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Measurements of the Size Distribution of Aerosols Produced by Ultrasonic Humidification

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Aerosol number and mass distributions produced by an ultrasonic room humidifier and an ultrasonic medical nebulizer were examined in a limited-scale study. Rapid droplet drying occurred at room humidities and under near saturated conditions. A model was tested describing the diameter of dried particles as a function of the dissolved mineral content of the water and the transducer frequency. Water containing 102 mg/L of dissolved minerals in a humidifier with a 1.6 MHz transducer produced droplets with a mass median diameter of 2.9 μm. The number median diameter of particles after drying was computed to be 0.11 μm. The distribution of particles in a nebulizer tube using a NaCl solution was shown to consist of a mixture of dried salt particles and droplets which included coagulated multiplets.

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BACKGROUND

Ultrasonic humidification has found widespread acceptance for increasing the humidity of air. Residential users operate them to increase room humidity for comfort and to alleviate respiratory problems. The ultrasonic technique generates relatively large volumes of water aerosol from a small, easy-to-operate device. Humidifiers using the same operating principle are also found routinely in medical applications for respiratory therapy. Their utility ranges from providing simple water aerosol humidification for selected areas of the respiratory tract to the generation of medicated solution aerosols for therapy. Humidifiers are usually referred to as nebulizers in the medical literature.

The production of aerosol droplets from an ultrasonic humidifier chamber as described by Spearman et al. (1982) is illustrated in Figure 1. The aerosol is formed at the surface of a water column emanating from a submerged high-frequency transducer. The water surface is shattered by the input energy into uniform droplets, which are carried to the outlet (room humidifier) or transfer tube (nebulizer) by the air stream from a small fan. Medical nebulizers function identically to room humidifiers, except that the solution to be dispersed can be isolated in a cup placed in a water bath.

A recent technical note by Highsmith et al. (1988) addressing indoor residential humidification showed that relatively large quantities of dry aerosols were produced by ultrasonic units when using selected samples of tap water. In one test, single room concentrations of fine aerosols (defined as those < 2.5-μm aerodynamic diameter size range), exceeded 6300 $\mu g/m^3$, when using tap water containing 300 mg/L dissolved minerals—a typical level of impurities. This indoor concentration exceeded both the allowable EPA PM10 ambient air standard (Federal Register, 1987) and the ACGIH occupational level for respirable particulates (ACGIH, 1984). Home ultrasonic humidifiers had previously been reported to produce a "white dust" (Consumer Reports, 1988) in the vicinity of the humidifier, implying a large settleable aerosol size. Highsmith et al. (1988) suggested that most of the particles were smaller than this coarse size, but provided only two

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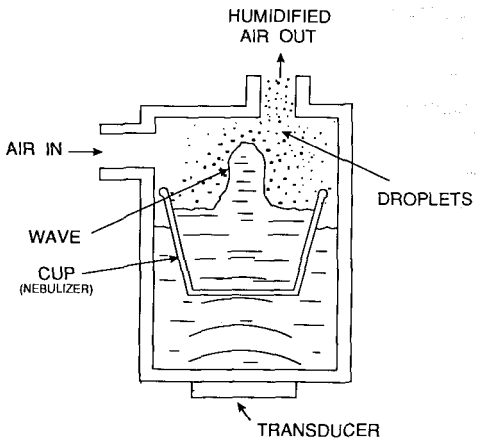

FIGURE 1. Ultrasonic transducer production of aerosols. From Spearman et al. (1982).

integrated categories for the size distribution.

The respiratory therapy literature (e.g., Spearman et al., 1982, Kacmarek et al., 1985) contains numerous descriptions of the application of nebulizers employing ultrasonic transducers. The use of humidifiers in therapy differs significantly from room humidification in that the aerosols are most often directed through a flexible tube to a face mask or tent used by a patient. This arrangement results in aerosol formation and transport processes under nearly saturated humidity conditions. This situation differs significantly from that occurring when the droplets are emitted into a room with a much lower relative humidity level. Since the generation processes are identical, it is useful to use both medical nebulizers and room humidifiers to study the two different humidity regimes. Additionally, the cup in the medical unit allows various test solutions to be used without contaminating the reservoir.

Swift (1980) describes possible rapid droplet evaporation if the output of a nebulizer is diluted significantly with dry air. Since ultrasonic nebulizers use relatively low carrier air flows of 20–25 L/min combined with large water consumption rates up to 6–9 g/min, saturated conditions are almost always present. Mercer et al. (1968) noted that both coagulation and drying of nebulized aerosols can occur under saturated conditions. They also showed that adding a solute to the water changes the equilibrium vapor pressure, affecting evaporation rates and ultimate droplet sizes.

In order to better understand the residential ultrasonic aerosol generation and transport processes, limited-scale tests were conducted simulating residential room humidification and nebulization into transport tubes at saturated humidities. This work included the determination of size distributions of liquid droplets and resultant dried particles in the range of 0.3–18 μm, and the testing of models to predict the characteristic sizes.

EXPERIMENTAL/RESULTS

A Sunbeam model 661 portable home humidifier and a DeVilbiss model 65 medical nebulizer were selected for testing, based on representativeness and availability. Mechanically identical versions of both units can still be purchased commercially. The laboratory test apparatus to study the size distributions consisted of a Plexiglas chamber with an adjustable vent and internal volume of 1.1 m^3. A ground wire was used to minimize the static surface charges on the chamber walls. The effectiveness of this measure was not quantified, although no visible deposits were noted on the walls after the tests were completed. A model CSASP-100 optical aerosol spectrometer made by Particle Measurement Systems (PMS) was used for all size distribution measurements. The three available size ranges (0.32–0.76, 0.5–2.75, and 1.0–12.25 μm (optical diameter) were used independently or in the composite mode, as needed. The PMS unit had been calibrated prior to the experiments with polystyrene latex spheres over the range of 0.5–3.2 μm. The ultrasonic home humidifier was operated in the chamber at room humidities with known quality water or prepared solutions. The aerosol cloud was well

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mixed, wet or dry as desired, and sampled representatively by the PMS monitor. The ultrasonic nebulizer stream was transported directly to the inlet of the PMS using corrugated 22-mm ID polyvinyl tubing supplied with the unit.

The dissolved solids content of Chapel Hill, NC tap water during the test was determined to be 102 mg/L, using an evaporation-to-dryness procedure essentially identical to ASTM D1888-78. The content of the laboratory distilled water was found to be 12 mg/L (a marginal quality for laboratory distilled water). The frequency of the piezoelectric crystal in the Sunbeam humidifier was measured and found to be approximately 1.6 MHz, superimposed on a 120-Hz carrier signal. Measurement of the input power to the crystal was not straightforward because of the design of the circuit board, and the power input in watts per centimeter of surface area was not determined. The frequency of the DeVilbiss nebulizer crystal was taken from the manufacturer's specifications to be 1.35 MHz since the transducer was not easily accessible.

The Sunbeam unit was first operated with tap water in the Plexiglas chamber, using four arbitrarily selected humidifier output settings. The PMS was operated in the lowest two ranges: 0.32–0.76 and 0.5–2.75 μm. Experiments were made with various chamber vent settings, door openings, and sampling line and humidifier locations to determine the conditions needed to produce liquid droplet evaporation. A scan with the PMS preceded each run to provide a background correction, which was typically very small. The humidifier was situated so that its supply air stream was drawn from the background room air. The drying conditions utilized the lower room humidities (40–50% RH) and longer aerosol transport times from the humidifier outlet to the measurement zone of the spectrometer. Computation using Langmuir's equation (Reist, 1984) of the drying time at a room relative humidity of 90%, showed that < 0.1 s was needed for an aqueous 3-μm droplet to completely dry.

An initial experiment was conducted with tap water to examine the effect of humidifier power setting on the size distribution. Substantial differences in particle concentrations were noted, but no measurable differences were detected in the size distributions. The majority of the count and mass distributions were, however, below the smallest detectable size of 0.45-μm aerodynamic diameter (0.32 μm optical) of the PMS. The largest particle size detected was about 1.0-μm aerodynamic diameter, assuming a dried particle density of 2.0 g/cm^3. This is a reasonable density, considering the typical dissolved constituents of tap water. As a comparison the DeVilbiss nebulizer was operated without an output hose attached and gave virtually identical distribution results with the tap water. A series of runs using the lab distilled water gave no counts detectable by the PMS above background for either humidifier.

A limited set of tests were conducted, assuming a shifting of the size distribution as the droplets dried to much smaller particles, representing their residual dissolved solids content. The output of the Sunbeam humidifier using tap water was injected directly into the inlet of the PMS using a 10-cm sampling line to maintain a high humidity and allow only minimal drying time. As expected the "wet" droplet count and mass distributions (Figure 2) were significantly larger than the previously measured dry versions. The mass distribution shows a bimodal shape with mode peaks at 2.4 and 4.4 μm. The count peak has a shoulder near 2.5 μm, indicating the presence of coalesced doublets. Plotting the cumulative data on log-probability paper (Figure 3) gives a count median diameter of 1.8 μm and a mass median diameter of 2.9 μm, with computed σ_g values ($[D_{84}/D_{16}]^{1/2}$) of 1.6 and 1.7, respectively.

Boucher and Kreuter (1968) describe a droplet formation equation derived by Lang (1962) and refined by Peskin and Raco

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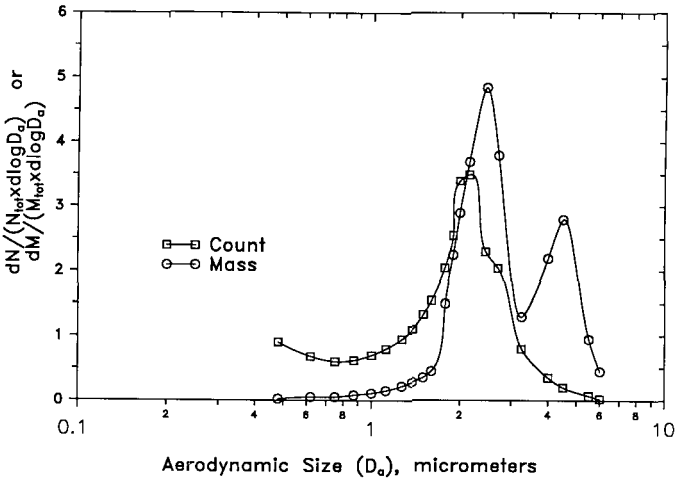

FIGURE 2. Size distribution of wet aerosol from a room humidifier using tap water.

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(1963). The equation predicts the number median aerodynamic diameter of aerosol droplets produced by an ultrasonic nebulizer for a given liquid surface tension and piezoelectric crystal operating frequency. It should be noted that the Boucher and Kreuter (1968) reference incorrectly describes this as the mass rather than the number median diameter. This equation:

$$D_{\text{Ndrop}} = 0.34\left(8\pi S/\left(\rho_{\text{liq}} F^2\right)\right)^{1/3}, \qquad (1)$$

where D_{Ndrop} = number median droplet diameter (cm); S = liquid surface tension (dynes/cm [72 dynes/cm for water and dilute aqueous solutions]); ρ_{liq} = density of liquid (g/cm^3); F = crystal frequency (Hz), applies to the initial liquid aerosol droplet formation process, without regard to transformations that may occur in transport tubes or the air. It predicts a liquid droplet number median size of 3.0 μm from a humidifier operating at 1.6 MHz. For lognormal distributions the mass median diameter could be computed by using the Hatch-Choate (1929) equation:

$$D_{\text{Mdrop}} = D_{\text{Ndrop}} \exp\left(3 \ln^2 \sigma_g\right), \qquad (2)$$

where D_{Mdrop} = mass median droplet diame-

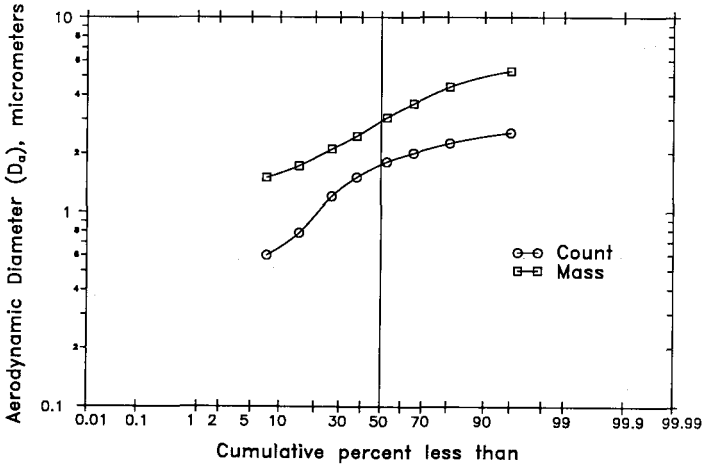

FIGURE 3. Cumulative distributions for Figure 2.

ter (cm); and σ_g = geometric standard deviation of the distribution. Equation (2) would predict a mass median diameter of 5.8 μm, using a σ_g of 1.6 and assuming a lognormal distribution. The disagreement with the measured median diameters in Figure 3 and the computed values could be expected, since partial drying had occurred between droplet generation and measurement by the PMS. In addition, the distributions deviate significantly from lognormality.

An earlier experiment with tap water had shown that the liquid droplets dry to nearly spherical shape (Hardy, 1988), supporting the derivation of the following equation which was based on Eq. (1). It computes the expected dry particle size from the operating parameters of the humidifier:

$$D_{\text{Ndry}} = \left[(KSC_w) / \left(\rho_{\text{sol}} \rho_{\text{liq}} F^2 \right) \right]^{1/3}, \qquad (3)$$

where D_{Ndry} = dry particle number median diameter (cm); K = constant = 9.877×10^{-7}; C_w = liquid dissolved solid content (mg/liter); ρ_{sol} = composite dry density of dissolved solids (g/cm^3).

The D_{Ndry} for the 102 mg/L tap water was computed from Eq. (2) to be 0.11 μm. This median size is well below the lower detectable range of the PMS. By combining Eqs. (1) and (3) the liquid droplet size can be related to the dry particle size:

$$D_{\text{Ndrop}} = D_{\text{Ndry}} \left(\rho_{\text{sol}} \times 10^6 / C_w \right)^{1/3} \qquad (4)$$

where D_{Ndrop} = droplet number median diameter (cm).

Liu (1976) gives a version of this equation to predict the characteristic particle size produced by the vibrating-orifice aerosol generator. The droplet formation process is, however, not the same for ultrasonic generation, producing broader distributions with σ_g's in the 1.4–1.6 range.

Using Eq. (4) solved for D_{Ndry} and the distribution data in Figure 3, an expected dry particle number median diameter of 0.14 μm was calculated. This is very close to the 0.11-μm value predicted based on Eq. (3). The computed D_{Ndry} for the distilled water was 0.05 μm.

In order to simulate larger dissolved solids content, a solution was made with 40,000 mg/L of NaCl in water. Using the DeVilbiss generator with a 43-cm length corrugated hose attached and allowing the emitted droplets to dry at room humidity in the chamber, the resultant size distribution was determined as shown in Figure 4. Unlike the tap and distilled water tests, the count and mass distributions were now well within the range of the PMS with modes of the predominant peaks at 0.9 μm for count and 1.6

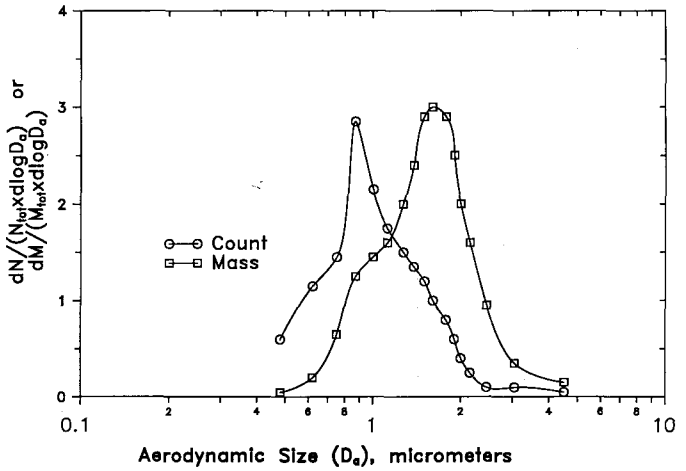

FIGURE 4. Size distribution of dry aerosol from a nebulizer using 40,000 mg/L solution of NaCl.

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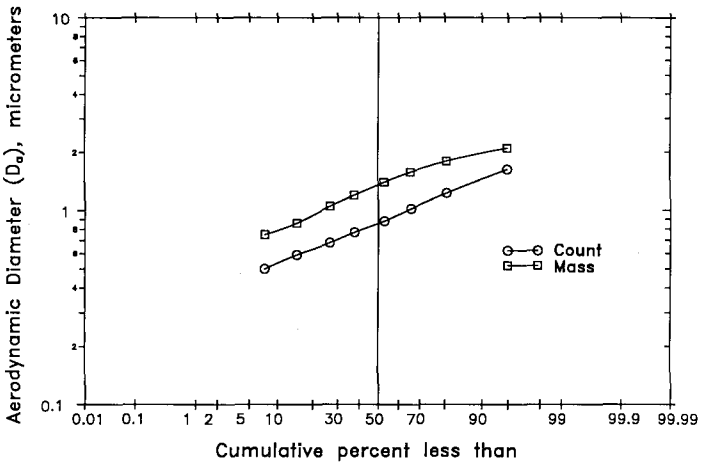

FIGURE 5. Cumulative distributions for Figure 4.

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μm for mass. The cumulative distributions (Figure 5) showed that the data were nearly lognormal and moderately monodisperse, both having σ_g's of 1.45. The D_{Mdry} computed using Eq. (2) and the Hatch-Choate equation and a NaCl density of 2.17 g/cm^3 gave a value of 1.2 μm. This is identical to the measured value of 1.3 μm within experimental error.

Because medical nebulizers are typically used with the aerosol at essentially 100% relative humidity, several tests were conducted to determine the effect of transport time (tube length) on particle size. Wet droplets produced by using distilled water in the DeVilbiss nebulizer were injected directly into the PMS. Sampling was conducted directly above the cup and at the outlets of three lengths (43, 103, and 198 cm) of 22-mm ID tubing attached to the nebulizer. With the tubing attached the DeVilbiss unit had flow velocities of 109, 100, and 86 cm/s, respectively, with corresponding residence times 0.4, 1.0, and 2.3 s.

The wet droplet count distribution results are shown in Figure 6 for each case, with the mass distributions shown in Figure 7. The count distributions show very pronounced peaks at 2.7 μm. The cup measurements gave a slightly different curve shape

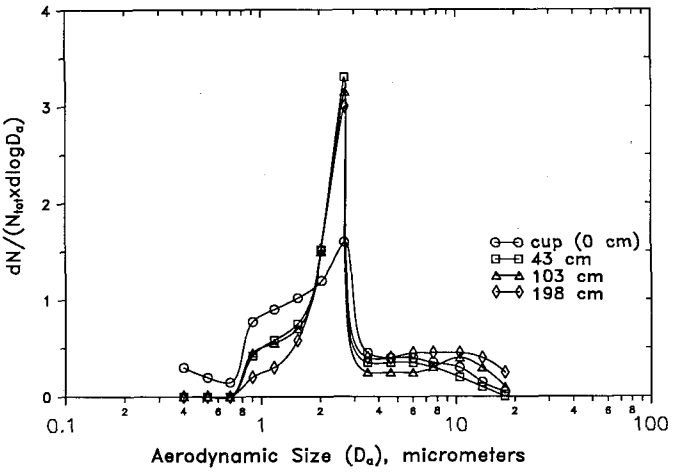

FIGURE 6. Count distributions for wet aerosol from a nebulizer using distilled water and an outlet hose.

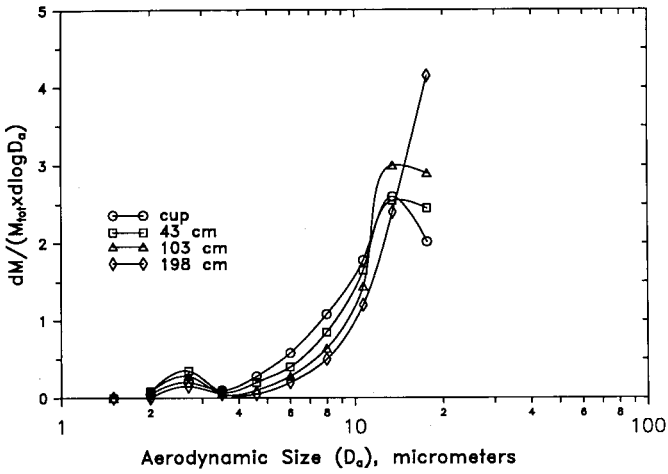

FIGURE 7. Mass distributions for wet aerosol from a nebulizer using distilled water and an outlet hose.

from the three attached tube cases. The mass distribution data in Figure 7 show a tiny peak at 2.7 μm, with most of the mass > 10 μm. As the transport times increased, the modal mass diameter also increased, reflecting droplet coalescence.

The experiment was then repeated using the 40,000 mg/L NaCl solution. The count distribution results for the three tube lengths are shown in Figure 8, with the mass distributions shown in Figure 9. The cup measurements using the PMS were unusable because of extremely high particle counts that overloaded the analyzer. The count data for the 43-cm case are very similar to the same case for the distilled water shown in Figure 6. The 103- and 198-cm cases, however, are substantially different, showing the formation of a dry NaCl particle peak at approximately 0.9 μm (see Figure 4). Droplet drying becomes significant in the nebulizer tubing between residence times of 0.4 and 1.0 s, even though the air stream is nearly saturated. Porstendorfer et al. (1977) showed a similar trend for NaCl solution droplets from a compressed air nebulizer. The mass distributions shown in Figure 9 show the tiny peak at 2.7 μm, but most of the mass is present in peaks with mode diameters from 8 to 14 μm. It should be noted that the mass distributions in Figure 9 are somewhat biased, since all particles were

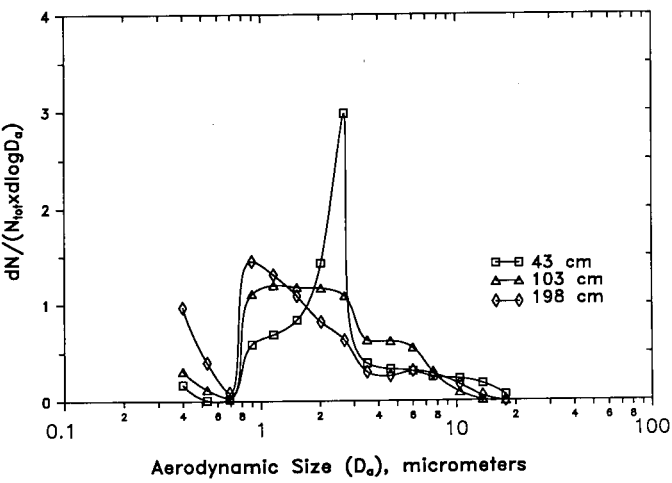

FIGURE 8. Count distributions for wet aerosol from a nebulizer using 40,000 mg/L NaCl and an outlet hose.

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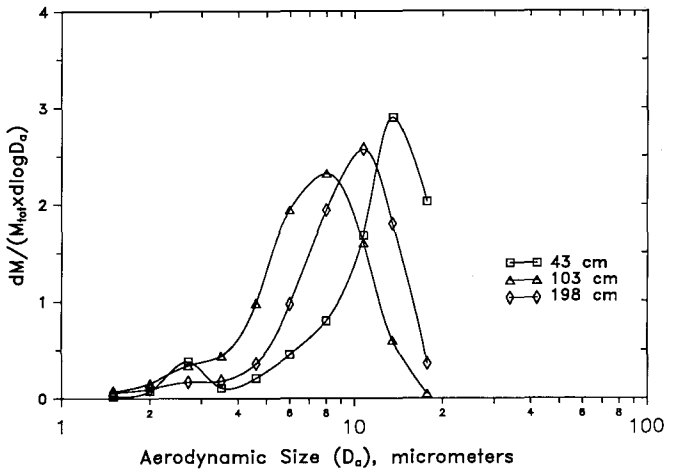

FIGURE 9. Mass distributions for wet aerosol from a nebulizer using 40,000 mg/L NaCl and an outlet hose.

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assumed for simplicity to have the density of the saline solution.

The peak diameters in Figure 9 did not change monotonically with tube length, but were a minimum at the 103-cm length. It is conjectured that this resulted from the competing processes of drying, coalescence and wall loss. After 0.4 s the predominant aerosol size is that of the generated 2.7-μm droplets. Figure 8 indicates, however, that a significant fraction of the droplets have already coalesced to larger sizes, shifting the mass median diameter to 10.9 μm. As the transport time increases to 1.0 s, drying becomes important, decreasing the mass median diameter to 6.3 μm. Coalescence is also occurring to produce a significant increase in droplets in the 3–6-μm range. A portion of the droplets > 10 μm have been lost to the walls or become smaller by drying. As the transport time increases to 2.3 s an even greater number of droplets have dried to the 0.9-μm size, but this is offset by the further coalescence of the 3–6-μm droplets to larger sizes. These factors combine to produce an increase in the mass median diameter at 2.3 s to 8.3 μm.

The results presented here assume that the PMS analyzer accurately responds to both water droplets and NaCl particles over the size range of interest. As pointed out by Pinnick and Auvermann (1979) the response of the PMS model CSASP to particle shape and refractive index must be considered. They showed a CSASP response to NaCl particles (refractive index of 1.544-0i) very similar to that of latex spheres, except in the range of 0.5–1.0 μm, where a positive bias of a factor of 2–3 could be expected. The response for the balance of the detectable CSASP size spectrum was within an estimated 20%. They also demonstrated that the response to water droplets (1.33-0i) was very close to Mie theory and similar to that of NaCl. Deviations were again noted in the 0.5–1.0-μm range, where positive biases of a factor of 3–4 were found.

The substantial CSASP response biases from 0.5–1.0 μm could seriously flaw distributions in this range. Of the distributions presented here, only Figures 4 and 5 describing the distribution and the slope (σ_g) of the dry particles resulting from the 40,000 mg/L NaCl solution are significantly affected by the range departing from Mie theory. The computed values and measured mass median diameters for these data, however, were identical within experimental error, indicating that refractive index bias was of little consequence in this case.

SUMMARY/IMPLICATIONS

Our experiments showed that the dried particles being generated at room humidities re-

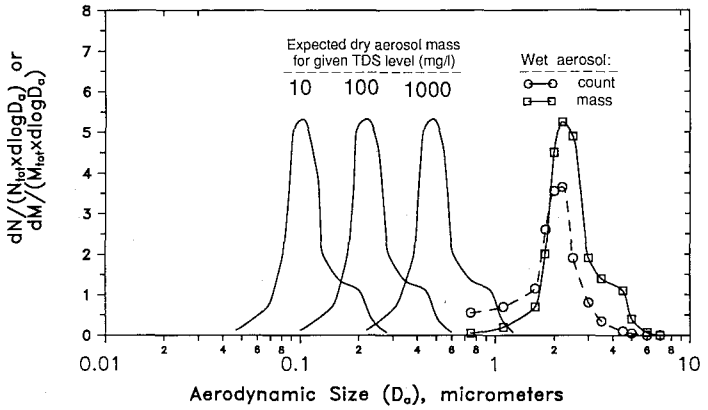

FIGURE 10. Estimated wet and dry aerosol size distributions for room humidifiers using tap water of various dissolved solids contents.

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sulted from the mass of dissolved material in each liquid droplet. The number median diameter of the dried particles produced from tap water with a dissolved solids content of 102 mg/L was predicted to be in the range of 0.11–0.14 μm. Applying Eq. (4) for tap water with dissolved solids contents of 10, 100, and 1000 mg/L produced the projected droplet and dry particle distributions shown in Figure 10. These three cases cover the range expected for most residential tap water, except in the hardest water locations. The distributions should be reasonably log-normal and monodisperse, with σ_g's from 1.4 to 1.6.

The (modified) equation of Boucher and Kreuter (1968) appears to give an accurate estimate of the characteristic liquid droplet size generated initially by either a room humidifier or a medical nebulizer. At room humidities the drying process is so rapid, however, that accurate size distribution measurements would be possible only with *in situ* techniques. Sampling the humidifier droplet output through a short sampling line into the PMS produced a reduced number median diameter as compared to the computed diameter. This approach not only allowed significant particle drying, but unrealistically increased the probability of particle coalescence. Sampling too close to the ultrasonic generation chamber produced particle count densities that often overloaded the PMS. More accurate validation of the droplet generation model would require either redesigning the optical sensing volume or incorporating high speed photography and optical counting.

At the saturated conditions of a nebulizer transport tube, droplet drying is less rapid than at room humidities. The data in Figure 7 would suggest that pure water humidification of the respiratory system using an ultrasonic nebulizer would deposit most of the mass in the mouth and nasal cavities. The effect of nebulizer tube length (residence time) on the mass median droplet diameter was insignificant. When NaCl was present, the count data in Figure 8 suggest that drying of the droplets to smaller particles becomes significant in a nebulizer tube when it is longer than about 100 cm. For residence times in excess of approximately 1.0 second, a variable mixture is present of dried NaCl particles less than 1.0 μm and coalesced multiplets of the primary salt solution droplet size. This would suggest that the mass and form of salt (or perhaps drug) that would be administered to various locations would not be uniform. In a transport tube environment the effect of solute concentrations on mass median diameter is then not

readily predictable from the models presented. The limited data from this study on nebulizers are not sufficient to develop a more appropriate model, but do suggest the need for further work.

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