Wavenumber/wavenumber region (cm^{-1})	Mode and wavenumber (cm^{-1})	Assignment
930-942	M1, M4: 940; M2, M3: 938; M4: 940; M5: 930	Protein (proline, α-helix)[1, 2]
950/952	M2: 950; M3, M5: 952	C-C stretch: protein (proline, valine, α -helix)[3, 4]
999-1000	M1, M4: 1000; M2, M3, M5: 999	Phenylalanine (breathing mode) [5]
1024-1027	M1: 1027; M2, M3, M4: 1026; M5: 1024	Phenylalanine[6-9]
1078/80/1100	M1: 1078; M2, M3, M4: 1080; M4: 1100	Lipids[4, 10-12]
1119-1124	M1: 1123; M2: 1124, M3, M5: 1119; M4: 1122	Proteins, lipids[11, 13]
1153/5	M1, M2, M5: 1153; M3, M4: 1155	Carbohydrates/carotenoids[5, 14, 15]
1172-1176	M1: 1176; M3, M5: 1172; M4: 1174	Tyrosine[11, 16]
1205	M1-5	Tyrosine, phenylalanine[2, 17]
1240-1300	M1: 1243; M4: 1247, M5: 1299	Proteins: amide III - 1243 β-sheet; 1266-1300 α-helix[18]
1309-1321	M1: 1321; M2: 1316; M3: 1309; M4: 1321	DNA/RNA, protein[19]
1335/7	M4: 1337; M5: 1335	CH_3CH_2 deformation, proteins/lipids[20]
1445-1450	M1, M5: 1448; M2: 1446; M3: 1445; M4: 1450	CH modes (CH_2 and CH_3 deformations: bending & scissoring) in proteins/lipids[5, 21]
1520-5	M2:1520; BSSMF-PCA: 1521; visit 1: 1523; visit 2: 1525	Carotenoids[22, 23]
1554/6	M1, M2, M4: 1554; M3: 1556; M5: 1555	Tryptophan, proteins[24, 25]
1652/6	M1, M3: 1656; M2: 1654; M4: 1653; M5: 1652	Proteins (amide I)[2, 26, 27]

Supplementary table 1. Tentative wavenumber assignments.

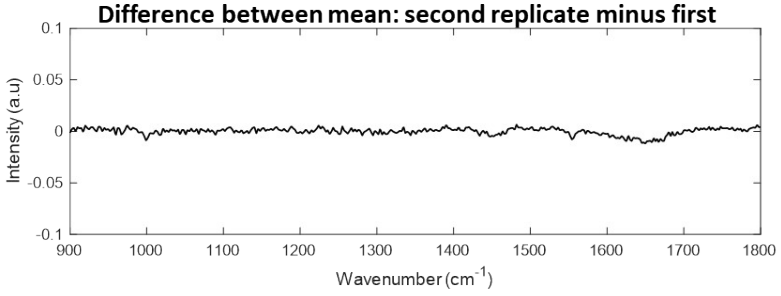
	M1	M2	M3	M4	M5	BSSMF-

						PCA
ALSFRS-R	*r 0.03* *p 0.8*	*r 0.17* *p 0.17*	*r -0.11* *p 0.37*	*r 0.09* *p 0.45*	***R -0.29*** ***p 0.01***	*r -0.19* *p 0.12*
DPR	*r -0.03* *p 0.8*	***r -0.31*** ***p 0.01***	*r 0.12* *p 0.3*	*r 0.09* *p 0.48*	*r 0.21* *p 0.09*	*r 0.2* *p 0.1*
FVC	***r -0.28*** ***p 0.02***	***r 0.39*** ***p 0.001***	*r -0.12* *p 0.34*	*r 0.19* *p 0.12*	***r -0.37*** ***p 0.002***	***r -0.4*** ***p 0.001***
NfL	*r -0.1* *p 0.39*	*r 0.04* *p 0.7*	*r -0.003* *p 0.9*	*r 0.08* *p 0.5*	*r -0.04* *p 0.7*	*r -0.07* *0.6*
Ferritin	*r 0.12* *p 0.3*	*r -0.04* *p 0.8*	*r 0.03* *p 0.8*	*r -0.15* *p 0.2*	*r 0.08* *p 0.5*	*r 0.1* *p 0.4*
CRP	*r -0.03* *p 0.8*	*r -0.2* *p 0.11*	*r 0.2* *p 0.11*	*r -0.19* *p 0.13*	***r 0.37*** ***p 0.003***	***r 0.29*** ***p 0.02***
C3	***r 0.28*** ***p 0.02***	***r -0.41*** ***p 0.0007***	***r 0.39*** ***p 0.001***	***r -0.32*** ***p 0.008***	*r 0.18* *p 0.14*	***r 0.55*** ***<0.0001***
C4	*r -0.05* *p 0.68*	*r -0.17* *p 0.16*	*r 0.2* *p 0.1*	*r -0.02* *p 0.86*	*r 0.06* *p 0.6*	*r 0.2* *p 0.11*

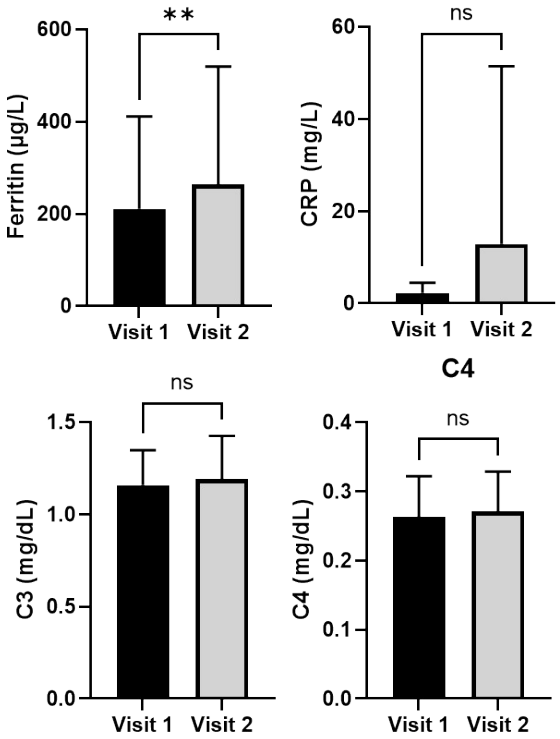
Supplementary table 2. Correlation of baseline BBSMF mode weights and clinical/biochemical markers of disease.

Mode	PC1 coefficients
M1	0.1930
M2	-0.6114
M3	0.6256
M4	-0.4003
M5	0.1931

Supplementary table 3. PC1 coefficients.



Supplementary figure 1. Difference between the mean plot for the technical replicates (2nd replicate minus the 1st).



Supplementary figure 2. Changes between the two study visits for additional biochemical markers of disease.

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