

1 **Developing A Custom-Built Metal ~~Cloud~~Aerosol Processing Chamber: Analysis of Aerosol**  
2 **Coagulation at Low Humidities.**

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4 Nevil A. Franco, Kyle J. Gorkowski, Katherine B. Benedict  
5 Earth and Environmental Science Division, Los Alamos National Laboratory, P.O. Box 1663  
6 Los Alamos, NM, US 87545-1663

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8 Corresponding Authors:

9 Kyle J. Gorkowski; [gorkowski@lanl.gov](mailto:gorkowski@lanl.gov)  
10 Katherine B. Benedict; [kbenedict@lanl.gov](mailto:kbenedict@lanl.gov)

11  
12 **Abstract**

13 We ~~are developing~~[have developed](#) an intermediate size (906 L) ~~cloud~~[aerosol processing](#)  
14 chamber, and this paper reports on the design and initial characterization of dry aerosol  
15 experiments. Specifically, we are determining wall-loss and coagulation correction factors using  
16 the observed size distribution measurements for surrogates of common aerosol classes: sodium  
17 chloride, sucrose, and ~~soot~~[biomass burning aerosol smoke](#). Results show that, on average,  
18 sodium chloride, sucrose, and ~~soot~~[smoke](#) wall-loss rates converge to similar values on relatively  
19 short time scales (<1 hour). The fitted coagulation correction factor,  $W_c^{-1}$ , for ~~soot~~[smoke](#)  
20 particles ( $1.23 \pm 0.312$ ), indicates [that on average](#) they adhere to each other more than sodium  
21 chloride ( $0.969 \pm 0.524$ ) and sucrose ( $1.16 \pm 1.38$ ). [The relative uncertainty is high for the](#)  
22 [coagulation correction, but it is consistent with our Monte Carlo error analysis.](#) This study lays

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the foundation for future experiments at elevated humidity and supersaturation conditions to characterize the influence of particle shape on coagulation and cloud parameters.

## 1 Introduction

Aerosol-cloud interactions remain one of the largest sources of uncertainty in the Earth's radiation budget. ~~By directly scattering, absorbing solar radiation and indirectly influencing cloud formation, aerosols affect longwave and shortwave radiation in the Earth's atmosphere<sup>1</sup>. Despite sustained research efforts, these impacts still pose significant challenges to our understanding of the aerosol cooling effect, estimated at  $-0.86 \pm 0.56 \text{ W/m}^2$ , and the effective anthropogenic radiative forcing of Earth's climate (estimated at  $-1.25 \pm 0.85 \text{ W/m}^2$ )<sup>1</sup>. By directly scattering, absorbing solar radiation and indirectly influencing cloud formation, aerosols affect longwave and shortwave radiation in the Earth's atmosphere (IPCC, 2023). Despite sustained research efforts, these impacts still pose significant challenges to our understanding of the aerosol cooling effect, estimated at  $-0.86 \pm 0.56 \text{ W/m}^2$ , and the effective anthropogenic radiative forcing of Earth's climate (estimated at  $-1.25 \pm 0.85 \text{ W/m}^2$ )(IPCC, 2023).~~ The complexity of aerosol sources, properties, and processing continues to hinder precise quantification of these forcing estimates.

~~A critical source of aerosols is wildfire smoke, which can influence radiative budgets up to a year depending on the transport and evolution of plumes<sup>2-4</sup>. Under extreme burning conditions, wildfires can generate pyrocumulonimbus clouds, lofting large concentrations of aerosol into the upper troposphere and lower stratosphere<sup>2,5,6</sup>. These smoke particles can exert prolonged effects on climate through chemical and physical processes such as condensation and coagulation<sup>7-9</sup>. The fractal nature of soot particles further complicates our understanding of their indirect effects on cloud formation and radiative properties<sup>10-12</sup>. For instance, during the Amazon~~

biomass burning season, Koren et. al.<sup>43</sup> reported a dramatic reduction in cumulus cloud cover—  
from 38% under cleaner conditions to 0% during heavy smoke. However, Kaufman & Koren et.  
al.<sup>44</sup> observed an increased cloud cover in regions with higher column aerosol concentrations.  
These discrepancies underscore the complexity of aerosol-cloud interactions, which depend on  
various factors such as aerosol composition, hygroscopicity, size distribution, supersaturation,  
and the prevailing atmospheric stability<sup>45</sup>. As wildfires increase in frequency and intensity due to  
climate change<sup>46</sup>, refining our knowledge of how these aerosols evolve and ultimately affect  
cloud development is crucial for improving climate models and future predictions.

Beyond large-scale aerosol effects, aging aerosols undergo microphysical transformations  
that can drastically alter their role in cloud processes. Condensation of organics and the mixing  
of sulfate with black carbon (BC) have both been shown to influence cloud dynamics<sup>47</sup>. Recent  
work indicates that larger BC agglomerates may form preferentially at cloud tops, while the  
heaviest-coated BC particles are most likely to be scavenged by cloud droplets<sup>48,49</sup>. Modeling  
these highly dynamic processes remains challenging, as it requires accurately representing  
particle growth, mixing states, and cloud interactions<sup>20–24</sup>.

Cloud chambers are valuable research tools for investigating microphysical mechanisms  
under well-controlled conditions<sup>25–28</sup>. Existing cloud chambers are their own institutional facility  
in the case of CLOUD at CERN<sup>29</sup>, AIDA Chamber EUROCHAMP<sup>30</sup>, and PI chamber at MTU<sup>26</sup>.  
All chambers however, come with artifacts—most notably, the loss of particles to chamber walls  
through gravity, diffusion, convection, and electrostatic forces<sup>21–24</sup>. Previous studies have  
highlighted the importance of accounting for both size-dependent and time-dependent wall  
losses<sup>25,26</sup>.

In this paper, we introduce the development of a Los Alamos National Laboratory (LANL) cloud chamber, which is specifically designed. A critical source of aerosols is wildfire smoke, which can influence radiative budgets up to a year depending on the transport and evolution of plumes (D'Angelo et al., 2022; Guimond et al., 2023; Yu et al., 2019). Under extreme burning conditions, wildfires can generate pyrocumulonimbus clouds, lofting large concentrations of aerosol into the upper troposphere and lower stratosphere (Leach and Gibson, 2021; Rodriguez et al., 2020; Yu et al., 2019). These smoke particles can exert prolonged effects on climate through chemical and physical processes such as condensation and coagulation (Fromm et al., 2022; Gorkowski et al., 2024; Reisner et al., 2023). The fractal nature of soot particles further complicates our understanding of their indirect effects on cloud formation and radiative properties (Cotton and Anthes, 2010; Das et al., 2021; June et al., 2022). For instance, during the Amazon biomass burning season, Koren et al. (2004) reported a dramatic reduction in cumulus cloud cover—from 38% under cleaner conditions to 0% during heavy smoke. However, Kaufman & Koren et al. (2006) observed an increased cloud cover in regions with higher column aerosol concentrations. These discrepancies underscore the complexity of aerosol-cloud interactions, which depend on various factors such as aerosol composition, hygroscopicity, size distribution, supersaturation, and the prevailing atmospheric stability (Feingold et al., 2001). As wildfires increase in frequency and intensity due to climate change (Cunningham et al., 2024), refining our knowledge of how these aerosols evolve and ultimately affect cloud development is crucial for improving climate models and future predictions.

Beyond large-scale aerosol effects, aging aerosols undergo microphysical transformations that can drastically alter their role in cloud processes. Condensation of organics and the mixing of sulfate with black carbon (BC) have both been shown to influence cloud dynamics (Ching et

al., 2018). Recent work indicates that larger BC agglomerates may form preferentially at cloud tops, while the heaviest-coated BC particles are most likely to be scavenged by cloud droplets (Taylor et al., 2014; Zanatta et al., 2023). Modeling these highly dynamic processes remains challenging, as it requires accurately representing particle growth, mixing states, and cloud interactions (Ching et al., 2016; Riemer et al., 2009; Yang et al., 2023; Yao et al., 2021; Zaveri et al., 2010).

Aerosol chambers are used to understand these chemical and microphysical transformation in controlled conditions (Becker, 2006; Doussin et al., 2023). Many were built for gas-phase and secondary organic aerosol experiments and feature large volumes with Teflon walls to reduce wall losses (Hynes et al., 2005; Shao et al., 2022b). Others are optimized for specific aerosol processes, like bioaerosols (Massabo, 2018). Cloud chambers are a class of chambers for investigating cloud microphysical mechanisms under well-controlled conditions (Chang et al., 2016; Khlystou et al., 1996; Niedermeier et al., 2020; Shao et al., 2022). Existing cloud chambers are their own institutional facility in the case of CLOUD at CERN (The Cloud Collaboration, 2001), AIDA Chamber EUROCHAMP (Wagner et al., 2006), and PI-chamber at MTU (Chang et al., 2016). These types of facilities are critical for advancing science but are often oversubscribed and require significant support to operate.

As outlined in many of the papers cited in the previous paragraph, all chambers however, come with artifacts—most notably, the loss of particles to chamber walls through gravity, diffusion, convection, and electrostatic forces (Corner and Pendlebury, 1951; Fotou and Pratsinis, 1993; Mahfouz and Donahue, 2020a; Wang et al., 2018). Previous studies have highlighted the importance of accounting for both size-dependent and time-dependent wall losses (Crump et al., 1982; Crump and Seinfeld, 1981).

In this paper, we introduce the development of a Los Alamos National Laboratory (LANL) aerosol processing chamber, which we use to investigate coagulation processes under simulated conditions. We present initial data from experiments where aerosols were injected in a dry environment to quantify losses to chamber walls, dilution, and coagulation effects. Different types of aerosols were examined to validate known aerosol behaviors and characterize coagulation. We further demonstrate the use of a python based aerosol package, Particula<sup>37</sup>, to model coagulation and wall loss rates. Through these studies, we aim to refine experimental design and advance understanding of how aerosols—particularly soot—undergo physical transformations that shape their role in cloud formation and climate forcing (Particula, 2025), to model coagulation and wall-loss rates. Through this study, we aim to characterize the behavior of aerosol in the dry chamber (influence of particle composition and shape) and determine conditions suitable for future studies at elevated humidity including supersaturation. In addition, we perform an uncertainty analysis on the coagulation correction retrieval to determine the range of aerosol concentrations that reduce uncertainty in coagulation corrections.

## 2 Chamber Development and Methods

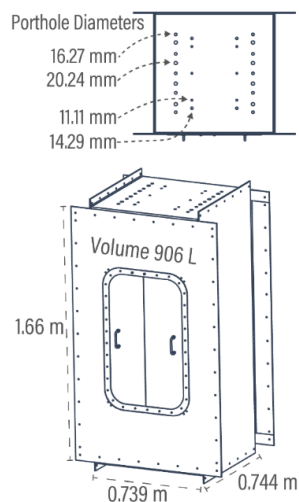
### 2.1 Setup of chamber and experiments

The LANL chamber is in the first phase of development with control of both temperature and humidity to be added in future work. The 906 L (0.906 m<sup>3</sup>, internal volume) chamber is made of 6 stainless steel walls which are inert and reduce the effects of electrostatic charge. The rectangular body and dimensions are shown in Figure 1. The chamber has an internal surface area of 6 m<sup>2</sup> with surface-to-volume ratio of 6.6 m<sup>-1</sup>. The chamber's joints are sealed with a fast cure marine adhesive caulk (Sika, Sikaflex 291) and the outside junctions where the walls

~~intersected were~~intersect are sealed using ZIP System™ Strech Tape (Huber Engineered Woods).

Portholes were made for the top and bottom plates where wires and probes be placed for measurements in the chamber and for aerosols to flow in and out of the chamber. Unused portholes are sealed with Swagelok caps and those used for probes and wires are sealed with a rubber gasket or a Teflon ferrule. A leak test was performed by pressurizing the chamber by feeding clean air in and sealing every outlet then seal any leak detected. [The chamber is designed to operate at ambient pressure.](#)

Copper tubing lines (3/8”) are used to supply aerosols to the chamber and deliver outflow sampling to instrumentation. Zero-air generators (T701 Teledyne Inc., USA) provide clean dry air to push aerosol to the chamber and additional dilution air using Teflon tubing (1/4”). Push flow enters at the bottom of the chamber, creating an upwards direction of flow. Aerosols are sampled from an outlet at the top of the chamber. A dilution flow is connected to the outlet line (88.9 mm from the outlet) to control aerosol concentrations and prevent overwhelming the sampling instruments. A minimum sampling flow rate of 1.5 L/min was needed to supply the instruments and we used a 1:5 ratio of push to dilution for the experiments presented here. This infers a residence timescale within the chamber of 604 minutes (10 hours) and half-life of 418 minutes (6.9 hours). The flow rates are controlled with mass flow controllers (MFC; Alicat). Prior to each experiment the chamber was flushed by pushing clean air with a flow of ~10 L/min for at least 3 hours to reach background (~0-10 cm<sup>-3</sup>).



**Figure 1. Schematic of LANL's 906 L chamber. The chamber has external dimensions of 1.66 m in height, 0.739 m in width, and 0.744 m in depth. The design includes 56 portholes with diameters ranging from 11.11 mm to 20.24 mm, shown across the top and mirrored on the bottom.**

## 2.2 Aerosol Generation and Instrumentation

Two aqueous solutions and controlled combustion of dried biomaterial were used as the sources of aerosols. Sodium chloride (NaCl; Sigma-Aldrich) was chosen because it is a well-understood compound in aerosol studies. Sucrose (Sigma-Aldrich) was used to act as a secondary organic aerosol surrogate, and it is also a well-studied aerosol. Each were dissolved in deionized water (Milli-Q, 18.2 MΩ) in separate solutions and were put on an Atomizer Aerosol Generator (3079, TSI Inc., USA). The particles coming out of the atomizer passed through a silica gel diffusion drier at a generation flow rate of ~2.4 L/min. The duration of aerosol injection varied based on the desired number concentration. To generate ~~soot~~smoke, 0.1 – 0.5 g samples of dried biomaterial *Poa pratensis* (Kentucky bluegrass) were weighed out, placed on a quartz boat and into a quartz-tube furnace (Carbolite Gero, TS1-1200, Verder Scientific, UK)

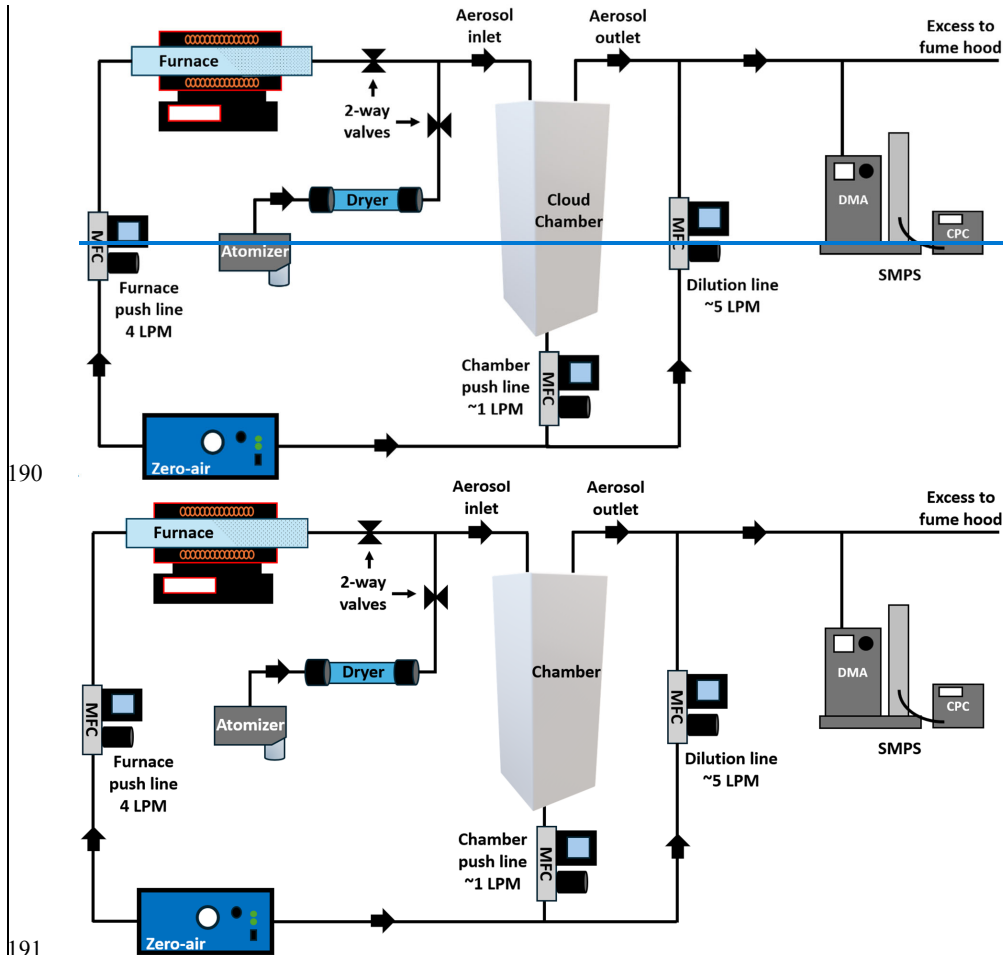
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that was set to 1000°C for a flaming combustion condition. ~~Soot particles were pushed to the chamber by zero air at 4 L/min for 5 minutes, an estimated time for~~ This identical setup was used in Benedict et al. (2024) which showed that at 1000°C burn the black carbon mass fraction averaged 17% for biomass fuels with a single scattering albedo of 0.35 (at 523 nm). We expect a similar smoke profile for the experiments presented here thus the smoke injected is a combination of soot, inorganic, and organic mass along with volatile vapors. Smoke particles were pushed to the chamber by zero-air at 4 L/min for 5 minutes, a time window used to ensure complete combustion of the sample.

Aerosol size and number distributions downstream of the ~~cloud~~ chamber were measured with a scanning mobility particle sizer (SMPS) that consists of a Differential Mobility Analyzer (3081 DMA, TSI Inc., USA) and a Condensation Particle Counter (3752, TSI Inc., USA). Measurement settings were set to continuously scan for 3 minutes/scan; 160 seconds recording with 20 seconds of purging, measuring sizes 15.7 – 764.5 nm. Our experimental matrix consisted of 5 repeats of NaCl, 4 repeats of sucrose and 6 ~~soot~~smoke experiments with varying biomaterial mass, they are outlined in Supplement Information Table 1. In all experiments the first 6 hours of data were used to analyze results.



**Figure 2. Schematic of experimental setup showing how aerosols are injected and sampled from the chamber.**

### 3 Theory on Chamber Processes

The processing of data from the LANL chamber experiments involved two key steps to analyze the underlying aerosol processes of coagulation, wall loss, and dilution (chamber push line).

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First, we determined the observed size-dependent particle rates:  $dN(D_p)/dt$ . The measured size distributions were fitted to a two-mode lognormal distribution. The lognormal distribution parameters were optimized using the Python library SciPy's optimization routines, with the mean squared error as the cost function. We used multiple minimization methods and selected the best fit for each timestep based on the highest Pearson R-squared value-with a minimum threshold of 0.85. The methods included Nelder-Mead (Simplex algorithm), Powell's method (Powell's conjugate direction method), L-BFGS-B (Limited-memory Broyden-Fletcher-Goldfarb-Shanno with Box constraints), TNC (Truncated Newton Conjugate-Gradient method), SLSQP (Sequential Least Squares Programming), and trust-constr (Trust Region Constrained method). We took this approach since the best fit varied with concentration and shape of the distribution. L-BFGS-B was typically the best for a lognormal distribution, but as the mode became broader (lower concentrations) then TNC, SLSQP or trust-constr would have a higher Pearson R-squared value. The transition of when this would occur was not an obvious concentration threshold. Therefore, we used all optimization routines for each lognormal distribution and selected the best fit based on the highest Pearson R-squared value.

~~Second, we fitted these observed rates to theoretical rates calculated from Particula<sup>22</sup>, a python-based aerosol microphysics package. The first step was to generate a new time series at a higher size resolution (log-spaced 250 bins), extrapolating to lower (20 nm) and upper (4  $\mu$ m) diameter limits. The size-dependent particle rate was then computed as the linear slope of 21 point moving window (10 before and 10 after). This final rate was subsequently used to fit the underlying aerosol processes in Equation 1 where  $N(D_p)$  represents the number concentration of particles of diameter,  $D_p$ ,  $K_{12}$  is the coagulation kernel,  $W_c^{-1}$  is the coagulation correction factor,~~

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$N_1$  and  $N_2$  are the concentrations of particles in the bins for  $K_{12}$ ,  $k_{flow}$  is the dilution rate, and  $\beta$  is the wall-loss rate.

Second, we fitted these observed rates to theoretical rates calculated from Particula (Particula, 2025), a python-based aerosol microphysics package. The first step was to generate a new time series at a higher size resolution (log-spaced 250 bins), starting at 20 nm and extrapolating the 746 nm SMPS upper limit to 4  $\mu\text{m}$ . The size-dependent particle rate was then computed as the linear slope of 21 point moving window (10 before and 10 after). The time window (60 min) was chosen through iteration, as shorter than 20 min had too much noise to have self-consistent results and longer than 90 min had increasing fit residuals. Our 60 min window results in a smoothed time evolution, which Mahfouz and Donahue (2020a) showed to be effective in coagulation analysis. Our moving window approach is different from smog chamber wall-loss experiments where the full 5 hours of the wall-loss experiment would be used to fit an apparent size-dependent, time-invariant wall-loss correction (Wang et al., 2018).

The resulting size-dependent rate was subsequently used to fit the underlying aerosol processes in Equation 1 where  $N(D_p)$  represents the number concentration of particles of diameter,  $D_p$ ,  $K_{12}$  is the coagulation kernel,  $W_c^{-1}$  is the coagulation correction factor,  $N_1$  and  $N_2$  are the concentrations of particles in the bins for  $K_{12}$ ,  $k_{flow}$  is the chamber flow coefficient, and  $\beta$  is the wall-loss rate.

$$\frac{dN(D_p)}{dt} = W_c^{-1} K_{12} N_1 N_2 - k_{flow} N(D_p) - \beta N(D_p) \quad \text{Equation 1}$$

The coagulation term is governed by a Brownian Coagulation kernel,  $K_{12}$ , that captures the collision frequency between bin number concentrations ( $N_1$  and  $N_2$ ). This kernel is described in Seinfeld and Pandis<sup>28</sup> (Section 13; Fuchs form with alpha efficiency form of 13.56); (Seinfeld

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and Pandis, 2016; Section 13 - Fuchs form with alpha efficiency 13.56), and calculated with Particula. Since  $K_{12}$  does not account for other interactions (e.g. Coulomb interactions) that may lead to coagulation,  $W_c^{-1}$ , the coagulation correction factor, was determined. In our analysis,  $W_c^{-1}$  is a free fit parameter to allow for un-modeled behaviors to be represented. The ~~dilution rate~~chamber flow coefficient,  $k_{flow} = Q/V$ , characterizes how the clean air flow rate ( $Q$ ) is used to push sample flow out of the chamber volume ( $V$ ). Finally, the wall-loss term,  $\beta N(D_p)$ , accounts for the size-dependent removal of particles to the chamber walls.

$$\beta = \frac{1}{LWH} \left( \frac{4H(L+W)\sqrt{k_e D}}{\pi} + v_p LW \times \coth \left( \frac{\pi v_p}{4\sqrt{k_e D}} \right) \right) \quad \text{Equation 2}$$

~~Equation 2 shows the wall loss rate ( $\beta$ ) varies with particle size, derived from a rectangular-chamber formulation adapted from Crump and Seinfeld<sup>25</sup> and Crump<sup>26</sup>. It incorporates both diffusion driven transport and gravitational settling. In this formulation,  $L$ ,  $W$ , and  $H$  denote the chamber's length, width, and height, respectively;  $k_e$  is the eddy wall diffusivity (a free fit parameter);  $D$  is the particle diffusion coefficient; and  $v_p$  is the particle gravitational settling velocity.~~

~~Equation 2 shows the wall-loss rate ( $\beta$ ) varies with particle size, derived from a rectangular-chamber formulation adapted from Crump and Seinfeld (1981) and Crump (1982). It incorporates both diffusion-driven transport and gravitational settling. In this formulation,  $L$ ,  $W$ , and  $H$  denote the chamber's length, width, and height, respectively;  $k_e$  is the eddy wall diffusivity (a free fit parameter);  $D$  is the particle diffusion coefficient; and  $v_p$  is the particle gravitational settling velocity. This physics-based wall-loss coefficient is different from Wang et~~

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al.'s (2018) method of apparent size-dependent wall-loss fit. In the apparent size-dependent wall-loss fit the rate equation is a two-term first-order rate equation, where there are no physical terms for the size of the chamber or particle settling velocity, in contrast to what we use in Equation 2. The apparent size-dependent wall-loss approach is common for smog chamber experiments (Doussin et al., 2023; Keywood et al., 2004; Loza et al., 2012; Nah et al., 2017; Ng et al., 2007) but would not work here since one of our goals is to specifically determine coagulation. In our case, we need a physics-based wall-loss rate equation, so we can determine if there are any coagulation corrections that could be applied. If we had used the apparent size-dependent wall-loss fit, then there would be little to no residuals for a coagulation correction analysis.

### **3.1 Volume Conservation Analysis**

If the corrected aerosol volumes remain consistent, within the noise measurement, we can infer that the processes described in Equation 1 accurately represent chamber behavior. Measured volume concentrations in our experiments were corrected by accounting for volume losses due to both wall loss and chamber flow. We calculated the cumulative lost volume and added it back to the measured values at each time point. In smog chamber experiments involving secondary organic aerosol formation, this volume conservation analysis provides a constraint on organic aerosol yields. Supplemental Figure S8 shows an example of our volume conservation plot from a smoke injection experiment. From this analysis, we conclude that volume is conserved and that no measurable condensation of biomass burning organic vapors occurs under our experimental conditions.

## **4 Results and Discussion**

### ***4.1 Example Analysis***

We show, in Figure 3, the L-BFGS-B optimization routine that was used on Equation 1 for experimental data from the smoke aerosol generated by combusting Kentucky bluegrass. Figure 3a shows the lognormal-fitted size distribution for the entire experiment, where particle growth is evident as the mode diameter shifts to larger sizes over the six-hour period. Figure 3b breaks down the observed rates after 1.5 hours into three calculated, time-varying, size-dependent components: coagulation, dilution, and wall-loss. At that time, coagulation dominates, reducing particles around 100 nm ( $\sim 0.16 \text{ cm}^{-3} \text{ s}^{-1}$ ) and forming larger particles around 200 nm. From these fits we are specifically interested in the kernel correction factor to better understand the importance of agglomeration of freshly emitted BC fractal-like particles and how it changes in time.

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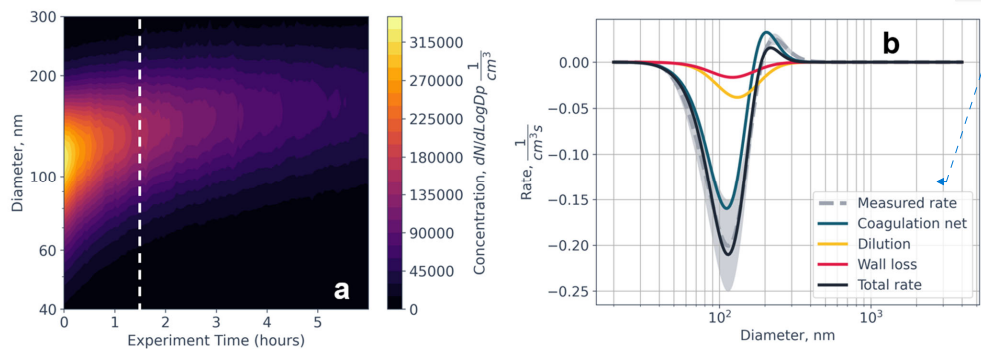
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**Figure 3. a) Time series of the lognormal-fitted size distribution and concentration for a smoke experiment. The dashed line marks a time slice at approximately 1.5 hours. b) At this time slice, particle loss rates are calculated, revealing both loss and gain of particles due to coagulation. (The time series of rates for individual aerosol species are provided in the Supplemental Figures S2-S4) In this panel, the dashed gray line represents the measured rate with uncertainty (shaded gray), while the blue, yellow, and pink lines correspond to the coagulation process, dilution, and wall loss, respectively.**

#### 4.2 Wall-loss Comparison

In Figure 4a we show the average wall-loss rates for our three different aerosol types based on 4–8 experiments each. Only results with valid optimizations and an R-squared above 0.85 were included. In the sucrose experiments, this filter led to data gaps during the later time periods (2–6 hours) for inclusion in the analysis. ~~To better compare with soot, we conducted additional NaCl experiments to have a more complete timeseries for one of the comparisons. The wall-loss rates during the first hour (< 1 hour) follow a similar trend apart from NaCl starting at a low wall-loss rate then rising close to a rate of  $2\text{ s}^{-1}$ . These initial wall-loss rates are consistent with the general observation in chamber studies that early mixing processes and injection conditions can dominate particle loss. Typical ranges reported in smog-chamber experiments span from  $< 1\text{ s}^{-1}$  to tens of  $\text{s}^{-1}$  depending on injection flow and the use of a fan<sup>29</sup>, particle species<sup>40,41</sup>, and chamber geometry<sup>40</sup>. Over longer times (>1 hours), all three aerosol types converge toward similar wall-loss rates, in agreement with the literature indicating that chamber turbulence and gravitational settling diminish~~ To better compare with smoke, we conducted additional NaCl experiments to have a more complete time series for one of the comparisons. The wall-loss rates during the first hour (< 1 hour) follow a similar trend apart from NaCl starting at a low wall-loss rate then rising close to a rate of  $2\text{ s}^{-1}$ . These initial wall-loss rates are consistent with the general observation in chamber studies that early mixing processes and injection conditions can dominate particle loss. Typical ranges reported in smog chamber experiments span from  $< 1\text{ s}^{-1}$  to tens of  $\text{s}^{-1}$  depending on injection flow and the use of a fan (Zong et al., 2023), particle species (Li et al., 2017; Wang et al., 2011), and chamber geometry (Wang et al., 2011). Over longer times (>1 hours), all three aerosol types converge toward similar wall-loss rates ( $0.1\text{ s}^{-1}$ ), in agreement with the literature indicating that chamber turbulence diminishes over time as mixing subsides.



Figure 4b shows the statistical distribution of the wall loss rates for each aerosol type during the first hour and the subsequent five hours. NaCl and sucrose do not exhibit a large variance in diffusivity for the first hour compared to soot which is  $1.12 \pm 1.55 \text{ s}^{-1}$ . NaCl, sucrose, and soot show mean wall loss rates of  $0.562 \pm 0.975 \text{ s}^{-1}$ ,  $0.233 \pm 0.286 \text{ s}^{-1}$ , and  $0.201 \pm 0.267 \text{ s}^{-1}$ , respectively. This convergence to relatively similar values is consistent with past observations in smog-chamber experiments, where turbulent mixing dissipates, and the system approaches a quasi-steady loss rate such as the CMU Teflon chamber<sup>23,34</sup>, the CESAM chamber<sup>40</sup>, and the ATR chamber<sup>20</sup>. However, NaCl and soot sucrose experiments display greater variability than soot, likely due to residual chamber turbulence and differences in particle surface charge stemming from their distinct generation methods (aerosolization vs. combustion).

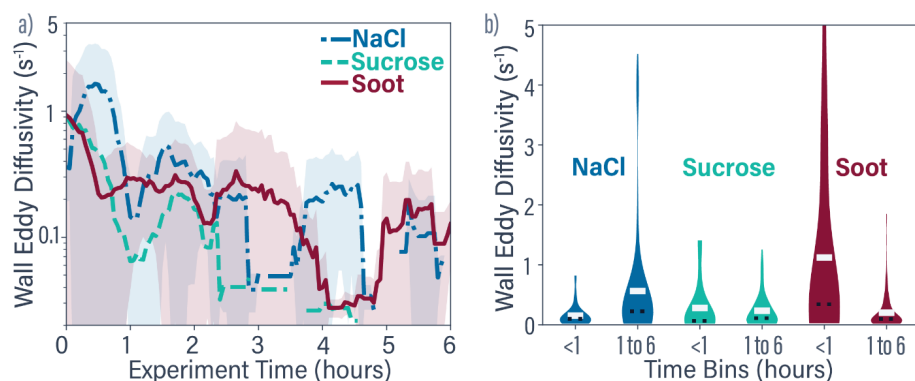
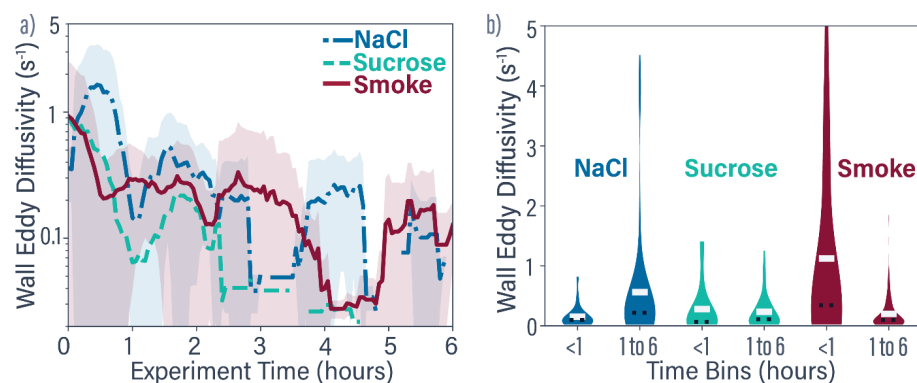


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**Figure 4. a) Average time series of the calculated wall eddy diffusivity for NaCl (blue), sucrose (green), and smoke (red). Only fits with valid optimizations and r-squared greater than 0.85 are included. b) Violin plots showing the mean (.), median (white bar) and overall distribution range of wall eddy diffusivity values for each aerosol type in two-time bins (<1 hour and 1–6 hours). The width of each colored region represents the relative density of data points at that value.**

### 4.3 Coagulation Corrections

To investigate the influence of interparticle forces on aerosol coagulation, we fitted a coagulation correction factor that would account for van der Waals forces, shape, and/or Coulomb interactions in the coagulation rate. When  $W_c^{-1} = 1$ , collisions are effectively “elastic,”

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with no net enhancement or inhibition. In contrast,  $W_C^{-1} > 1$  indicates that coagulation is enhanced (e.g. due to attractive forces, favorable particle morphology, or turbulence), whereas  $W_C^{-1} < 1$  implies reduced coagulation (e.g. electrostatic repulsion or other inhibiting effects).

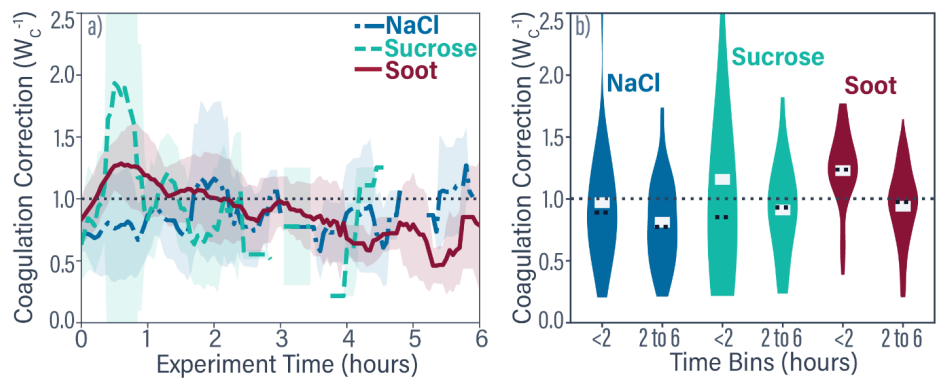
In figure 5a., the soot experiments show an initial period where  $W_C^{-1} > 1$ , which may be explained by the fractal nature of soot aggregates that can promote sticking or chain formation upon collision. By the third hour in all experiments, accounting for the variation the average coagulation corrections extend above and below 1. During this later phase, particle concentrations ( $< 3 \times 10^3 \text{ cm}^{-3}$ ) no longer sustain significant coagulation losses, consistent with prior studies showing that coagulation becomes negligible under low concentration conditions<sup>34,42,43</sup>.

In figure 5a., the smoke experiments show an initial period where  $W_C^{-1} > 1$ , which may be explained by the fractal nature of soot aggregates that can promote sticking or chain formation upon collision. By the third hour in all experiments, accounting for the variation the average coagulation corrections extend above and below 1. During this later phase, particle concentrations ( $< 10^4 \text{ cm}^{-3}$ ) no longer sustain significant coagulation losses, consistent with prior studies showing that coagulation becomes negligible under lower concentration conditions (Hussein et al., 2009; Mahfouz and Donahue, 2020b; Yu et al., 2022).

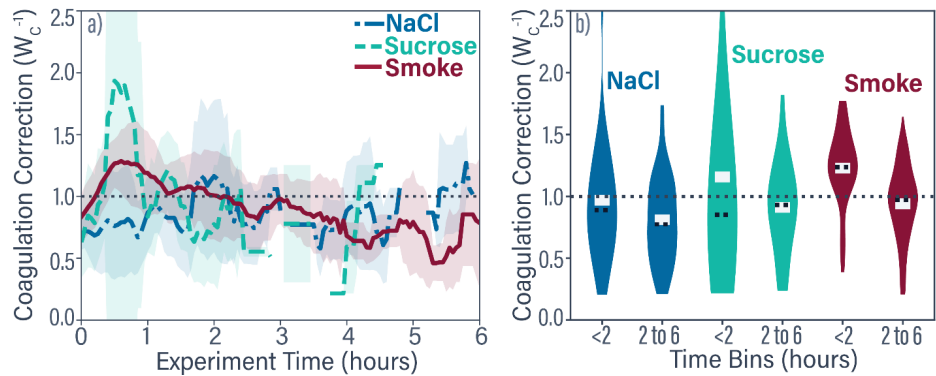
Figure 5b shows the distribution of coagulation corrections for these time periods. All three aerosols show a mean  $W_C^{-1}$  value around 1 ( $0.969 \pm 0.524$  for NaCl,  $1.16 \pm 1.38$  for sucrose, and  $1.23 \pm 0.312$  for ~~soot~~smoke), suggesting a slight repulsion or negligible net sticking among particles. However, the standard deviations do encompass  $W_C^{-1} = 1$ . ~~Soot~~Smoke exhibits a slightly higher coagulation corrections initially followed by reduced values ( $0.941 \pm 0.307$ ) in later periods. These observations align with the notion that both particle morphology (e.g., fractal

386 soot structures) and injection-induced turbulence can transiently enhance coagulation, but the  
387 effect diminishes as particles coagulate.

388



389



390 **Figure 5. a) Average time series of the calculated coagulation correction for NaCl (blue),**  
391 **sucrose (green), and ~~soot~~smoke (red). Only fits with valid optimizations and r-squared**  
392 **greater than 0.85 are included. b) Violin plots showing the mean (.), median (white bar)**  
393 **and range of the calculated coagulation correction for each aerosol type averaged across**  
394 **replicate experiments for the indicated time bins (similar to Figure 4b).**  
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#### 4.4 Monte Carlo Error Analysis

To understand the large standard deviations that emerged from our fits of wall loss and coagulation correction, we performed a Monte Carlo error analysis. We began by constructing three number-size distributions, each formed by the sum of two log-normal modes with equal particle numbers. In the first case both modes were centered at 100 nm but differed in geometric standard deviation, i.e., the distribution is summation of a 100 nm mode with a GSD of 1.4 plus a 100 nm mode with a GSD of 1.8. The resulting distribution reflects the broad distributions we observe in our measurements. The second case repeated this structure at 200 nm. The third case was a hypothetical experiment that combined narrow 100 nm and 300 nm modes (both GSD = 1.2) to test the response to a bimodal aerosol distribution.

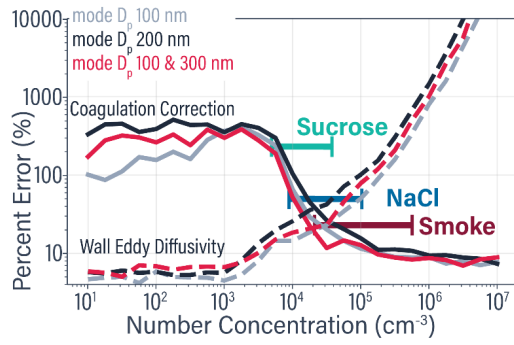
For every distribution we calculated Equation 1 assuming a wall-eddy diffusivity of  $0.1 \text{ s}^{-1}$  and a coagulation correction factor ( $W_C^{-1}$ ) of 1.0. This is a null case in which no additional correction to the Brownian coagulation kernel is required. We then superimposed random noise of  $\pm 20\%$  on both the size spectrum and the rate. This noise mirrors uncertainties reported in instrument intercomparisons of  $\pm 10\%$  error between 20 nm and 200 nm and up to  $\pm 30\%$  above 200 nm (Wiedensohler et al., 2012). Thus,  $\pm 20\%$  is a middle point across the range we measured. Applying the same noise to the rate represents the best-case scenario for our analysis pipeline.

With these noisy data sets created, we refit the wall-eddy diffusivity and coagulation correction 80 times at each total number concentration shown in Figure 6. From the ensemble of fits we calculated the percent error in each retrieved parameter and averaged the results (Figure 6). The results in Figure 6 reveal a clear trend percent error. When total number concentration exceeds roughly  $10^4 \text{ cm}^{-3}$ , the uncertainty in the coagulation correction begins to fall. This is

419 consistent with the fact that Brownian coagulation scales with particle number squared and can  
420 be distinguished from measurement noise only at higher concentrations. Conversely, the error in  
421 the wall-eddy diffusivity grows with concentration. Once coagulation dominates the particle loss  
422 budget, the data contains too little information to constrain the comparatively low wall loss sink,  
423 increasing the relative uncertainty. In other words, when coagulation governs the system  
424 dynamics, the wall-loss term becomes a minor, poorly resolved correction.

425 The three analyzed distributions exhibit similar percent errors in the coagulation  
426 correction. The slightly lower error for the 100 nm mode compared to the 200 nm mode is  
427 consistent with the behavior of the Brownian coagulation kernel, where smaller particles have  
428 higher coagulation coefficients and therefore undergo more frequent collisions. This leads to a  
429 greater rate of change in the distribution for a given number concentration, resulting in better  
430 signal-to-noise. The hypothetical bimodal distribution generally shows the lowest uncertainty  
431 among the three cases (in our experimental range), although the improvement is modest.

432 Annotations in Figure 6 mark the concentration ranges for the three chamber campaigns,  
433 sucrose, NaCl, and smoke aerosols. They also indicate the measured coefficient of variation in  
434 the mean coagulation correction for each case. The agreement between these annotated  
435 uncertainties and the Monte Carlo error analysis confirms that the observed variability is  
436 consistent with the measurement noise.



**Figure 6: Percent error in fitting of coagulation correction (solid line) and wall eddy diffusivity (dashed line) as a function of number concentration. Lines represent mean errors for size distributions with different modal diameters: 100 nm (gray), 200 nm (black), and a bimodal 100 & 300 nm distribution (red). Annotated markers indicate representative number concentration ranges for Sucrose, NaCl, and Smoke experiments, along with the coefficient of variation (standard deviation divided by the mean) in the coagulation correction, reflecting relative uncertainty rather than bias.**

## 5 Discussion

Our initial experiments in this new ~~cloud~~ chamber focused on dry conditions and a set of aerosols to quantify how particles evolve in the absence of humidity (<10% relative humidity). Despite the relatively simple setup—no temperature or humidity control—two key insights will be used in future humidified experiments. First, the wall-loss rates converged to similar values across all aerosol types after the first hour, indicating that early differences largely arose from injection flow conditions and subsequent turbulence. Over time, these chamber conditions stabilized, reinforcing the well-documented notion that particle wall losses approach a quasi-steady state as mixing subsides.

A second important finding is that coagulation within the chamber is most pronounced during the initial phase of each experiment. Though this is more uncertain due to larger relative errors, ~~Soot~~[Smoke](#) showed signs of coagulation enhancement, potentially attributable to its

fractal structure. Once total number concentrations fell below a few thousand particles per cubic centimeter, coagulation slowed considerably, consistent with the literature. Collectively, these observations highlight the dynamic interplay between wall loss, particle morphology, and injection protocols in shaping the early stages of aerosol evolution in chamber studies.

Our results also shed light on the influence of particle composition and shape. While aerosols like NaCl and sucrose exhibited expected behavior—~~initial~~[average](#) collision enhancements near unity—~~soot~~[smoke](#) displayed additional complexity. Early-time coagulation factors for ~~soot~~[smoke](#) were moderately elevated, suggesting that [soot](#)-fractal aggregates [within smoke](#) can ~~promote sticking or have an~~ increased collisional radius. Over longer times, the coagulation rates for all three aerosols converged to near unity or below, indicating negligible net enhancement under steady-state conditions. These observations set the stage for more detailed investigations of fractal-like particles under high humidity environments (>90% relative humidity).

Although these initial experiments focused on low humidities, the chamber design allows for temperature and humidity control to be integrated in future work. [The Monte Carlo error analysis points to using number concentrations above  \$10^6 \text{ cm}^{-3}\$  for reducing the percent error in future coagulation correction experiments.](#) Extending to more complex atmospherically relevant aerosol mixtures—such as ~~soot~~[smoke](#) mixed with organic vapors or inorganic salts—will further elucidate aerosol ~~aging pathways and cloud~~[coagulation](#) interactions. Additionally, the use of more advanced aerosol instrumentation will improve the characterization of particle morphologies and mixing states that evolve during cloud processing.



## 6 Conclusion

The custom-built 906 L stainless-steel chamber provided reproducible measurements of particle size distributions under dry conditions, confirming its suitability for controlled aerosol research. Although initial turbulence drove high wall-loss rates, these converged to stable values across NaCl, sucrose, and ~~soot—undersearings~~smoke. This underscores that injection protocols and mixing strongly influence early aerosol behavior. The chamber's intermediate size and flexible design for future temperature and humidity controls make it a useful platform to investigate aerosol-cloud interactions more comprehensively. Integrating additional measurements of particle shape, chemical composition, and mixing state will further clarify the complexities of aerosol aging and cloud formation. Building on these dry experiments, upcoming work at higher humidity will reveal how aerosol coagulation and phase changes affect cloud processes such as droplet activation and scavenging. By disentangling coagulation, dilution, and wall-loss mechanisms, this chamber ultimately enables rigorous study of aerosol transformations—, particularly for ~~fractal soot—~~smoke, in cloud-relevant environments, helping advance both scientific understanding and climate prediction.

*Author contributions.* NAF performed the experiments, contributed to the analysis of the results, and wrote the manuscript. KJG was responsible for the design of the chamber and the study, development of the data analysis methods, and contributed to the writing of the manuscript. KBB contributed to the design of the study, analysis of the results, and contributed to the writing of the manuscript.

*Competing interests.* The authors declare no competing interests.

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503 *Code/Data availability.* The python data analysis code is available from corresponding authors.

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505 *Acknowledgments.* The authors would like to thank Hannah Brink, Ryan Farley, James E. Lee,

506 and Matt Nelson for their feedback on the manuscript. Release Number LA-UR-25-22994.

507

508 *Financial support.* Research presented in this article was supported by the Laboratory Directed

509 Research and Development program of Los Alamos National Laboratory under project number

510 20230248ER. This work was supported by the U.S. Department of Energy through the Los

511 Alamos National Laboratory. Los Alamos National Laboratory is operated by Triad National

512 Security, LLC, for the National Nuclear Security Administration of U.S. Department of Energy

513 (Contract No. 89233218CNA000001).

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