Electronic Supplementary Material (ESI) for Soft Matter.

Microheterogeneity metrics for diffusion in soft matter: Supplemental Material

<u>Fractional Brownian motion model of HA</u>

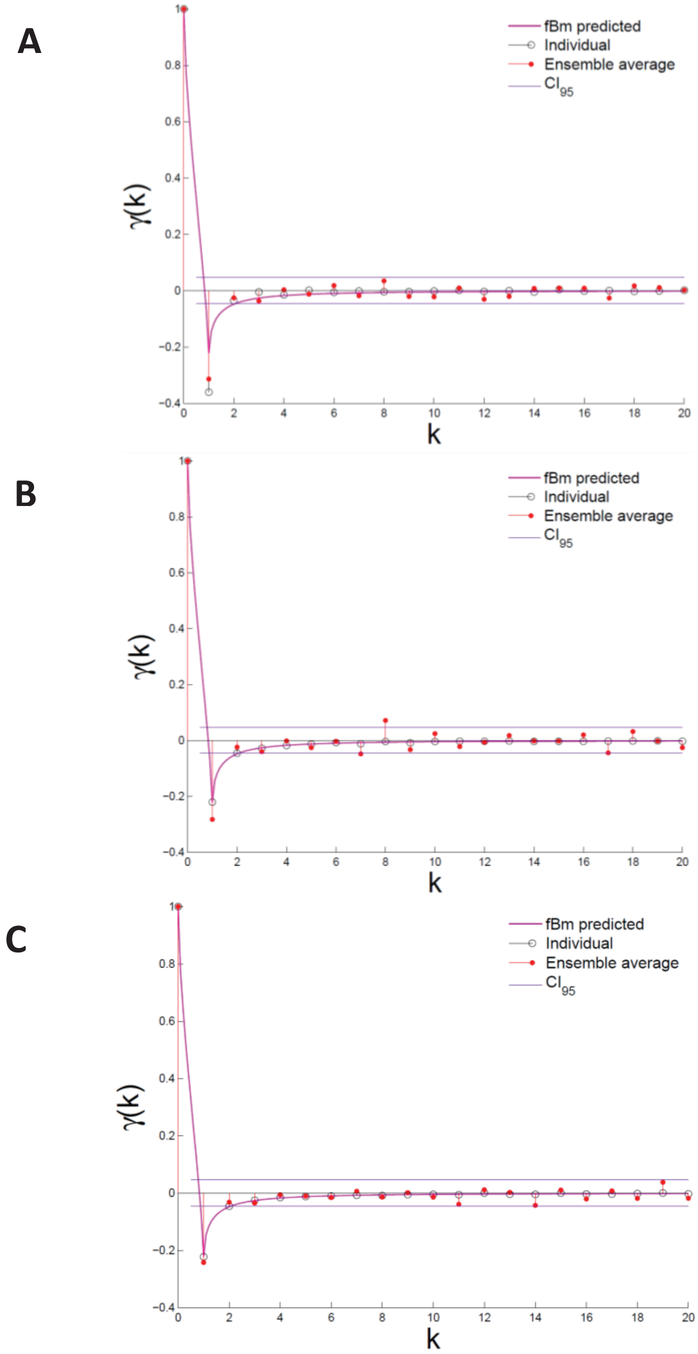

Figure S1: Autocovariance of 10mg/ml HA experimental data (S1A) and two simulated data sets (S1B,S1C). In **B**, the data were generated using $\alpha = 0.638$ and $D_{fBm} = 9.58E^{(-5)}\ \mu m^2 / s^{\alpha}$. These are the best-fit fractional Brownian parameters from the data in **A**. In **C**, the data were generated using $\alpha = 0.638$ and $D_{fBm} = 1E^{(-5)}\ \mu m^2 / s^{\alpha}$. Each data set contains 175 particle paths.

Metric Comparison on two-cluster data sets

Table S1: Metric comparison on 2-cluster data sets described in Sec. 4.2.2. The second through fourth column show the values of the non-Gaussian parameter, excess kurtosis and heterogeneity ratio, respectively, for each of the four 2-cluster data sets. The fifth through seventh columns show the relative contributions of the 10%, 25% and 50% highest values of the individual compliance to the ensemble mean, respectively. The final column shows the results of the Stage 2 metric of Valentine *et al.*[1] described in Section 2.2.

	NG (Eqn. 2)[2]	Ku (Eqn. 3)[3]	HR (Eqn. 4)[4]	Tseng 10%[5]	Tseng 25%	Tseng 50%	F-test[1]
Δ=0.050	$9.11E^{-2}$	$2.73E^{-1}$	$8.44E^{1}$	$1.12E^{1}$	$2.72E^{1}$	$5.28E^{1}$	2-3
Δ=0.075	$3.25E^{-1}$	$1.08E^{-1}$	$9.88E^{1}$	$1.13E^{1}$	$2.74E^{1}$	$5.29E^{1}$	2-3
Δ=0.10	$3.57E^{-1}$	$1.19E^{-1}$	$1.14E^{2}$	$1.15E^{1}$	$2.77E^{1}$	$5.34E^{1}$	2-3

Homogeneous Data: Simulated and Experimental

Simulated Newtonian

Table S2: Results for simulated homogeneous Newtonian data. Superscript s identifies the values used to generate the data and superscript p the values predicted by the clustering algorithm.

	Power law exponent			Fractional Diffusion Coefficient $\left(\mu m^2/s^\alpha\right)$		
	$\boldsymbol{\alpha^s}$	$\boldsymbol{\alpha^p}$	$\boldsymbol{CI_{95}}$	$\boldsymbol{D^s_{fBm}}$	$\boldsymbol{D^p_{fBm}}$	$\boldsymbol{CI_{95}}$
σ_x Cluster 1	$1.00E^0$	$9.66E^{-1}$	$(9.33E^{-1}, 9.98E^{-1})$	$1.61E^{-2}$	$1.60E^{-2}$	$(1.59E^{-2}, 1.62E^{-2})$
σ_y Cluster 1	$1.00E^0$	$1.02E^0$	$(9.91E^{-1}, 1.06E^0)$	$1.61E^{-2}$	$1.58E^{-2}$	$(1.56E^{-2}, 1.60E^{-2})$

Experimental Newtonian

Table S3: Results for experimental homogeneous Newtonian data. Superscript e identifies the expected values and superscript p the values predicted by the clustering algorithm.

	Power law exponent			Fractional Diffusion Coefficient $\left(\mu m^2/s^\alpha\right)$		
	$\boldsymbol{\alpha^e}$	$\boldsymbol{\alpha^p}$	$\boldsymbol{CI_{95}}$	$\boldsymbol{D^e_{fBm}}$	$\boldsymbol{D^p_{fBm}}$	$\boldsymbol{CI_{95}}$
σ_x Cluster 1	$1.00E^0$	$9.81E^{-1}$	$(9.62E^{-1}, 1.00E^{-1})$	$1.61E^{-2}$	$1.37E^{-2}$	$(1.36E^{-2}, 1.38E^{-2})$
σ_y Cluster 1	$1.00E^0$	$9.56E^{-1}$	$(9.20E^{-1}, 9.92E^{-1})$	$1.61E^{-2}$	$1.44E^{-2}$	$(1.42E^{-2}, 1.45E^{-2})$

Experimental Newtonian (α fixed)

Table S4: Results for experimental homogeneous Newtonian data when fBm exponent is fixed at $\alpha = 1$. Superscripts same as Table S2.

	Diffusion Coefficient $\left(\mu m^2/s\right)$		
	$\boldsymbol{D^e_{fBm}}$	$\boldsymbol{D^p_{fBm}}$	$\boldsymbol{CI_{95}}$
σ_x Cluster 1	$1.61E^{-2}$	$1.36E^{-2}$	$(1.35E^{-2}, 1.37E^{-2})$
σ_y Cluster 1	$1.61E^{-2}$	$1.43E^{-2}$	$(1.40E^{-2}, 1.45E^{-2})$

Simulated Non-Newtonian

Table S5: Results for simulated homogeneous non-Newtonian data. Superscripts same as Table S1.

	Power law exponent			Fractional Diffusion Coefficient $\left(\mu m^2/s^\alpha\right)$		
	$\boldsymbol{\alpha^s}$	$\boldsymbol{\alpha^p}$	$\boldsymbol{CI_{95}}$	$\boldsymbol{D^s_{fBm}}$	$\boldsymbol{D^p_{fBm}}$	$\boldsymbol{CI_{95}}$
σ_x Cluster 1	$5.76E^{-1}$	$5.16E^{-1}$	$(4.45E^{-1}, 5.88E^{-1})$	$9.30E^{-5}$	$9.69E^{-5}$	$(9.35E^{-5}, 1.00E^{-4})$
σ_y Cluster 1	$5.76E^{-1}$	$5.92E^{-1}$	$(5.68E^{-1}, 6.16E^{-1})$	$9.30E^{-5}$	$9.51E^{-5}$	$(9.40E^{-5}, 9.61E^{-5})$

Experimental Non-Newtonian

Table S6: Results for experimental homogeneous non-Newtonian data. Superscripts same as Table S2.

	Power law exponent			Fractional Diffusion Coefficient $\left(\mu m^2/s^\alpha\right)$		
	$\boldsymbol{\alpha^e}$	$\boldsymbol{\alpha^p}$	$\boldsymbol{CI_{95}}$	$\boldsymbol{D^e_{fBm}}$	$\boldsymbol{D^p_{fBm}}$	$\boldsymbol{CI_{95}}$
σ_x Cluster 1 Cluster 2	 N/A N/A	 $5.76E^{-1}$ $4.41E^{-1}$	 $(5.08E^{-1}, 6.45E^{-1})$ $(3.71E^{-1}, 5.11E^{-1})$	 N/A N/A	 $9.63E^{-5}$ $1.52E^{-4}$	 $(9.32E^{-5}, 9.94E^{-5})$ $(1.46E^{-4}, 1.58E^{-4})$
σ_y Cluster 1 Cluster 2	 N/A N/A	 $5.73E^{-1}$ $4.37E^{-1}$	 $(4.91E^{-1}, 6.55E^{-1})$ $(3.15E^{-1}, 5.59E^{-1})$	 N/A N/A	 $9.09E^{-5}$ $1.21E^{-4}$	 $(8.74E^{-5}, 9.44E^{-5})$ $(1.13E^{-4}, 1.29E^{-4})$

Heterogeneous Data: Simulated and Experimental

Simulated Newtonian

Table S7: Results for simulated heterogeneous Newtonian data. Superscripts same as Table S1.

	Power law exponent			Fractional Diffusion Coefficient $\left({\mu m^2}/{s^\alpha}\right)$		
	α^s	α^p	CI_{95}	D^s_{fBm}	D^p_{fBm}	CI_{95}
σ_x						
Cluster 1	$1.00E^0$	$9.72E^{-1}$	$(9.41E^{-1}, 1.00E^0)$	$8.05E^{-3}$	$7.81E^{-3}$	$(7.73E^{-3}, 7.90E^{-3})$
Cluster 2	$1.00E^0$	$1.02E^0$	$(9.97E^{-1}, 1.03E^0)$	$1.61E^{-2}$	$1.60E^{-2}$	$(1.59E^{-2}, 1.61E^{-2})$
σ_y						
Cluster 1	$1.00E^0$	$9.51E^{-1}$	$(9.24E^{-1}, 9.79E^{-1})$	$8.05E^{-3}$	$7.46E^{-3}$	$(7.39E^{-3}, 7.54E^{-3})$
Cluster 2	$1.00E^0$	$9.98E^{-1}$	$(9.59E^{-1}, 1.04E^0)$	$1.61E^{-2}$	$1.56E^{-2}$	$(1.54E^{-2}, 1.59E^{-2})$

Experimental Newtonian

Table S8: Results for experimental heterogeneous Newtonian data. Superscripts same as Table S2.

	Power law exponent			Fractional Diffusion Coefficient $\left({\mu m^2}/{s^\alpha}\right)$		
	α^e	α^p	CI_{95}	D^e_{fBm}	D^p_{fBm}	CI_{95}
σ_x						
Cluster 1	$1.00E^0$	$9.84E^{-1}$	$(9.60E^{-1}, 1.01E^{-1})$	$1.61E^{-2}$	$1.28E^{-2}$	$(1.27E^{-2}, 1.29E^{-2})$
Cluster 2	$1.00E^0$	$9.52E^{-1}$	$(9.08E^{-1}, 9.97E^{-1})$	$8.05E^{-3}$	$8.58E^{-3}$	$(8.44E^{-3}, 8.72E^{-3})$
σ_y						
Cluster 1	$1.00E^0$	$9.50E^{-1}$	$(9.05E^{-1}, 9.95E^{-1})$	$1.61E^{-2}$	$1.37E^{-2}$	$(1.35E^{-2}, 1.39E^{-2})$
Cluster 2	$1.00E^0$	$9.46E^{-1}$	$(9.04E^{-1}, 9.87E^{-1})$	$8.05E^{-3}$	$8.71E^{-3}$	$(8.58E^{-3}, 8.85E^{-3})$

Experimental Newtonian (α fixed)

Table S9: Results for experimental heterogeneous Newtonian data when fBm exponent is fixed at $\alpha = 1$. Superscripts same as Table S2.

	Diffusion Coefficient $\left(\mu m^2/s\right)$		
	D^e_{fBm}	D^p_{fBm}	CI_{95}
σ_x			
Cluster 1	$1.61E^{-2}$	$1.28E^{-2}$	$(1.27E^{-2}, 1.29E^{-2})$
Cluster 2	$8.05E^{-3}$	$8.52E^{-3}$	$(8.36E^{-3}, 8.68E^{-3})$
σ_y			
Cluster 1	$1.61E^{-2}$	$1.36E^{-2}$	$(1.33E^{-2}, 1.38E^{-2})$
Cluster 2	$8.05E^{-3}$	$8.65E^{-3}$	$(8.48E^{-3}, 8.81E^{-3})$

Simulated non-Newtonian

Table S10: Results for simulated heterogeneous non-Newtonian data. Superscripts same as Table S1.

	Power law exponent			Fractional Diffusion Coefficient $\left(\mu m^2/s^\alpha\right)$		
	α^s	α^p	CI_{95}	D^s_{fBm}	D^p_{fBm}	CI_{95}
σ_x						
Cluster 1	$6.40E^{-1}$	$6.55E^{-1}$	$(6.29E^{-1}, 6.80E^{-1})$	$1.00E^{-4}$	$1.02E^{-4}$	$(1.01E^{-4}, 1.04E^{-4})$
Cluster 2	$7.20E^{-1}$	$6.61E^{-1}$	$(6.16E^{-1}, 7.06E^{-1})$	$4.20E^{-4}$	$4.13E^{-4}$	$(4.05E^{-4}, 4.21E^{-4})$
σ_y						
Cluster 1	$6.40E^{-1}$	$6.18E^{-1}$	$(5.83E^{-1}, 6.53E^{-1})$	$1.00E^{-4}$	$1.04E^{-4}$	$(1.02E^{-4}, 1.06E^{-4})$
Cluster 2	$7.20E^{-1}$	$7.00E^{-1}$	$(6.83E^{-1}, 7.18E^{-1})$	$4.20E^{-4}$	$4.09E^{-4}$	$(4.06E^{-4}, 4.12E^{-4})$

Experimental Non-Newtonian

Table S11: Results for experimental heterogeneous non-Newtonian data. Superscripts same as Table S2.

	Power law exponent			Fractional Diffusion Coefficient $\left(\mu m^2/s^\alpha\right)$		
	α^e	α^p	CI_{95}	D^e_{fBm}	D^p_{fBm}	CI_{95}
σ_x						
Cluster 1	N/A	$6.41E^{-1}$	$(5.91E^{-1}, 6.91E^{-1})$	N/A	$1.03E^{-4}$	$(1.01E^{-4}, 1.05E^{-4})$
Cluster 2	N/A	$6.98E^{-1}$	$(6.20E^{-1}, 7.77E^{-1})$	N/A	$4.40E^{-4}$	$(4.25E^{-4}, 4.55E^{-4})$
σ_y						
Cluster 1	N/A	$6.44E^{-1}$	$(6.08E^{-1}, 6.80E^{-1})$	N/A	$9.88E^{-5}$	$(9.72E^{-5}, 1.00E^{-4})$
Cluster 2	N/A	$7.97E^{-1}$	$(7.75E^{-1}, 8.20E^{-1})$	N/A	$4.18E^{-4}$	$(4.14E^{-4}, 4.22E^{-4})$

Experimental Agarose

Table S122: Results for experimental agarose data. Superscripts same as Table S2.

	Power law exponent			Fractional Diffusion Coefficient $\left({\mu m^2}/{s^\alpha}\right)$		
	α^e	α^p	CI_{95}	D^e_{fBm}	D^p_{fBm}	CI_{95}
σ_x						
Cluster 1	N/A	$1.10E^{-1}$	$(6.38E^{-2}, 1.57E^{-1})$	N/A	$1.05E^{-4}$	$(1.00E^{-4}, 1.10E^{-4})$
Cluster 2	N/A	$9.36E^{-1}$	$(8.40E^{-1}, 1.03E^{0})$	N/A	$2.61E^{-1}$	$(2.51E^{-1}, 2.71E^{-1})$
Cluster 3	N/A	$2.72E^{-1}$	$(1.36E^{-1}, 4.08E^{-1})$	N/A	$3.22E^{-3}$	$(2.91E^{-3}, 3.53E^{-3})$
Cluster 4	N/A	$1.08E^{0}$	$(8.24E^{-1}, 1.33E^{0})$	N/A	$1.03E^{-1}$	$(9.32E^{-2}, 1.14E^{-1})$
σ_y						
Cluster 1	N/A	$1.28E^{-1}$	$(4.29E^{-2}, 2.12E^{-1})$	N/A	$1.09E^{-4}$	$(1.00E^{-4}, 1.18E^{-4})$
Cluster 2	N/A	$9.27E^{-1}$	$(8.53E^{-1}, 1.00E^{0})$	N/A	$2.68E^{-1}$	$(2.60E^{-1}, 2.76E^{-1})$
Cluster 3	N/A	$2.07E^{-1}$	$(7.27E^{-3}, 4.07E^{-1})$	N/A	$2.41E^{-3}$	$(2.02E^{-3}, 2.81E^{-3})$
Cluster 4	N/A	$9.11E^{-1}$	$(7.60E^{-1}, 1.06E^{0})$	N/A	$9.97E^{-2}$	$(9.37E^{-2}, 1.06E^{-1})$

Experimental HBE Mucus

Table S133: Results for experimental HBE mucus. Superscripts same as Table S2.

	Power law exponent			Fractional Diffusion Coefficient $\left({\mu m^2}/{s^\alpha}\right)$		
	α^e	α^p	CI_{95}	D^e_{fBm}	D^p_{fBm}	CI_{95}
σ_x						
Cluster 1	N/A	$8.83E^{-1}$	$(8.73E^{-1}, 8.94E^{-1})$	N/A	$6.61E^{-2}$	$(6.59E^{-2}, 6.64E^{-2})$
Cluster 2	N/A	$3.69E^{-1}$	$(2.68E^{-1}, 4.69E^{-1})$	N/A	$6.14E^{-3}$	$(5.77E^{-3}, 6.52E^{-3})$
Cluster 3	N/A	$5.87E^{-1}$	$(5.27E^{-1}, 6.46E^{-1})$	N/A	$2.34E^{-2}$	$(2.28E^{-2}, 2.41E^{-2})$
σ_y						
Cluster 1	N/A	$8.71E^{-1}$	$(8.35E^{-1}, 9.06E^{-1})$	N/A	$6.53E^{-2}$	$(6.44E^{-2}, 6.62E^{-2})$
Cluster 2	N/A	$3.65E^{-1}$	$(2.43E^{-1}, 4.87E^{-1})$	N/A	$6.27E^{-3}$	$(5.81E^{-3}, 6.74E^{-3})$
Cluster 3	N/A	$6.10E^{-1}$	$(5.81E^{-1}, 6.38E^{-1})$	N/A	$2.49E^{-2}$	$(2.45E^{-2}, 2.52E^{-2})$

References

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