# The Behavior of Large, Low-Surface-Tension Water Drops Falling at Terminal Velocity in Air

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ABSTRACT

A vertical wind tunnel was constructed to study the behavior of large, low-surface-tension drops in free fall. The tunnel is simple, but provides a low turbulence (0.7%) flow which stably supports large water drops falling at terminal velocity. The influence of reduced surface tension on maximum drop size, drop terminal velocity, and drop shape was investigated. It was found that drops of low surface tension break up at a smaller size than drops with normal surface tension, are more deformed than drops of equal mass having normal surface tension, and have a lower terminal velocity than drops of equal mass and normal surface tension. Drops only partially coated with surfactant cannot be stably supported and undergo violent oscillations. Before any field testing of possible cloud modification by reducing rainwater surface tension is warranted, further investigation of the behavior of low-surface-tension drops should be undertaken and, in particular, the behavior of drops only partially coated with surfactant should be studied.

## 1. Introduction

For more than 50 years, the behavior of large water drops in free fall has been examined. To study the terminal velocity, shape, evaporation, circulation and breakup of raindrop-sized water drops, investigators have used a variety of vertical wind tunnels and drop towers or drop tubes (Lenard, 1904; Schmidt, 1909; Flower, 1928; Laws, 1941; Blanchard, 1949; Gunn and Kinzer, 1949; Fournier d'Able and Hidayetulla, 1955; Magarvey and Taylor, 1956; Komabayasi *et al.*, 1964; Cotton and Gokhale, 1967; Pruppacher and Beard, 1970).

However, of all the studies, only a few included experiments to study the effect changes in surface tension have on the various water drop properties. Blanchard (1949) observed low-surface-tension water drops in a wind tunnel and found these drops to be more deformed than drops with normal surface tension, but concluded that slight changes in raindrop surface tension would not affect the raindrop behavior. Kachurin and Morachevski (1965) cited a relation between maximum drop diameter ($D_{\max}$) before drop breakup and surface tension ($\sigma$) of the form

$$D_{\max}=K\sigma^{\frac{1}{2}}, \tag{1}$$

but gave no experimental details.

Interest in the behavior of low-surface-tension drops is largely a result of the hypothesis that precipitation processes might be modified by reducing the rainwater surface tension. Morachevski and Kiriukhin (1968) claimed success in modifying the so-called "chain reaction" process postulated by Langmuir (1948) and claimed the behavior of developing convective clouds was changed by the introduction of surfactants (surface tension reducing chemicals). Ryan (1970) suggested that changes in rainwater surface tension would change the raindrop size distribution and increase the sweeping action of rain.

Since there are surfactants now available which are capable of reducing water surface tension by a factor of 4 with surfactant concentrations of only 10 ppm, it is conceivable that a significant reduction in rainwater surface tension could be obtained for a short time interval.

To quantitatively investigate the behavior of low-surface-tension drops, a vertical wind tunnel was constructed in which large water drops could be suspended in free fall and studied.

## 2. Experimental equipment

The vertical wind tunnel constructed is shown in Fig. 1. The tunnel is open ended (the air is not recirculated) and is similar to the tunnel described by Pruppacher and Neiburger (1968) in that the air is sucked rather than blown through the tunnel. While this tunnel is much less sophisticated than the UCLA tunnel, low-turbulence flow suitable for the study of large drops was successfully attained.

The entrance area is a 60 cm diameter inflated rubber truck tire tube which presents a smooth curved entrance to the tunnel to minimize the generation of eddies as air flows into the tunnel.

The honeycomb section is a 30 cm deep, 3 mm diameter cell, expanded aluminum piece made by Hexcel Corporation. The section is deeper than is necessary to assure smooth flow at the honeycomb exit.

The contraction section is constructed of spun aluminum with a contraction ratio of 20:1. Fine-mesh screens at the contraction section entrance insure smooth flow into this section of the tunnel. The cross-sectional profile of the contraction section is similar to contraction sections used in very low turbulence aerodynamic wind tunnels, and in hindsight a cross section specifically designed for this tunnel and velocity range might have improved performance somewhat.

Maybank and Briosi (1961) and Pruppacher and Neiburger (1968) used a tapered plexiglas section as an observation section; we chose a similar design to provide a condition of vertical stability for the large free floating drops we wished to study. The observation section has a 3° taper and various taps for hot film probes, pilot tubes, drop injection, and a photographic port.

The air pump is a relatively simple squirrel cage fan which is vibrationally isolated from the rest of the tunnel.

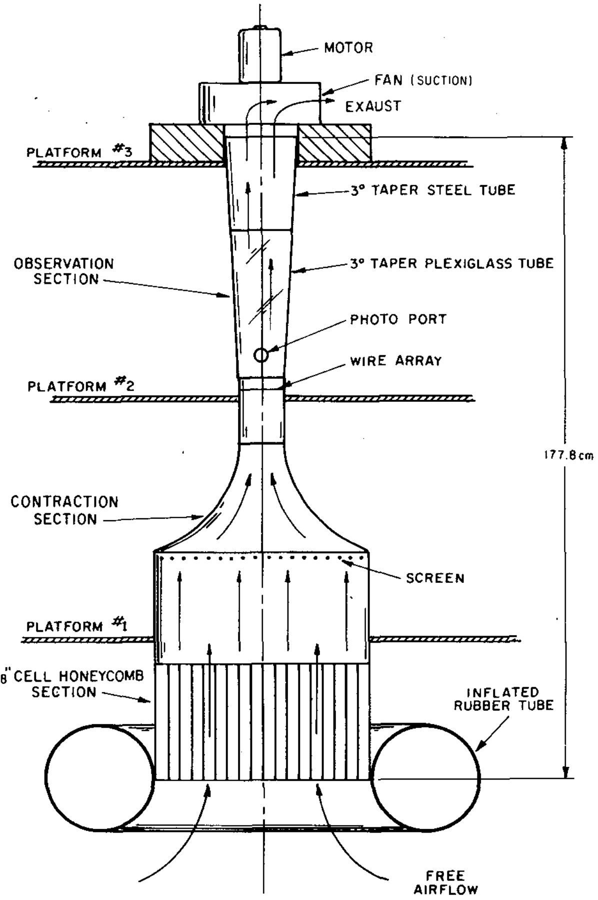


FIG. 1a. Schematic diagram of vertical wind tunnel.

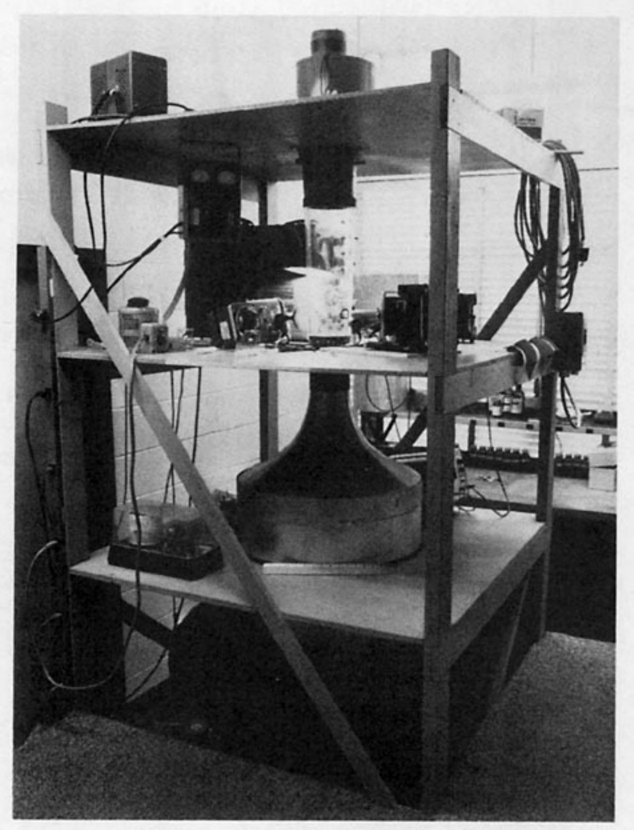


FIG. 1b. Photograph of vertical wind tunnel.

Velocity measurements were carried out using a Hill miniature Pitot tube and micromanometer. Turbulence measurements, and some velocity measurements, were carried out with a Thermo-Systems Inc. hot-film probe and anemometer power supply.

The tunnel velocities of the experiments were in the range 5–10 m $s^{-1}$. The turbulence level of the air stream was relatively constant over the range of velocities and was less than 0.7%.

Large drops injected into the tunnel would quickly drift from the center of the tunnel and impact the walls. To prevent this, it was necessary to develop a position of horizontal stability in the tunnel. Blanchard (1948) found this can be done by forming a velocity dip or minimum in the velocity profile. Previous investigators have formed this velocity dip by inserting screens, shaped inserts or rods in the tunnel below the observation section. However, all such devices generate turbulence in the air stream and defeat the purpose of a low-turbulence tunnel.

Water drops were injected into the tunnel through long hypodermic needles and, on occasion, it was noticed that the injected drop would be captured in the needle wake and be stably supported for short intervals. Further experiments revealed that over the velocity range of interest, wire as thin as 25 μm would develop a wake which provided a stable drop position, and yet no induced turbulence could be detected down-

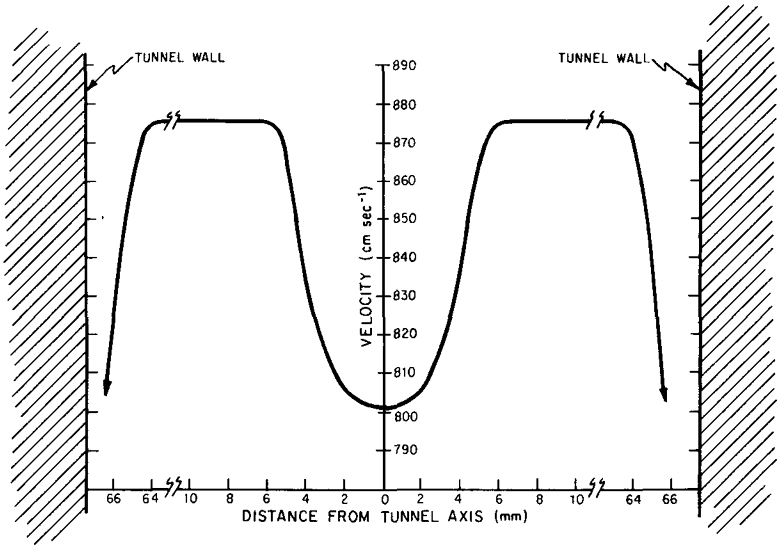


Fig. 2. Wind tunnel velocity profile.

stream of the wire. After much experimentation, a wire array was developed which formed a non-turbulent velocity dip to provide a position of horizontal stability for drops in the range 3–14 mm horizontal dimension. Three 50 $\mu$m diameter wires support two wire rings of 50 $\mu$m diameter wire. The large ring is 7.5 mm in diameter and the smaller ring 4.5 mm. The turbulence level at the observation area above the wires is no greater than in the tunnel free stream ($<0.7\%$). The velocity profile at a typical tunnel operating velocity is shown in Fig. 2. Some qualitative experiments indicated that drops in the range 1–3 mm diameter could be stably supported in the dip formed by the coincidence of the three support wires with the concentric rings removed. The wire array was very fragile so that great care was necessary in properly aligning the array and maintaining tension on the support wires. Any relaxation in the wire tension allowed the wire to vibrate in the air stream and introduced turbulence in the observation section which prevented stable support of the drops.

A small flat glass photographic window was inserted in the curved plexiglas observation section. Drop photographs were made using a special optical arrangement and Polaroid type 52 film with a Polaroid holder in a Crown Graphic camera body. With this system, it was possible to ensure that a useful drop photograph was obtained while the drop was still supported in the tunnel.

It was planned to measure the tunnel velocity to determine the terminal velocity of stably supported drops. To investigate the influence the tunnel wall effects had on the velocity of the supported drops, we followed a procedure similar to that of Beard (1970). The water drops we studied were in the size range of 2–9 mm equivalent spherical diameter with corresponding Reynolds numbers in the range 860–5400. The tunnel diameter at the measurement level was 135 mm so that the ratio $A/R$ (where $A$ is the drop radius and $R$ the tunnel radius) is 0.1 for the largest drops studied. Fidleris and Whitmore (1961) showed that for spheres falling in cylindrical vessels, the retarding effect of the wall was less than 1% for an $A/R$ ratio of 0.1 and a Reynolds number exceeding 30. Most of the experiments were done with drops where $A/R<0.06$ and thus it was concluded that errors in the drop terminal velocity due to the tunnel wall effects were less than 1%.

## 3. Experiments

Studies with the wind tunnel were made on the effect surface tension has on 1) maximum drop size, 2) drop terminal velocity, and 3) drop shape.

### *a. Surfactants*

The surfactants we chose for the experiments were 3M Company fluorochemical surfactants FC-98, FC-126, FC-128 and FC-170. Surfactants FC-128 and 170 were used to obtain very low surface tensions ($\sim$20 dyn cm$^{-1}$), while FC-98 and 126 were used for intermediate surface tensions (30–50 dyn cm$^{-1}$). Chemically pure distilled deionized water was used in experiments with water at normal surface tension and also in stock solutions of water with various surfactant concentrations. The surface tension of the stock solutions was measured using a Du Nouy tensiometer. During these measurements, it was noted that the surface tension of a slightly agitated solution would be 10–20% more than the surface tension of a solution which had been allowed to set for some time. It was concluded that the surface concentration of surfactant was increasing with time (as might be expected due to the molecular structure of the chemical), and thus the measured surface tension was decreasing with time. The greatest variation was

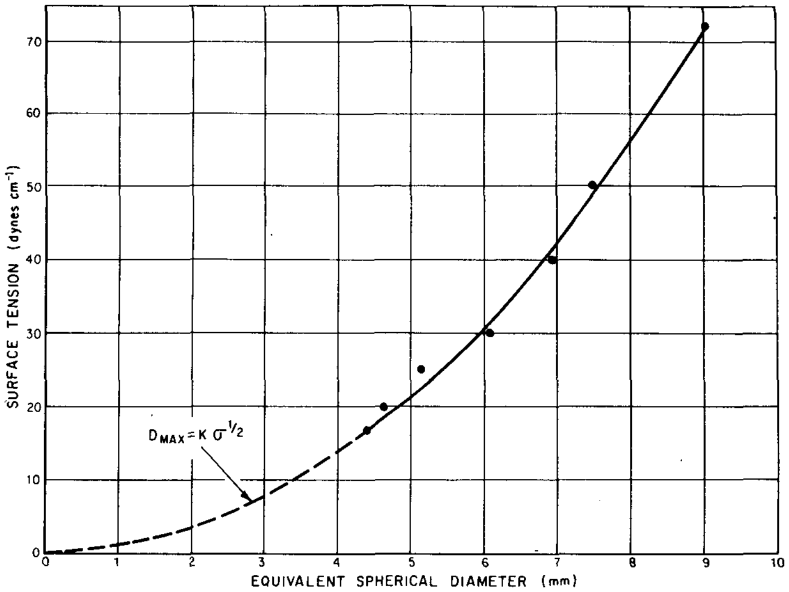


FIG. 3. Maximum water drop size at breakup vs surface tension.

found in solutions of intermediate surface tension. The drops supported in the wind tunnel are in agitation because of the drop oscillations and internal circulations. Therefore, the stock solution surface tension was recorded after slight agitation. The solutions were also slightly agitated before withdrawing any solution into hypodermic syringes for injection into the tunnel.

*b. Maximum drop size*

To form the largest free floating drop of a particular water surface tension, the slightly agitated stock solution was injected through a long hypodermic needle into the wind tunnel dip to form a drop which was stable and free floating. Then a very fine hypodermic needle was used to carefully inject small drops ($<1$ mm diameter) of the solution into the wake of the large drop at a downstream distance of usually no more than several millimeters. The drops gently coalesced and the drop size increased. With experience, one could tell when the maximum drop size had been approached. Each time a droplet was added, a drop photo was taken until such time as the drop broke up from instability. It appears that the drop coalescence was gentle enough that the drop size was limited by instability rather than breakup because of drop collisions.

The maximum drop size was determined by overlaying a calibrated grid on the drop photo and calculating the drop volume and equivalent spherical radius ($R_0$) by a grid slice method at vertical intervals of 0.5 mm. There is some error involved in this technique in that just before breakup, the drops are oscillating and one cannot be sure that the drops photographed are radially symmetrical. Many drop photos were obtained for each surfactant solution used, but only those photos were chosen for graphic size determination in

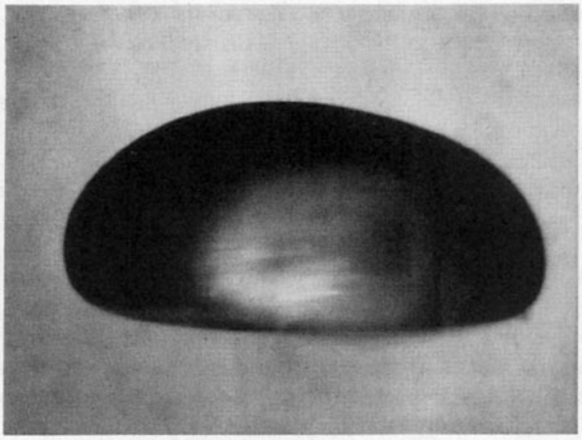


FIG. 4. Drop shape for $\sigma = 72$ dyn cm$^{-1}$ ($R_0 = 4.52$ mm).

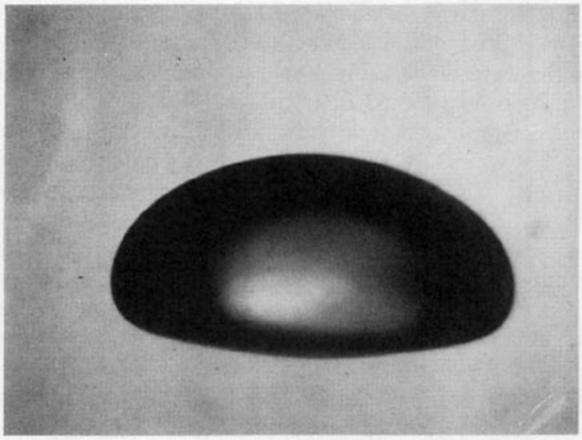


FIG. 5. Drop shape for $\sigma = 50$ dyn cm$^{-1}$ ($R_0 = 3.74$ mm).

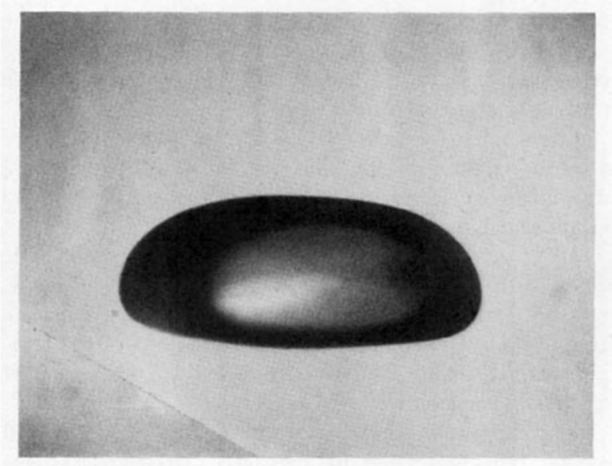


Fig. 6. Drop shape for $\sigma=33$ dyn cm$^{-1}$ ($R_0=3.12$ mm).

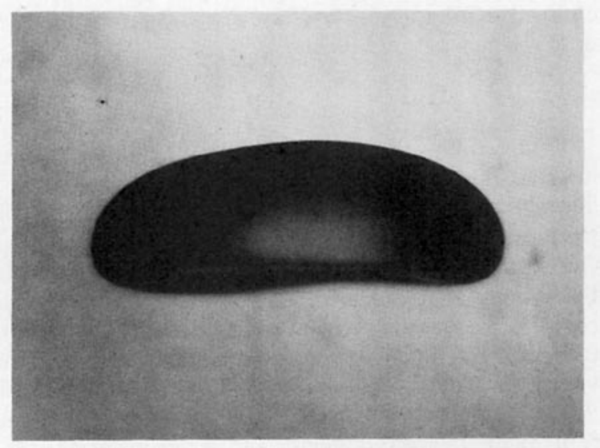


Fig. 8. Low-surface-tension drop with a concave base: $\sigma=20$ dyn cm$^{-1}$.

which the image of the backlight source was symmetrical; therefore, one could be confident that the drop was reasonably symmetrical and the technique as accurate as possible. However, it would have been possible to remove the drop from the tunnel and weigh it after the photo had been taken and this technique might have proved more accurate than determining drop size graphically.

The maximum equivalent spherical drop diameter as a function of surface tension is shown in Fig. 3 and representative photos of maximum size stable drops are shown in Figs. 4–7. Notice that the bases of the drops are distorted and, in some cases, the bases were dramatically concave. This has been reported and discussed elsewhere (Koenig, 1965; Pruppacher and Pitter, 1971) and can be clearly seen in Figs. 7 and 8. Fig. 8 is a drop photo with an oblique base view of a large, very distorted, low-surface-tension drop.

The experiments were done with several different surfactants where the surfactant concentration was relatively high (>5000 ppm) and each data point in Fig. 3 represents the average value of the maximum drop size for one particular surfactant and surface tension.

The maximum stable drop size for distilled water drops ($\sigma=72$ dyn cm$^{-1}$) is 9.1 mm diameter and agrees well with previous observations and theoretical calculations of Pruppacher and Pitter (1971).

The data curve of Fig. 3 agrees with Eq. (1). The dashed portion of the curve is the extension of $D_{\max}=K\sigma^{\frac{1}{2}}$ below our experimental data. The value of $K$ determined from the experimental data is 0.109 cm$^{\frac{3}{2}}$ dyn$^{-\frac{1}{2}}$.

A quantitative study of the actual drop breakup was not made, but some observations are interesting. At times, the breakup was observed to be of the violent "bag mode" type, but, at other times, breakup was observed to be a relatively nonviolent action. As the maximum drop size was approached, the drop oscillation would increase and finally the drop would take on a dumbell shape, the neck would rupture, and two approximately equal sized drops would be left to drift away and impact the tunnel wall. Fig. 9 shows the

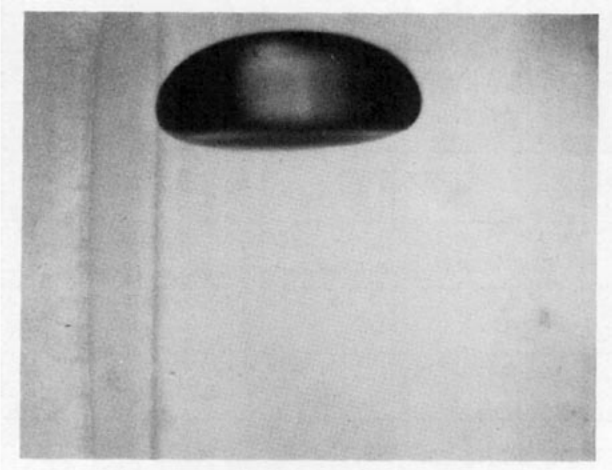


Fig. 7. Drop shape for $\sigma=17$ dyn cm$^{-1}$ ($R_0=2.23$ mm).

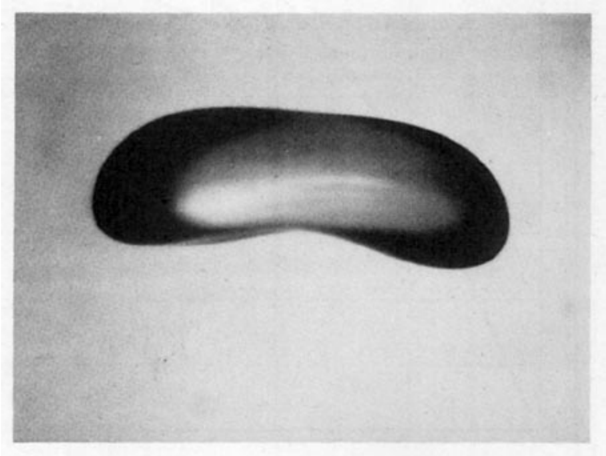


Fig. 9. Highly distorted low-surface-tension drop seconds before breakup.

dumbell shape and concave base of a low-surface-tension drop seconds before breakup.

### *c. Terminal velocity*

To determine the terminal velocity, the drops were injected through a hypodermic needle into the tunnel dip as in the previous experiment. Needles of varying thickness were used to control the injected drop size. Once the drop was at a stable shape and position in the tunnel, a drop photograph was taken, and, if acceptable, the drop was removed and the miniature Pitot tube and micromanometer were used to measure the velocity at the point the drop had been photographed.

These measurements were made for drops of 2–8 mm equivalent spherical diameter and for drop surface tensions of 17, 33, 50, and 72 dyn $\mathrm{cm}^{-1}$. The family of curves in Fig. 10 summarizes the results of these experiments.

There is an error in measuring the tunnel velocity and equating it with the drop terminal velocity. The drop is stabilized in the velocity dip in the tunnel and it is the velocity at the center of the dip which is measured. Because of the shape of the dip and the size of the drop, the actual drop terminal velocity is some value between the minimum at the dip and the maximum at the free stream beyond the dip. To correct for this error, correction factors were determined for the measured velocity which take into account the geometry of the velocity dip and the drop cross section. The flow is assumed to be incompressible and the actual terminal velocity is equal to the mean velocity of the portion of velocity dip beneath the supported drop cross section. In all cases, differences between the measured and corrected velocities are less than 6%.

The results obtained by Gunn and Kinzer (1949) are also shown in Fig. 10. The present results for $\sigma = 72$ dyn $\mathrm{cm}^{-1}$ are lower by 2–4% than those obtained by Gunn and Kinzer.

An effort was made to conduct all experiments at 20°C, 1013 mb and 50% relative humidity, but this was not possible because of day-to-day changes in the tunnel room environment. These variations in the temperature, pressure and humidity led to variations of about 1% in the measured velocity.

The experimental data for the curves of $\sigma = 72$ and $\sigma = 17$ dyn $\mathrm{cm}^{-1}$ show little scatter. However, we encountered difficulty in obtaining consistent experimental data for drops of the same size for $\sigma = 33$ and $\sigma = 50$ dyn $\mathrm{cm}^{-1}$. The surfactants used with these intermediate surface tensions were not in the high concentrations of the surfactant used in the experiments at $\sigma = 17$ dyn $\mathrm{cm}^{-1}$, and, as discussed previously, time variations were noted in the measured surface tension. These variations in the surface tension probably accounted for the variation in our experimental data.

It is felt the velocity data for $\sigma = 17$ and $\sigma = 72$ dyn $\mathrm{cm}^{-1}$ are correct within 5%. However, because of the errors mentioned, the curves for $\sigma = 33$ and $\sigma = 50$ dyn $\mathrm{cm}^{-1}$ may be only within 10% of the true terminal velocity.

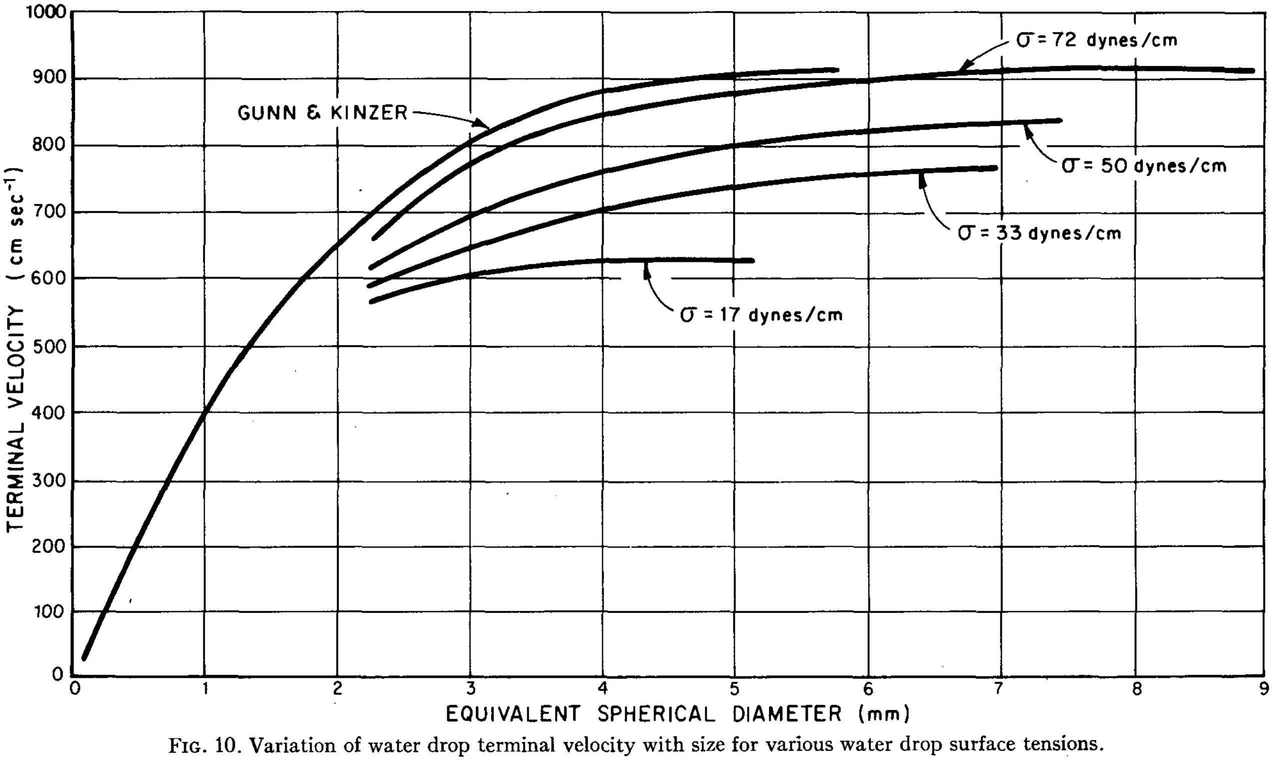


FIG. 10. Variation of water drop terminal velocity with size for various water drop surface tensions.

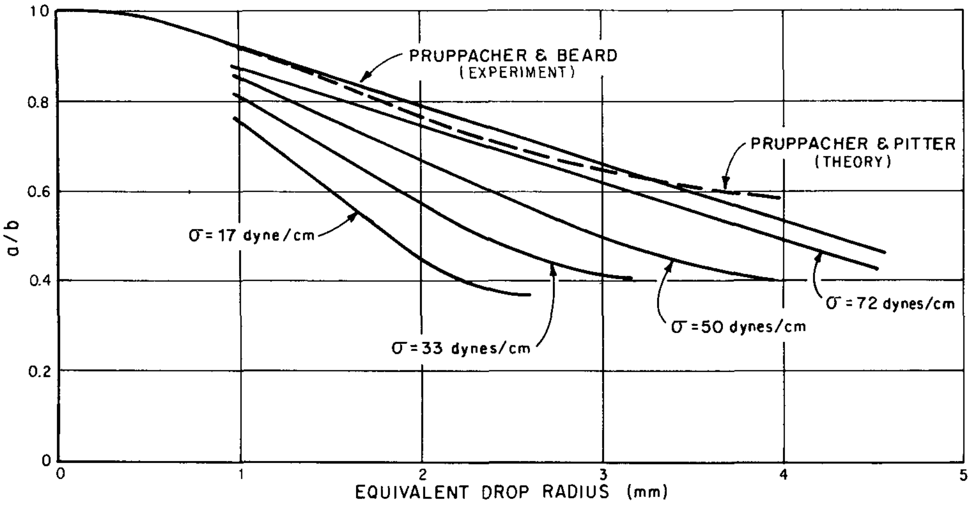


FIG. 11. Drop deformation vs size for various water drop surface tensions.

*d. Drop shape*

The drop photographs that were taken during the velocity experiments were analyzed to examine the effect that surface tension had on drop shape as a function of drop size. Following previous convention, the ratio of the drop minor axis dimension to major axis dimension ($a/b$) is plotted in Fig. 11 as a function of equivalent spherical radius for drop surface tensions of 17, 33, 50 and 72 dyn cm$^{-1}$. The experimental results of Pruppacher and Beard (1970) for $\sigma = 72$ and a curve of modeled drop deformation (Pruppacher and Pitter, 1971) are also shown.

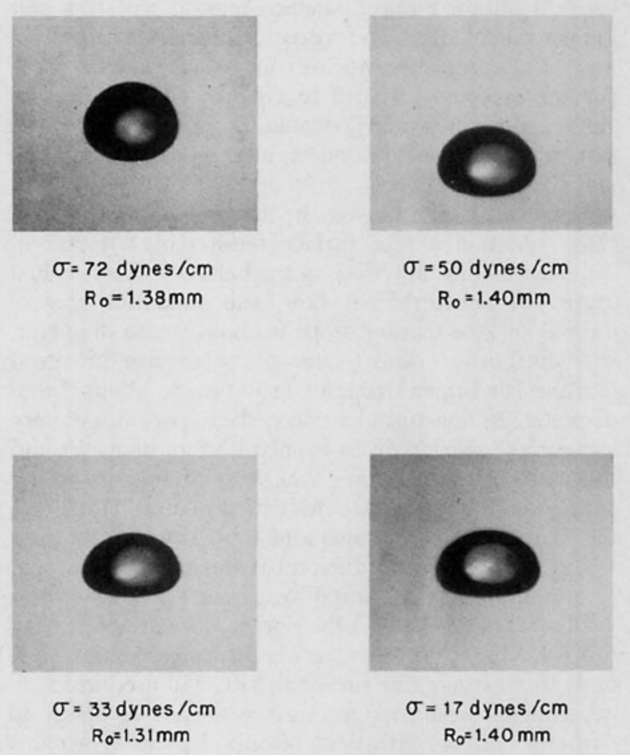
Photographs of drops of equivalent spherical diameter of about 3 mm and different surface tensions are shown in Fig. 12. As in the previous section, data for $\sigma = 72$ and $\sigma = 17$ dyn cm$^{-1}$ are consistent. Because of a variability in surface tension, there is a significant scatter in the data for $\sigma = 33$ and 50 dyn cm$^{-1}$. Because of this scatter, the curves for these latter surface tensions may be in error by as much as 10%.

## 4. Discussion of the results and observations

The experiments show the behavior of low-surface-tension drops to be dramatically different than drops of normal surface tension. As the drop surface tension decreases, the drop deformation increases, the drop falls slower, and breaks up at a smaller size than the normal surface tension drop.

Pruppacher and Pitter (1971) have discussed the development of concave bases in water drops of normal surface tension as being a necessary condition for drop breakup. It is observed that this concave depression develops in low-surface-tension drops at a much smaller size than in drops with normal surface tension. It appears that the development of this concave feature is related to drop deformation. That is, when the deformation ratio is about 0.75 the base becomes concave. The reduction in drop surface tension increases the drop deformation, the concave feature develops at a smaller drop size, and the concavity enlarges so that the critical size for breakup discussed by Pruppacher and Pitter (1971) is reached at a much smaller size with drops of low surface tension.

The greater deformation with decreasing surface tension seen in Fig. 11 increases the drag on water drops of the same mass but different surface tension. The increased drag decreases the terminal velocity as seen



$\sigma = 72$ dynes/cm
$R_0 = 1.38$ mm

$\sigma = 50$ dynes/cm
$R_0 = 1.40$ mm

$\sigma = 33$ dynes/cm
$R_0 = 1.31$ mm

$\sigma = 17$ dynes/cm
$R_0 = 1.40$ mm

FIG. 12. Variation in drop shape with surface tension for equivalent spherical diameter drops of about 3 mm.

in Fig. 10. The curves in Fig. 10 are converging at small drop sizes and, extrapolating the curves, we find the convergence occurs at about 0.5 mm radius. The smaller the drop, the less the influence of low surface tension and, at sizes smaller than about 0.5 mm radius, the drops are essentially spherical and one would expect the terminal velocities to be equal and independent of surface tension. The curves of Fig. 11 can also be seen to converge which also indicates that drops smaller than about 0.5 mm radius are essentially spherical regardless of surface tension.

Fig. 11 indicates there is a limit to drop deformation. While there is scatter in the data, we do see indications of a change in the slope of the low-surface-tension curves as the limiting drop size is approached. This limiting deformation value is about 0.4.

Drops having diameters of 6–9 mm, such as have been studied in the present experiments, have never been observed in the atmosphere. The largest drops observed in nature are of the order of 5 mm diameter. Thus, the current experiments lend further credence to the now generally accepted belief that raindrop size is limited by breakup caused by drop interaction rather than breakup from individual drop instability (Cotton and Gokhale, 1967; List *et al.*, 1970).

List *et al.* (1970) studied the temporary collisions of drops with lowered surface tension ($\sigma = 52$ dyn cm$^{-1}$) and concluded the reduction in surface tension had no effect on the collision and breakup of the pairs of 2–4 mm diameter drops used in the experiments. Milk was used to obtain reduced surface tension and it is not known how density and viscosity changes in the fluid may have contributed to the conclusions; however, the current experiments tend to confirm that changes in surface tension by 30% would not necessarily cause significant changes in raindrop interaction and breakup and size distribution.

However, it can be seen in the present experiments that reduction of the surface tension by a factor of 3–4 has a significant effect on the behavior of individual drops. In non-turbulent flow, the maximum size of normal surface tension drops is about 9 mm diameter, with drop interactions (collision, coalescence, breakup) limiting the upper size limit in nature to about 5 mm diameter. In non-turbulent flow, the upper limit of very low surface tension drops is only 4.5 mm diameter and it is not illogical to assume that drop interactions would also lower the upper size limit in this case. Thus, it is felt that drop interactions and drop size distributions would be significantly different if the drop surface tension was lower than normal by a factor of 3–4.

Observations of the behavior of low-surface-tension drops having a very low surfactant concentration were most interesting. The surfactant FC-170 used in many experiments would reduce surface tension to about 40 dyn cm$^{-1}$ in concentrations of only 2 ppm. When this stock solution was agitated, however, and a drop injected into the tunnel, the drops underwent violent oscillations and could not be balanced for more than a few seconds; after this time they were thrown out of the velocity dip and impacted the tunnel wall. Drops of similar surface tension but having a higher surfactant concentration were not observed to exhibit such behavior.

It is concluded that, while the surface tension of the bulk solution was measured to be 40 dyn cm$^{-1}$ with the tensiometer, the surfactant was in such a low concentration that a drop suspended in the tunnel was not completely covered with the surfactant. Thus, because of the very low surfactant concentration, and the fact that internal drop circulations were constantly exposing fresh drop surface, a system had formed where part of the drop surface had a surface tension of 72 dyn cm$^{-1}$ and part a much lower surface tension, perhaps as low as 20 dyn cm$^{-1}$ where there was a film of the surfactant. This is a highly unstable system and violent behavior of the drop would be expected. This behavior was also noticed in relatively small drops (3 mm diameter) treated with very low concentrations of highly active surfactant.

### IMPLICATIONS

The hypothesis of using surfactants for cloud modification has been that it would be necessary to achieve a large bulk reduction in the cloud and rainwater surface tension to cause changes in the drop breakup, sweep-out, coalescence, etc. The results of these experiments suggest that introduction of only minute quantities of highly active surfactants on the water drop surface would dramatically affect the behavior of the drop because of the instability created. The resultant effects might be more dramatic than if surfactants were added in quantities large enough to lower the surface tension of the entire drop surface by a factor of 4.

## 5. Conclusions

The first extensive quantitative study of the behavior of low-surface-tension drops has been conducted. Dramatic changes in the maximum size shape and terminal velocity of low surface tension drops were observed.

The results of this study indicate that further study of the behavior of drops, only partially coated by surfactant, should prove most interesting and may have greater applications to the possible modification of precipitation processes than the study of uniform surface tension drops. Field experiments, using surfactants to test such modification hypotheses before laboratory and numerical experiments further explore and define what might be expected in field experiments, are unwarranted at this time.

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## REFERENCES

Beard, K. V., 1970: A wind tunnel investigation of the terminal velocities, collection kernels, and ventilation coefficients of water drops freely falling in air. Ph.D. dissertation, University of California, Los Angeles.

Blanchard, D. C., 1948: Observations of the behavior of water drops at terminal velocity in air. General Electric Res. Lab., Occas. Rept. No. 7, Project Cirrus, 13 pp.

——, 1949: Experiments with water drops and the interactions between them at terminal velocity in air. General Electric Res. Lab. Occas. Rept. No. 17, Project Cirrus, 29 pp.

Cotton, W. R., and N. R. Gokhale, 1967: Collision, coalescence and breakup of large water drops in a vertical wind tunnel. *J. Geophys. Res.*, **72**, 4041–4049.

Fidleris, V., and R. L. Whitmore, 1961: Experimental determination of the wall effect for spheres falling axially in cylindrical vessels. *Brit. J. Appl. Phys.*, **12**, 490–494.

Flower, W. D., 1928: The terminal velocity of drops. *Proc. Phys. Soc.*, **40**, 179–181.

Fournier d'Able, E. M., and M. S. Hidayetulla, 1955: The breakup of large water drops falling at terminal velocity in still air. *Quart. J. Roy. Meteor. Soc.*, **81**, 610–613.

Gunn, R., and G. D. Kinzer, 1949: The terminal velocity of fall for water drops in stagnant air. *J. Meteor.*, **6**, 243–248.

Kachurin, L. G., and V. C. Morachevski, 1965: Kinetics of phase transitions of water in the atmosphere. Leningrad, Izdatvo Universiteta. [Translated by Z. Lerman, The Israel Program for Scientific Translations, 1966.]

Koenig, R., 1965: Drop freezing through drop breakup. *J. Atmos. Sci.*, **22**, 448–451.

Kombayasi, M., T. Gonda and K. Kono, 1964: Lifetime of waterdrops before breaking and size distribution of fragment drops. *J. Meteor. Soc., Japan*, Ser. 2, **42**, 330–340.

Langmuir, I., 1948: The production of rain by a chain reaction at temperatures above freezing. *J. Meteor.*, **5**, 175–192.

Laws, J. O., 1941: Measurements of the fall-velocity of waterdrops and raindrops. *Trans. Amer. Geophys. Union*, **22**, 709–721.

Lenard, P., 1904: Ueber Regen. *Meteor. Z.*, **21**, 249–260.

List, R., C. F. MacNeil and J. D. McTaggart-Cowan, 1970: Laboratory investigations of temporary collisions of raindrops. *J. Geophys. Res.*, **75**, 7573–7580.

Magarvey, R. H., and B. W. Taylor, 1956: Free fall breakup of large drops. *J. Appl. Phys.*, **27**, 1129–1135.

Maybank, J., and G. K. Briosi, 1961: A vertical wind tunnel. Suffield Tech. Paper No. 202, Suffield Experimental Station, Ralston, Alberta, Canada, 8 pp.

Morachevski, V. C., and B. V. Kiriukhin, 1968: On the artificial control of the chain process of raindrop growth in clouds. *Proc. Intern. Conf. Cloud Phys.*, Toronto, 683–687.

Pruppacher, H. R., and M. Neiburger, 1968: The UCLA cloud tunnel. *Proc. Intern. Conf. Cloud Phys.*, Toronto, 389–392.

——, and K. V. Beard, 1970: A wind tunnel investigation of the internal circulation and shape of water drops falling at terminal velocity in air. *Quart. J. Roy. Meteor. Soc.*, **96**, 247–256.

——, and R. L. Pitter, 1971: A semi-empirical determination of the shape of cloud and raindrops. *J. Atmos. Sci.*, **28**, 86–94.

Ryan, R. T., 1970: The possible modification of convective systems by the use of surfactants. *Preprints Sec. Nat. Conf. Weather Modification*, 6–11 April, Santa Barbara, Amer. Meteor. Soc., 393–396.

Schmidt, W., 1909: Eine umittelbare Bestimmung der Fallgeschwindigheit von Regentropfer. *Meteor. Z.*, **26**, 183–184.