Supplementary Information

Impactor Apparatus for the Study of Particle Rebound: Relative Humidity and Capillary Forces

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S1. Semi-Empirical Design Constraints for Good Impactor Performance

Design criteria as a function of impactor transmission function were investigated theoretically by Marple et al. (1976) and subsequently tested against measurements (Marple and Willeke 1976). The design setpoint aerodynamic diameter $d_a^\dagger$ is related to the design particle Stokes number $\text{Stk}^\dagger$ by Equation 1 of the main text. For an impactor having straight orifice circular nozzles, the particle size having $\text{Stk}^\dagger$ of 0.20 corresponds to the particle size of 50% transmission efficiency (Jurcik and Wang 1995).

The Cunningham slip correction factor of Equation 1 is calculated as follows (Hinds 1999):

$$C_c = 1 + \text{Kn} \left[1.142 + 0.558 \exp(-0.999 \text{Kn}^{-1}) \right] \quad (\text{S1})$$

where Kn is the Knudsen number, defined as twice the gas mean free path divided by particle diameter. For simplification, we use d_a for particle diameter.

The average gas velocity of Equation 1 is calculated following the method outlined by Hering et al. (1987) for an assumption of adiabatic flow:

$$\bar{v} = \beta \left[1 - \bar{v}^2 \left[\frac{\gamma - 1}{2\gamma R T_0} \right] \right] \quad (\text{S2})$$

in which $\beta = (4Q/\pi\zeta D^2)(P_2/P_1)$, γ is the ratio of the heat capacity at constant pressure to that at constant volume, R is the ideal gas constant, and T_0 is the upstream temperature. β is the jet velocity for isothermal conditions, P_1 is the upstream pressure, and P_2 is the downstream pressure. Equation S2 is solved iteratively.

As alternative to a statement of impactor performance by the measured setpoint aerodynamic diameters d_a^* , a measured Stk^* can be related to d_a^* by Equation 1. For the measured setpoint aerodynamic diameters d_a^* of 84.9 and 277.7 nm for the α and β impactors, respectively, the

corresponding Stk^* values are 0.179 and 0.183 by Equation 1. For comparison, commercial impactors such as ELPI obtained Stk^* values between 0.177 and 0.233.

An impactor is characterized by a transmission function $\Omega(d_a)$. Typical optimized designs seeks to make the slope of the function as steep as possible at d_a^* . Steep slopes are typical for a Reynolds numbers between 500 and 3000 (Marple and Liu 1974; Marple and Willeke 1976). The Reynolds number is a dimensionless parameter defined as the ratio of inertial forces to viscous forces of the gas flow through the nozzle:

$$Re = \frac{\rho_g \bar{v} D}{\eta} \quad (S3)$$

where ρ_g is the gas mass density. The Reynolds number for the α and β impactors for the conditions of this study is 500.

For multiple nozzles, independent behavior is essential to modeling and understanding the data sets. The spacing among the nozzles must therefore be sufficiently apart that interactions do not occur among the flow fields. For this purpose, a dimensionless cross-flow parameter X can characterize the interactions among multiple nozzles, as follows (Fang et al. 1991a):

$$X = \frac{\zeta D}{4D_c} \quad (S4)$$

in which D_c is the nozzle cluster diameter (m). The nozzle cluster diameter is defined as the diameter for which all nozzles are inside the circle. Satisfying the condition of $X < 1.2$ obviates significant interactions among nozzle flows (Fang et al. 1991a; Kwon et al. 2002). For the α and β impactors, X is 0.03.

Additional design considerations include the spacing L between the nozzles and the impaction plate as well as the thickness T of the nozzles in relation to nozzle diameter D . Represented in dimensionless form as L/D and T/D , $L/D > 1.0$ and $T/D > 1.0$ should be satisfied (cf. Table 1) such that

the design equation based on $Stk^\dagger$ holds (i.e., Equation 1) (Marple and Liu 1974; Marple and Willeke 1976).

With the foregoing design criteria in mind, two types of nozzle plates were drilled, the first having 32 holes of nominal dimensions D of 0.084 mm diameter (referred to hereafter as the α plate) and the second having 15 holes of nominal dimensions of 0.180 mm diameter (referred to hereafter as the β plate) (cf. Table 1). The housing and impaction plates were fabricated from aluminum. The nozzle plate was micro-drilled from stainless steel (National Jet Company). The distance L between the nozzle plate and the impaction plate was controlled through the use of stacked brass shims that allowed stepwise changes in L of 50 μm . The precise distance L was not critical to the performance of a jet impactor (Marple and Liu 1974; Hinds 1999). The plates and housing were sealed by Viton o-rings.

During the course of the experiments, the inflow rate to each impactor was measured using a Gilibrator (Sensidyne), and the pressure drop across each impactor was measured using a differential pressure gauge (Dwyer, DM-1112). The pressure drop was varied by using a downstream needle valve. This data set allowed the impactor stages to be used as orifice-based flowmeters for the actual experiments (i.e., by measurement of pressure drop). Most of the pressure drop was obtained by a critical orifice housed in a downstream condensation particle counter (CPC; TSI 3010) connected to external vacuum (Seifert et al. 2004). Modulation of the pressure drop and hence additional small flow was achieved by adjusting a needle valve connected to vacuum that was in a parallel configuration with the CPC. The downstream pressures of the α and β nozzle plates were 87.5 ± 0.3 kPa and 98.5 ± 0.3 kPa, respectively (Table 1). The standard deviation arises from slight differences in the manufacturing of the multiple α and β nozzle plates.

The flow and the pressure of each CPC were measured daily. Particle concentrations were adjusted by up to 10% using the ratio of the actual flow at actual pressure to a nominal flow of 1 Lpm at

standard pressure. Following Zhang et al. (1991) and Seifert et al. (2004), the intrinsic counting efficiency of the CPC above 30 kPa did not depend on pressure.

S2. Flow Dynamics and Relative Humidity

Several possibilities should be considered concerning the possible effects of flow dynamics on relative humidity.

A. Pressure drop

Because of the pressure drop in some impactors, such as in cascade impactors (e.g., ELPI Stage #3 and MOUDI Stage #9 of Table 1), there is a decrease in relative humidity upstream of the impactor stage of interest relative to the ambient relative humidity. The change in upstream pressure results in reduced RH according to the following equation:

$$RH_0 = RH_{amb} \frac{P_1}{P_{amb}} \quad (S5)$$

where RH_0 is the upstream relative humidity, RH_{amb} is the ambient relative humidity, P_1 is the upstream pressure, and P_{amb} is the ambient pressure. As an example, the corresponding RH_0 , implying the maximum RH values that can be reached for $RH_{amb} = 100\%$, are 66% RH and 89% RH for the ELPI Stage #3 and MOUDI Stage #9. The α and β impactors by comparison can be operated using $P_1 = P_{amb}$ such that up to 100% RH can be used in the experiments.

B. Jet Flow

As the flow expands and accelerates through the nozzle, both temperature and pressure drop isentropically. The temperature change is related to the speed of the fluid, as follows:

$$T_n = T_{amb} \left(1 + \frac{k-1}{2} Ma^2 \right)^{-1} \quad (S6)$$

where T_n is the temperature in the nozzle, T_{amb} is the ambient temperature, k is the ratio of heat capacity at constant pressure to that at constant temperature for air, and Ma is the Mach number of the jet. The

decrease in temperature within the jet decreases the saturation vapor pressure of water, favoring an increase in relative humidity. The drop in pressure, however, decreases the partial pressure of water, favoring a decrease in relative humidity. For the α impactor operated according to the conditions of Table 1, the relative humidity dropped by approximately 10% RH because of the pressure drop and increased by approximately 33% RH because of the temperature drop. The respective values were 2% RH and 5% RH for the β impactor.

These changes in gas phase water activity provide a thermodynamic driving force for evaporation or condensation of water. An aqueous particle traversing through an impactor can either shrink or grow depending on the overall net effect of the pressure drop and temperature drop on relative humidity (Fang et al. 1991b). Possible changes in particle diameter would introduce systematic errors in $\Omega(d_a)$. In order to assess the importance of the change of thermodynamic driving force on particle diameter, however, the kinetics of evaporation or condensation must be considered.

The characteristic time required for equilibration between gas and particle phase water is estimated by the following equation (Fang et al. 1991b):

$$\tau_{equi} = \frac{R \rho J_c}{8 D_v Ma J} \frac{d_{equi}^2 - d_0^2}{P_1/T_1 - P_2/T_2} \quad (S7)$$

where R is the universal gas constant, ρ is the particle material density, d_{equi} is the equilibrium diameter, d_0 is the initial particle diameter, D_v is the binary diffusion coefficient for water in air, Ma is the Mach number of the jet, P_1 is the partial pressure of water at the particle surface (previous stage pressure), T_1 is the temperature at the particle surface (previous stage temperature), P_2 is the local partial pressure of water in the nozzle, T_2 is the local temperature in the jet, and J/J_c is a correction factor for the Fuchs effect (Fuchs and Sutugin 1970; Seinfeld and Pandis 1998). For the calculation of a lower limit to τ_{equi} , the initial diameters were chosen so that the equilibrium diameters would correspond to a jet RH of 95%. The initial diameters corresponded to 76 and 91 % RH for the α and β impactor, respectively. The

initial and equilibrium diameters were estimated based on the growth factor of ammonium sulfate. Results, corresponding to lower limits of $\tau_{equi} = 48 \mu s$ and $\tau_{equi} = 360 \mu s$ for the α and β nozzle plate respectively, are tabulated in Table S3.

To assess the possibility of kinetic limitations, the lower limits of the equilibration times should be compared to the residence times. The residence times are calculated from the length of the nozzle and velocity of the jet, as follows:

$$\tau_{res} = \frac{T + L}{v_0} \quad (S8)$$

for terms as defined in Tables 1 and 2. The residence times in the jet are $\tau_{res} = 8 \mu s$ and $\tau_{res} = 26 \mu s$ for the α and β impactors, respectively. The implication is that $\tau_{res} < \tau_{equi}$, implying an absence of diameter change.

S3. Preparation and Conditioning of Test Aerosols

Ammonium sulfate particles (Fluka, Analytical, $\geq 99.9999\%$) were produced by atomization (TSI 3076) of 0.01% or 1.0% (w/w) aqueous solutions in nanopure water (18.1 M Ω cm). Polystyrene latex particles were atomized from an aqueous suspension prepared using 64 drops of 400 nm PSL spheres (Thermo Scientific, NIST traceable) in 100 mL water. Oleic acid particles were generated by vaporization-condensation below 100 °C in a U-tube configuration (Sigma Aldrich, $\geq 99\%$ (GC)) (Kuwata and Martin 2012). Flows were regulated using mass flow controllers (MKS Instruments M100B) and powered by an analog box (MKS Instruments 247D). Particle-free dry air was produced using a commercial pure air generator (AADCO, 737 series).

Once produced, the ammonium sulfate and PSL aerosols flowed through a 2-L chamber to allow any large droplets to separate by gravitation and then passed through two diffusion dryers to decrease RH to $< 5\%$. For oleic acid, the drying treatment was not needed. After drying (as needed), 0.4 Lpm was passed into a 10-L mixing chamber and was diluted with 10 - 20 Lpm of dry air. The number-diameter

distributions of the particle populations were measured by a scanning mobility particle sizer (TSI, model 3080) without a pre-impactor (Wang and Flagan 1990).

The sample flow was conditioned to an RH of < 5% up to 95% by passage through a Nafion tube (Perma Pure Nafion tube, MD 110). The RH system utilized proportioning of dry and wet air flows (Rosenoern et al. 2009). An RH probe (Rotronic Hygroclip, HC2-S) was located in-line at the exit of the Nafion tube. The RH probe was calibrated using saturated salt solutions consisting of LiCl (11.3%), MgCl₂ (33.01%), NaCl (75.3%), KCl₂ (85.11%), and KNO₃ (94.5%). A pressure drop of 1 kPa occurred across the Nafion tubing, setting an upper limit of 99% RH for the impactor experiments.

S4. Effect of Collection Time on Surface Loading

The impaction plates were employed either uncoated or coated with high-vacuum grease (Dow Corning). The plates were coated by applying grease with a spatula and then placed in an oven overnight to produce a smooth surface. The uncoated plates (when clean) allowed particle rebound to occur whereas the coated plates eliminated particle rebound (Turner and Hering 1987; Marple et al. 1991; Pak et al. 1992). The estimated coating thickness was 45 μm based on gravimetric analysis.

The effect of exposure time on transmission efficiency was evaluated in control experiments having high particle concentrations ($\geq 1500 \text{ cm}^{-3}$). The control experiments were conducted using the setup shown in Figure 1 (gray boxes bypassed), and a dry ammonium sulfate test aerosol was used. The transmission functions of both the coated and uncoated plates are plotted in Figure S7 as a function of collection time and fractional surface loading S_L in the impaction region, defined as the ratio of the total projected area of deposited particles to the nozzle area, as follows:

$$S_L = \frac{N d_p^2}{\zeta D^2} \quad (\text{S9})$$

in which N is the number of deposited particles based on the drop in CPC concentration (Wang and John 1987).

For the coated plates, the transmission function did not change for exposure times of up to 90 min for typical experimental conditions (Figure S7). There were significant effects, however, of collection time on the transmission function for the initially clean, uncoated impaction plates. The transmission function decreased (indicating reduced rebound) for operation times longer than 20 min (Figure S7). The reduced rebound arose from the build-up of a particle film on the impaction plate. Several factors reduced rebound, including increased adhesion at impact, an uneven surface, and other factors (Wang and John 1987). This problem of plate aging was avoided in the actual experiments by following a protocol that maintained particle number concentrations below 1000 cm^{-3} and replaced the uncoated plate every five experiments (ca. 20 min each).

S5. Conversion from DMA Voltage to Aerodynamic Diameter

The setpoint particle electric mobility Z^* of the DMA is calculated from the voltage applied to the inner collector rod as follows:

$$Z^* = \frac{Q}{2\pi \phi L} \ln\left(\frac{R_2}{R_1}\right) \quad (\text{S10})$$

in which Q is the sheath air flow rate, R_1 is the inner radius of the flow channel, R_2 is the outer radius of the flow channel, ϕ is the rod voltage, and L is the effective length of the rod (Knutson and Whitby 1975). Mobility diameter d_m is related to electric mobility, as follows:

$$Z = \frac{q e C_c(d_{m,q})}{3\pi \eta d_{m,q}} \quad (\text{S11})$$

in which q is the number of elementary charges on the particle and e is the elementary charge. Equating the two equations at the setpoint and solving for the particle diameter gives. The following equation is obtained on combining Equations S10 and S11:

$$\frac{d_{m,q}}{C_c(d_{m,q})} = \frac{2 q e \phi L}{3 \eta Q \ln(R_2/R_1)} \quad (\text{S12})$$

The aerodynamic diameter is related to mobility diameter as follows (Kelly and McMurry 1992; Hinds 1999):

$$C_c(d_a) d_a^2 = \frac{\rho_p}{\rho_0} \frac{C_c(d_m)}{d_m} d_{me}^3 \quad (\text{S13})$$

for the mass-equivalent diameter d_{me} . There is the relationship that $C_c(d_m)/d_m = C_c(\delta d_{me})/\kappa \delta d_{me}$ for a porosity factor δ , an envelope shape factor κ , and a mass-equivalent diameter d_{me} . With this equation, for a limiting approximation of a nonporous (i.e., $\delta = 1$) and spherical (i.e., $\kappa = 1$) particle, $d_m = d_{me}$.

Equation S13 reduces to the following:

$$d_a = \left(\frac{\rho_p}{\rho_0} \frac{C_c(d_m)}{C_c(d_a)} \right)^{1/2} d_m \quad (\text{S14})$$

in which d_a is the aerodynamic diameter, ρ_p is the particle material density, ρ_0 is the reference material density (1000 kg m^{-3}). Combining equations S12 and S14 leads to the following result for relating aerodynamic diameter to the voltage applied to the DMA rod:

$$d_{a,q} = \left(\frac{\rho_p}{\rho_0} \frac{C_c^3(d_{m,q})}{C_c(d_a)} \right)^{1/2} \frac{2 q e \phi L}{3 \eta Q \ln(R_2/R_1)} \quad (\text{S15})$$

In practice, given the presence of the term $C_c(d_m)$ in Equation S15, this equation is not solved directly. Rather Equations S12 and S14 are numerically solved stepwise. An example of the sequence of conversions of the data set from DMA voltage to particle aerodynamic diameter is plotted in Figure S8 for $q = +1$, including RH-dependent corrections of particle material density.

After the DMA, the particles underwent RH conditioning prior to entering the impactor apparatus. Adjustments are made in the analysis for changes in particle diameter. The RH-dependent hygroscopic growth factors for ammonium sulfate are taken from Biskos et al. (2006). For $<80\%$ RH, the ammonium sulfate particles are solid ($\rho = 1770 \text{ kg m}^{-3}$) and taken as spherical. At 70% RH, a particle growth factor of 1.01 is taken into account for surface-adsorbed water and to estimate the

material density ($\rho = 1747 \text{ kg m}^{-3}$). Above 80% RH, the particles deliquesce and are aqueous. For the aqueous particles, because of hygroscopic growth and the Kelvin effect the material density is adjusted both for diameter and relative humidity (cf. Table 1 of Biskos et al. (2006), including erratum that 0.0234 therein should be 0.0177).

PSL spheres are insoluble in water and do not undergo hygroscopic growth for $\text{RH} < 100\%$, except for adsorption that does not change diameter significantly. Experimental results from tandem DMA experiments to measure the hygroscopic growth factor of PSL spheres at 70% and 85% RH indicate that the growth factor is unity within uncertainty (Ye et al. 2009; Tan et al. 2013). PSL spheres grow only for supersaturated conditions (i.e., as CCN) (Kim et al. 2011). Although this growth could occur for β nozzles for $\text{RH} > 96\%$, the particle residence time in supersaturated conditions is not sufficient to allow significant condensation (cf. §SS2).

S6. Analysis including Multiply Charged Particles

The particle population passing the DMA and entering the impactor was dominated in number concentration by $d_{m,+1}$ because of the relative charging efficiencies of +1, +2, and +3 charge particles. Even so, there were sufficient $d_{m,+2}$ particles, as well as $d_{m,+3}$ particles to a lesser extent, that data corrections for multiply charged particles were needed. The multiply charged particles had the same electric mobility (thus passing through the DMA together for a single voltage setting) but had progressively larger aerodynamic diameters of $d_{a,+1}$, $d_{a,+2}$, and $d_{a,+3}$. The number concentration of each subpopulation was $N(d_{a,+1})$, $N(d_{a,+2})$, and $N(d_{a,+3})$, respectively, for a total particle number concentration of N . The particle subpopulations had transmission functions $\Omega(d_{a,+1})$, $\Omega(d_{a,+2})$, and $\Omega(d_{a,+3})$ through the impactor. The observed transmission function $\Omega(d_m)$ then had several contributing terms, as follows:

$\Omega(d_m) = (N(d_{a,+1})\Omega(d_{a,+1}) + N(d_{a,+2})\Omega(d_{a,+2}) + N(d_{a,+3})\Omega(d_{a,+3}))/N$. The data corrections sought to obtain $\Omega(d_{a,+1})$ from measurements of $\Omega(d_m)$.

The procedure for data correction to compensate for multiply charged particles was as follows. The number concentrations $N(d_{a,+1})$, $N(d_{a,+2})$, and $N(d_{a,+3})$ of the +1, +2, and +3 subpopulations passing the DMA were simulated using the *AerosolCalculator* (www.seas.harvard.edu/AerosolCalculator), taking into account the number-diameter distribution of the total particle population measured by the SMPS and conversion of mobility diameter to aerodynamic diameter (Equation S14). For a specific DMA setting of $d_{m,+1} = i$, the observed transmission function Ω^i through the impactor was described as follows:

$$\Omega^i = \frac{1}{N^i} \begin{bmatrix} \dots 0 \dots & N^i(d_{a,+1}) & \dots 0 \dots & N^i(d_{a,+2}) & \dots 0 \dots & N^i(d_{a,+3}) & \dots 0 \dots \end{bmatrix} \begin{bmatrix} \Omega(\dots) \\ \Omega(d_{a,+1}) \\ \Omega(\dots) \\ \Omega(d_{a,+2}) \\ \Omega(\dots) \\ \Omega(d_{a,+3}) \\ \Omega(\dots) \end{bmatrix} \quad (\text{S16})$$

More generally, measurements $\Omega^i(d_m)$ across all DMA settings of $i \in \{1, \dots, p\}$ can be represented as follows:

$$\begin{matrix} \mathbf{A} & = & \mathbf{B} & \mathbf{C} \end{matrix}$$

$$\begin{bmatrix} \Omega^1(d_m) \\ \vdots \\ \Omega^p(d_m) \end{bmatrix} = \begin{bmatrix} N^1(d_{a,+1})/N^1 & \dots 0 \dots & N^1(d_{a,+2})/N^1 & \dots 0 \dots & \dots 0 \dots \\ \dots & \dots & \dots & \dots & \dots \\ \dots 0 \dots & N^p(d_{a,+1})/N^p & \dots 0 \dots & N^p(d_{a,+2})/N^p & \dots 0 \dots \end{bmatrix} \begin{bmatrix} \Omega^1(d_a) \\ \vdots \\ \vdots \\ \vdots \\ \Omega^j(d_a) \end{bmatrix}$$

for a set of aerodynamic diameters j for $j \in \{d_a^{\min}, \dots, d_a^{\max}\}$ across the interval $\{d_a^{\min}, d_a^{\max}\}$. Given that

$\mathbf{A}$ and $\mathbf{B}$ are known, the transmission functions $\Omega^j(d_a)$ are then be obtained by taking the pseudoinverse

of matrix $\mathbf{B}$, i.e., $\mathbf{C} = \mathbf{B}^{-1} \mathbf{A}$. In the typical analysis, $\mathbf{A}$ had 9 rows and $\mathbf{C}$ had 15 rows for the α impactor and $\mathbf{A}$ had 15 rows and $\mathbf{C}$ had 35 rows for the β impactor. Data sets with corrections for multiply charged particles appear in Figures S9c and S9d, Figures S10c and S10d, and Tables 2, S1, and S2. Other figures and tables present uncorrected data.

Examples of the analysis, including both original and corrected data, are shown in Figures S9 and S10 for particle populations of oleic acid and ammonium sulfate, respectively. Smaller diameters have more proximate multiply charged diameters than do larger diameters. A prominent example of the contribution and subsequent correction for multiply charged particles is apparent in the interval of 60 to 80 nm in Figure S9a (no correction) compared to Figure S9c (corrected). The apparent step function in Figure S9a arises from the +2 particles. This step function is absent in Figure S9c because of the data correction. The correction applied for multiply charged particles is generally greater for the α compared to the β impactor because of the number-diameter distribution of the test aerosol.

S7. Numerical Simulations of Flow Fields and Particle Trajectories

The flow fields inside the impactors were simulated numerically using the *Non-Isothermal Flow* (NITF) module of the software package *COMSOL Multiphysics*. Simulation conditions included compressible flow, an absence of turbulence ($\text{Re} = 500$ and $\text{Ma} < 0.4$), an axisymmetric geometry of the nozzle plate over the impaction plate, no slip at the walls, and a macroscopic temperature of 293 K. The input and output boundary conditions conformed to the upstream and downstream pressures. The simulation grid included 247000 and 339000 points for the α and β impactors, respectively. Figure S2 shows false-color plots of the simulated flow speeds. Figure S3 shows vector plots of the flow. The fields are similar to those presented in Arffman et al. (2011) for a similar configuration. The average jet velocity and temperature were calculated by integration across the nozzle exit surface and are listed in Table 2.

Particle trajectories were simulated in the flow field by a Euler method (Arffman et al. 2011). Particle bins were introduced radially across the entrance of the nozzle. Thirty positions and twenty-four particle sizes were used. The coated-plate transmission function corresponded to that fraction of particles that had non-intersecting trajectories with the impaction plate. The simulated transmission function for uncoated impaction plates took into account the impact velocity in conjunction with either Equation 9 or 13. Examples of the normally incident impact velocities are plotted in Figure S4 to show the dependence on radial position at the nozzle entrance. Depending on flow conditions, the maximum velocities can occur at the centerline (Figure S4a) or on the edges (Figure S4b) (Arffman et al. 2012). Examples of the resulting probability density function of normally incident impact velocities (i.e., taking into account the particle flux along each radial streamline of the nozzle) are plotted in Figure S5.

S8. Offsets for Coated compared to Uncoated Impaction Plates

There is a systematic offset in d_a^* of up to 10% between the transmission functions of the coated compared to the uncoated plates (Figures S9 and S12; Tables S1 and S2). This offset can be seen, for example, in Figure S9b. The transmission functions in the coated experiments (closed symbols) are shifted to smaller diameters compared to transmission functions for the uncoated experiments (open symbols). The systematic offset can be at least partially explained by differences in the distance from nozzle exit and the impaction plate due to the thickness of the grease film applied to the impaction plates. The nozzle-to-plate distance L shortened by approximately 10% for the coated compared to uncoated plates.

There can also be systematic errors associated in the estimate of aerodynamic diameter, especially at high RH as suggested by the outlier values at 95% RH (Table S2). Although the particles were selected dry by the DMA, they were subsequently grown to a larger diameter at elevated RH. This diameter was estimated by a hygroscopic growth model (Biskos et al. 2006). Uncertainties in the model

or in the measurement or control of RH near 95% RH can contribute to errors in the estimated wet particle diameter.

In addition to the systematic differences between coated and uncoated plates, there is also random variability on order of 5% among the data sets of a single plate type. The random variability of 5% lies within that of previous literature results (Kelly and McMurry 1992; Stein et al. 1994). One source of random error was the use of different nozzle plates during the course of experiments. Although each nozzle plate was nominally identical, in actuality there were differences in their manufacture, and the nozzle positions and diameters varied sufficiently to affect the data sets. The importance of a consistent use of the same nozzle plate to reduce this source of 5% variability was discovered only after the original data sets had been collected.

Because of variation in d_a^* , as apparent in Figures S9 and S12 as well as Tables S1 and S2, the transmission function as plotted in Figure 4 is an empirical adjustment to the original data shown in Figure S12 by translation of each data as necessary along the abscissa to the experimentally determined average value of d_a^* listed in Table 2.

S9. Capillary Adhesion Energy

At the time of impact, a meniscus is assumed to form between the particle and the surface separated by distance z_0 . The particle has zero velocity at this point, and the meniscus is assumed to have time to fully form. As the particle separates ($z > z_0$), the volume of the meniscus is assumed to stay constant, implying an assumption that the rebound time is shorter than the characteristic time of evaporation/condensation. The water volume then necks as the particle separates.

For this description, the capillary force between a sphere and a flat surface is to good approximation as follows (Butt and Kappl 2009):

$$F = 2\pi \sigma c d_p \left(1 - \left(1 + \frac{2 V_0}{z^2 \pi d_p} \right)^{-1/2} \right) \quad \text{for } z \geq z_0 \quad (\text{S17})$$

Terms σ and c are defined in the main text. Term V_0 is a fixed volume of water in the meniscus. The volume depends on the initial curvature r_0 of the meniscus:

$$V_0 = (\pi/2) (4 r_0^2 c^2 - z_0^2) d_p \quad (\text{S18})$$

The initial curvature produced at the point of impact z_0 depends on relative humidity, as follows:

$$r_0 = -\frac{2 \sigma V_m / R T}{\ln(\text{RH}/100)} \quad (\text{S19})$$

in which V_m is the molar volume of water (Yang et al. 2010).

The RH-dependent adhesion energy arising from capillary forces is given by integration of Equation S17, as follows (§SS9)

$$\begin{aligned} E_a &= \int_{z_0}^{\infty} F dz \\ &= 2\pi \sigma c d_p \int_{z_0}^{\infty} \left(1 - \left(1 + \frac{2 V_0}{z^2 \pi d_p} \right)^{-1/2} \right) dz \\ &= 2\pi \sigma c d_p \left(-1 + \sqrt{1 + \frac{2 V_0}{z_0^2 \pi d_p}} \right) z_0 \end{aligned} \quad (\text{S20})$$

A value of $z_0 = 0.2$ nm is typically estimated (Israelachvili 2011). Under the approximation of $z_0 \rightarrow 0$ for Equation S20, the following simplified equation is obtained:

$$\begin{aligned} E_a &\approx 2\pi \sigma c d_p \lim_{z_0 \rightarrow 0} \left[\left(-1 + \sqrt{1 + \frac{2 V_0}{z_0^2 \pi d_p}} \right) z_0 \right] \\ &= 2 \sigma c \sqrt{2\pi d_p V_0} \\ &= -\frac{8\pi \sigma^2 c^2 d_p V_m}{R T \ln(\text{RH}/100)} \end{aligned} \quad (\text{S21})$$

For the range of diameters of the present study, the difference in results between Equations S20 and S21 for E_a ranges from $< 1\%$ for 95% RH up to 45% for 5% RH. Thus, for higher RH values, Equation S21 is an acceptable approximation of the adhesion energy arising from capillary forces. For the low RH values, the calculated E_a is negligibly small so that the relative difference between Equations S20 and S21 is not important with respect to the influence of capillary forces on bounce, i.e., van der waals forces dominate at low RH. For these reasons, the simplification of Equation S21 is used in the present analysis.

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Relative Humidity	α Impactor				β Impactor			
	Assuming Singly Charged Particles		Adjusted for Multiply Charged Particles		Assuming Singly Charged Particles		Adjusted for Multiply Charged Particles	
	d_a^* (nm)	δ (nm)	d_a^* (nm)	δ (nm)	d_a^* (nm)	δ (nm)	d_a^* (nm)	δ (nm)
Uncoated	64.1 $\pm$ 1.9	11.8 $\pm$ 1.7	78.4 $\pm$ 1.8	11.1 $\pm$ 1.4	277.0 $\pm$ 0.3	12.1 $\pm$ 0.2	293.5 $\pm$ 0.5	3.9 $\pm$ 0.4
	66.8 $\pm$ 2.3	13.0 $\pm$ 2.1	82.6 $\pm$ 1.9	11.6 $\pm$ 1.6	264.2 $\pm$ 0.3	13.0 $\pm$ 0.3	285.6 $\pm$ 0.7	9.2 $\pm$ 0.7
	73.9 $\pm$ 1.4	9.5 $\pm$ 1.3	90.6 $\pm$ 0.7	4.4 $\pm$ 0.7	268.0 $\pm$ 0.5	12.8 $\pm$ 0.5	288.4 $\pm$ 0.8	7.8 $\pm$ 0.7
Coated	56.4 $\pm$ 3.7	8.0 $\pm$ 3.2	81.3 $\pm$ 2.7	5.0 $\pm$ 2.0	260.2 $\pm$ 0.2	15.1 $\pm$ 0.3	281.5 $\pm$ 0.8	11.8 $\pm$ 0.8
	60.2 $\pm$ 2.9	9.6 $\pm$ 2.6	84.1 $\pm$ 2.5	7.2 $\pm$ 1.9	241.7 $\pm$ 0.3	20.6 $\pm$ 0.3	262.1 $\pm$ 0.9	20.3 $\pm$ 0.8
	70.5 $\pm$ 1.1	10.4 $\pm$ 1.0	87.2 $\pm$ 0.9	6.8 $\pm$ 0.8	256.8 $\pm$ 0.9	20.4 $\pm$ 1.0	276.5 $\pm$ 1.3	17.2 $\pm$ 1.2
Mean	65.4 $\pm$ 5.8	10.4 $\pm$ 5.2	84.0 $\pm$ 4.6	7.7 $\pm$ 3.6	261.3 $\pm$ 1.1	15.7 $\pm$ 1.2	281.3 $\pm$ 2.1	11.7 $\pm$ 2.0

Table S1. Measured setpoint aerodynamic diameters across three sets of replicate measurements for uncoated and grease-coated impaction plates using liquid spherical particles (oleic acid, $\rho = 895 \text{ kg m}^{-3}$, cf. Figure S9). The setpoint aerodynamic diameter d_a^* and the width parameter δ are calculated from fitting the data sets to Equation 2. One-sigma uncertainties to the parameter fits, taking into account uncertainties in the data points, are listed.

Relative Humidity	α Impactor				β Impactor			
	Assuming Singly Charged Particles		Adjusted for Multiply Charged Particles		Assuming Singly Charged Particles		Adjusted for Multiply Charged Particles	
	d_a^* (nm)	δ (nm)	d_a^* (nm)	δ (nm)	d_a^* (nm)	δ (nm)	d_a^* (nm)	δ (nm)
5	88.8 $\pm$ 1.7	12.1 $\pm$ 1.5	90.2 $\pm$ 1.5	11.5 $\pm$ 1.5	247.2 $\pm$ 1.5	22.0 $\pm$ 1.2	249.7 $\pm$ 1.2	15.9 $\pm$ 1.1
25	88.1 $\pm$ 1.1	10.9 $\pm$ 1.0	89.4 $\pm$ 1.0	10.5 $\pm$ 0.9	279.4 $\pm$ 1.6	23.6 $\pm$ 1.5	284.7 $\pm$ 0.7	19.0 $\pm$ 0.7
50	86.6 $\pm$ 1.6	10.7 $\pm$ 1.4	87.9 $\pm$ 1.4	10.0 $\pm$ 1.3	256.6 $\pm$ 1.0	25.3 $\pm$ 0.9	262.0 $\pm$ 1.1	20.9 $\pm$ 1.0
70	83.5 $\pm$ 1.0	10.2 $\pm$ 0.9	86.2 $\pm$ 1.5	11.1 $\pm$ 1.2	265.4 $\pm$ 1.2	29.9 $\pm$ 1.1	271.5 $\pm$ 1.2	25.5 $\pm$ 1.0
80	84.5 $\pm$ 0.7	7.1 $\pm$ 0.8	85.3 $\pm$ 0.7	6.5 $\pm$ 0.7	289.4 $\pm$ 1.9	21.5 $\pm$ 1.9	294.1 $\pm$ 1.2	15.8 $\pm$ 1.1
95	75.5 $\pm$ 0.1	6.3 $\pm$ 0.1	76.0 $\pm$ 0.1	6.3 $\pm$ 0.1	277.4 $\pm$ 0.6	17.7 $\pm$ 0.4	282.4 $\pm$ 0.7	16.3 $\pm$ 0.7
Mean	84.5 $\pm$ 2.8	9.5 $\pm$ 2.5	85.8 $\pm$ 2.8	9.3 $\pm$ 2.6	269.2 $\pm$ 3.3	23.3 $\pm$ 3.1	274.1 $\pm$ 2.6	18.9 $\pm$ 2.3

Table S2. Measured setpoint aerodynamic diameters at different relative humidities for a test aerosol of ammonium sulfate (cf. Figure S10). The setpoint aerodynamic diameter d_a^* , the width parameter δ , and the one-sigma uncertainties are calculated from fitting the data sets to Equation 2. For <80% RH, the ammonium sulfate particles are solid ($\rho = 1770 \text{ kg m}^{-3}$) and taken as spherical. At 70% RH, a particle growth factor of 1.01 is taken into account for surface-adsorbed water and to estimate the material density ($\rho = 1747 \text{ kg m}^{-3}$). Above 80% RH, the particles deliquesce and are aqueous. For the aqueous particles, because of hygroscopic growth and the Kelvin effect the material density is adjusted both for diameter and relative humidity (cf. Table 1 of Biskos et al. (2006), including erratum that 0.0234 therein should be 0.0177).

Nozzles	$\bar{v}$ (m s ⁻¹)	Ma	d_0 (nm)	d_{equi} (nm)	τ_{equi} (μ s)	τ_{res} (μ s)
α	106.5	0.47	95	115	48.4	7.6
β	44.8	0.21	320	326	360	26.4

Table S3. Summary of terms and results for the equilibration time compared to the residence time in the jet. See further discussion in §SS2.

Particle Diameter (nm)	Relaxation Time (μs)	Nozzle Exit		First Impact		Second Impact	
		Velocity (m s^{-1})	Stopping Distance (μm)	Velocity (m s^{-1})	Stopping Distance (μm)	Velocity (m s^{-1})	Stopping Distance (μm)
76.4	0.063	157	9.9	7.2	0.5	0.9	0.1
81.4	0.068	157	10.7	11.2	0.8	1.1	0.1
86.4	0.073	157	11.5	15.5	1.1	1.2	0.1
91.4	0.078	157	12.3	20.1	1.6	1.2	0.1
96.4	0.084	157	13.1	23.9	2.0	1.2	0.1
101.4	0.089	157	14.0	28.7	2.6	1.3	0.1
106.4	0.095	157	14.9	32.4	3.1	1.3	0.1
111.4	0.100	157	15.8	36.5	3.7	1.4	0.1
116.4	0.106	157	16.7	40.5	4.3	1.5	0.2
121.4	0.112	157	17.6	44.3	5.0	1.7	0.2
126.4	0.118	157	18.6	47.9	5.7	2.0	0.2
131.4	0.125	157	19.6	51.3	6.4	2.4	0.3
141.4	0.138	157	21.6	58.0	8.0	3.7	0.5
156.4	0.158	157	24.8	66.8	10.6	6.6	1.0
181.4	0.195	157	30.7	79.1	15.4	12.0	2.3
206.4	0.236	157	37.0	89.4	21.1	17.7	4.2

Table S4. Velocities and stopping distances at nozzle exit (center), for first impact, and for second impact associated with Figure S6.

Supplementary Figures

Figure S1. Cross-section view of impactors. The scale of the interior region is expanded in the vertical for ease of viewing. The impactor can hold either one stage or two sequential stages. Measurements of the present study were conducted using only one single stage.

Figure S2. False-color plot of the numerically simulated spatial field of the flow speed. For clarity of presentation, the aspect ratios of the panel are not unity compared to actual dimensions. The α -stage and β -stage have displayed aspect ratios of 2:1.1 and 3:1, respectively. The simulation used the parameters of Table 1 in the Computational Fluid Dynamics module of the software package *COMSOL Multiphysics* (4.3.0.151, Stockholm, Sweden).

a. α - Nozzle Plate

b. β - Nozzle Plate

Figure S3. Vector plot of the velocity field of Figure S2.

Figure S4. Simulated normally incident first-bounce impact velocities depending on initial radial position in the nozzle. Results are shown for several different particle diameters. (a) α Impactor. (b) β Impactor.

Figure S5. Probability density function of normally incident first-bounce impact velocities for (a) the α impactor and (b) the β impactor. Results are shown for several different particle diameters. For clarity of presentation, the probability density functions are smoothed across the underlying bin distributions.

Figure S6. (a) Simulated particle trajectories near the impaction plate for particles of $d_a = 181$ nm for several initial radial positions in the α impactor. In this simulation, upon collision with the impaction plate, the z -component of the particle velocity v_z reverses (i.e., $e = 1.0$ implying elastic collision). Particle rebound occurs for $v_z > v_c$ (Equation 9). For some of the trajectories, a particle collides a second or third time with the impaction plate. In the case that the second or third collision occurs with $v_z < v_c$, the particle adheres. The relaxation time and stopping distance of each bin included in the simulation are listed in Table S4 for the first and second impact. (b) Simulated rebound fraction taking into account multiple collisions. Cases taking into account up to one, two, and three impacts are plotted. The single-impact case corresponds to the unscaled simulations of Figure 5. The uncertainty expressed by the hatched region results from the error in ξ (Table 3). The simulation results are compared to experimental data collected for dry conditions ($RH < 5\%$).

Figure S7. Effect of exposure time on the transmission functions of coated and uncoated impaction plates for a test aerosol of ammonium sulfate using the α impactor. Fractional surface loading is defined by Equation S9. Results are shown for several different particle number concentrations. Dashed lines represent exponential fits to the data.

Figure S8. Example of the conversion from $\Omega(Z^*)$, to $\Omega(d_m)$, to $\Omega(d_a)$ in panels a, b, and c, respectively (cf. §SS5). Results are shown for a test aerosol of ammonium sulfate particles impinging onto coated impaction plates for several different RH values. Particles of +1 charge are assumed for the shown panels (cf. §SS6).

Figure S9. Transmission functions $\Omega(d_a)$ of oleic acid particles for uncoated (open symbols) and coated (solid symbols) impactation plates. Data are segregated by symbols for nominally identical nozzle plates (see text). (a) α -Impactor, no corrections to data for multiply charged particles (i.e., all assumed to have +1 charge). (b) β -Impactor, no corrections for multiply charge particles. (c) α -Impactor, data corrected for contribution by multiply charged particles. (d) β -Impactor, data corrected for contribution by multiply charged particles.

Figure S10. Similar to Figure S9 but for a test aerosol of ammonium sulfate. The colored lines correspond to experiments at different RH, using the same color scheme as Figure S8.

Figure S11. Number-diameter distributions of test aerosols used in the experiments underlying Figure 6. (a) Polystyrene latex (PSL) particles and (b) ammonium sulfate particles. Dashed lines represent the impactor setpoints used in the associated experiments.

Figure S12. Original data of Figure 4 in absence of translation of data sets along abscissa (i.e., as done to match d_a^* of Table 2).