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The Influence of Relative Humidity on Nanoparticle Concentration and Particle Mass Distribution Measurements by the MOUDI

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A humidity control system was operated upstream of two collocated MOUDIs (micro-orifice uniform deposit impactors) for sampling ambient aerosol particles. One MOUDI used silicone-grease-coated aluminum foils (ALs) as the impaction substrates and was considered as the reference impactor, while the other used uncoated ALs or uncoated Teflon filters (TFs) as the impaction substrates for quantifying the effect of different relative humidities (RHs) and impaction substrates on the $PM_{0.1}$ concentrations and mass distributions of ambient PMs. Test results showed that decreasing RH in general increased particle bounce from uncoated substrates with the bounce from uncoated ALs being more severe than that from uncoated TFs. Particle bounce did not influence the overall mass distribution of ambient fine particles when RH ranged between 40% and 80%, whereas it led to undersampling of particles greater than 2.5 μm in aerodynamic diameter severely. Oversampling of $PM_{0.1}$ occurred by as much as 95%–180% or 25%–55% when the MOUDI used uncoated ALs or TFs, respectively, as RH was reduced from 50% to 25%. Particle bounce was found to be negligible, and $PM_{0.1}$ and $PM_{2.5}$ could be sampled accurately with less than 5% error at the RH of 75%–80% or 65%–80% when uncoated ALs or TFs were used, respectively.

INTRODUCTION

Both ambient ultrafine particles (UFPs) and engineered nanoparticles (NPs) may pose health risks to human beings when they are inhaled or ingested. UFPs and NPs are referred to as $PM_{0.1}$, or particles with diameter smaller than 0.1 μm, in this study. NPs induce lung injury because of their ability to generate reactive oxygen species. Using the RAW 264.7 phagocytic cell line to compare the cellular effects of ambient UFPs with four other engineered NPs, it was found that ambient UFPs and engineered cationic polystyrene NPs showed clear evidence of cellular toxicity as compared to the other engineered NPs. Ambient UFPs had an additional effect to induce proinflammatory responses (Xia et al. 2006). Therefore, it is important to understand the pollutant sources of NPs and adopt proper control measures to avoid human exposure to both UFPs and engineered NPs.

The micro-orifice uniform deposit impactor (MOUDI, MSP Corp., Shoreview, MN, USA) is one of the most commonly used devices for sampling NPs (Chow and Watson 2007). The mass concentration of NPs determined by gravimetric analysis and species concentration determined by subsequent chemical analysis help identify the pollutant sources (Cass et al. 2000). Uncoated substrates provide minimum interference to chemical analysis of collected samples. However, ambient solid particles may bounce easily when impacting the uncoated substrates or previously deposited particles (Tsai and Cheng 1995). Particles that bounce from the upper impactor stages to the lower stages with smaller cutsizes will lead to overestimation of particle mass concentrations in the lower stages. Applying a sticky substance or low viscosity oil on the substrates was recommended to reduce bounce (Gulijk et al. 2003; Pak et al., 1992; Turner and Hering 1987), but interference with the chemical analysis of particulate organic carbon could occur. Besides, using a sticky substance would be ineffective in high-temperature sampling (Cheng and Yeh 1979). Moreover, evaporation of oil and subsequent adsorption by filters may also occur, which create errors in determining particle mass and chemical species concentrations. Thus, uncoated substrates sometimes are used in field sampling studies, and this could be the reason why the MOUDI oversampled $PM_{0.1}$ severely as compared to the calculated $PM_{0.1}$ concentrations from the scaning mobility particle sizer (SMPS) data (Khlystov et al. 2004; Shen et al. 2002).

Ambient aerosol particles are known to be hygroscopic due to the presence of inorganic salts and organic acid, leading to

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water vapor absorption as relative humidity (RH) increases. This suggests that sampling ambient particles at a controlled and high RH condition without using coated impaction substrates is possible since liquid particles are much less bouncy than solid particles. Vasiliou et al. (1999) used an RH conditioner made of 16 ceramic tubes and filled with saturated sodium chloride (NaCl) solution to maintain the RH of the aerosol flow entering the uncoated ten-stage MOUDI (MSP Model 11) at about 75% ± 2% during field samplings at Las Vegas with low RHs of about 10%–30%. The ratio of the percent of sulfate mass on the after filter (or $PM_{0.056}$) to the sum of all stages of the conditioned MOUDI was compared with that of a collocated unconditioned MOUDI. It was found that the bounce of fine ambient particles in the conditioned MOUDI was largely eliminated when the sampled aerosol flow was above ∼70% RH. However, there was a tendency for the sulfate loading at the after filter to increase with decreasing RH due to particle bounce. The average percentage of sulfate mass on MOUDI after filter was 2% and 4%, respectively, for the conditioned and unconditioned MOUDIs implied that the unconditioned MOUDI oversampled $PM_{0.056}$ by as much as 100%. They concluded that an accurate mass distribution of ambient particles could be obtained by the MOUDI when RHs were kept at above ∼70% but below 80% for fear that flow-induced sizing errors might occur (Fang et al. 1991).

Stein et al. (1994) used a tandem differential mobility analyzer (TDMA) with an RH conditioner to study the bounce of atmospheric 0.25-μm particles in a one-stage 0.1-μm cutsize impactor in which uncoated aluminum foils (ALs) were used as the impaction substrates. They found that particle bounce increased sharply as RH decreased below 60%–70%. At the RHs of 50%, 40%, 30%, and 20%, about 10%, 20%, 40%, and 70% of incoming 0.25-μm particles bounced. Less than 3% of particles bounced when the RH was kept at 70%–80%, when the surface loading effect on bounce was also found to be negligible. On the other hand, Dzubay et al. (1976) studied the bounce of larger particles by using the Anderson 2000 cascade impactor, which had the size cuts of 7.0, 3.3, 2.0, and 1.1 μm. In the study, two of the impactors were compared in parallel on the roof of a 4-m high building near a freeway in downtown Durham, NC, one of which used Dow Corning high-vacuum silicone-grease-coated ALs as impaction substrates while the other used uncoated ALs. The grease-coated impactor was regarded as the reference sampler. They found that the mass median aerodynamic diameter (MMAD) of the uncoated impactors was 2–5 times smaller than the coated impactor due to particle bounce. However, the RH data were not reported and related to the bounce effect on the distortion of MMAD.

The main objective of this article is to study the influence of the RH of the inlet aerosol flow on the UFP concentration collected by the after filter of the MOUDI with different uncoated substrates. In addition, the mass distribution as well as $PM_{2.5}$ and PM_{10} fractions were also studied. A humidity control system was operated upstream of two parallel MOUDIs, one of which used silicone-grease-coated ALs (M1, reference MOUDI) and the other used uncoated ALs (M2) or uncoated Teflon filters (TFs, M3) as the impaction substrates. The present experimental setup enabled the study of particle bounce at different conditioned RHs (10%–98%), when conditioned aerosol flow was introduced simultaneously into both MOUDIs to exclude the effect of particle loss in the conditioner on the comparison study since the same inlet was used. The goal of this study is to find out the RH value at which particles do not bounce from uncoated substrates, and measurements of mass distributions and $PM_{0.1}$, $PM_{2.5}$, and PM_{10} concentrations are not influenced by particle bounce.

METHODS

The field comparison tests were conducted 10 m high at the 3rd floor of the Environmental Engineering Building of National Chiao Tung University in Hsinchu, Taiwan. In total, more than forty 24-h samples were taken from January 2009 to March 2010.

A humidity control system consisting of a humidity conditioner (FC 200-780, Perma Pure LLC, Toms River, NJ, USA) with a proportional–integral–differential (PID) controller was used upstream of two collocated MOUDIs as shown in Figure 1. Fixed RHs of 10%–98% at the inlet aerosol flow with the variation within ±2% RH were set by adjusting the flow rates of the moist and dry sheath air of the humidity conditioner through the automatic PID controller. Normally, the set RH was stabilized within 10 min.

In the MOUDIs, the nozzle plates of 3.2-μm cutsize were replaced with those of 2.5-μm cutsize, and the 56-nm cutsize nozzle plates (stage 10) were removed so that $PM_{0.1}$ was collected by the after filter; that is, the cutsizes of the MOUDIs in this study were 18, 10, 5.6, 2.5, 1.8, 1.0, 0.56, 0.32, 0.18, and 0.1 μm at sampling flow rate of 30 L/min. Teflon filters (TefloR2PL047, Pall Corp., Washington, NY, USA) were used at the after-filter stage of each tested MOUDI, while three different impaction substrates were used in stages 0–9 of three different MOUDIs, respectively, including grease-coated ALs (M1, reference sampler), uncoated ALs (M2), and uncoated TFs. The impaction plates were rotated against the nozzle plates by 18° for 3 s after every 20-s pause during the operation of all MOUDIs. Conditioned aerosol flow was introduced into M1 and M2 simultaneously, or M1 and M3 simultaneously, and the effect of particle bounce on PM measurement was determined by comparing the PM mass concentration collected at every stage of the MOUDIs as

$$\frac{|(C_{iM1} - C_{iM2})|}{C_{iM1}} \text{ or } \frac{|(C_{iM1} - C_{iM3})|}{C_{iM1}}, \qquad [1]$$

where C_{iM1}, C_{iM2}, and C_{iM3} are the PM mass concentrations at stage i of M1, M2, and M3, respectively ($i = 0$–9 for impaction stages, $i = 10$ for the after-filter stage). The effect of particle bounce on $PM_{2.5}$ and PM_{10} concentrations was also examined,

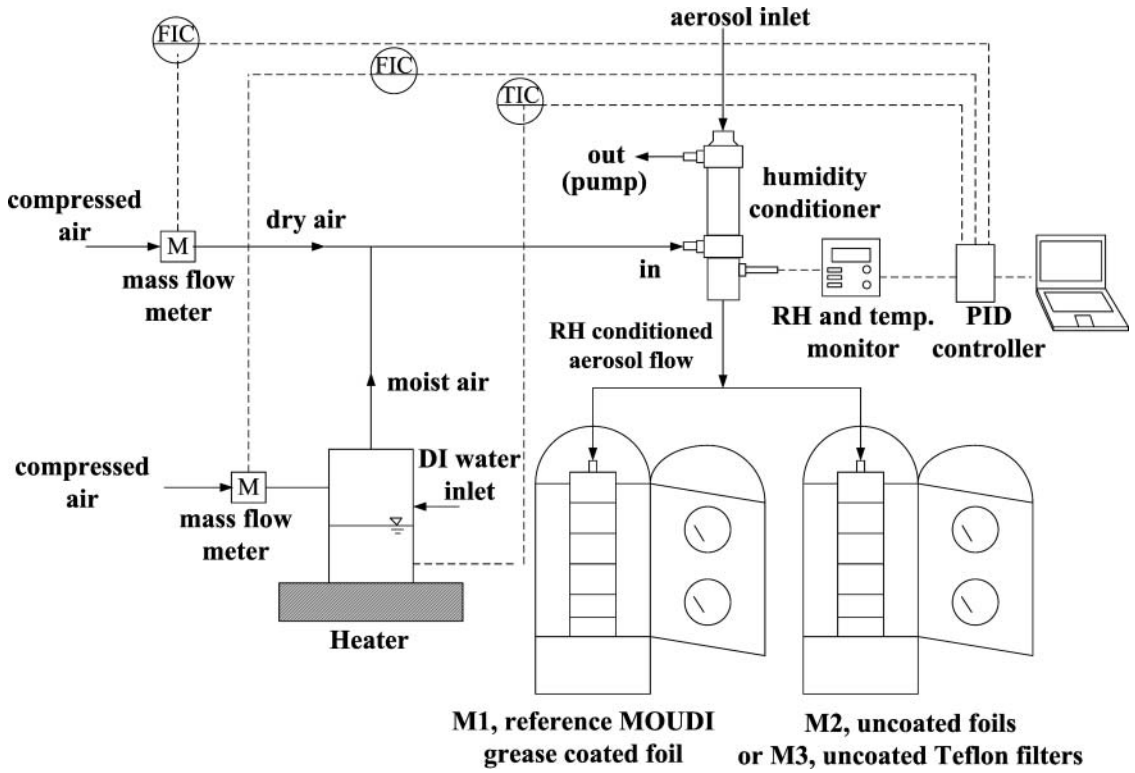

FIG. 1. Schematic diagram of the present experimental system.

in which $PM_{2.5}$ and PM_{10} were calculated based on the mass distributions of MOUDI and the collection efficiency curves of the EPA $PM_{2.5}$ Well Impactor Ninety-Six (Peters et al. 1997) and the Hi-Vol Sampler (McFarland et al. 1984), respectively. The MMADs of the accumulation and coarse particle modes, which can be used to examine the distortion of the particle mass distribution, were calculated by the DISFIT software (Whitby and Whitby 1989) in this study.

In our previous study, a MOUDI using coated ALs substrates and the Teflon after filter (similar to the reference sampler M1 in this study) was applied to obtain the chemical mass closure of $PM_{0.1}$ at different atmospheric environments successfully (Chen, Tsai, Chou, et al. 2010; Chen, Tsai, Huang, et al. 2010). The $PM_{0.1}$ concentration measured by the MOUDI was found to agree with that converted from the number concentration of an SMPS (Model 3936, TSI, Inc., Shoreview, MN, USA) by using the effective density obtained by Chen, Tsai, Chou, et al. (2010). Both $PM_{2.5}$ and PM_{10} concentrations of the grease-coated M1 were also found to be in very good agreement with those of a collocated dichotomous sampler (Model SA-241, Andersen, Inc., GA, USA), with an average relative difference of less than 7.5% (Chen, Tsai, Huang, et al. 2010).

Before the particle bounce test, three MOUDIs (M1, M2, and M3), all of which used grease-coated ALs in stages 0–9 and TF in the after filter, were collocated to compare the mass distributions of atmospheric aerosols to ensure no bias between them. In total, five 24-h measurements were conducted. In addition, two MOUDIs (M1 and M2), one (M1) with a humidity conditioner and the other (M2) without a humidity conditioner at its inlet, were collocated in the comparison tests to examine particle loss in the humidity conditioner. Five 24-h measurements were conducted during the cloudy and rainy days of high RH of 80%–95% when particle bounce was not expected to occur (Vasiliou et al. 1999). Loss of submicron and ultrafine particles with the diameter smaller than 0.6 μm in the humidity conditioner was examined in more detail by introducing polydisperse NaCl particles into the humidity conditioner. The particles were produced by atomizing an NaCl solution using an atomizer (Model 3076, TSI, Inc., MN, USA), into the humidity conditioner. Subsequently, the loss was determined by comparing the size distributions of NaCl particles at the upstream and downstream of the humidity conditioner as measured by the TSI model 3936 SMPS with a long DMA (TSI model 3081).

In every sampling run, at least two TFs and two uncoated and silicone-grease-coated ALs were used as laboratory and field blanks, respectively, for gravimetric analysis. All filter samples were conditioned at least for 24 h in a temperature and RH controlled room (21.5°C $\pm$ 1°C, 40% $\pm$ 5% RH) before and after sampling. For coated ALs, 0.3–0.5 mg of silicone grease (KF-96-SP, Topco Technologies Corp., Taiwan) was applied uniformly on the foils (Chen, Tsai, Huang, et al. 2010; Pak et al. 1992). After coating, the foils were baked in

an oven at 65°C for 90 min (Marple et al. 1991). The electrostatic charge of the TFs was eliminated by an ionizing air blower (Model CSD-0911, MEISEI, Japan) before weighing (Tsai et al. 2002). A microbalance (Model CP2P-F, Sartorius, Goettingen, Germany) was used to weigh the filters after they were conditioned.

The possibility that the coated silicone grease might evaporate from stages 0–9 during sampling and be subsequently absorbed by the collected $PM_{0.1}$ particles or the after filter was examined before the sampling campaign. The weight of the coated ALs before and after sampling clean filtered air by a HEPA (high efficiency particulate air) filter for 24 h was compared to check if evaporation occurred from the coated ALs. Results showed that the weight difference was <2 μg for each foil of stages 0–9, indicating evaporation was negligible during sampling. That is, use of silicon grease in stages 0–9 did not interfere with the $PM_{0.1}$ mass concentrations in this study.

RESULTS AND DISCUSSION

QA/QC Results

The gravimetric analysis of all laboratory and field blanks showed that weight differences between presampling and postsampling were less than 3 μg, which was relatively low compared to the weight of 43 μg for the typical $PM_{0.1}$ concentration of 1 μg/m^3 in urban areas (Chen, Tsai, Huang, et al. 2010). The comparison of mass concentrations of ambient particles between three collocated MOUDIs showed very good agreement for all size intervals, especially for the last three stages ($PM_{0.18–0.32}$, $PM_{0.1–0.18}$, and $PM_{0.1}$) as shown in Figure 2. Four repeat tests, conducted at different days, showed similar results. Particles at the present site were seen to be bimodal including the accumulation and coarse modes, which enabled us to investigate the bounce of UFPs, fine and coarse particles at the same time using different impaction substrates at different RHs.

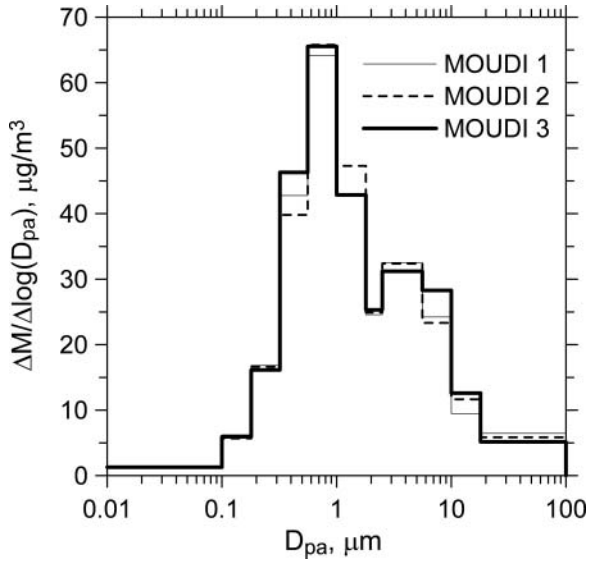

FIG. 2. Comparison of particle mass distributions between three collocated MOUDIs.

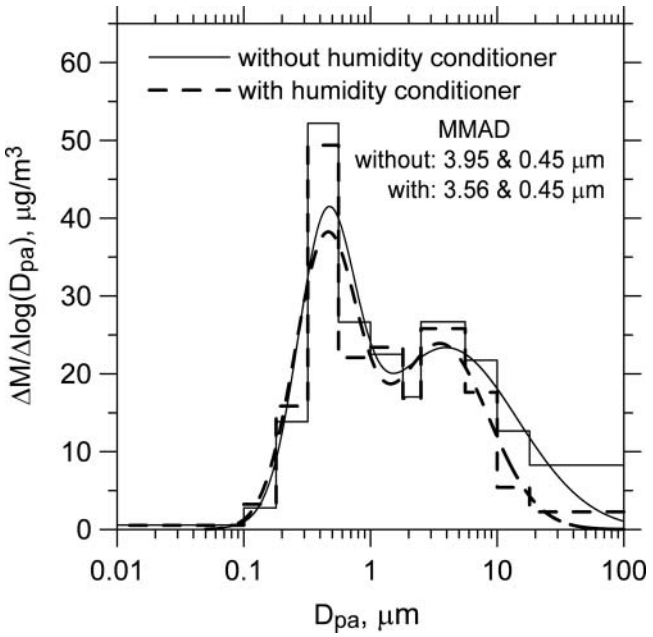

FIG. 3. Comparison of particle mass distributions between two collocated MOUDIs with and without humidity conditioner, ambient RH = 80%–95%. Note: There was no control on the RH of the inlet aerosols for the MOUDI with the humidity conditioner.

Particle Loss in the Humidity Conditioner

Figure 3 shows the comparison of mass distributions obtained by the MOUDI with the humidity conditioner installed but not running and the other MOUDI without the conditioner. There was no control of the RH for the MOUDI with the humidity conditioner, and both MOUDIs sampled aerosols at the RH of 80%–95%. The MMADs of the coarse and accumulation modes of the MOUDI with the conditioner were found to be 3.56 and 0.45 μm, respectively, while the corresponding MMADs of the other MOUDI without the conditioner were 3.95 and 0.45 μm, respectively. The same accumulation-mode MMADs and similar mass concentrations in each stage below the 2.5-μm cutsize stage between the two MOUDIs indicated that particles loss in the humidity conditioner was low for fine particles. However, a smaller coarse-mode MMAD (with a difference of $\sim$10%) and a lower mass concentration in each stage above the 2.5-μm stage of the MOUDI with the conditioner than that without conditioner indicate that a loss of coarse particles occurred in the humidity conditioner. Loss in the conditioner for $PM_{>18}$ (stage 0), $PM_{10–18}$ (stage 1), $PM_{5.6–10}$ (stage 2), and $PM_{2.5–5.6}$ (stage 3) was 72.5%, 52.3%, 20.9%, and 5.3%, respectively, leading to an underestimation of the corresponding mass concentrations of these large PM fractions. In comparison, $PM_{0.1}$ concentrations of the two collocated MOUDIs, which were 0.56

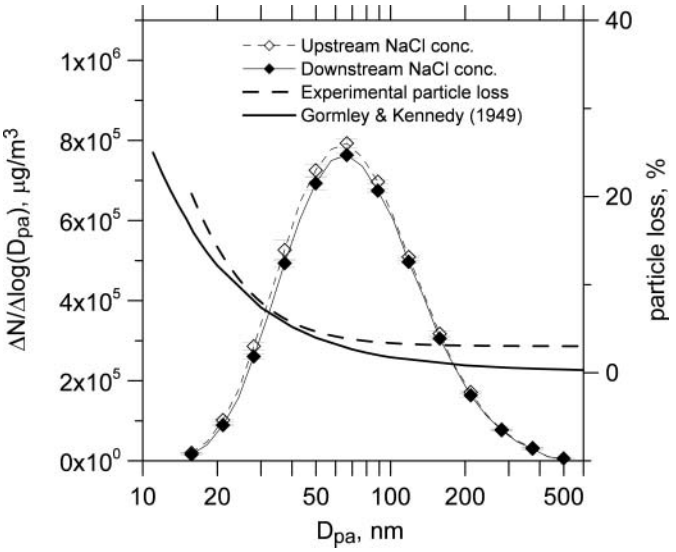

FIG. 4. Comparison of particle loss in the humidity conditioner for particles smaller than 600 nm between the experimental data and the predicted values of Gormley and Kennedy (1949) equation.

and 0.53 μg/m^3, respectively, were in very good agreement with a difference of only 5.4%. Four other repeated measurements showed similar results with the average difference of 5.8% $\pm$ 2.1% (average $\pm$ standard deviation) between the two MOUDIs.

For the humidity conditioner, Figure 4 compares the experimental data of particle loss for particles smaller than 0.6 μm in aerodynamic diameter with the theoretical values obtained from the equation of Gormley and Kennedy (1949). In the figure, the aerodynamic diameter of NaCl was converted from the mobility diameter of the SMPS by using an effective density of 1.9 g/cm^3 for NaCl particles, which was obtained by comparing the volume concentration determined from the SMPS using the AIM software with the mass concentration from the gravimetric analysis of the collected filter samples of the generated NaCl particles. As can be seen in the figure, the experimental data for particle loss are in very good agreement with the theoretical values with a maximum difference of ~4%. The experimental particle loss increased from 3.4% to 20.3% as the particle diameter was decreased from 100 to 15 nm. For particles larger than 100 nm, slightly higher experimental losses than the predicted values were observed, because of the fact that the effects of inertial impaction and gravity were not considered in the theory. From the particle loss data and density of NaCl particles, the loss of $PM_{0.1}$ concentration was calculated to be about only ~5%. This small loss of $PM_{0.1}$ is similar to the small difference of 5.4% between the two MOUDIs (one with the conditioner and the other without the conditioner) described previously in Figure 3. In summary, installing the humidity conditioner at the MOUDI inlet does not lead to the losses of fine and nanosized particles, but the losses for coarse particles are substantial.

Particle Bounce Effect on Mass Distribution

Figure 5 compares the mass distributions and the MMADs of M1 (coated ALs) with those of M2 (uncoated ALs) or M3 (uncoated TFs) when the inlet aerosols for both collocated MOUDIs were conditioned to 25%, 50%, and 75% RHs. The RHs covered dry, moderate, and wet conditions. Both accumulation- and coarse-mode MMADs of M2 and M3 were found to be in good agreement with those of M1 at 50% and 75% RHs (Figure 5c–f). However, at RH = 25%, shifting of the mass distribution curve to the smaller diameter occurred, which led to about 9%–12% (M2) or 6%–9% (M3) smaller accumulation- and coarse-mode MMADs, respectively. Even more shifting of the curve toward the smaller size was found at RH = 10% for M2 when the reduction of accumulation- and coarse-mode MMADs was 21% and 15%, respectively.

It was found that M1 sampled much more $PM_{2.5–10}$ mass than M2 and M3 at both RHs of 25% and 50%. M2 and M3 undersampled $PM_{2.5–10}$ by 22.5% and 14.2%, respectively, at the RH of 25%, while undersampling at 50% RH was less (11.0% and 10.2% respectively) for M2 and M3. That is, more particles bounced from uncoated ALs than from uncoated TFs at RHs <50%. Besides, M2 and M3 were also found to undersample particles at upper 0th, 1st, and 2nd stages even at an RH as high as 75% (Figure 5e–f), with 30.5%, 7.0%, and 8.4% underestimation for M2 and 26.5%, 6.4%, and 8.4% underestimation for M3, respectively. Particle bounce was found to be reduced further at RHs of 80%–98%, and undersampling was reduced to less than 3% for both M2 and M3. The observation that bounce occurred for particles larger than 2.5 μm was in agreement with that observed by Dzubay et al. (1976). The bouncing particles were lost on the inner wall of the MOUDI rather than bounced to the lower stages, as it was evident that the sum of PM mass at lower 4–9 stages did not increase in M2 and M3. This speculation was also observed by Cheng and Yeh (1979) who found that wall loss of 7, 5, 3, and 1.8-μm particles was about 58%, 54%, 40%, and 20%, respectively, in a ungreased Sierra radial slit jet cascade impactor (Model 216, Sierra Instruments, Inc., CA, USA).

Ratios of M2 and M3 PM concentrations to those of M1 at different RHs are compared in Figure 6 to examine the effects of RHs and different substrates on particle bounce. The linear regressions of $PM_{0.1}$ and PM_{10} ratios are also shown in the figure. Note that the regression of PM_{10} ratios was obtained from pooling both M2 and M3 data since they showed a similar trend. From the figure, it is observed that the ratios of M2 and M3 to M1 are close to 1 for all PMs at high RHs of 75%–98%. Surprisingly, the ratios of M2 and M3 to M1 for $PM_{2.5}$ were both found to be near 1 even when RH was decreased from 75% to 10% or 20%, respectively. However, the PM_{10} ratios were decreased to ~0.75 and ~0.8 for M2 and M3, respectively, at these low RHs. This was due to the loss of coarse particles ($PM_{2.5–10}$) at the low RHs as discussed previously. Although M2 and M3 sampled $PM_{2.5}$ concentrations close to those of M1 even at low RH of 10% or 20%, their mass distributions and

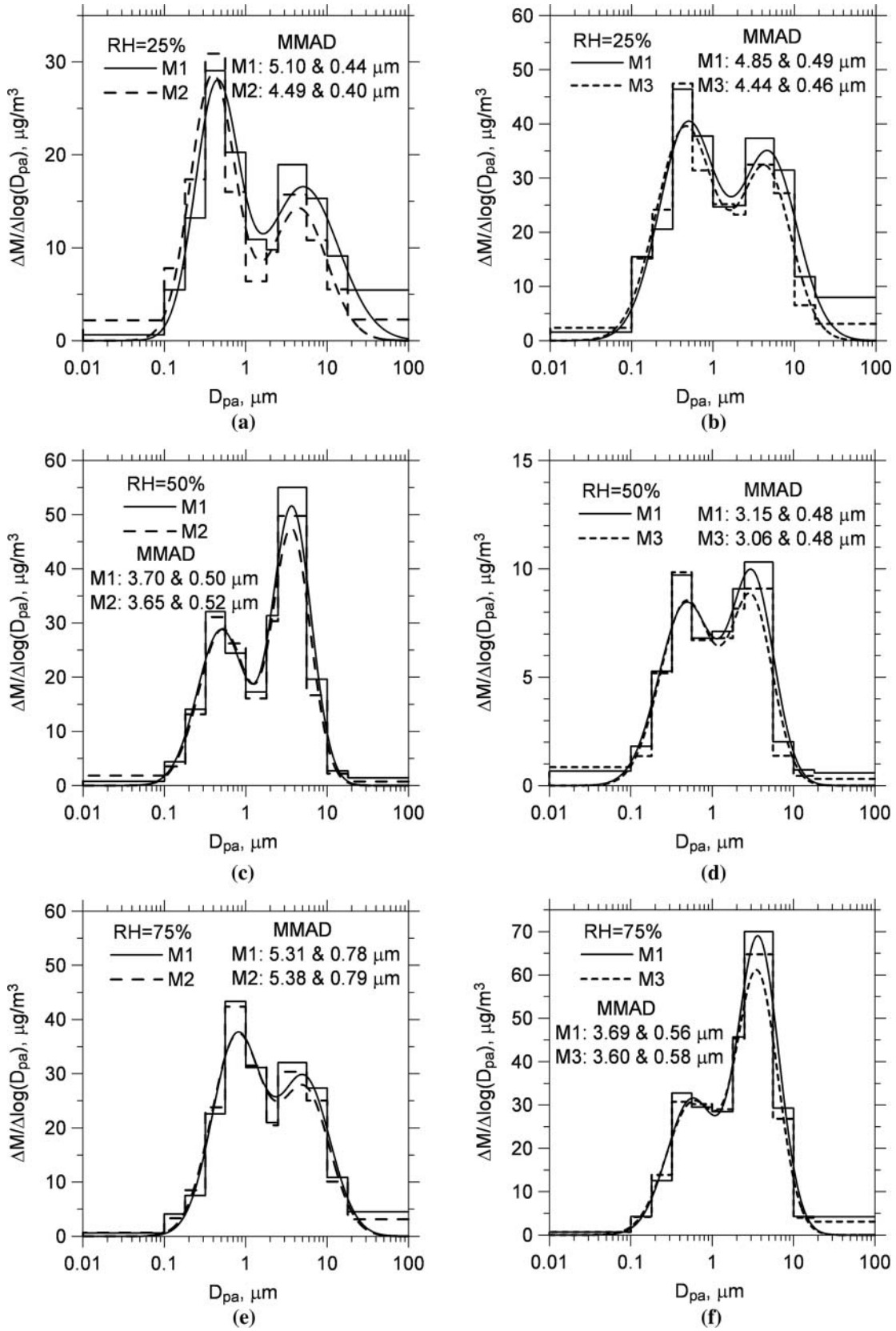

FIG. 5. Comparison of particle mass distributions and the accumulation and coarse MMADs of M2 or M3 with those of M1 at the conditioned RH of 25%, 50%, and 75%.

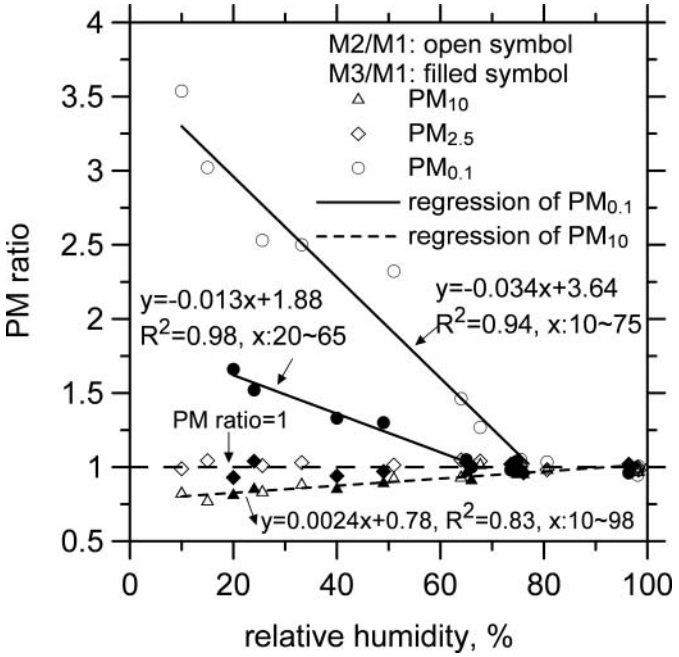

FIG. 6. Ratio of PM_{10}, $PM_{2.5}$, and $PM_{0.1}$ by M2 or M3 to that by M1 at different conditioned RHs.

accumulation MMADs deviated severely from those of M1, which can be seen in Figure 5a and b and Figure 7, respectively. This is because fine particles bounced to the lower stages rather than being lost in the inner wall, as was the case for coarse particles.

Particle Bounce Effect on $PM_{0.1}$ Concentration

The average $PM_{0.1}$ determined by M1 for all forty samples was 0.8 ± 0.3 $\mu g/m^3$ (average $\pm$ standard deviation), which was comparable with those at the urban areas in Los Angeles, CA (Cass et al. 2000). As was mentioned previously, the present M1 determined a comparable $PM_{0.1}$ concentration to that converted from the SMPS data. The $PM_{0.1}$ of M1 was therefore used as a reference for quantifying the influence of particle bounce on $PM_{0.1}$ of M2 and M3.

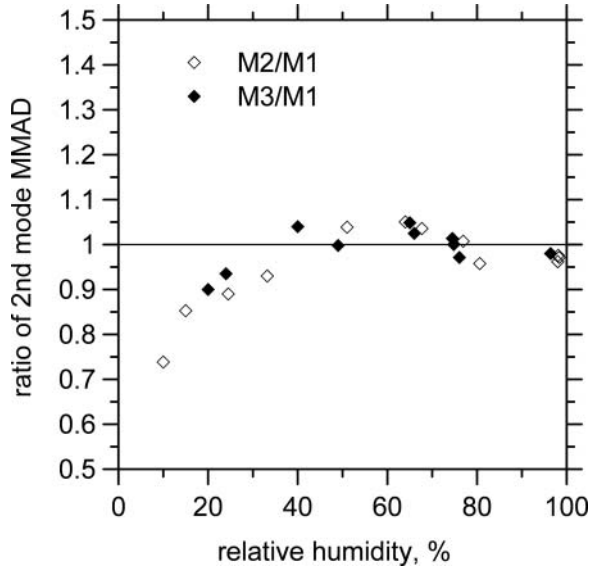

FIG. 7. Ratio of accumulation-mode MMADs of M2 or M3 to those of M1 at different conditioned RHs.

It was found that both M2 and M3 collected $PM_{0.1}$ concentrations very close to those of M1 at the RH of 75% (Figure 5e and f). However, bouncing particles from stages 8 and 9 to the after filter ($PM_{0.1}$) led to oversampling of $PM_{0.1}$ by more than 30% at RHs of 25% and 50% (Figure 5a–d). Oversampling of $PM_{0.1}$ by 55% at 25% RH in M3 was very close to that found in Vasiliou et al. (1999). Particles could also bounce from 4 to 6 stages and be collected at lower stages (stages 7–9 and the after filter) at the RH of 25% for M2 (Figure 5a and b). Less severe bounce in M3 (TFs) than that in M2 (ALs) at the RH of 25% or 50% was found because of the softer filter surface and the additional filtration mechanism of M3.

Figure 6 shows bounce starts to occur at RHs below 75% and 65% in M2 and M3, respectively, which was also found by Stein et al. (1994) and Vasiliou et al. (1999). Both M2 and M3 sampled $PM_{0.1}$ concentrations close to those of M1 at RHs $>\sim$75 and $>\sim$65%, respectively, with a difference of less than 5%. However, $PM_{0.1}$ was oversampled by M2 by as much as 95% and 180% at the RH of 50% and 25%, respectively, while oversampling by M3 was less, which was 25% and 55%, respectively. On the basis of the present results, it can be concluded that particle bounce is negligible at the RH of $>$65% or $>$75% when uncoated TFs (M3) or uncoated ALs (M2) are used as the impaction substrates of the MOUDI.

In order to know the bounce effect on the distortion of the mass distribution, the ratio of the accumulation-mode MMADs of M2 and M3 to those of M1 at the RH from 10% to 98% was compared and shown in Figure 7. As can be seen, the MMADs of both M2 and M3 are in good agreement with those of M1 at RH $>\sim$40% with a difference of less than $\pm$5%. However, the difference increased with decreasing RH, and the ratio was decreased to $\sim$0.75 for M2 at the RH of 10% and to $\sim$0.9 for M3 at the RH of 20%. That is, accumulation-mode particles can be collected accurately by the MOUDI using uncoated substrates as RH $>$40%.

CONCLUSION

The effects of RH of the incoming aerosols and different substrates on particle bounce in the MOUDI were examined in this study. The tested RH ranged from 10% to 98%, which was achieved by a humidity conditioner, and the tested substrates were silicone-grease-coated ALs (M1), uncoated ALs (M2), and uncoated TFs (M3). Before the bounce test, particle loss in the conditioner was examined first, and results showed that the conditioner did not lead to losses of fine and nanosized particles, but losses for coarse particles were severe.

Because of particle bounce, M2 oversampled $PM_{0.1}$ by as much as 95% and 180% at the RH of 50% and 25%, respectively, while $PM_{0.1}$ oversampling by M3 was 25% and 55%, respectively, at these two RHs. Less severe bounce in M3 (TFs) than in M2 (ALs) at the RH of 25% and 50% was caused possibly by the more rigid surface of ALs than TFs. It was found that the accumulation-mode particles could be collected accurately by the MOUDI using uncoated substrates when RH > 40%. Both M2 and M3 sampled $PM_{2.5}$ comparably to M1 even when RH was decreased from 75% to 10% or 20%, respectively, because bouncing fine particles were collected by the lower stages of the MOUDI. In comparison, larger bouncing particles of $PM_{>18}$ (stage 0), $PM_{10–18}$ (stage 1), $PM_{5.6–10}$ (stage 2), and $PM_{2.5–5.6}$ (stage 3) were lost on the inner wall of M2 and M3 rather than bounced to the lower stages. On the basis of the present experimental results and the previous finding that flow-induced sizing errors occurred at RH > 80% (Fang et al. 1991), it is concluded that $PM_{0.1}$ and $PM_{2.5}$ can be obtained accurately with less than 5% error at RHs of 65%–80% and 75%–80%, respectively, when uncoated TFs and uncoated ALs are used in MOUDIs.

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