Supplemental Information

S1 WFIMS Configuration

Q_a	Aerosol Flow	0.3 l/min
Q_{sh}	Sheath Flow	16.5 l/min
ΔT_{con}	Conditioner	-12 °C
ΔT_{ini}	Initiator	+28 °C
ΔT_{mod}	Moderator	-10 °C
a	Distance between separator electrodes	0.91 cm
b	Width of flow channel = Entrance slit width	12.70 cm
	Entrance slit gap	0.25 mm
l_s	Separator length	14.18 cm
x_{view}	Range in x-direction of viewing area	0.18 cm–0.73 cm
y_{view}	Range in y-direction of viewing area	-3.50 cm− +3.50 cm

S2 HFIMS configuration during laboratory characterization

WFIMS	V	Separating Voltage	1000 V	
	$D_{p,min}$	Minimum wet particle diameter	44.8 nm	
	$D_{p,max}$	Maximum wet particle diameter	160.8 nm	
DMA	Type	NDMA		
	Q_a	Aerosol Flow	1.3 l/min	
	Q_{sh}	Sheath Flow	13 l/min	
	V	Classifying Voltage	5332 V	
	$D_{p,0}$	Dry Particle Diameter	50 nm	
RH Control			Deliquescence	Efflorescence
	RH_a	Aerosol RH	20 %−85 %	85 %
	RH_{sh}	Sheath RH		18.8 %−79.9 %
	RH	Mixed RH		20 %−80 %
HFIMS	$D_{p,min}/D_{p,0}$	Minimum Growth Factor	0.9	
	$D_{p,max}/D_{p,0}$	Maximum Growth Factor	3.2	
CPC	Q_a	Aerosol Flow	1 l/min	

S3 Effective Voltage Calibration

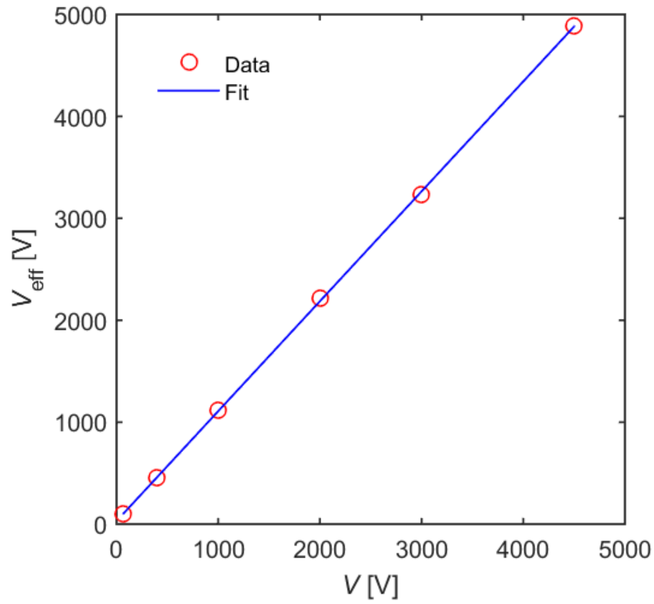

Figure S1 Calibration curve for the separator voltage of HFIMS. *V* and V_{eff} are applied and effective voltage, respectively.

To account for non-idealities of the electric field between WFIMS separator electrodes, the applied voltages V were replaced with effective voltages V_{eff} when calculating instrument response mobilities Z_p^*. The V_{eff} values were derived from WFIMS measurements of DMA classified particles at low RH (<10 %). For particles classified with DMA centroid mobilities $Z_{p,DMA}$ ranging from $9.2 \cdot 10^{-7}$ m²/Vs at V =70 V to $2.3 \cdot 10^{-8}$ m²/Vs at V = 4500 V (see Sect. S4), the values of V_{eff} (see Eq. (2)) were found such that Z_p^* calculated based on measured particle positions (Eq. (1)) and V_{eff} will reproduce $Z_{p,DMA}$. Resulting effective voltages are plotted as a function of applied voltages in Fig. S1 (open red circles). It can be seen that for the entire applied voltage range, i. e. 70−4500 V, V_{eff} exceeds V, and the relationship between V_{eff} and V is linear. Hence following parametrization was used to obtain a general expression for V_{eff} based on applied voltages:

$$V_{eff}(V) = a(V + b). \quad (S1)$$

By fitting V_{eff} derived from measurements parameters a and b were determined as 1.1 and 27.8 V, respectively. The 27.8 V represents a constant offset, and might be due to an offset of the WFIMS HV power supply, which has a full scale of 10000 V. The scaling factor a might be related to voltage dependent edge effects of the electric field.

S4 HFIMS Configuration during ambient tests

WFIMS	Separating Voltage [V]	Minimum wet particle diameter ($D_{p,min}$) [nm]	Maximum wet particle diameter ($D_{p,max}$) [nm]
	70	12.8	40.4
	400	28.1	94.1
	1000	44.8	160.8
	2000	64.9	253.3
	3000	81.3	338.3
	4500	102.5	461.4
	4500	102.5	461.4
DMA	Q_a	Aerosol Flow	0.3 l/min
	Q_{sh}	Sheath Flow	3 l/min
	Type	Classifying Voltage [V]	Dry Particle Diameter [nm]
	NDMA	115	15
	NDMA	599	35
	NDMA	1181	50
	NDMA	2202	70
	NDMA	4923	110
	LDMA	1148	165
	LDMA	2355	265
RH Control	RH_a	Aerosol RH	85%
	RH_{sh}	Sheath RH	
	RH	Mixed RH	
HFIMS	$D_{p,0}$ [nm]	Minimum Growth Factor ($D_{p,min}/D_{p,0}$)	Maximum Growth Factor ($D_{p,max}/D_{p,0}$)
	15	0.9	2.7
	35	0.8	2.7
	50	0.9	3.2
	70	0.9	3.6
	110	0.7	3.1
	165	0.6	2.8
	265	0.4	1.7
CPC	Q_a	Aerosol Flow	1.0 l/min
	Q_{Tr}	Transport Flow	5.4 l/min

S5 Conditioning of aerosol sample RH by the WFIMS sheath flow

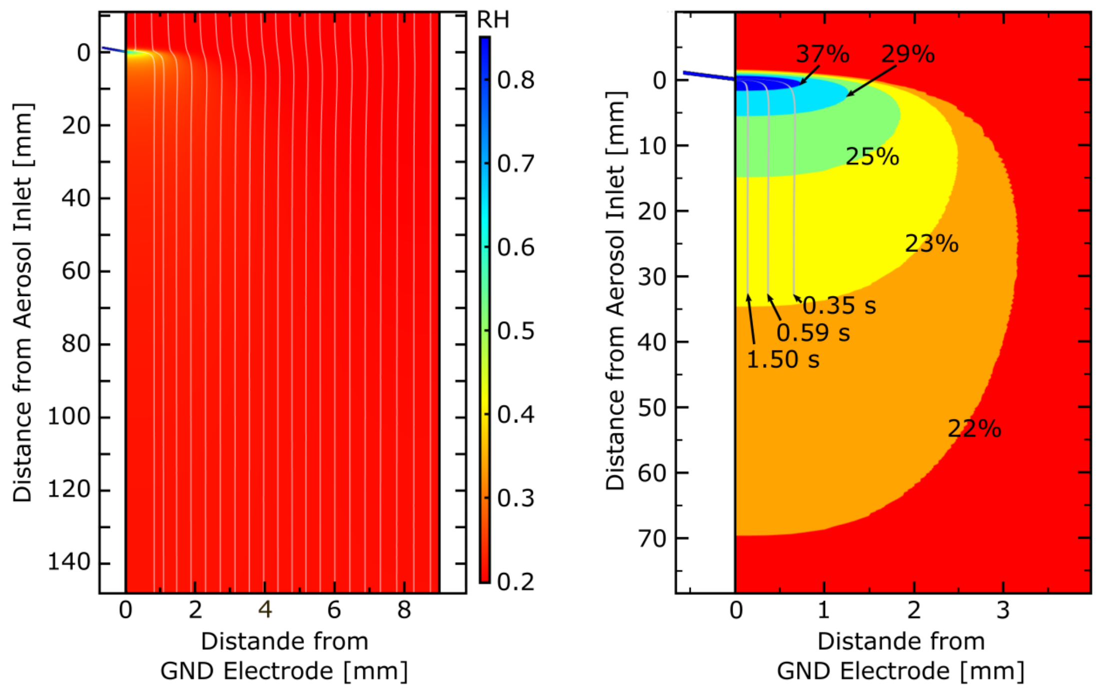

Figure S2 (a) Offset electrode used in HFIMS, with aerosol inlet on left. (b) Enlargement of inlet region showing relative humidity profiles when entering aerosol is at 85 % and sheath flow at 20 %.

The single high voltage electrode was configured with an offset such that the high voltage region begins slightly downstream (32 mm) of the introduction of aerosol into the mobility separator. This allows the sheath flow to provide the final humidity conditioning of the aerosol flow prior to mobility classification as the aerosol flow is just 2 % of the total flow. The electrode offset is illustrated in Fig. S2a, where the dimension along the width of the channel is shown enlarged relative to the length. Fig. S2b shows modeled relative humidity profiles at the aerosol inlet obtained using COMSOL Multiphysics®, a numerical modeling package. For instance when the entering aerosol is at 85% RH (0.3 l/min) and the entering sheath flow is at 20 % (16.5 l/min) then at the 32 mm position downstream where the mobility separation begins, the aerosol has reached 23 % RH, within 2.1 % of the desired RH 21.1 % =(0.3·85 %+16.5·20 %)/(0.3+16.5) after a complete mixing of aerosol and sheath flows. The equilibration time for the aerosol ranges from 0.35 to 1.50 s, as shown Fig. S2b. This approach allows the final RH adjustment of the aerosol to be made within the WFIMS system.