**Microheterogeneity metrics for diffusion in soft matter: Supplemental Material**

Fractional Brownian motion model of HA

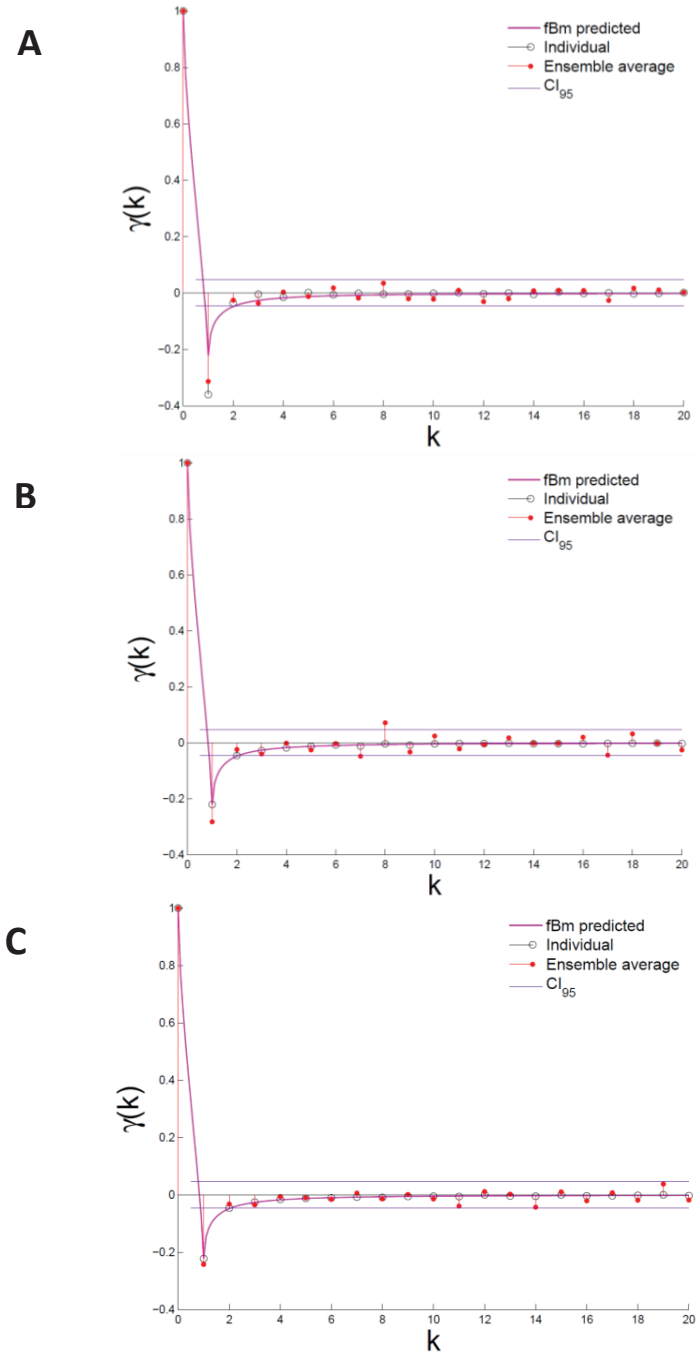


Figure S1: Autocovariance of 10mg/ml HA experimental data (S1A) and two simulated data sets (S1B,S1C). In **B**, the data were generated using $\alpha = 0.638$ and $D\_fBm = 9.58E\text{^}(-5)\ \mu m\text{^}2\ /\ s\text{^}\alpha$ . These are the best-fit fractional Brownian parameters from the data in **A**. In **C**, the data were generated using $\alpha = 0.638$ and $D\_fBm = 1E\text{^}(-5)\ \mu m\text{^}2\ /\ s\text{^}\alpha$. 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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^s}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^s_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ Cluster 1 | $1.00E^0$ | $9.66E^{-1}$ | $(9.33E^{-1}, 9.98E^{-1})$ | $1.61E^{-2}$ | $1.60E^{-2}$ | $(1.59E^{-2}, 1.62E^{-2})$ |
| $\sigma_y$ 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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^e}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^e_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ Cluster 1 | $1.00E^0$ | $9.81E^{-1}$ | $(9.62E^{-1}, 1.00E^{-1})$ | $1.61E^{-2}$ | $1.37E^{-2}$ | $(1.36E^{-2}, 1.38E^{-2})$ |
| $\sigma_y$ 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 ($\alpha$ fixed)

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

| | Diffusion Coefficient $\left(\mu m^2/_{s}\right)$ | | |
|---|---|---|---|
| | $\boldsymbol{D^e_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ Cluster 1 | $1.61E^{-2}$ | $1.36E^{-2}$ | $(1.35E^{-2}, 1.37E^{-2})$ |
| $\sigma_y$ 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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^s}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^s_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ Cluster 1 | $5.76E^{-1}$ | $5.16E^{-1}$ | $(4.45E^{-1}, 5.88E^{-1})$ | $9.30E^{-5}$ | $9.69E^{-5}$ | $(9.35E^{-5}, 1.00E^{-4})$ |
| $\sigma_y$ 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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^e}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^e_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ | | | | | | |
| Cluster 1 | N/A | $5.76E^{-1}$ | $(5.08E^{-1}, 6.45E^{-1})$ | N/A | $9.63E^{-5}$ | $(9.32E^{-5}, 9.94E^{-5})$ |
| Cluster 2 | N/A | $4.41E^{-1}$ | $(3.71E^{-1}, 5.11E^{-1})$ | N/A | $1.52E^{-4}$ | $(1.46E^{-4}, 1.58E^{-4})$ |
| $\sigma_y$ | | | | | | |
| Cluster 1 | N/A | $5.73E^{-1}$ | $(4.91E^{-1}, 6.55E^{-1})$ | N/A | $9.09E^{-5}$ | $(8.74E^{-5}, 9.44E^{-5})$ |
| Cluster 2 | N/A | $4.37E^{-1}$ | $(3.15E^{-1}, 5.59E^{-1})$ | N/A | $1.21E^{-4}$ | $(1.13E^{-4}, 1.29E^{-4})$ |

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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^s}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^s_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ | | | | | | |
| 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 | $1.00E^0$ | $1.02E^0$ | $(9.97E^{-1}, 1.03E^0)$ | $1.61E^{-2}$ | $1.60E^{-2}$ | $(1.59E^{-2}, 1.61E^{-2})$ |
| $\sigma_y$ | | | | | | |
| 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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^e}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^e_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ | | | | | | |
| 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 | $1.00E^0$ | $9.52E^{-1}$ | $(9.08E^{-1}, 9.97E^{-1})$ | $8.05E^{-3}$ | $8.58E^{-3}$ | $(8.44E^{-3}, 8.72E^{-3})$ |
| $\sigma_y$ | | | | | | |
| 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 ($\alpha$ fixed)

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

| | Diffusion Coefficient $\left(\mu m^2/_s\right)$ | | |
|---|---|---|---|
| | $\boldsymbol{D^e_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ | | | |
| Cluster 1 | $1.61E^{-2}$ | $1.28E^{-2}$ | $(1.27E^{-2}, 1.29E^{-2})$ |
| Cluster 2 | $8.05E^{-3}$ | $8.52E^{-3}$ | $(8.36E^{-3}, 8.68E^{-3})$ |
| $\sigma_y$ | | | |
| 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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^s}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^s_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ | | | | | | |
| 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 | $7.20E^{-1}$ | $6.61E^{-1}$ | $(6.16E^{-1}, 7.06E^{-1})$ | $4.20E^{-4}$ | $4.13E^{-4}$ | $(4.05E^{-4}, 4.21E^{-4})$ |
| $\sigma_y$ | | | | | | |
| 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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^e}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^e_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ | | | | | | |
| 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 | $4.40E^{-4}$ | $(4.25E^{-4}, 4.55E^{-4})$ |
| $\sigma_y$ | | | | | | |
| 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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^e}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^e_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ | | | | | | |
| 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 | $1.03E^{-1}$ | $(9.32E^{-2}, 1.14E^{-1})$ |
| $\sigma_y$ | | | | | | |
| 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 $\left(\mu m^2/_{s^\alpha}\right)$ | | |
|---|---|---|---|---|---|---|
| | $\boldsymbol{\alpha^e}$ | $\boldsymbol{\alpha^p}$ | $\boldsymbol{CI_{95}}$ | $\boldsymbol{D^e_{fBm}}$ | $\boldsymbol{D^p_{fBm}}$ | $\boldsymbol{CI_{95}}$ |
| $\sigma_x$ | | | | | | |
| 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 | $2.34E^{-2}$ | $(2.28E^{-2}, 2.41E^{-2})$ |
| $\sigma_y$ | | | | | | |
| 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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