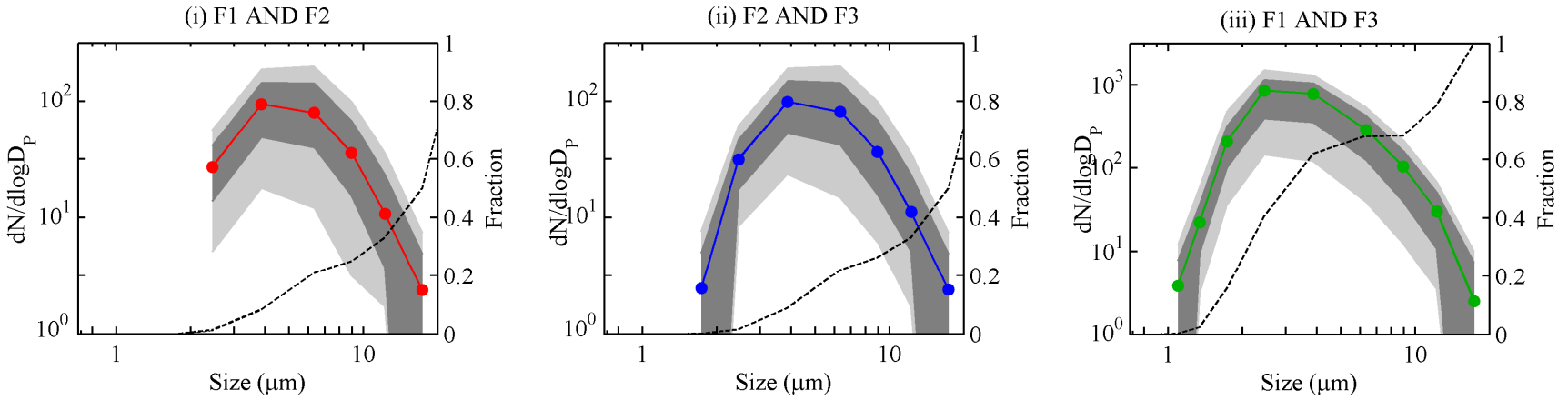


Figure S1. Size distributions of particles exhibiting dual-channel fluorescence in Manchester

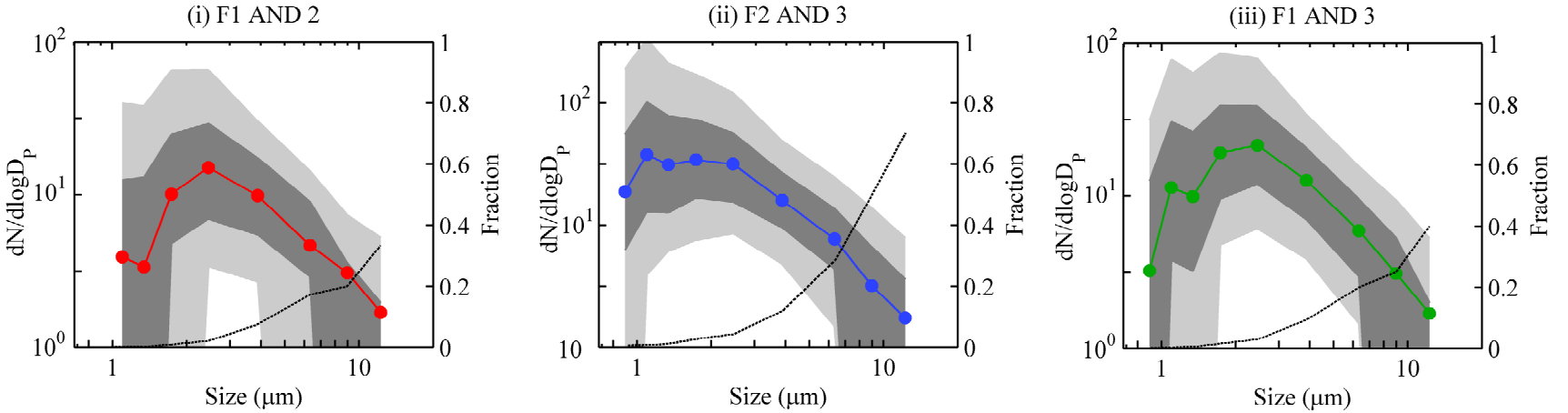


Figure S2. Size distributions of particles exhibiting dual-channel fluorescence in Borneo

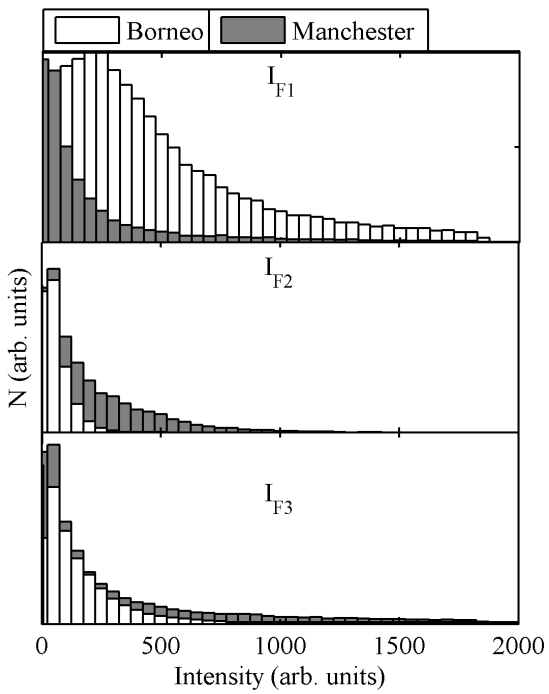


Figure S3. Histograms of fluorescence intensity at each site. Borneo N is scaled so as not to obscure the distribution behind. Minimum fluorescence intensity thresholds are subtracted before plotting.

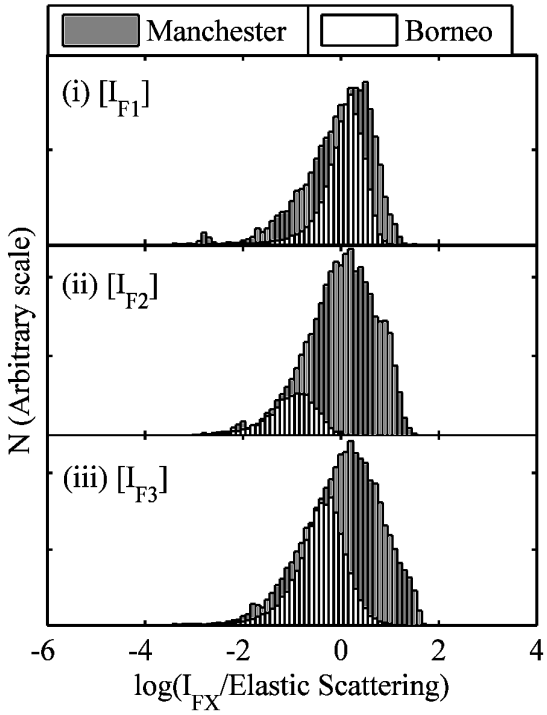


Figure S4. Histograms of fluorescence intensity divided by elastic scattering intensity for each site. Borneo N is scaled so as not to obscure the distribution behind.

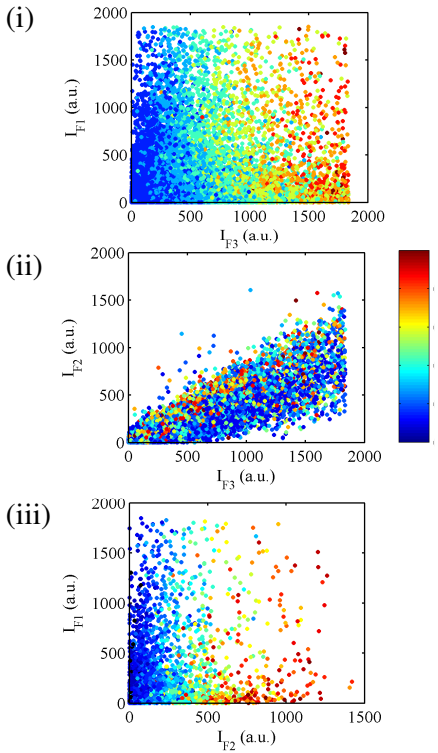


Figure S5. (i)F1 vs F2, (ii)F1 vs F3, (iii)F2 vs F3 scatter plots for Manchester. Each plot is coloured according to the intensity of fluorescence in the channel not on the x or y axes.

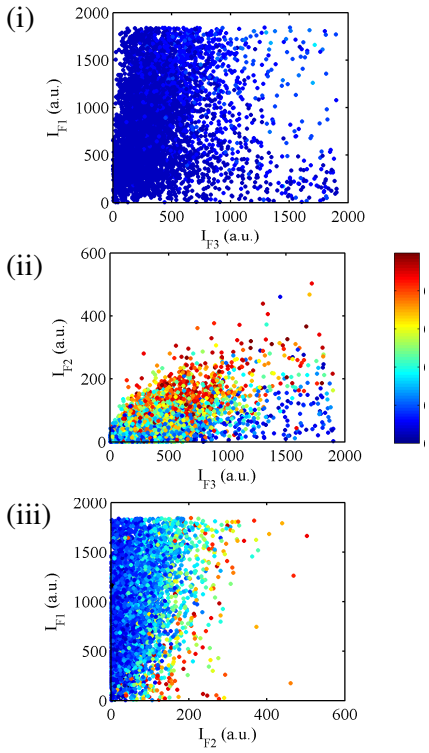


Figure S6. (i)F1 vs F2, (ii)F1 vs F3, (iii)F2 vs F3 scatter plots for Borneo. Each plot is coloured according to the intensity of fluorescence in the channel not on the x or y axes.

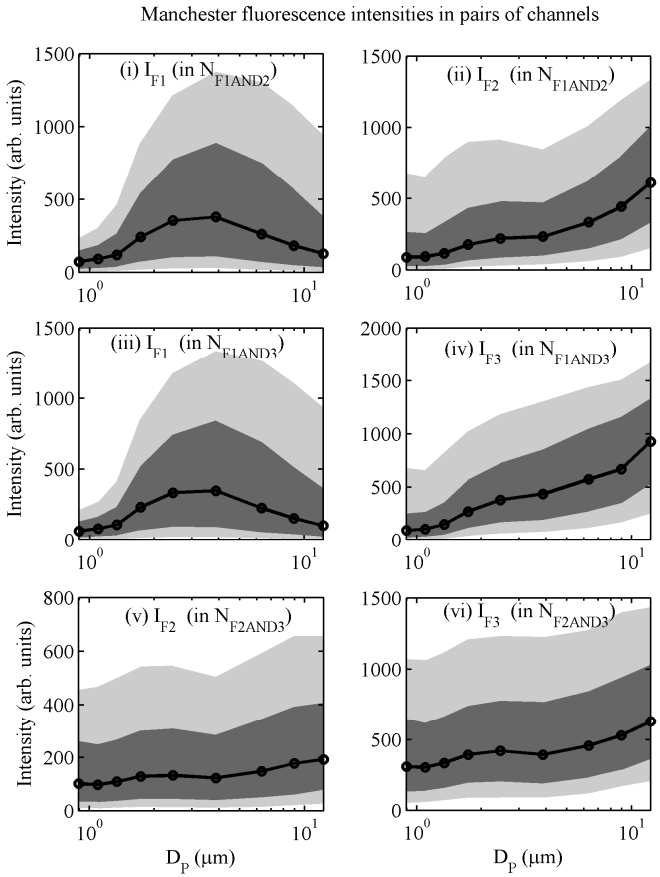


Figure S7. Manchester I_{F1}, I_{F2}; (ii) I_{F1}, I_{F3}; (iii) I_{F2}, I_{F3} vs D_P plots when particles exhibiting dual-channel fluorescence are selected from the dataset.

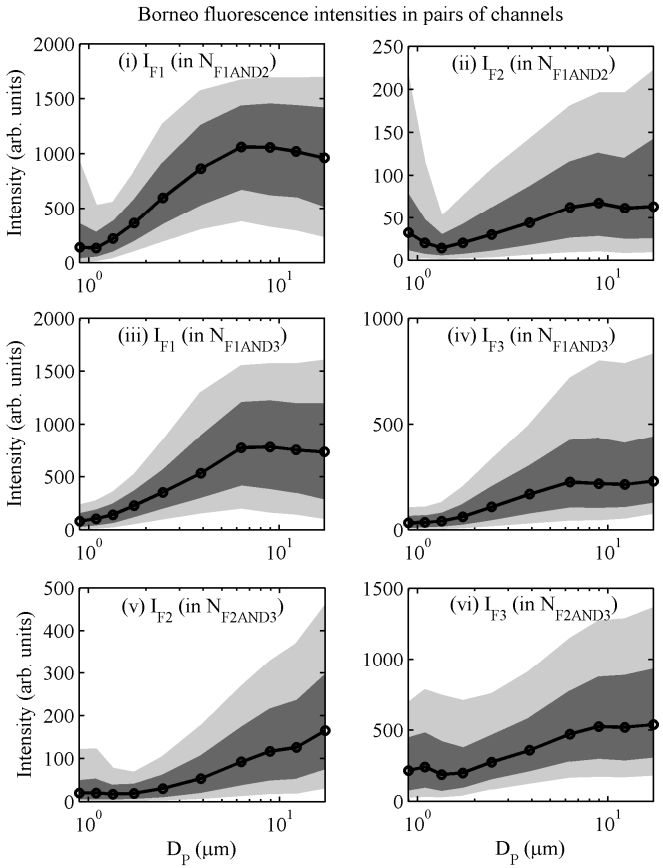
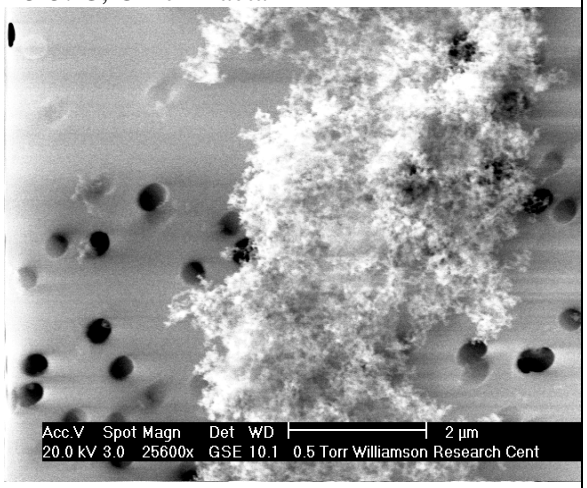


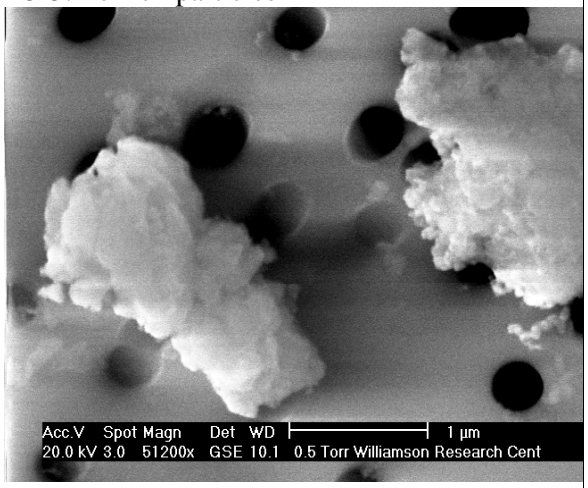
Figure S8. Borneo I_{F1}, I_{F2}; (ii) I_{F1}, I_{F3}; (iii) I_{F2}, I_{F3} vs D_P plots when particles exhibiting dual-channel fluorescence are selected from the dataset.

Figure S9. ESEM images of the more common coarse mode aerosol particle morphologies found in Manchester with major EDX peaks labelled. Non-PBA that resemble PBA are included, as are PBA candidates.

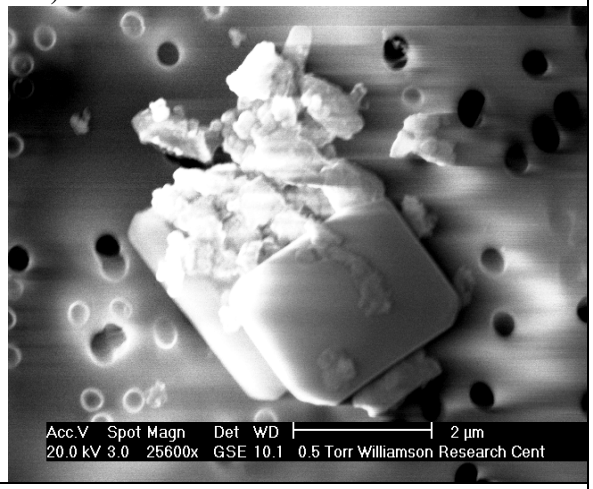
R3-3: C, O rich fractal



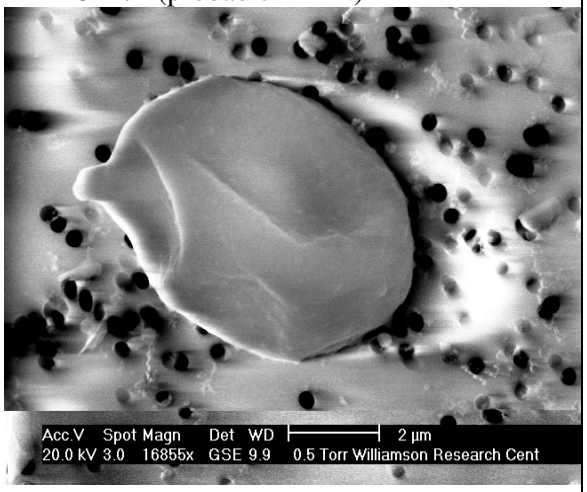
R3-5: Fe-rich particles



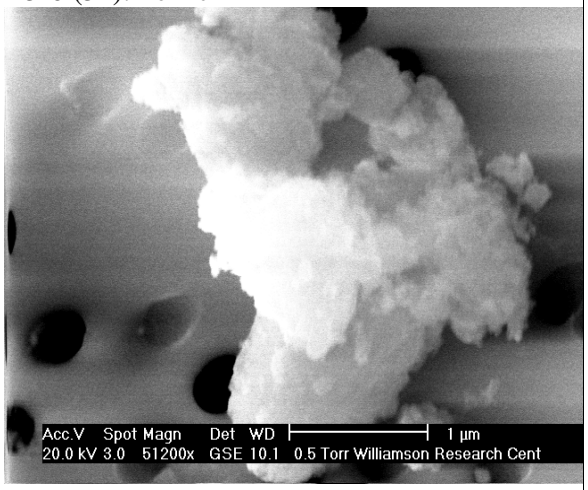
R5-2: Cl and Si dominated mixture (Part sea salt)



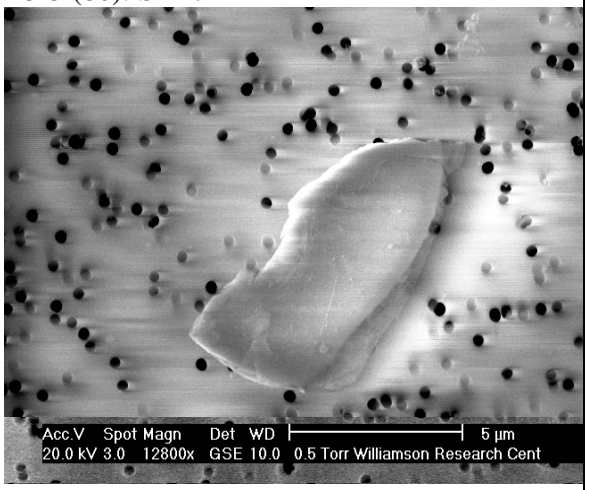
R1-4 C-rich (probable PBAP)



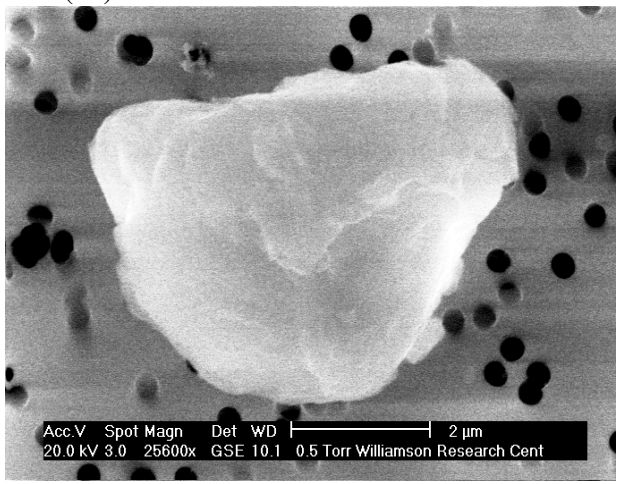
R3-6 (51): Fe-rich



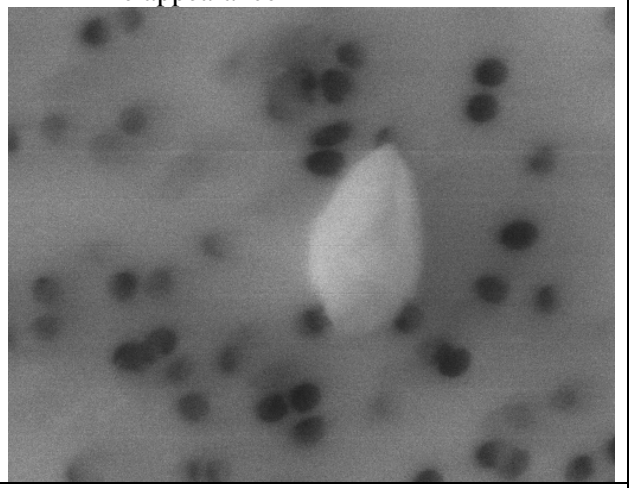
R5-5 (80): Si-rich



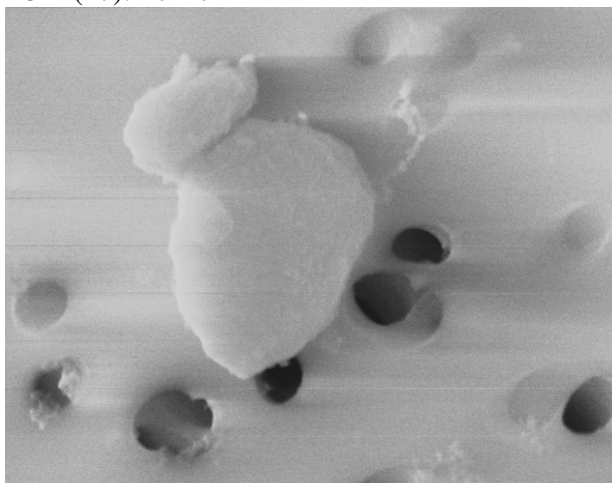
r5-1 (75): Si-rich



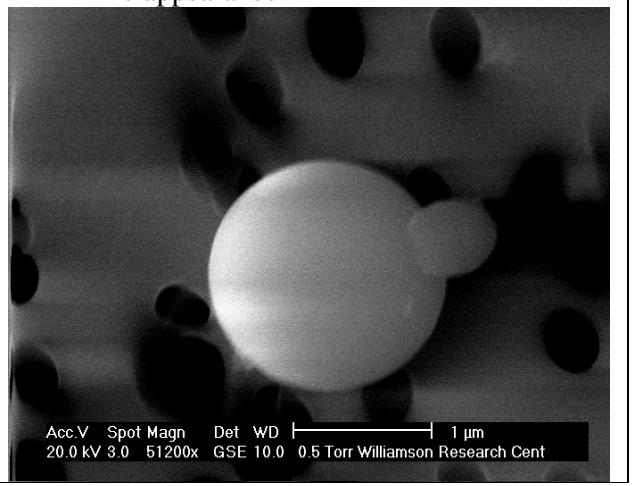
R2-A9: 2µm x 1µm Si-rich
"PBA-like appearance"



R3-2 (47): Fe-rich



R5-3 (78): Si-rich (probable alumo-silicate)
"PBA-like appearance"



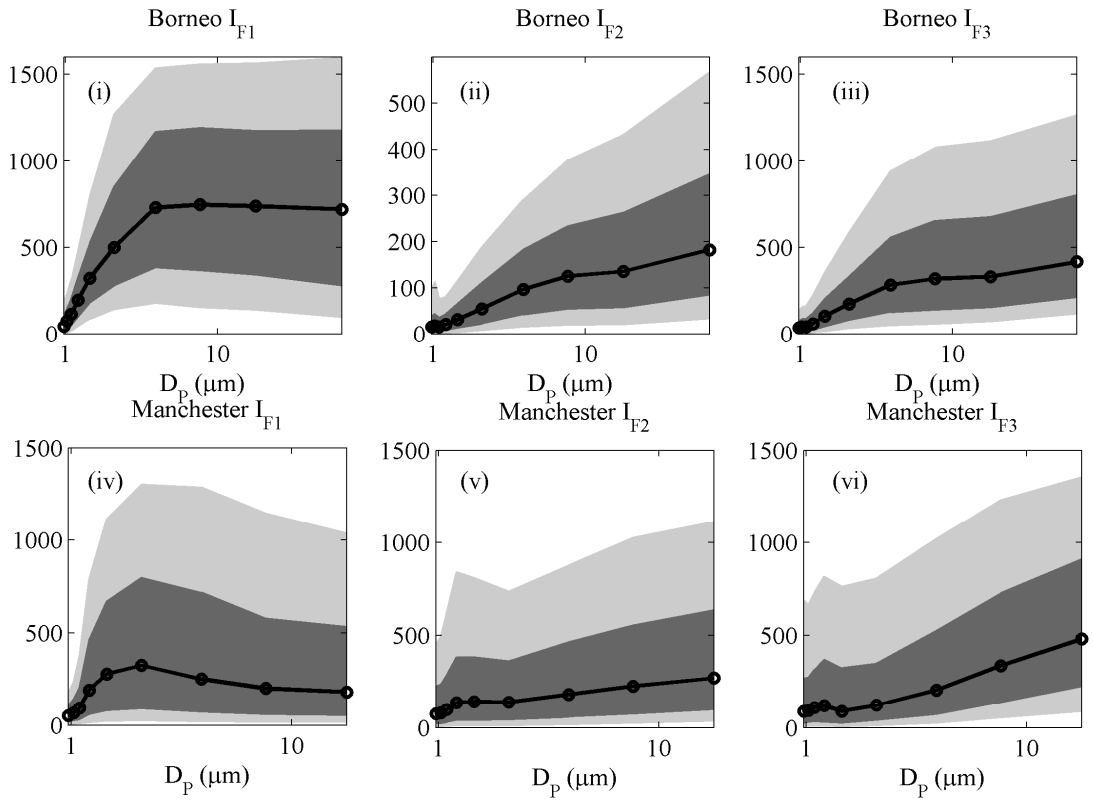


Figure S10 Plots of fluorescence intensity versus D_P on a linear scale using the same data as Figure 8 in the manuscript. Light grey shading denotes the 10th and 90th percentiles, dark grey shows the inter-quartile range and solid lines represent the median fluorescence intensity.

Table S1 The contribution of particles that saturate the fluorescence detector

	Contribution to N_{TOT} (%)			Contribution to N_{FL} (same channel)		
Location	F1	F2	F3	F1	F2	F3
Borneo	3%	0.008%	0.34%	5%	0.05%	0.6%
Manchester	0.13%	0.28%	0.6%	3.2%	5.1%	5.8%