

Influence of Organic Films on the Hygroscopicity of Ultrafine Sulfuric Acid Aerosol

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In ambient air, organic chemicals are common constituents of aerosol particles, and many of them are surface-active. If these compounds are present as surface films on aqueous droplets, mass transfer of gaseous molecules into or out of the droplets could be impeded, and the hygroscopic growth of acidic droplets could be reduced. In this work, the effects of organic films on the hygroscopic growth rate of ultrafine H₂SO₄ aerosols (40–120 nm) were investigated under relative humidity conditions of 20–85% and over time scales of 6 and 10 s. Lauric (C₁₂, saturated), stearic (C₁₈, saturated), and oleic (C₁₈, unsaturated) acids were chosen as the film coating materials. The experimental results showed that the presence of monolayer films of some fatty acids retards the hygroscopic growth rate of H₂SO₄ aerosol due to the obstructive effect of the films on the permeation of water vapor. The retardation effects depend on the original film coating thickness. Molecular structure of the film-forming compounds is an important factor for determining the effectiveness of the film. Linear molecules, such as lauric and stearic acids, have stronger effects on mass transport of water vapor than those of nonlinear molecules, such as oleic acid.

Introduction

Knowledge of the hygroscopicity of atmospheric aerosols containing H₂SO₄ is essential to a better understanding of the atmospheric chemistry and human pulmonary responses to such aerosols. The hygroscopicity of sulfuric acid aerosols affects droplet size distribution and the pH of the droplets. Consequently, it may affect the particles' atmospheric lifetimes, their chemical reactivity, and their physical properties (such as the light scattering and absorption properties) (1). Theoretical calculation show that, under the conditions prevalent in human airways (elevated temperature and relative humidity), sulfuric acid aerosols tend to grow. Such growth affects the particle deposition efficiency and site as well as the droplet acidity and neutralization by endogenous ammonia (2, 3).

In ambient air, organic chemicals are common constituents of aerosol particles. A great variety of organic compounds have been detected in atmospheric aerosols. Many

of these are surface-active organic compounds and their precursors, such as long-chain aliphatic hydrocarbons (C ≥ 10) and their acid, amine, alcohol, aldehyde, ketone, and ester derivatives (4–7). These compounds have natural as well as man-made origins. Over 100 compounds were identified in the natural fraction, including C₉–C₂₂ straight-chain acids. The presence of these compounds in sufficient quantity on aerosol particles implies the potential for the presence of surface films (8). The organic films presumably coalesce and thicken as the particles reside in the atmosphere (4, 9) and could impede mass transfer between gaseous molecules and the particles.

The effectiveness of organic films as barriers to mass transfer is determined by the properties of the film-forming compounds, such as the length of the hydrocarbon chain, the geometry of the molecule, and the type and position of the functional group. Blank observed that the absorption of CO₂, O₂, H₂O, N₂O, and SO₂ by aqueous solutions can be considerably slowed by films of C₁₆ and C₁₈ alcohols or C₁₈ acid and C₁₇ amines (10, 11). Significant retardation effects of monolayer organic films on the evaporation/growth rate of large size acid droplets (diameter in several micrometers to 100 μm) were reported by many investigators (12–15) and summarized by Barnes (16). However, there is a lack of data in the submicrometer particle size range, while the particles with radii less than 1 μm are richer in organic matter than those with larger sizes (6).

In this study, the effects of organic films on the hygroscopic growth rate of ultrafine sulfuric acid aerosols were experimentally examined in relation to film thickness and particle diameter, in the diameter range of 40–120 nm and in relative humidities (RH) up to 85%. The high RH simulates the conditions in summertime ambient air and in human airways. The effectiveness of films of C₁₂ and C₁₈ fatty acid monolayers were investigated with respect to the chain length and molecular structure (saturated or unsaturated) of the film-forming compounds. C₁₂ and C₁₈ fatty acids were chosen as the film-forming compounds for investigating the film effectiveness of fatty acid monolayers. These compounds are not only highly surface active but also exist as the dominant fatty acids in the ambient aerosols, based on the analyses of aerosol, surface, and subsurface particulate samples collected from different places in the tropical Atlantic Ocean by Marty and Salot (17).

Experimental Section

A schematic diagram of the experimental system for measuring the hygroscopic growth rate of organic film coated H₂SO₄ droplets is shown in Figure 1. The system consists of a condensation H₂SO₄ aerosol generation system, an organic film coating system, a constant flow clean air (particle and organic free) generator, an air humidify conditioner, a steady-state flow reactor, and a precise particle sizing system—the scanning tandem differential mobility particle spectrometer (STDMPs).

All chemical compounds used in this study were reagent grade and were used without further purification.

Scanning Tandem Differential Mobility Particle Spectrometer (STDMPs). The STDMPs system was built for generating monodisperse H₂SO₄ aerosols, for determining the thickness of organic coatings on particle surfaces, and for measuring the particle hygroscopic size change in relation to the film thickness. The system consists of two differential mobility analyzers (DMA; model 3071, TSI, MN), a condensation particle counter (CPC; Model 3010S, TSI, MN) equipped with a scanning electrical mobility spectrometer

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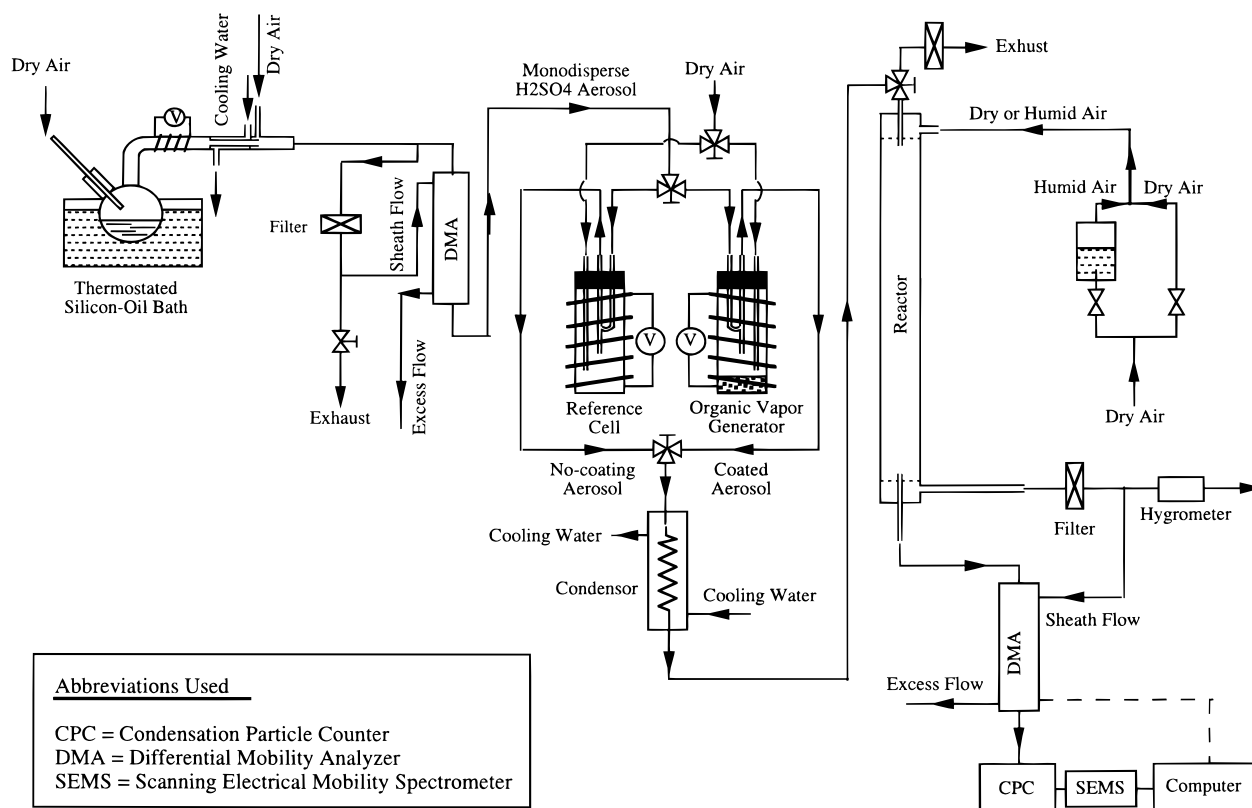


FIGURE 1. Schematic diagram of the experimental system for measuring hygroscopic characteristics of organic coated ultrafine sulfuric acid droplets.

chip and software (SEMS; Model 390089, TSI, MN) (18), and an interfacing personal computer for system automation and data acquisition. Particle diameter changes are determined by precisely measuring the change in the electrical mobility of singly charged particles that results from their growth or shrinkage. Rader and McMurry (19) showed that diameter changes of about 0.3% could be resolved with this instrumentation. It is suitable for studies of particles in the 0.01–0.2 μm diameter range, with a corresponding resolution of 0.03–0.6 nm. This technique was successfully used by Zhang et al. (20) in the study of the evaporation rate of monodisperse DOP particles. To measure the coating thickness, a diffusion battery/CNC system was used by Däumer et al. (21). The size resolution of this technique was reported in a range of 0.5 nm for particles smaller than 0.04 μm , with the sensitivity dropping sharply for particles larger than 0.04 μm (22). Therefore, for determining the coating thickness as well as the hygroscopic size change, the STDMPs has an overwhelming advantage with regard to measurement range, precision and resolution.

The entire system was adjusted and calibrated using pure H_2SO_4 droplets (uncoated) as the test material. The measurement data of hygroscopic growth factors were compared with the theoretical data calculated using a correction for the Kelvin effects (23). For all droplet sizes collectively, the ratio of the experimental to the theoretical values was 1.005 ± 0.003 (mean \pm SD).

Condensation H_2SO_4 Aerosol Generation System. The ultrafine H_2SO_4 aerosol was produced in a condensation aerosol generator based on the design principle of Fuchs and Sutugin (24). Preheated particle-free, ammonia-free, moisture-free air passes over the surface of H_2SO_4 that is retained in a thermostated silicone oil bath. The mixture of air and H_2SO_4 vapor then passes through a thermostated water-cooled condenser to form ultrafine aerosol. The size of H_2SO_4 droplets can be controlled by varying the evaporating

temperature of liquid H_2SO_4 and the carrier gas flow rate. The particles generated in this system are nearly monodispersed. The geometric standard deviations (GSDs) are about 1.1, 1.5, and 1.6 for particles with diameters of 20, 40, and 100 nm, respectively. The concentration range is from 10^4 to $10^6/\text{cm}^3$. To maximize the monodispersity of the aerosols, high-purity H_2SO_4 was used, and the constancy of the evaporating temperature as well as the gas flow rate were maintained during each experiment. The whole system is made of Pyrex glass. All the ground glass joints were sealed using Teflon sleeves instead of lubricant to prevent the system being contaminated by organic impurities.

The generator has a very stable output with regard to particle size, with average relative GSDs of 0.01 and 0.03 over time scales of 1 and 6 h, respectively. The particle number concentration produced can be as high as $10^7/\text{cm}^3$. The generated aerosol can be classified by the DMA under the same RH condition. The extracted particles from the DMA are highly monodispersed, with a GSD within 1.05.

Organic Film Coating System. The organic film coating system used for coating the monodisperse acidic aerosols with the organic vapor was designed and built using the heterogeneous condensation technique (25). Organic vapor is generated by heating the substance in a temperature-controlled vessel. A constant flow of the ultrafine monodisperse sulfuric acid aerosol passes through the top of the vessel and mixes immediately with the organic vapor. The gas and particle mixture then flows through a condenser, where heat exchange between the mixture and coolant (cold water) occurs and organic vapor is rapidly condensed on the droplet surface. The film coating thickness can be controlled by varying either the evaporating temperature or the flow rate of the aerosol.

If the size of particle core (the acid portion only) remains unchanged during the coating process and if the droplets are spherical, the film thickness (γ_0) can be determined by

TABLE 1. List of Highly Surface-Active Compounds To Be Selected as the Film Coating Materials

compound	formula	molecular weight	boiling point ^a (°C)
Aliphatic Acids			
lauric (dodecanoic) acid	<i>n</i> -C ₁₁ H ₂₃ COOH	200.3	131 (1 mmHg)
stearic (octadecanoic) acid	<i>n</i> -C ₁₇ H ₃₅ COOH	284.5	360 (d), 232 (15 mmHg)
Unsaturated Acids			
oleic (<i>cis</i> -9-octadecenoic) acid	C ₁₇ H ₃₃ COOH	282.5	228–9 (15 mmHg) 286 (100 mmHg)

^a Data are from ref 31.

measuring the particle size changes before and after coating. However, the size of sulfuric acid droplets is very sensitive to its surrounding environmental temperature. The particle core shrinks slightly due to temperature elevation inside the organic vapor generator and cannot always fully recover to its original size after being coated. For precise measurement of the film thickness, noncoated droplets were exposed to the same conditions as the coated ones. They were passed through a reference vessel, which was identical to the organic vapor generator but contained no organic chemicals, as shown in Figure 1.

Dry and Humidified Clean Air Generation System. All of the air used in this study was laboratory room air that was pretreated to dry and clean conditions before entering the experimental system. The pretreatment system consists of three columns packed with citric acid, silica gel, and charcoal, respectively, for removing ammonia, moisture, and organic compounds, and a high-efficiency particle filter (HEPA) for removing all particles.

Humidified air was obtained by passing a constant flow of the dry and clean air through a cluster of 10 12-in.-long Nafion tubes (Perma Pure, Inc., Toms River, NJ) immersed in distilled water. Nafion is a highly water hydration material that is able to absorb up to 22% of water by weight. Water continuously passes through the Nafion membrane and evaporates into the air that flows through the inside of the tubing. The humidified air then mixes with dry air in a specific ratio to reach the desired humidity. The temperature and relative humidity are monitored on-line by a thermocouple and a dew point monitor (Hygro M1, General Eastern, Woburn, MA).

Steady-State Flow Reactor. A cylindrical steady-state flow "reactor" was designed for measuring the hygroscopic growth rate of H₂SO₄ aerosols. It consists of a stainless steel tube (7/8 in. i.d. and 117 cm in length), inlets and outlets for the particle flow and sheath flow, and associated connectors. A "dry" monodisperse acid aerosol flow passes through the reactor at the center of the reactor. The sheath flow consists of dry or humidified air with a constant RH. The monodisperse aerosol flow and the sheath flow were set to travel at nearly equal longitudinal velocity in the reactor. The hygroscopic growth rate was determined by applying the STDMPs system to measure the size changes of the particles between the entrance and the exit of the reactor.

A laminar flow (Reynolds number < 2000) with a constant flow rate has to be maintained in the reactor to maintain stable profiles of the aerosol along the reactor length. For a reactor with fixed length and diameter, the aerosol residence time (t_r) in the reactor is controlled by changing the volumetric flow rate (Q). In typical experimental conditions, the Reynolds number in the reactor is below 400, thus safely under the laminar flow critical limit. However, the laminar flow creates a parabolic velocity profile that causes a radial dependency of the residence time of droplets moving through the tube. To reduce the nonuniformity of the residence time, only a small portion of sample was extracted from the core of the flow. For the case of our experimental conditions, where the sampling flow rate (Q_s) at the core is less than

one-tenth of the total flow rate (Q), the calculated longest and shortest transit times of droplets differ less than 3% from the mean. The time needed for flow to become fully developed is about 0.5 s under the experimental conditions. Therefore, the mean residence time of the flow (\bar{t}_r) can be approximately estimated by applying the laminar flow theory, as

$$\bar{t}_r = \frac{\pi R^2 L}{Q_s} \left(1 - \sqrt{1 - \frac{Q_s}{Q}} \right) \quad (1)$$

where R and L are the radius and length of the reactor, respectively.

The accuracy of eq 1 was examined by Rader et al. (26) with a TDMA/CNC system. The measured residence times should be longer than those predicted values, due to the additional times needed for the flow to pass through the DMAs and the connection tubings.

Selection of Film-Forming Organic Compounds. Film-forming organic compounds are all polar–nonpolar in type. The molecules are elongated and steeply oriented to the surface. Any polar sections of the organic molecules (i.e., COOH or OH group) are likely to be preferentially oriented toward the core and to interact strongly with the substrate as a result of hydrogen bonding, while the nonpolar sections (long chains of hydrocarbon) interact with each other and spread out as a monolayer film on the surface (27, 28). Straight-chain hydrocarbon derivatives, such as acids, alcohols, etc., can be made to exhibit the various monolayer states by a suitable choice of chain length and temperature. The films most likely to have a significant influence on atmospheric chemistry are solid or liquid condensed films that result with closely packed, specially oriented molecules.

The C₁₂ and C₁₈ fatty acids, listed in Table 1, were chosen for study in this work since these compounds are not only highly surface-active but also have been detected in the atmospheric aerosols (5, 7, 29, 30). These choices also allow us to examine some of the properties of the film-forming compounds that may affect the effectiveness of organic films, such as length of the hydrocarbon chain and the molecular structure of saturated and unsaturated acids.

Film Thickness. The coating thickness is an important factor that determines the effectiveness of a surface film. The film thickness of the aerosol particles, γ_0 , can be determined by comparing the diameters of the droplets with or without coatings, $(d_0)_c$ and d_0 , measured by the STDMPs:

$$\gamma_0 = \frac{[(d_0)_c - d_0]}{2} \quad (2)$$

Hygroscopic Growth Factor. Hygroscopicity depends on the RH. The droplet hygroscopic growth factor, α , is defined as a ratio of the hygroscopic diameter at a certain RH, d_h , to the droplet diameter at 0% RH, d_0 :

$$\alpha = \frac{d_h}{d_0} \quad (3)$$

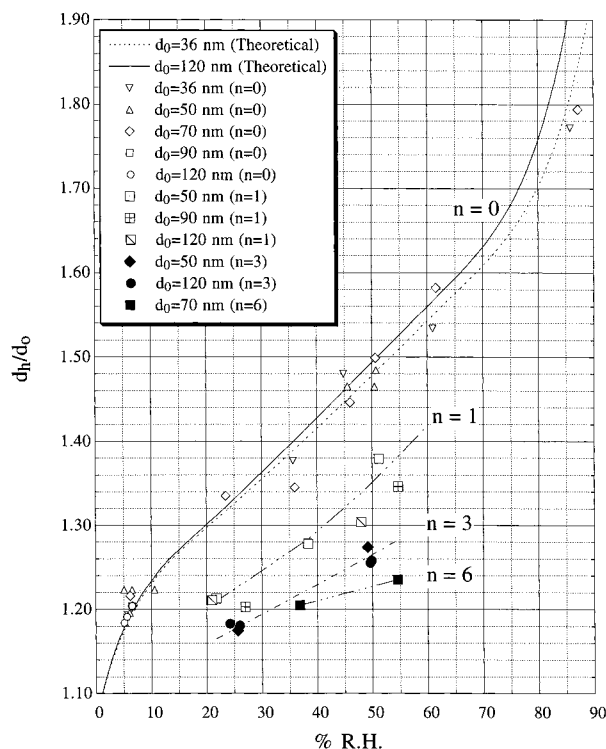


FIGURE 2. Hygroscopic growth factor ($\alpha = d_t/d_0$) of H_2SO_4 droplets with or without lauric acid ($n\text{-C}_{11}\text{H}_{23}\text{COOH}$) coating. [$d_0 = 36\text{--}120$ nm, $T = 25^\circ\text{C}$, $t_r = 6$ s, $n = \text{number of coating layers}$.]

Under our experimental conditions, the lowest RH (dry) condition that can be reached was about 5%. The value of d_0 can be derived from the theoretical value of α (α_{theo}) and the measurement of the droplet diameter at this dry condition (d_d):

$$d_0 = \frac{d_d}{\alpha_{\text{theo}}} \quad (4)$$

Results and Discussion

The hygroscopic growth rates of the organic coated H_2SO_4 aerosols were studied in the 40–120 nm diameter range with respect to the dry size, d_0 , under RH conditions of 20–85% over a time intervals of 6 and 10 s, respectively. Two saturated fatty acids, lauric acid ($n\text{-C}_{11}\text{H}_{23}\text{COOH}$) and stearic acid ($n\text{-C}_{17}\text{H}_{35}\text{COOH}$), and an unsaturated fatty acid, oleic acid ($\text{C}_{17}\text{H}_{33}\text{COOH}$), were chosen as the film coating materials. The coating thickness varied from 2.2 to 30 nm (equivalent to about 1–13 monolayers).

The hygroscopic growth rates of coated H_2SO_4 droplets were compared to those of uncoated H_2SO_4 droplets, as experimentally measured and theoretically calculated. The results, expressed as a function of the particle hygroscopic growth factor versus the growth time and the relative humidity conditions for each selected organic compound and initial film thickness, are shown in Figures 2–5.

The experimental results show that the monolayer films of the saturated fatty acids (lauric acid and stearic acid) retard the hygroscopic growth rate of H_2SO_4 aerosols (Figure 2–4). According to literature data, the thickness of a single monolayer organic film (γ_s) is about 2.4 nm (27). After a monolayer film is formed on the surface of the droplets, the film can thicken by the continuing vapor deposition to form multilayers and polymeric membranes. Multilayers behave more and more like a separate bulk phase through which gases must pass if they are to reach the aqueous surface of the acid droplet. The experimental data clearly show the

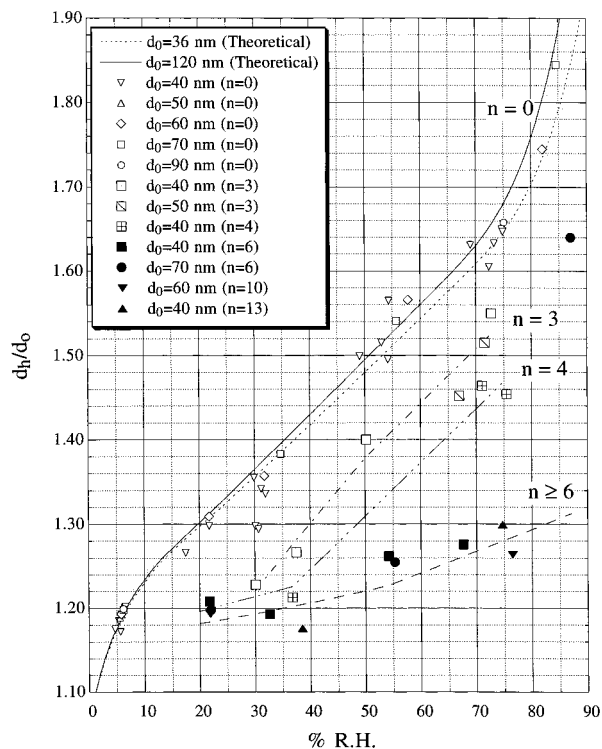


FIGURE 3. Hygroscopic growth factor ($\alpha = d_t/d_0$) of H_2SO_4 droplets with or without lauric acid ($n\text{-C}_{11}\text{H}_{23}\text{COOH}$) coating. [$d_0 = 36\text{--}120$ nm, $T = 25^\circ\text{C}$, $t_r = 10$ s, $n = \text{number of coating layers}$.]

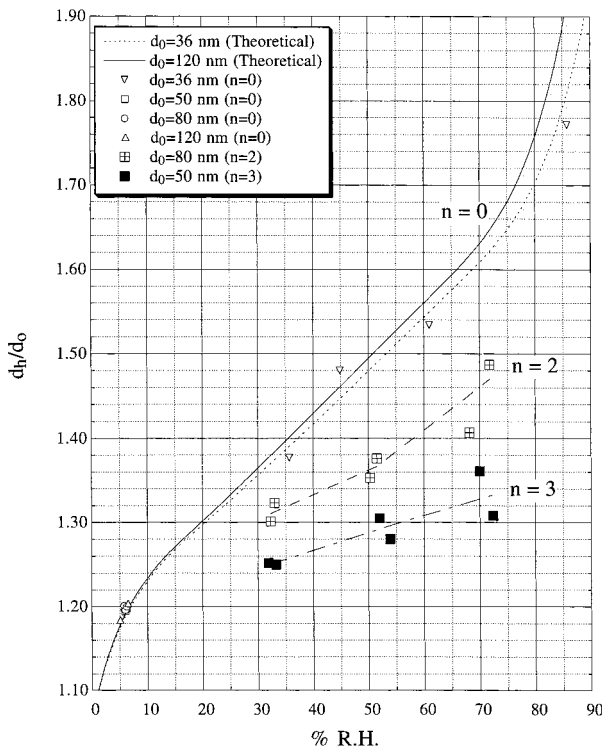


FIGURE 4. Hygroscopic growth factor ($\alpha = d_t/d_0$) of H_2SO_4 droplets with or without stearic acid ($n\text{-C}_{17}\text{H}_{35}\text{COOH}$) coating. [$d_0 = 36\text{--}120$ nm, $T = 25^\circ\text{C}$, $t_r = 6$ s, $n = \text{number of coating layers}$.]

decrease in hygroscopic growth rates as the films thicken. For example, at 50% RH condition and over a time scale of 6 s, the average growth factors of lauric acid coated H_2SO_4 particles with single-, triple- and six-layer film coatings were reduced to 90.2%, 84.8%, and 81.5% of the values of uncoated H_2SO_4 particles, respectively.

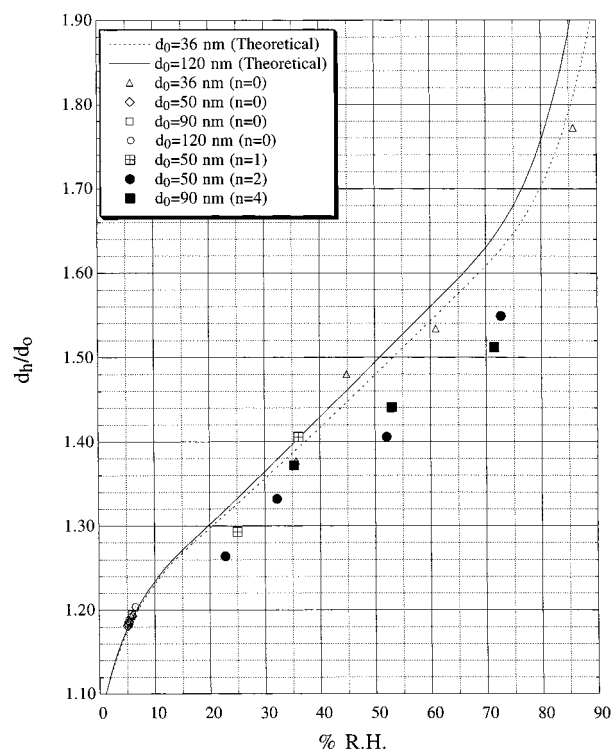


FIGURE 5. Hygroscopic growth factor ($\alpha = d_h/d_0$) of H_2SO_4 droplets with or without oleic acid ($C_{17}H_{33}COOH$) coating. [$d_0 = 36$ – 120 nm, $T = 25^\circ C$, $t_r = 6$ s, n = number of coating layers.]

For lauric acid films, with coating layers less than 6, the slopes of hygroscopic growth factor–RH curves decrease significantly as the number of layers increase and increase as reaction time increases from 6 to 10 s (Figures 2 and 3). These results strongly suggest that the retardation was mainly attributable to the obstructive effects of the film on the permeation of water vapor. However, for films equal to or thicker than 6 layers, the growth factor–RH curve remains the same regardless of the film coating layers (varying from 6 to 13) and the residence time, varying from 6 to 10 s. The obstructive effects of films on water uptake may reach a limit once the number of coating layers reaches a critical value, say $n = 6$ for lauric acid film. This saturation effect may be related to the nonuniformity of the film coating, i.e., the film may not be uniformly distributed over the particle surface. The coating thickness reported in this work is based only on the average value of surface coverage. Some of the surface area may be covered only by a thin layer of film, or even not covered by film, resulting in less retardation effects than the overall growth rate indicates. Increasing the coating layer may achieve a more complete film coating. At this time, due to the limited data now available, a full analysis with respect to the kinetics and thermodynamics is premature. However, there can be little doubt that certain fatty acid films influence the hygroscopicity of ultrafine acid aerosol.

The molecular structure of the film-forming compounds is an important factor for determining the effectiveness of the film. Oleic acid, which has same number of carbon atoms with stearic acid but a double bond, forms fluid surface films. While linear molecules form tightly packed surface films around the droplets, which strongly impede the mass transfer rates of gaseous molecules across the air–droplets interface, the data show that there were no observable retardation effects for a single monolayer oleic acid coating as well as much smaller effects for multilayers than those produced by the lauric acid (C_{12}) and stearic acid (C_{18}) films (Figure 5).

The hygroscopicity of each organic film coating material was tested by using its particle form (without H_2SO_4 acid)

TABLE 2. Experimental Results of Hygroscopicity of Film Coating Materials in Aerosol Phase

organic compound	RH (%)	d_p (nm) (mean \pm SD)
lauric acid	5.6	35.41 \pm 0.58
	62.9	35.74 \pm 0.17
	5.8	31.77 \pm 0.11
	64.2	31.43 \pm 0.18
stearic acid	6.3	57.25 \pm 0.17
	52.7	57.25 \pm 0.26
	68.6	57.28 \pm 0.26
	4.4	58.90 \pm 0.16
oleic acid	33.0	59.55 \pm 0.01
	46.2	59.54 \pm 0.03

under the same experimental conditions for testing the acid droplets. No observable hygroscopic growth was found for lauric acid and stearic acid, and slight size increases were observed for oleic acid as shown in Table 2. We presume that the organic films behave similarly to pure particles of the same composition and that the film thickness will not grow as RH increases. However, the film may become thinner or be broken due to acid particle hygroscopic growth with increasing RH. Therefore, the film retardation effects on the particle hygroscopic growth could be reduced above a certain RH level. Such circumstances did not appear under our experimental conditions (below 85% RH). However, the measurement may not be accurate at higher RH conditions due to significant change of the film thickness.

The retardation effects of lauric acid film on hygroscopic growth of sulfuric acid droplets were significantly increased as the growth period decreased from 10 to 6 s. By analogy, the influence of organic film on the hygroscopicity of acid aerosols should be more significant in the human airway system where the residence time of inhaled particles is much shorter. Thus, the organic film may be responsible for impeding the hygroscopic growth of the acid particles in human airway system. Consequently, it may affect the human health impact as (a) the coated acid particles have smaller sizes that may penetrate deeper than uncoated particles in humid respiratory airways and (b) the coated acid particles have higher H^+ concentration or acidity that may produce greater pulmonary responses than those produced by the acid particles without such coatings.

Acknowledgments

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