

## Supplementary Material

**Table S1.** List of items added to the UHSAS flow system

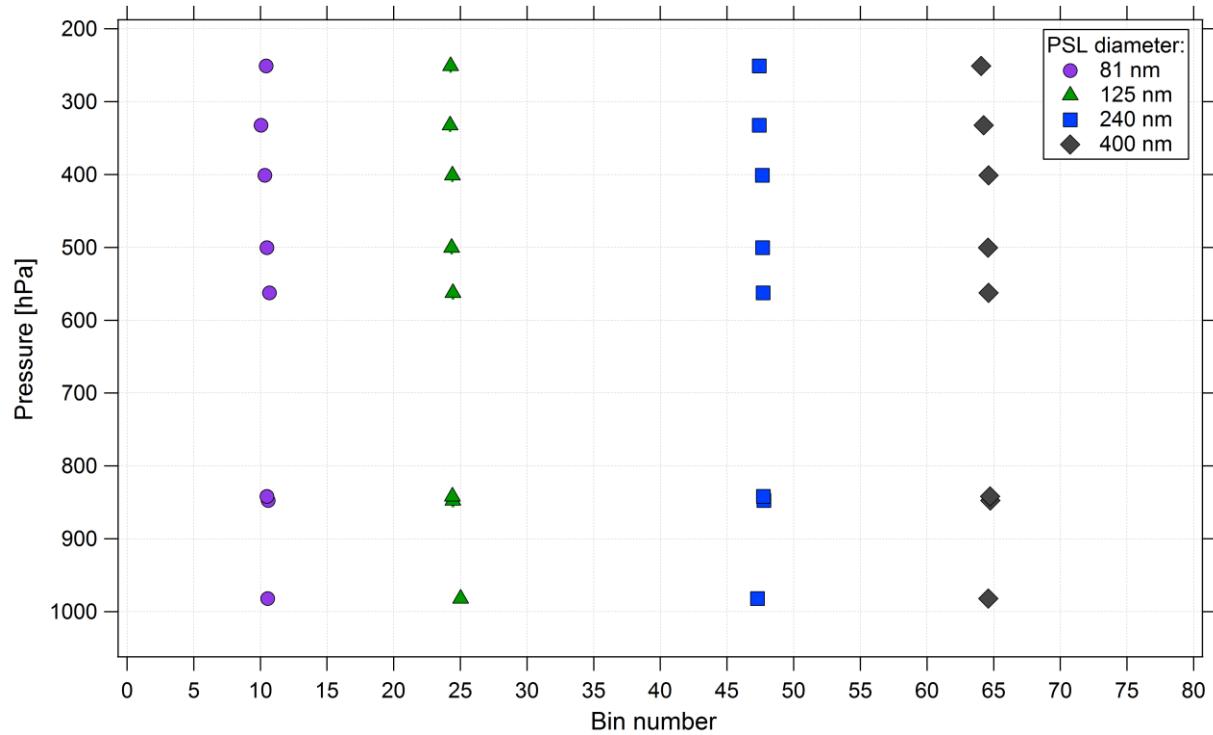
Part of the flow system	Item added	Specifications
Sample flow	Laminar flow element (LFE)	ID=0.069 cm Length=17.5 cm Laminar flow range: 0-0.1 L min <sup>-1</sup>
	Differential pressure transducer	Alicat, P-5in-dp, range: 0-2" H <sub>2</sub> O
Sheath flow	Volumetric flow controller (VFC) replaces needle valve	Alicat, MC-15LPM, operated in volume flow control mode

**Table S2.** Thermo denuder components and dimensions

Part	Model	Manufacturer
Temperature controller	CN32PT-224-C24	Omega Engineering Inc., Stamford, CT, USA
Solid state relay	120D25	OPTO 22, Temecula, CA, USA
Platinum resistance temperature detector (RTD)	HEL-707-U-1-12-00	Honeywell Sensing and Control, Golden Valley, MN, USA
Cylindrical cartridge heater	HDC00066	Honeywell Sensing and Control, Golden Valley, MN, USA
Housing tube	stainless steel, ID=1.12 cm	
Perforated tube insert	PS4542A: Stainless Steel 17-4 PH	Xometry, Inc., Gaithersburg, MD, USA
Activated carbon fiber cloth	FlexZorb Thickness 0.3 mm	Calgon Carbon Corporation, Moon Township, Pennsylvania, USA

**Table S3.** Estimated relative uncertainties in aerosol number ( $\sigma_N$ ), surface ( $\sigma_S$ ) and volume ( $\sigma_V$ ) at STP conditions. Uncertainties are calculated for representative size distributions in the marine boundary layer (MBL) and free troposphere (FT) using different averaging times.

Date and location	Integration time (s)	Counts	N (cm <sup>-3</sup> )	S (μm <sup>2</sup> cm <sup>-3</sup> )	V (μm <sup>3</sup> cm <sup>-3</sup> )	P (hPa)	$\sigma_N$ (%)	$\sigma_S$ (%)	$\sigma_V$ (%)
2017/02/05	1	31	34.1	2.376	0.074	1101	$\pm 18$	+29.5/-43.0	+35.0/-55.8
MBL	10	297	27.4	2.888	0.177	1101	$\pm 5.9$	+17.4/-29.7	+22.9/-40.5
32 °S	60	1654	27.4	2.81	0.14	1104	$\pm 2.6$	+14.1/-26.6	+19.7/-38.0
2017/02/10	1	134	138.2	19.614	1.636	1061	$\pm 8.7$	+20.2/-31.6	+25.7/-42.2
MBL	10	1582	150.3	26.065	2.779	1058	$\pm 2.7$	+14.2/-25.2	+19.7/-36.0
59.77 °S	60	8705	145.9	25.585	2.628	1061	$\pm 1.4$	+12.9/-23.9	+18.5/-34.7
2017/01/26	1	6	35.5	0.938	0.015	294	$\pm 40.8$	+52.4/-70.8	+57.9/-86.3
FT	10	40	21.7	0.458	0.007	293	$\pm 15.8$	+27.4/-47.1	+32.9/-62.9
4 °N	60	255	24.0	0.597	0.01	294	$\pm 6.3$	+17.9/-36.4	+23.4/-50.9
2017/01/26	1	53	442.4	10.119	0.156	256	$\pm 13.8$	+25.3/-44.4	+30.8/-59.5
FT	10	541	411.9	12.775	0.262	256	$\pm 4.4$	+15.9/-33.1	+21.4/-46.3
18.7 °N	60	3208	438.9	13.994	0.361	256	$\pm 2.0$	+13.5/-30.5	+19.0/-41.7



**Fig.S1.** UHSAS-2 sizing response to pressure change as a function of bin number for a mixture of four PSL particle sizes. Bin uncertainty, obscured by the symbols, is based on the uncertainty in a Gaussian fit to the size distribution histogram.