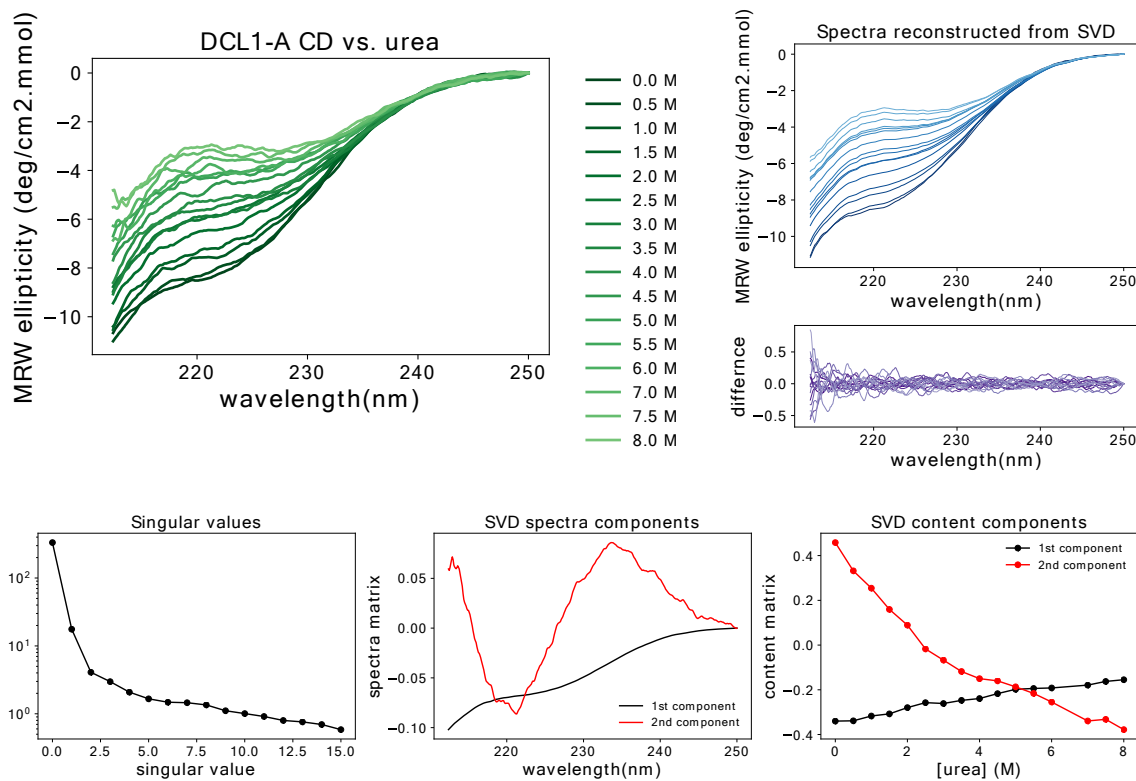

Supplementary figure 2

Supplementary Figure 3

Experimental SAXS data. Top left, raw SAXS data. Top right, Kratky plot. Bottom, linear fit of the Guinier region

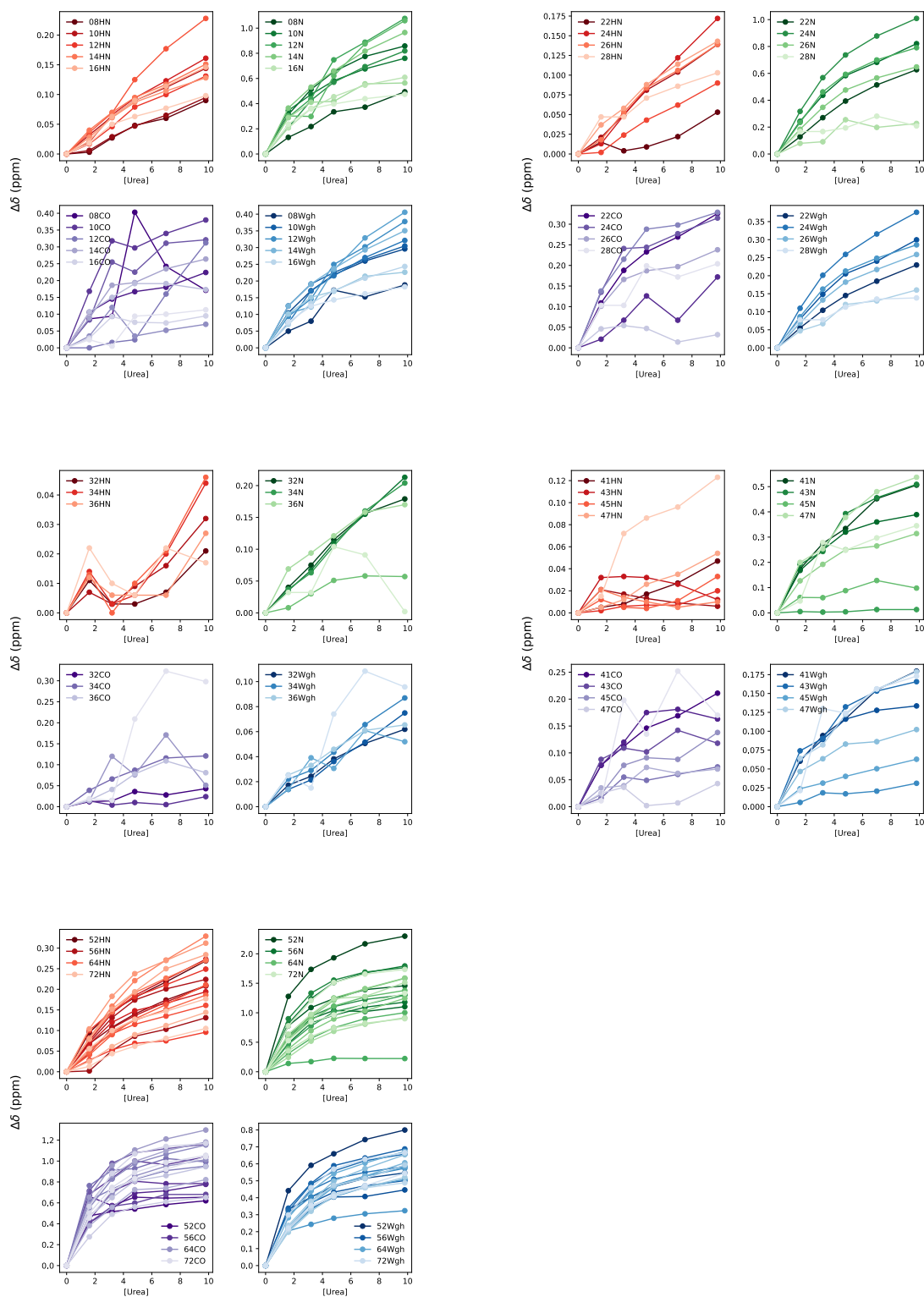

Supplementary figure 3

Supplementary Figure 4

Urea titration of DCL1-A followed by CD spectroscopy. Spectra are shown down to 212 nm, as the absorption of 8M urea is too high below this wavelength (up, left). Singular value decomposition of the spectra matrix shows two significant components (down, left). All spectra in the titration can be reconstructed with these first two components (up, right). The first component corresponds mostly to the disordered protein base spectrum, whereas the second, with a minimum at ca. 220 nm, corresponds mostly to a helical feature (down, center). The contribution of the second component reduces as urea concentration increases (down, right).

Supplementary figure 4.
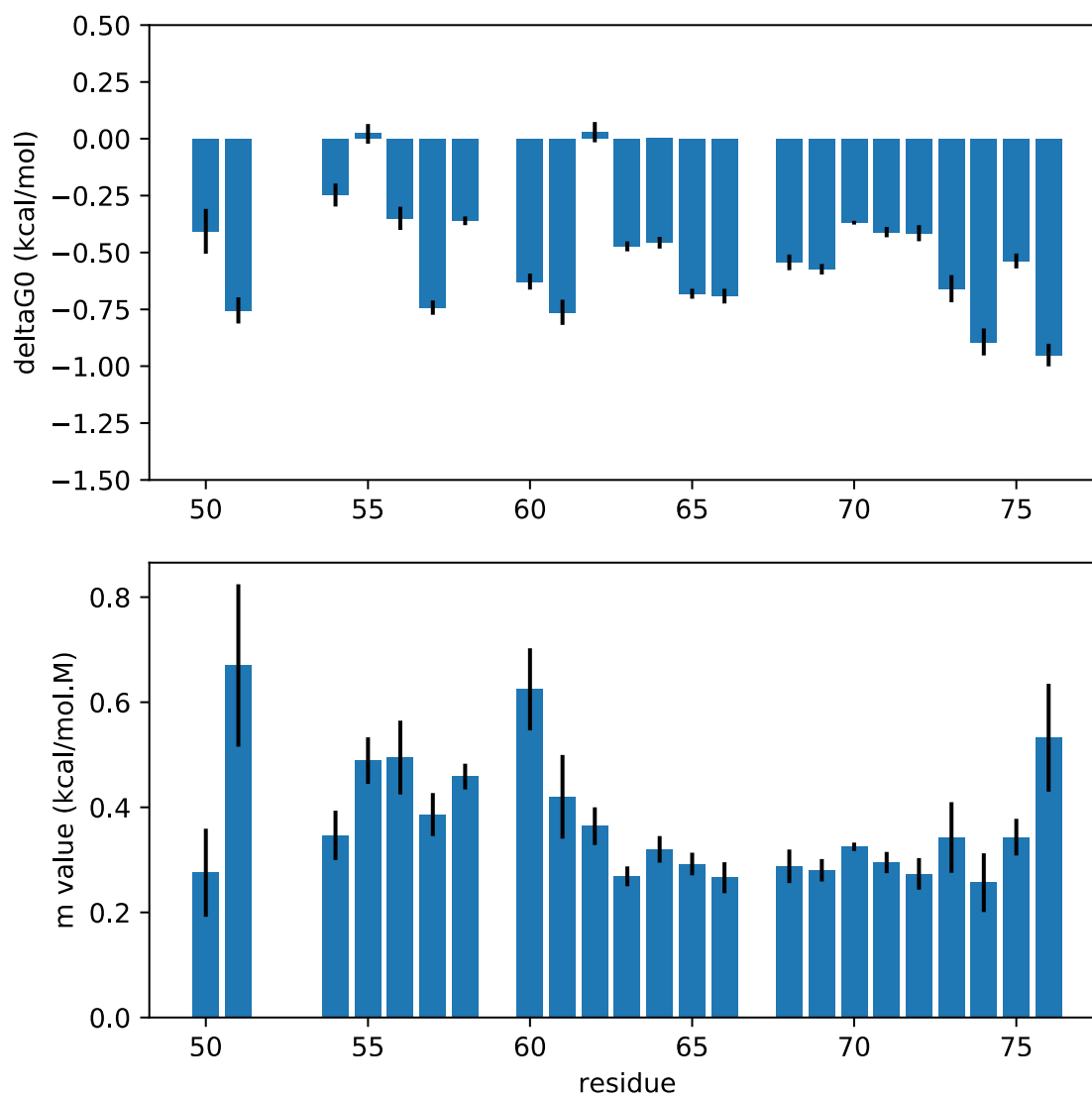
Supplementary Figure 5

Individual and weighed chemical shifts obtained on the urea titration of DCL1-A. Curves are grouped by regions corresponding to the secondary structure of the folded form.

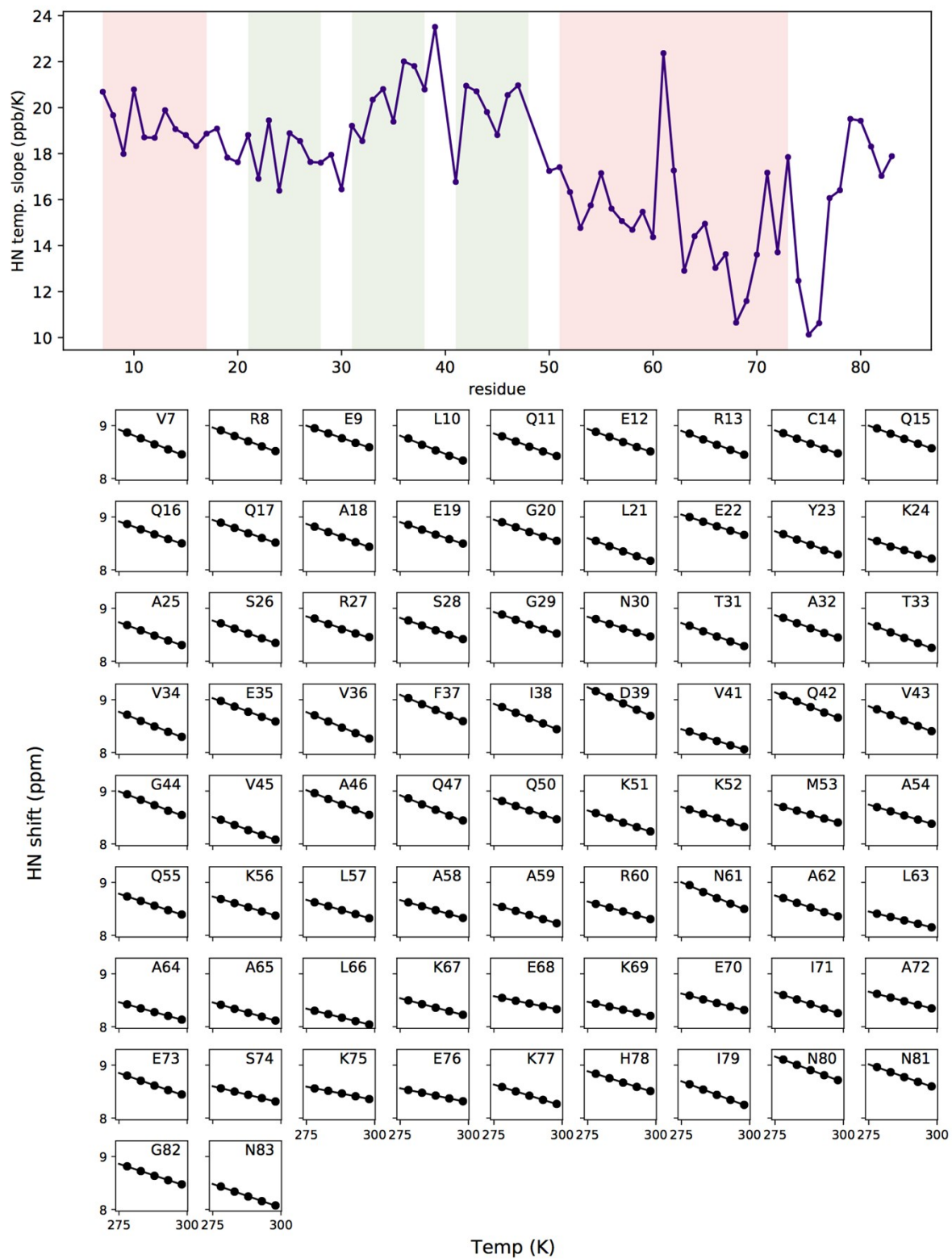Supplementary figure 5.

Supplementary Figure 6

Parameters obtained for individual fits of urea titrations using the linear extrapolation model for residues along helix 2

Supplementary figure 6.
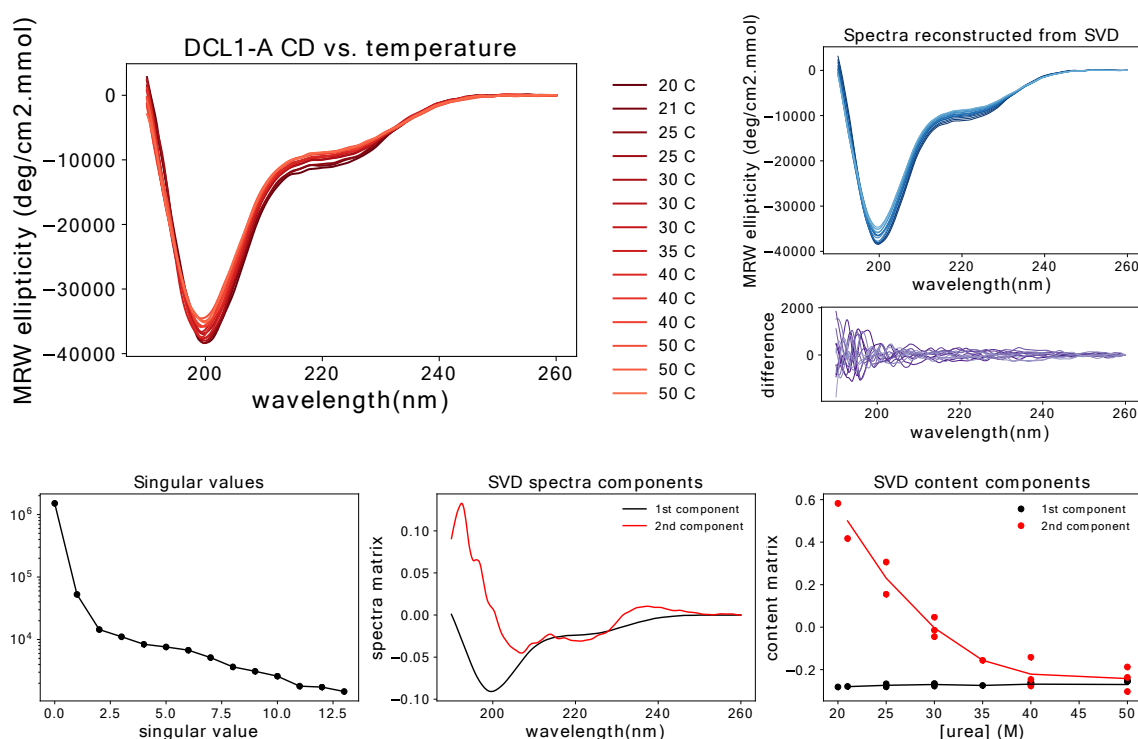
Supplementary Figure 7

Amide proton temperature coefficients and fits for individual residues

Supplementary figure 7.

Supplementary Figure 8

Temperature dependence of DCL1-A far UV CD spectra (up, left). Singular value decomposition of the spectra matrix shows two significant components (down, left). All spectra in the titration can be reconstructed with these first two components (up, right). The first component corresponds mostly to the disordered protein base spectrum, whereas the second, with minima at ca. 205 nm and 220 nm and positive CD below 200 nm, corresponds mostly to a helical feature (down, center). The contribution of the second component increases as temperature decreases (down, right). The continuous line follows the average of SVD content components at each temperature.
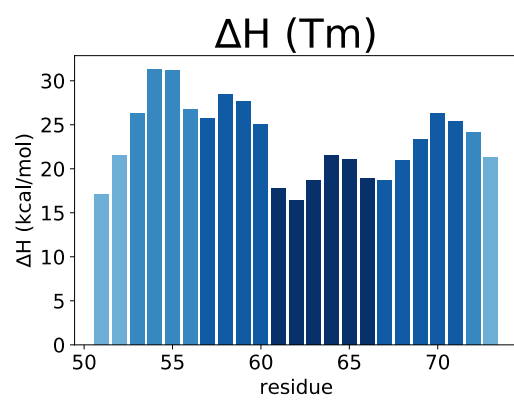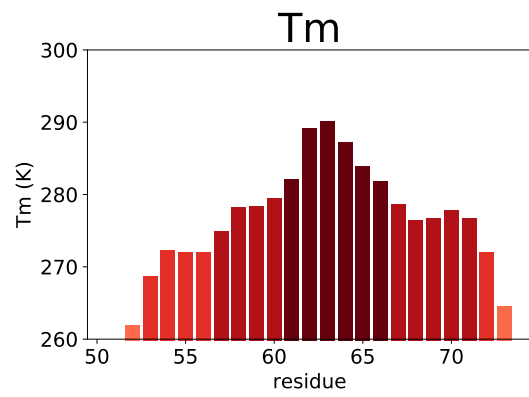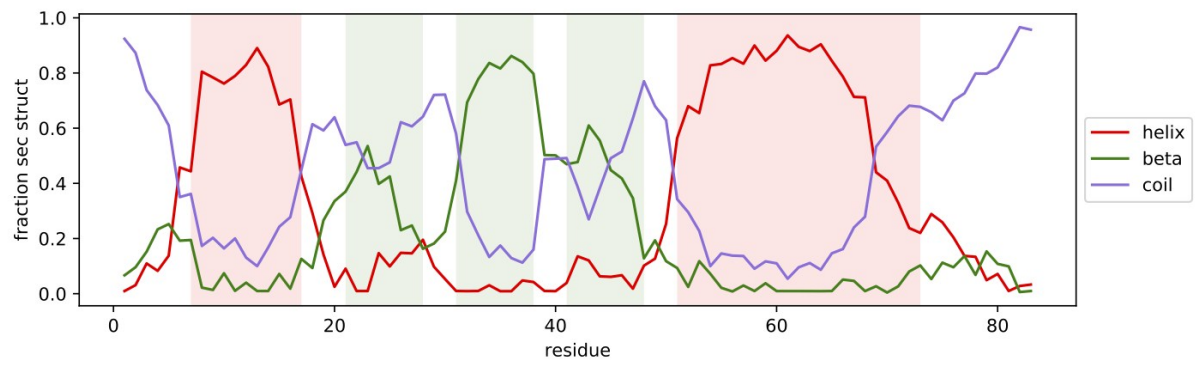

Supplementary figure 8.

Supplementary Figure 9

Values of ∆H(Tm) and Tm obtained fixing ∆CP to zero

Supplementary figure 9.

Supplementary Figure 10

S2D calculation of DCL1-A secondary structure population

Supplementary figure 10.