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A Device to Measure the Size of Volatile Droplets Utilizing a Hot-film Sensor

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ABSTRACT. A device has been developed that is useful for the size measurement of volatile liquid droplets. In this device, droplets are sampled into a tube, where they are allowed to collide with a hot-film sensor, which has originally been designed for use in a constant-temperature anemometer. Upon a collision of each droplet, a pulse is formed in the voltage applied to the sensor, and the diameter of the droplet is obtained directly from the pulse area; no calibration with any standard is necessary. The accuracy of the hot-film method was proved through test experiments in which water droplet sizes measured by the present device were compared with those measured by a microscopic photographic or a gravitational settling method. By evaluating the detection efficiency of droplets, the number concentration of droplets was also obtainable. The device was shown to be applicable to ethanol droplets as well as to water droplets. A field test to observe a fog event was carried out, and it was found that the variation of the liquid water content could be tracked with a measurement interval of about 3 min. AEROSOL SCIENCE AND TECHNOLOGY 26:505–515 (1997)

INTRODUCTION

It is generally difficult to measure the size of volatile droplets, since it is highly susceptible to variation during the measurement. The droplets should be sampled without change of the ambient temperature and pressure. These requirements are satisfied most readily by optical methods, and indeed several sorts of them are commonly used in the field of cloud physics and of two-phase flow engineering; in the former field, the impaction-replicating methods are also in some use. Among all of those, the most powerful one seems to be the phase-based optical method such as the phase Doppler particle analysis (Durst and Zare, 1976). This technique eliminates the need for the calibration procedure, which is indispensable in the more traditional intensity-based optical method. The absolute measurement is highly desirable since the calibration with volatile droplets of known sizes is an extremely laborious task.

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While the phase-based method is superior to others in many respects, it requires elaborate instrumentation, toilsome optimization of various parameters and complicated data processing procedure, and these make it unsuitable for handy use, for example, in field observations. A simpler method was proposed by Medecki et al. (1975), who showed that a hot-wire device used for measuring the cloud water content was usable also for droplet sizing. Their method, however, was not free from the calibration problem. This study proposes another version of hot-wire sizing device that affords the absolute measurement while retaining the simplicity in construction and operation. It utilizes a hot-film sensor, which has been used so far in a constant temperature anemometer, and it produces signals directly connected to the diameter of individual droplets, thus eliminating the necessity of calibration. Moreover, the sensor can be mounted in a sampling tube compact enough to conserve the droplet size.

PRINCIPLE OF MEASUREMENT

The droplet sizing method developed here is based on measurement of the heat of vaporization of a droplet colliding with a heated filament. A commercially available hot-film sensor (Kanomax Model 1210-60W), which has originally been designed for the measurement of water flow rate, is applied for this purpose. The sensor is a quartz filament, 152 μm in diameter and 3 mm in length, stretched between two branches of a Y-shaped supporter. The filament is coated with platinum film, so that it can be electrically heated. Sensor voltage is applied to the platinum film from a constant temperature anemometer unit (CTA unit; Kanomax Model 1010), which functions to keep the sensor at a constant temperature. If the sensor temperature is set above the boiling point of the liquid, a droplet is warmed up upon collision with the sensor, and it evaporates by absorbing the necessary amount of heat from the sensor. Accordingly the output voltage from the CTA unit increases so as to compensate for the heat lost by the evaporation, and it returns to the initial level after the evaporation is completed. As a result the voltage forms a pulse upon a collision of a single droplet. If the initial level and the increase of the voltage are denoted by V and ΔV, respectively, the increase in electric power is given by

$$\Delta P = (V + \Delta V)^2/R - V^2/R \sim 2V\,\Delta V/R, \tag{1}$$

where R is the electric resistance of the sensor in the heated condition. Then the amount of heat, Δq, absorbed by a droplet colliding with the sensor is

$$\Delta q = \int \Delta P\,dt = (2V/R)\int \Delta V\,dt. \tag{2}$$

Here t denotes the time and the integration extends over a single pulse duration. Therefore $\int \Delta V dt$ is the area of a voltage pulse. On the other hand, Δq is related to the diameter, D, of the droplet by

$$D = (6\Delta q/\pi\rho c)^{1/3}. \tag{3}$$

Here ρ is the density of the liquid, and c is the heat required to warm up and vaporize the unit mass of the liquid initially kept at the air temperature, and is given by

$$c = C_L(t_b - t_0) + H + C_v(t_s - t_b), \tag{4}$$

where C_L and C_v are the specific heat of the liquid and its vapor, respectively; H is the heat of vaporization; t_b is the boiling point; t_0 is the temperature of the air; and t_s is the temperature of the sensor. If one is aware of what kind of liquid droplets are being sampled, ρ and c appearing in Eq. (3) are obtainable from physicochemical data, so that D can be calculated from Δq directly. Namely no calibration is required; the present method thus has capability of an absolute measurement as long as the droplet material is known.

On the principle that the amount of heat lost during droplets' collision with a heated wire is proportional to the mass of the droplet, at least three devices have been

constructed for the measurement of the liquid water content of clouds. They are Johnson–William liquid water probe (Neel and Steinmetz, 1952; Neel, 1955), "nimbiometer" (Merceret and Schricker, 1975), and CSIRO liquid water probe (King et al., 1978), and the characteristics of these devices have been examined in detail (Baumgardner, 1983; Bradley and King, 1979; King et al., 1981; Knollenberg, 1972; Spyers-Duran, 1968). Following the same principle, the present study attempts to extend the performance of the hot-wire device to the measurement of the droplet size spectra by resolving pulses in the sensor voltage caused by collision of individual droplets. A droplet-sizing device based on the pulse observation was developed by KDL Associates, Inc. (Medecki et al., 1975, 1979; Magnus et al., 1979), but its operation is somewhat different from the present one. Their device utilizes a hot-wire sensor in a constant current mode: collision and subsequent evaporation of a droplet causes a decrease in the sensor temperature, and it leads to a decrease in the electric resistance, which in turn brings about a downward pulse of the voltage. Since the temperature decrease is dependent on the heat capacity of the sensor, it is necessary to determine this quantity in advance by a calibration procedure using droplets with known sizes.

EXPERIMENTAL SETUP AND PERFORMANCE TESTS

The hot-film sensor was mounted in a sampling tube as shown in Figure 1. The tube was entirely made of glass with one of its ends formed into a streamlined nozzle, behind which the sensor filament was set at a right angle to the tube axis. The opening of the nozzle was about 1 mm in diameter. The exit end was connected to a pump via a mist trap and a flowmeter. The whole experimental setup used for the performance test is shown in Figure 2. Water droplets were generated by an ultrasonic nebulizer. Air was inhaled through the nozzle by the pump and the droplets were sucked into the tube so that some of them collided with the filament. The voltage supply line to the sensor was branched out to a digital oscilloscope (Tektronix Model 2430) so as to monitor the pulses produced on collisions of the droplets. The CTA unit employed here accepted an input value for the operating resistance, R, of the sensor; if one puts R equal to, say, 6.9 Ω, then the sensor temperature was kept, through the resistance-temperature relationship, at 140 °C. An example of the oscilloscope display in such a condition is shown in Figure 3(a). In this case, V was about 1.3 V and ΔV was 10 mV on the average. The oscilloscope had 1024 channels for picking-up digitalized signals in the course of a single scan. In order to obtain accurate pulse area, it was necessary to pick up more than ten points from each pulse, whereas the typical pulse width was 500 μs as shown in Figure 3(a). Therefore an appropriate picking-up interval was about 40 μs, and accordingly the scanning time of the oscilloscope was set to be 40 ms. The digitalized voltage signal was transferred to a personal computer (NEC PC 9801 VM with a 16-bit CPU and a 10-MHz

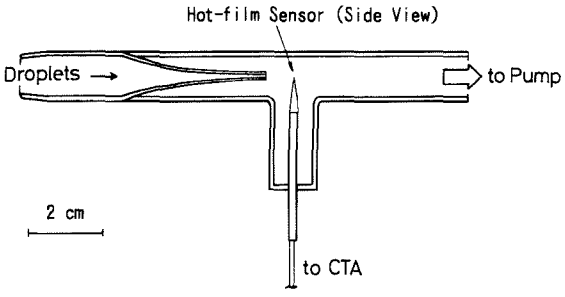

FIGURE 1. Structure of the sampling tube.

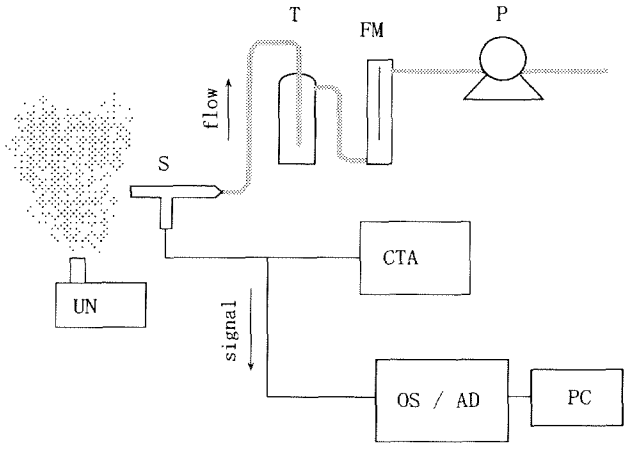

FIGURE 2. Experimental set up for the performance test. UN: ultrasonic nebulizer, S: sampling tube, CTA: constant-temperature anemometer unit, OS/AD: digital oscilloscope or AD converter, PC: personal computer, T: mist trap, FM: flowmeter, P: pump.

clock), and there noise was removed by smoothing [Figure 3(b)]. Recognition of each pulse was facilitated by deriving the differential waveform [Figure 3(c)]. Subsequently the area of each pulse was calculated and converted into the droplet diameter through Eqs. (2) and (3). The number of pulses were classified according to the diameter and stored in the computer memory. This cycle was repeated until sufficient number of signals were accumulated for deriving a statistically significant size distribution. Figure 4(a) shows a size distribution obtained in the test experiment; in this example, 633 droplets were detected in 75 cycles and classified according to a diameter interval of 0.2 μm. While the net sampling period was 40 ms, it took 7.2 s to run a cycle because of the time required for the data transfer and processing, and hence the fraction of the sampling period was 0.0056 ($= 40$ ms/7.2 s). Although this system was used in the test experiments described below, it was found that the fraction was too small particularly in the field observation, where sometimes very thin fogs were to be measured. Therefore another data acquisition system was prepared, in which a high-speed AD converter (Proside PCL-814 PG) was used instead of the digital oscilloscope, and it was controlled by a computer of higher performance (Pro-side 486C33 with a 32-bit CPU and a 33-MHz clock). With this system the signal was picked up at an interval of 20 μs, and the data were processed every 0.1 s. The time for a cycle was

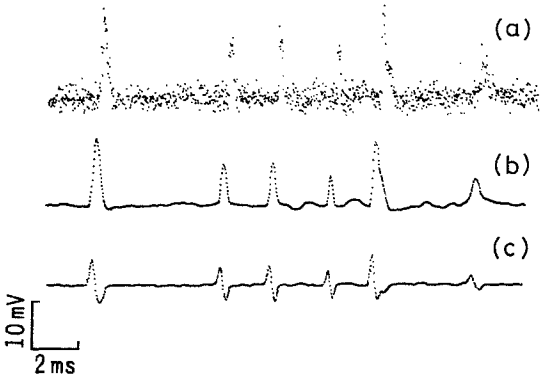

FIGURE 3. Output voltage signal from CTA; (a) Raw data, (b) after smoothing, (c) after differentiation.

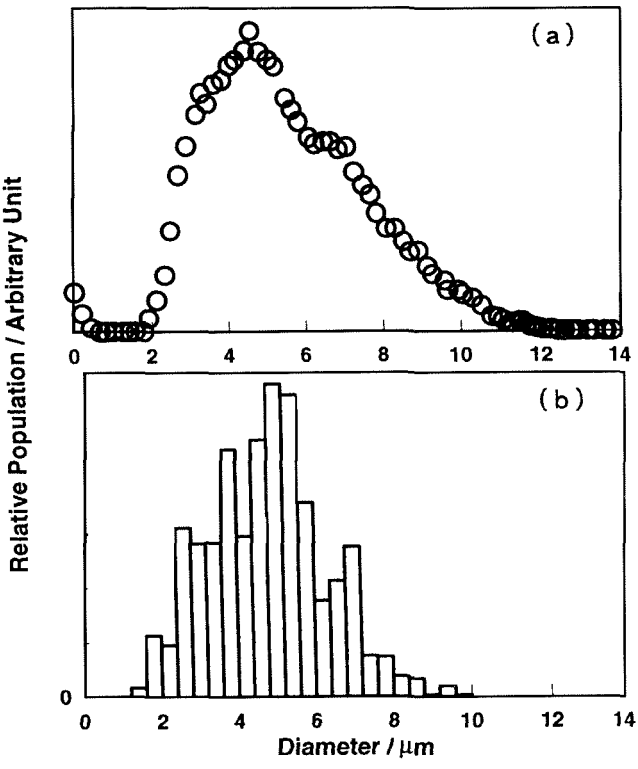

FIGURE 4. Size distribution obtained by (a) the hot-film sensor and (b) the microscopic photographic method.

3 s, so that the fractional sampling period was now 0.033, considerably larger than in the original system.

Optimization of the Sensor Temperature

The response of the sensor was examined at several different temperatures to find the optimum value. When the temperature was as low as 70°C, the voltage pulses were broad and so low that they were hardly recognizable from the background noises. As the temperature of the sensor was set higher, pulses got sharper and higher, and accordingly the sensitivity was improved. When the temperature exceeded 200°C, however, the pulse width no longer got narrower, while the lifetime of the sensor filament seemed to be shorter. From these observations, the optimum temperature of the sensor was found to be about 140°C for water droplets.

Selection of Sampling Flow Rate

For a constant concentration, the number of droplets hitting the sensor increased, of course, as the sampling flow rate increased. Therefore larger flow rate was favorable for obtaining a statistically meaningful size distribution in a shorter period. However, if the flow rate was too large, pulses from the droplets tended to overlap with each other causing a positive systematic error in the size measurement. Moreover, there arose turbulence in the air flow coming out of the nozzle, resulting in increased background noise, which significantly lowered the sensitivity. On the basis of several trial measurements, it was found that the flow rate should not exceed 1.0 ℓ min^{-1} in order to detect small signals from droplets of a few μm in diameter. On the other hand, it was feared that, if the flow rate was too small, very fine droplets failed to collide with the sensor because of their small inertia, and conse-

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quently the size distribution was biased to the larger size range. In order to examine this possibility, the size distribution was measured with several flow rates, and the variation of the average diameter was inspected. The measurement was done under two conditions differing in the droplet density; the results are shown in Figure 5. In both conditions, the average diameters were observed to get larger for the smaller flow rates, and this was consistent with the above anticipation. It was found that the average diameter almost leveled out for flow rates larger than about 0.7 ℓ min^{-1}. Taking these results into consideration, the sampling flow rate was selected to be in the range 0.7 ~ 1.0 ℓ min^{-1} and was adjusted to obtain as many overlapped pulses as possible. At a flow rate of 0.7 ℓ min^{-1}, the smallest detectable size was about 2 μm; in Figure 4(a), a slight increase in the population is discernible in the size range less than 1 μm, but it must be false due to the background turbulence.

The flow rate selected on the basis of the above consideration does not always permit the isokinetic sampling from the ambient air. In the anisokinetic condition, the sampling efficiency depends on the size of particles, and this effect has to be allowed for to derive the size distribution. In this study, the sampling efficiency formula proposed by Zhang and Liu (1989) was used for checking the anisokinetic effect. The ultrasonic nebulizer used in the test experiment generated an air stream with the velocity of 30 cm s^{-1}, while the flow velocity at the entrance of the sampling tube was 15 cm s^{-1} for a flow rate of 0.7 ℓ min^{-1}. In such a condition, the sampling efficiency should be larger for larger particles. However, it was figured out that the efficiency for the largest droplet (about 12 μm) observed in this test experiment was only 6% higher than unity, and accordingly the anisokinetic effect was not significant in comparison with the random fluctuation in the droplet concentration.

It is appropriate to insert a few notes here about the gravitational settling of droplets during the sampling. For a droplet with a diameter of, say, 10 μm, the settling velocity is 0.3 cm s^{-1}; on the other hand, the flow velocity is calculated, on the basis of the flow rate and the size of the sampling path, to be 15 cm s^{-1} and 15 m s^{-1}, respectively, at the sampling tube entrance and at the nozzle exit, when the flow rate is 0.7 ℓ min^{-1}. From these figures, the loss in the number of droplets is estimated to be 0.7%. For a 20-μm droplet, the loss comes out to be 5%. It is not difficult to prepare a correction curve as a function of droplet diameter. For the particular results obtained in this study (Figure 4, and Figure 10 to be shown below), however, the correction was found to be hardly significant.

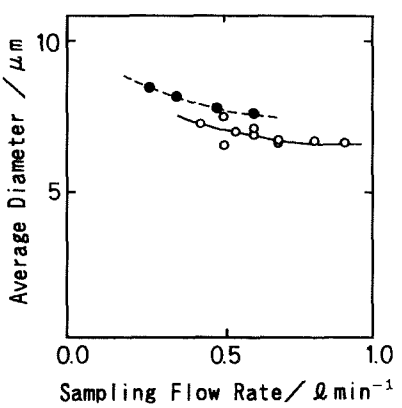

FIGURE 5. Dependence of the average diameter on the sampling flow rate. ●——●: sample with the liquid water content = 4.5 g m^{-3}, ○——○: sample with the liquid water content = 0.5 g m^{-3}.

Test on Accuracy

As mentioned in the Introduction, the conservation of droplet sizes during the measurement is a crucial point that influences the accuracy. In the present device, the sampled air is blown against the sensor through a very short path, and the sampling tube is wholly placed in the ambient air. Therefore it is expected that little change is caused in the temperature and humidity of

the air, and consequently the size of the droplets is conserved. In order to check the accuracy, two test measurements were carried out in which the droplet sizes obtained by the present method were compared with those from other methods based on entirely different principles.

(1) A glass plate was set on the specimen stage of an optical microscope, after coating it with a 1 : 1 mixture of white Vaseline and liquid paraffin. The hot-film sampling tube was placed close to the glass plate. Water droplets were blown against the plate from the ultrasonic nebulizer, and photographs of droplets collected onto it were taken simultaneously with the hot-film operation. Looking through the microscope, it was observed that shrinkage due to evaporation of the collected droplets became recognizable after the lapse of several tens of seconds. Therefore the photographing was finished within 30 s after the start of the collection. Then diameters of the collected droplets were measured geometrically on the photograph. The size distribution derived in this way is shown in Figure 4(b) in comparison with the distribution obtained by the hot-film sensor. There resulted good agreement between the two observations.

(2) Test equipment was constructed in which the measurement of the gravitational settling velocity and the size measurement by the hot-film were done for one and the same droplet. Figure 6 illustrates such equipment. Its essential part is a vertically placed glass cylinder, which is 700 mm in height and 50 mm in diameter; the cylinder has two horizontal collinear side tubes near the bottom. Water droplets from a vibrating orifice aerosol generator (Kanomax Model 3050) were led to the top of the cylinder, and they were left to settle by free gravitation. A light beam from a He–Ne laser passed across the cylinder through the side tubes, and the hot-film sensor was placed at 5 mm below the light beam. Under these arrangements, part of the falling droplets crossed the laser beam and scattered the light, and then collided with the sensor filament after a period dependent on the settling velocity of the droplet. The scattered light intensity was monitored by a

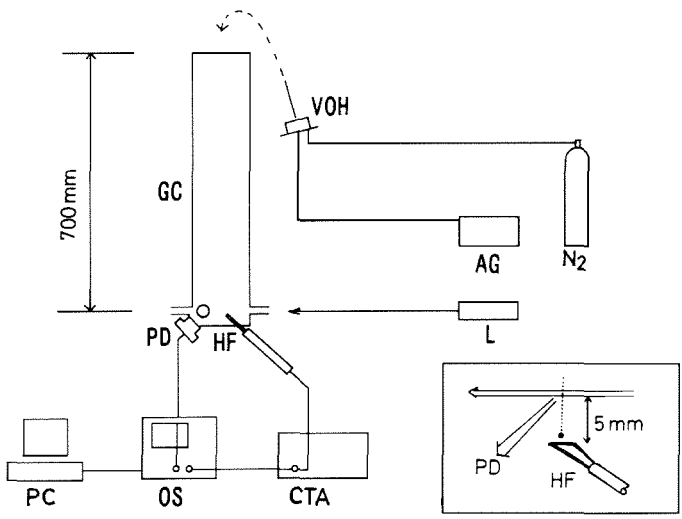

FIGURE 6. Test equipment for the terminal settling velocity measurement. GC: glass cylinder, AG: aerosol generator, VOH: vibrating orifice head (taken off from the aerosol generator and placed close to the top of the glass cylinder), N_2: nitrogen gas container for the carrier gas flow, L: laser, HF: hot-film sensor, PD: photodiode, CTA: constant temperature anemometer unit, OS: oscilloscope, PC: personal computer; insert below shows the configuration of the incident light beam, hot-film sensor, a falling droplet and light scattered to the photodiode.

photodiode, and its output signal was fed into one of the two channels of the digital oscilloscope; the other channel received the signal from the hot-film sensor. Figure 7 shows two examples of the oscilloscope record, which enabled one to determine the time required for a droplet to reach the sensor filament after it crossed the light beam. The distance between the light beam and the filament was measured precisely by a cathetometer. Therefore, it was possible to calculate the settling velocity, and then to convert the velocity into the diameter of each droplet. In this conversion, the deviation from the Stokes' law was taken into account by using the terminal velocity–size relationship reported by Beard and Pruppacher (1969). The diameter thus obtained is plotted in Figure 8 against that derived from the hot-film sensor in the range from 30 to 100 μm. The diameters determined by the two different methods were found to agree within the uncertainty of the experiment.

As a result of these test experiments, it was proved that the present method based on the heat of vaporization measurement could give accurate size of droplets without any calibration procedure.

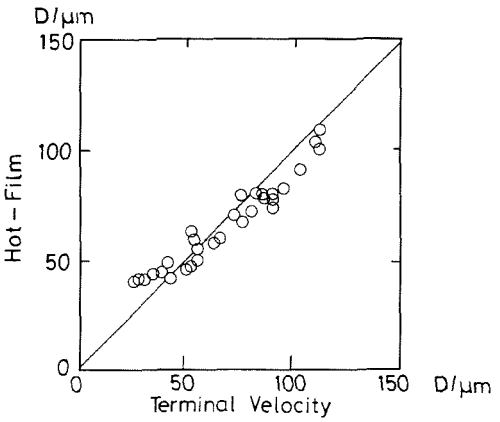

FIGURE 8. Comparison of the droplet diameter, D, obtained from the hot-film with that from the terminal settling velocity measurement.

CONCENTRATION MEASUREMENT

Besides the size distribution measurement, the present method can be used for concentration measurements as well, if the detection efficiency of the droplets coming into the sampling tube is evaluated in advance. In this study the efficiency was determined by a gravimetric method in the following way. A mist trap, which was a small cylinder filled with glass beads, was inserted between the sampling tube and the pump to collect all the droplets passing through the tube. After an operation, say, for an hour, the mass increase, ΔM, of the trap was determined along with the size distribution function $f(D)$. By placing another hot-film sensor between the trap and the pump, it was confirmed that no droplet escaped from the trap. Water droplets were generated in large excess of those to be sampled. Therefore the relative humidity (RH) of the air was maintained at 100%, and accordingly there was expected no evaporation loss in the trap. This was confirmed by the following check procedure: after collecting droplets for some time and weighing the trap, a subsidiary mist trap was inserted between the nebulizer and the main trap, and the sampling was continued

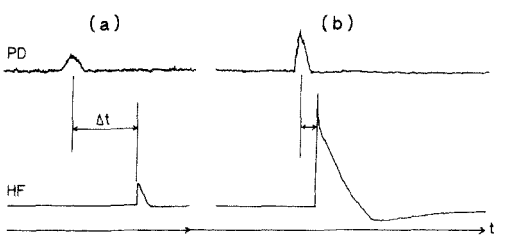

FIGURE 7. Signals from the test equipment. The upper curves show the scattered light signals from the photodiode, while the pulses from the hot-film sensor are shown on the lower curves; Δt is the time of transit of a droplet from the light beam to the sensor filament. (a) and (b) are the signals from the smaller and the larger droplets, respectively.

for some additional time. The subsidiary trap removed all the droplets but kept the relative humidity of the air at 100%. Subsequently the main trap was weighed again and little change was found. This procedure simultaneously showed that there was no condensation gain either. The loss in the connecting line must be negligible, since it was only a few cm in length.

Then the detection efficiency, E, was calculated by

$$E = N\int f(D)(\pi D^3/6)\rho\, dD/\Delta M, \qquad (5)$$

where N is the total number of detected droplets. For the original data acquisition system with the digital oscilloscope, E was found to be 0.005 at a flow rate of 0.7 ℓ min^{-1}, whereas $E \sim 0.03$ for the improved system with the AD converter. Roughly speaking, E is given by a product of three factors, i.e., the fraction of the sampling period, the sampling efficiency, and the collision efficiency of the droplet with the sensor. As stated before, the fraction of the sampling period is 0.0056, and 0.033 with the original and the improved system, respectively, while the sampling efficiency was calculated, by Zhang and Liu's formula, to be 1.02 as averaged over the size range of the droplets from the nebulizer. Therefore the above values of E mean that the collision efficiency is close to unity.

Once the efficiency E is determined for a particular sampling tube, the mass and number concentration of the droplets are obtained by the following formulas:

$$C_m = N\int f(D)(\pi D^3/6)\rho\, dD/(EQ_sT_s), \qquad (6)$$

$$C_n = N/(EQ_sT_s). \qquad (7)$$

Here Q_s and T_s are the sample air flow rate and the sampling period, respectively, and hence Q_sT_s is the volume of air that has passed through the tube.

APPLICATIONS

A measurement on ethanol droplets was tried to check the response of the sensor against a liquid that has a heat of vaporization considerably smaller than that of water. It was found that the height of pulses was much the same as in the water case, but pulse width was smaller. Size distributions of ethanol and water droplets from the nebulizer were very similar.

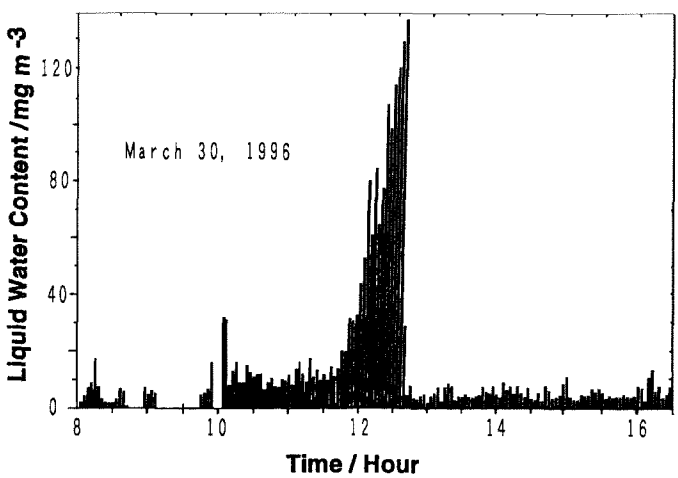

FIGURE 9. Variation of the liquid water content observed in a fog event, March 30, 1996, at Tsukuba.

One of the strong points of the present system is its portability, which makes it suitable for field observations. The sampling tube (Figure 1) is so small that it can be placed at nearly any position. Other instruments, the CTA unit, a digital oscilloscope, a computer and a pump, need not be placed close to the sampling point; they can be separated by more than 10 m to a place most convenient for the operation. Taking advantage of this feature, a field test was carried out for fogs formed around our institute. In this test observation, the sampling tube was mounted at the end of a 1.5-m rod stretched out from a 2nd-floor window of a building. With the improved data acquisition system, the signals were accumulated for 60 cycles, or in other words, the size distribution and the liquid water content were obtained every 3 min. Figure 9 shows the liquid water content observed in this way; thus variations of the fog density were tracked in fair detail. Figure 10 is the number-based size distribution of the droplets detected in the entire observation. During the observation, the wind velocity was measured but the wind direction was not. Accordingly, the correction for the anisokinetic effect was done only tentatively on the basis of the average wind velocity, 0.8 m s^{-1}, with an assumption that the wind direction was constant. Undoubtedly more detailed recording of the wind condition was required to obtain an accurate size distribution, since the correction was nearly 200% for 20 μm droplets.

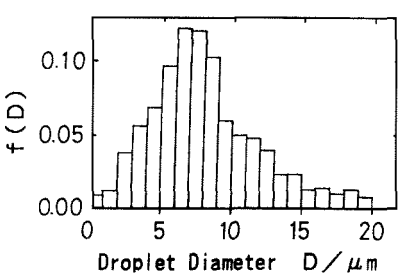

FIGURE 10. Number-based size distribution, f(D), of fog droplets; f(D) is normalized by $\int f(D)\, dD = 1$.

SUMMARY AND ADDITIONAL REMARKS

By applying a hot-film sensor of a constant temperature anemometer, a device has been developed for the size measurement of volatile liquid droplets. Its capability is as follows:

(1) The changeability of droplet size during the measurement is averted by short-path sampling through a compact tube placed in the midst of the sample air. The accuracy of the present method has been proved by parallel measurements based on different principles.

(2) Absolute measurement is possible as long as one is aware of the liquid properties: the diameter is directly obtained from the pulse area and the heat data without any calibration procedure.

(3) The lower limit of the size measurement depends on the background noise due to the turbulence of the air flow. With a flow rate of 0.7 ℓ min^{-1}, a droplet as small as 2 μm was detected. On the other hand, the largest measurable diameter is probably limited by the condition that the evaporation of a droplet should be completed quickly enough to produce a single voltage peak. The hot-film sensor itself was found to have the capability of sizing a droplet with a diameter up to 100 μm in the case of water. Needless to say, however, the measurement of such a large droplet requires great care to the anisokinetic sampling effect as well as to the gravitational settling and the inertial deposition during the sampling.

(4) The number concentration of droplets can be measured by the present device, if the detection efficiency of droplets is evaluated.

(5) When the number concentration of droplets is, for example, on the order of 10^8 m^{-3}, the size distribution can be obtained for every 3 min. Thus the device can measure a fog event with nearly real-time response.

(6) It is useful for sizing droplets, not only of water, but also of other volatile liq-

uids; it was shown to be applicable to ethanol, and the experience of this study suggests the applicability to any liquid whose boiling point is below 200°C.

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