NOTES AND CORRESPONDENCE

Concentrations of Sea-Spray Droplets at Various Wind Velocities: Separating Productions through Bubble Bursting and Wind Tearing

JIN WU

Institute of Hydraulic and Ocean Engineering, National Cheng Kung University, Tainan, Taiwan, and Air–Sea Interaction Laboratory, Graduate College of Marine Studies, University of Delaware, Lewes, Delaware

20 May 1998 and 30 March 1999

ABSTRACT

There are two sets of comprehensive measurements of sea spray: de Leeuw and Smith et al. Their data are interpreted to describe similar productions of film and jet drops by bursting bubbles. For measurements of Smith et al., those droplets associated with the second peak at large sizes of the production spectrum are shown to be spume drops generated by the wind tearing of wave crests. Together with physical understanding, representations of field data are deduced for film and jet drops and are discussed for spume drops.

1. Introduction

Effort has been intensified toward evaluating the influence of sea spray on optical extinction (Gathman 1983), global warming (Sligo 1990), and air–sea heat exchanges (Andreas 1992). All of these depend critically on the parameterization of droplet concentrations under various wind velocities. As pointed out by Andreas et al. (1995), large discrepancies still exist among several commonly referred proposals. Moreover, most of these models were not verified with field measurements. Two sets of comprehensive oceanic data, however, are available, provided earlier by de Leeuw (1986) and recently by Smith et al. (1993). These data are compared to examine the uncertainty of understanding spray productions under various wind velocities. The results are very encouraging. Through the synthesis of these two sets of measurements, production spectra of film and jet drops by the bursting of bubbles are deduced, and generation functions of spume drops by the wind tearing are further explored.

2. Measurements

a. de Leeuw

Spray particles were measured by de Leeuw (1986) using an impaction method. Two groups of samplers were used near the Station Lima in the Atlantic Ocean: one group was mounted on decks of the MS *Cumulus* at elevations of 4, 6, and 11 m above the sea surface and the other on a wave rider at elevations of 0.2, 0.5, 1, and 2 m. Measurements at the elevation of 11 m were the most comprehensive, covering 18 wind speeds from 3 to 17 m s^{-1}. Droplet concentrations were collected in eight diameter bands, equally spaced on a logarithmic scale between 10 and 100 μm. Measured concentrations for small and large droplets were shown by Wu (1990b) to vary with the droplet diameter in accordance with two distinctly different functional forms. The division between two ranges varies with the wind velocity, but centers around 30 μm in diameter.

Preobrazhenskii (1973) reported the most comprehensive set of spray data at small sizes. Judging from his results, the concentration of droplets per unit diameter band, dN/dD, for small sizes could be represented by (Wu 1990b)

$$(dN/dD)/(dN/dD)_u = \exp(-D/D_u), \qquad (1)$$

where D is the droplet diameter at a relative humidity of 80%, $(dN/dD)_u$ was obtained by extending on a semilogarithmic plot the straight line fitted to measured concentrations at various diameters to $D = 0$ μm, and D_u is the diameter characterizing the size distribution of droplets. Values of $(dN/dD)_u$ and D_u obtained by Wu (1990b) from de Leeuw's data are reproduced in Figs. 1a,b, where U_{10} is the wind velocity at 10 m above the mean sea surface. The characteristic diameter D_u is seen in Fig. 1b to increase continuously with the wind velocity. As the wind velocity increases, the characteristic concentration $(dN/dD)_u$ is seen in Fig. 1a to decrease

Corresponding author address: Dr. Jin Wu, Institute of Hydraulic and Ocean Engineering, National Cheng Kung University, Tainan, Taiwan.
E-mail: jinwu@mail.ncku.edu.tw

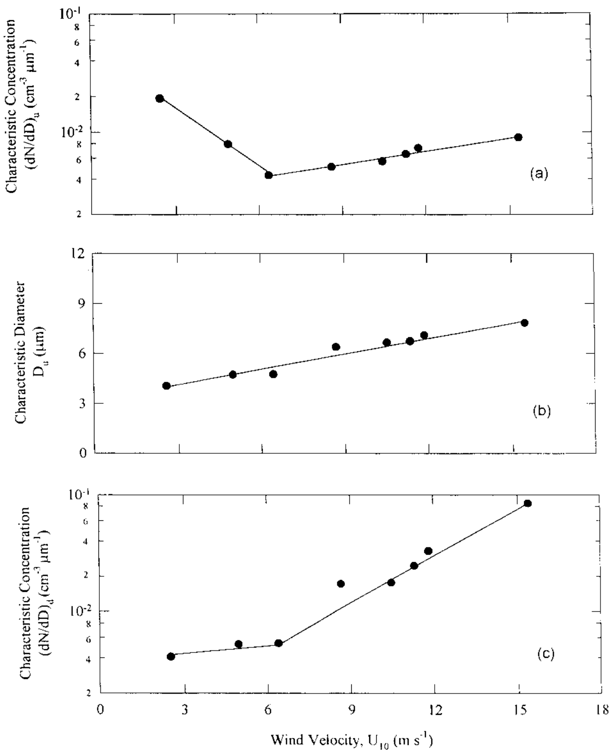

FIG. 1. Characteristic parameters associated with exponential and powers laws describing the data of de Leeuw (1986).

at low winds indicating that relatively more larger droplets are produced at higher wind velocities, and to increase steadily at high winds representing an overall increase of spray concentrations with the wind velocity. Lines fitted to the results in Figs. 1a,b, can be expressed as

$$\ln(dN/dD)_u = -3.034 - 2.675U_{10} \qquad U_{10} < 6.2 \text{ m s}^{-1}$$

$$\ln(dN/dD)_u = -5.995 + 0.084U_{10} \qquad U_{10} > 6.2 \text{ m s}^{-1} \tag{2}$$

$$D_u = 3.240 + 0.304U_{10}, \tag{3}$$

where D_u and U_{10} are in μm and m s^{-1}.

The de Leeuw (1986) data for large droplets have been interpreted by Wu (1990b) to follow a power-law-type dropoff for sizes larger than 20 μm in diameter at low winds and 30 μm in diameter at high winds. This portion of the data was described by

$$dN/dD = (dN/dD)_d(D/10)^{-5.5}, \tag{4}$$

where $(dN/dD)_d$ is the droplet concentration obtained by extending the above power law fitted to the data to $D = 10$ μm, and D again is expressed in μm. Values of $(dN/dD)_d$ are also reproduced from Wu; see Fig. 1c. At low wind speeds ($U_{10} < 7$ m s^{-1}) where waves break infrequently, the rate of droplet production increases very slowly with the wind speed. The production rate is seen to increase rapidly at higher winds, under which wave breaking intensified and the atmospheric surface layer became aerodynamically rough (Wu 1981) to make the upward transport of droplets more effective. The data in Fig. 1c can be represented by

$$\ln(dN/dD)_d = -5.636 + 0.068U_{10} \qquad U_{10} < 6.2 \text{ m s}^{-1},$$

$$\ln(dN/dD)_d = -6.959 + 0.296U_{10} \qquad U_{10} > 6.2 \text{ m s}^{-1}. \tag{5}$$

b. *Smith et al.*

Measurements of marine aerosols were conducted by Smith et al. (1993) on an island, 100 km off the west coast of Scotland, over a wide range of wind velocities spanning between 0 and 34 m s^{-1}. Particle concentrations were obtained with two optical particle counters mounted at 14 m above the mean sea surface. Their data covered the droplet-radius range 1–23.5 μm. Relative humidity varied during their measurements, having a mean value of about 77% with some hourly averages exceeding 90%. The droplet radius R reported by Smith et al. is then considered as that at a relative humidity of 80%.

Droplet concentrations, dN/dR, measured by Smith et al. (1993) at wind speeds of 5, 10, 15, and 20 m s^{-1} are reproduced from their article in Fig. 2; these wind velocities were also measured at the 14-m elevation. For the equilibrium condition with the gravitational deposition of particles being balanced by their production, Smith et al. suggested

$$dF_m/dR = wdN/dR, \tag{6}$$

in which dF_m/dR expressed in m^{-2} s^{-1} μm^{-1} is the droplet flux per unit area of the sea surface and w is the deposition velocity. Incorporating the deposition velocity derived by Slinn and Slinn (1981), Smith et al. described their data sorted into unit wind speed bands with

$$dF_m/dR = \sum_{i=1,2} A_i \exp[-f_i \ln^2(R/r_i)] \tag{7}$$

for $\log A_1 = 0.0676U_{14} + 2.43$ and $\log A_2 = 0.959U_{14}^{1/2} - 1.476$, where constants f_1 and f_2 have values of 3.1 and 3.3, respectively; the radii r_1, and r_2 are 2.1 and 9.2 μm; and U_{14} (expressed in m s^{-1}) is the wind velocity at 14 m above the mean sea surface.

3. Production spectra of marine aerosols

a. *Sea spray produced by bursting bubbles: Film and jet drops*

Results, shown in Eqs. (1)–(5) deduced from the de Leeuw (1986) data at wind speeds corresponding to those under which measurements of Smith et al. (1993)

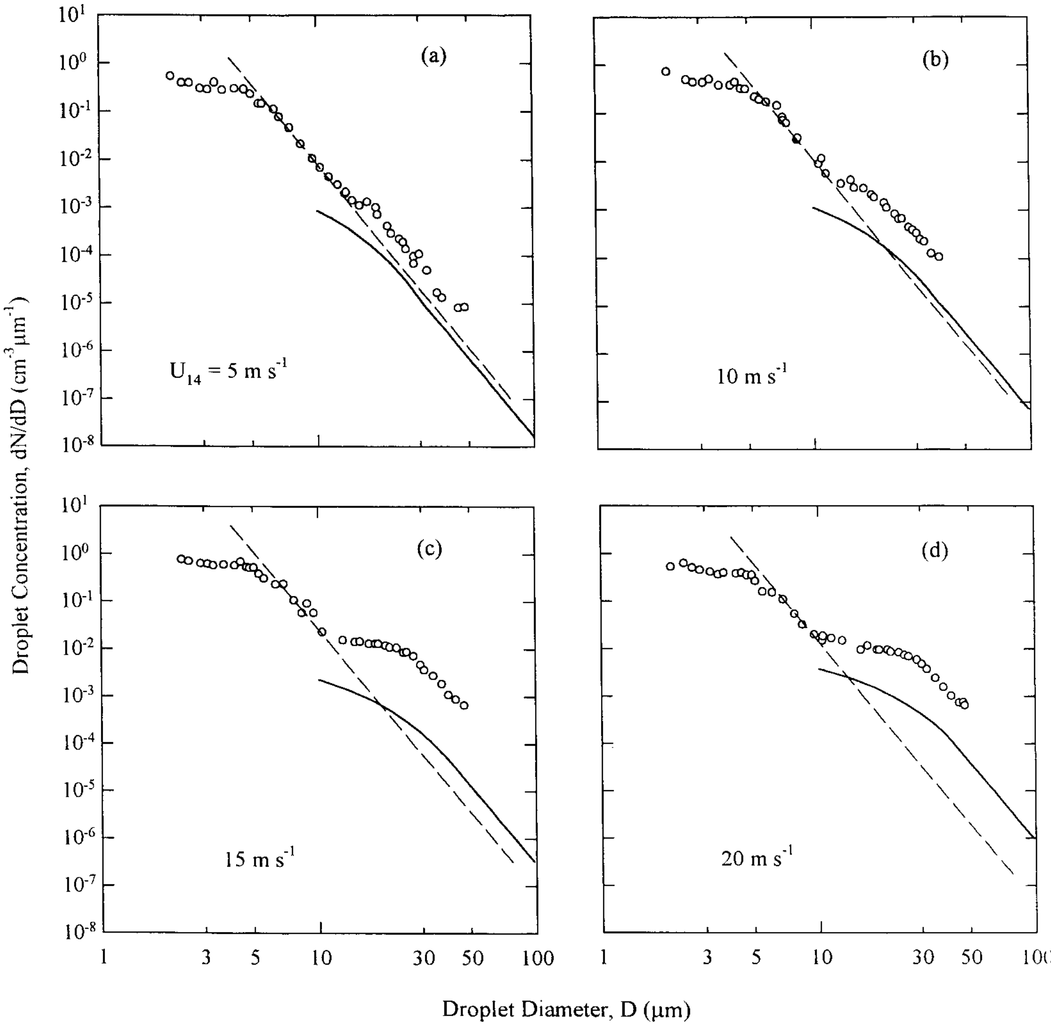

FIG. 2. Droplet concentrations measured by Smith et al. (1993) and de Leeuw (1986) and the comparison of their results. The former measurements are represented by open circles and the latter by solid lines; the dashed lines correspond to $dN/dD \sim D^{-5.5}$.

were performed are also diagramed in Fig. 2. The wind speed U_{10} used in these equations to obtain the lines shown in the figure are determined from U_{14} through the logarithmic wind profile. Droplet concentrations were measured by de Leeuw at an 11-m elevation and by Smith et al. at a 14-m elevation. The concentration at a higher elevation is, of course, smaller; the difference between droplet concentrations at two elevations should also vary with the droplet diameter, with a greater difference for larger droplets; these variations, however, have not been quantified. More on this will be discussed.

At all wind speeds, de Leeuw's (1986) measurements over small sizes apparently provide much smaller droplet concentrations than those reported by Smith et al. (1993); the difference becomes smaller over larger sizes. Taking both groups of results together, we see nonetheless a number of interesting features. A power-law-type dropoff, displayed as a linear trend in Fig. 2, also appears to exist for measurements of Smith et al. not only for large diameters, but also for smaller sizes indicated by the dotted line. Moreover, for both size ranges their measurements follow well the same $D^{-5.5}$ dropoff deduced from de Leeuw's data. Over small sizes, dropoffs of measurements by Smith et al. at two low wind speeds of 5 and 10 m s^{-1} almost coincide with those representing de Leeuw's measurements. They start to deviate systematically from each other at two high wind speeds of 15 and 20 m s^{-1}.

Let us examine further the results shown in Fig. 2 over three size regions: the power-law dropoff indicated as the dashed and solid lines, and those on either side of the dropoff. As mentioned earlier, droplet concentrations reported by de Leeuw (1986) over the small-size region were lower than those reported by Smith et al. (1993). The low concentrations can be attributed to a reduction in the sampling efficiency of de Leeuw's instrument at the small droplet end of the measured size spectrum (Vrins and Hofschreuder 1983). In fact, this was the reason that a lower bound of 10 μm in diameter was imposed by de Leeuw for his measurements. The falloff in sampling efficiency is apparently quite significant, especially toward smaller sizes, to cause the leveling off of the measured spectra. As for the power-law region, de Leeuw's results represented by the solid line are below those reported by Smith et al. at the lowest wind velocity; then the results increase systematically to go above the dashed line representing the extension of Smith et al.'s data at small to large sizes as the wind velocity increases. The closeness of these two sets of data at the lowest wind speed indicates that the droplet concentration differs little at elevations of 10 and 14 m. The results measured by Smith et al., although at a higher elevation, are seen to be slightly greater, possibly due to the fact that the spray production in the nearshore region is somewhat greater than that in the open sea. The systematic increase of de Leeuw's results with respect to the extension of Smith et al.'s represented by the dashed line will be attributed later to the contribution of spume drops.

Accepting the above, the dropoff of measurements by Smith et al. (1993) represented by the dashed lines over small sizes is apparently associated with film and jet drops. Such an interpretation is consistent with the fact that this dropoff at the lowest wind speed as mentioned earlier can almost be superimposed upon that of de Leeuw's (1986) measurements. It should be noted that the production of spume drops started at a speed of about 11 m s^{-1} (Monahan et al. 1983) and became significant at much higher wind speeds. In summary, it is very encouraging to see that the closeness of these two comprehensive sets of spray data may finally provide a basis for quantifying the production of spray droplets by oceanic bubbles, say, below a wind speed of 11 m s^{-1}, which covers well prevailing sea conditions. The difference between the measurements of de Leeuw and of Smith et al. is seen in Fig. 2 to become larger at high winds. The large deviation at a wind speed of 20 m s^{-1}, however, can be due simply to the overextension of de Leeuw's results. As shown in Fig. 1, the highest wind velocity encountered in his measurements was only slightly above 15 m s^{-1}. The sea becomes very stormy beyond this wind speed; both measurements of de Leeuw and Smith et al. might be affected.

In summary, both sets of measurements, de Leeuw (1986) and Smith et al. (1993), appear to describe nearly the same production of film and jet drops. The former set parameterized in Eqs. (4) and (5) were deduced from data collected in the open sea. However, de Leeuw's measurements were affected by the sampling efficiency for small sizes, say, less than 10 μm in diameter as discussed earlier. The portion of the spectrum at small sizes, on the other hand, can be supplemented very nicely by the measurements of Smith et al. (1993) and are parameterized as the first term in Eq. (7) with the coefficient A_1. Taken together, we see that there are little differences between the production of film and jet drops by bursting bubbles in the open sea and in coastal areas. Their production can be described by the almost-connected dropoffs displayed by de Leeuw's data at large sizes and extended by those of Smith et al. to smaller sizes.

b. Sea spray produced by wind tearing: Spume drops

Spectra of droplets obtained by Wu (1973) at high wind speeds in a wind–wave tank demonstrated a rather distinct pattern of spray production; the spectra were narrowly peaked at large radii. These features were later associated with the production of spume drops (Monahan et al. 1983). Subsequently, droplets within the lowest meter above the undulating sea surface were observed by Wu et al. (1984) in clusters separated roughly by the length of dominant waves; in addition, the size spectrum of these droplets was similar to that of presumably spume drops observed in the tank (Wu 1973). In the meantime, the de Leeuw (1986) measurements displaying a maximum droplet concentration away from the sea surface were suggested earlier to provide evidence of the surface tearing (Wu 1990a). At the crest of breaking waves, the profile is the sharpest while water particles race to approach the phase velocity of dominant waves; these are the most favorable conditions for the wind tearing.

The production of spume drops was estimated by Wu (1993) on the basis of spray measurements in a wind–wave tank (Wu 1973) and the field (Wu et al. 1984). Shapes of the spectra obtained from the tank and field were actually the same on the large-radius side, following R^{-8}; this constituted the most critical portion of the spectrum. Averaging field and laboratory results, the lower bound of the R^{-8}-segment was placed at $R = 150$ μm. For smaller sizes, a segment of $R^{-2.8}$ suggested by Wu et al. (1984) was adopted by Andreas (1992) in his model; he also added another segment of R^{-1}. These segments were also adopted by Wu (1993) with slightly rounded-off exponents to R^{-3} and R^{-1}. The radius dividing these two regions was chosen to be 75 μm. Finally, a sharp cutoff was located at the radius of 37.5 μm, which was very close to the value adopted by Andreas of 31.5 μm. In summary, the spectra of both models have three size regions; each region has a different spectral dropoff. The spectral shapes displayed by de Leeuw's (1986) and Smith et al.'s (1993) measurements do not appear to contradict these dropoffs.

The key element of spume-drop modeling is the rate of its production per unit area of the sea surface. In the Andreas (1992) model, this rate was obtained from the extension of a spectrum proposed by Miller (1987) primarily for film and jet drops. While in the model of Wu (1993), rates of droplet fluxes obtained at various wind friction velocities in the laboratory were applied to the field. The total production rates of spume drops for the

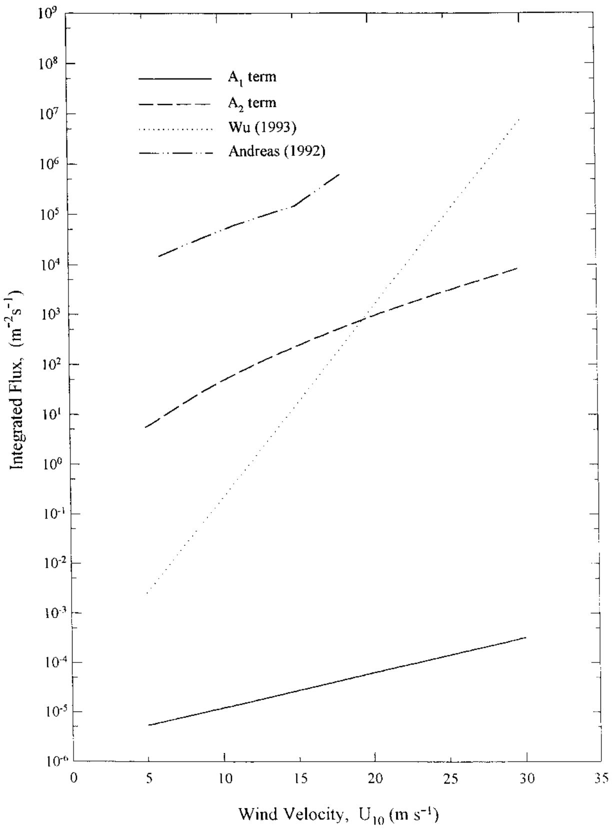

FIG. 3. Comparison of two-spray production rates reported by Smith et al. (1993), A_1 and A_2, with those of spume drops proposed by Andreas (1992) and Wu (1993).

radius range of 75–500 μm offered by both models at various wind velocities are reproduced from Wu (1993) in Fig. 3. There is apparently a slight error in Wu's earlier computation of Andreas's rate; the results shown in the figure are correct.

The elevated concentration over large sizes near 20 μm is the most important feature displayed by the measurements of Smith et al. (1993); this elevated portion becomes larger as the wind velocity increases. Such rather sudden increases of droplet concentrations signify a new source of spray production; their occurrences principally at large droplet diameters and also more intensively at higher wind velocities further suggest a probable association with spume drops. The production starts to elevate at the diameter of about 10 μm, which is actually the lower bound of de Leeuw's (1986) measurements. Consequently, an elevated production is not clearly seen from de Leeuw's results represented by the solid lines in Fig. 2. Nevertheless, these lines should include the production of spume drops. Note also that a greater production of spume drops is expected in experiments by Smith et al. conducted in the coastal region, where choppy dominant waves induce a more widespread breaking at even low winds. In Fig. 2, the additional production measured by Smith et al. above the dashed line is considered herewith mainly as the contribution from spume drops. (As noted by one of the referees, without a detailed knowledge of their experimental site, surf production on offshore rocks and shoals should not be completely ruled out.)

Empirical expressions, Eq. (7), suggested by Smith et al. (1993) to describe droplet concentrations at various wind speeds, consist of two portions. As discussed in the previous section, the portion represented by the first term with the coefficient A_1 responsible for covering primarily small sizes and its extension to large sizes is associated with film and jet drops produced by bursting bubbles. The elevated portion associated with the coefficient A_2, excluding the portion associated with A_1, then represents spume drops produced by the tearing of waves by the wind. Production rates associated with A_1 and A_2 are also presented in Fig. 3. Here, we have used the formula suggested by Fitzgerald (1975) to transfer the radius R at a relative humidity of 80% in Eq. (7) to R_{100} at a relative humidity of 100%; the integration is for the radius (R_{100}) range of 75 to 500 μm. Note that the measurements of Smith et al. were extended to these large sizes, thus facilitating the present quantitative comparison. It is then interesting to compare this production with those suggested in spume-drop models of Andreas (1992) and Wu (1993). With respect to the measurements of Smith et al. (1993), the Andreas (1992) model of spume drops may overestimate the production by surface tearing while Wu's (1993) formulation may overestimate the increase of spume-drop production rate with wind velocity. The production rate suggested by Andreas is three orders of magnitude greater than that deduced from measurements by Smith et al. This is not surprising as his model was constructed on the basis of the production of film and jet drops, by extending it to large sizes. Note that Andreas et al. (1995) also compared over smaller sizes ($R < 100$ μm), the Andreas (1992) estimates and measurements of Smith et al.; in this case, as expected, the difference is only about one order of magnitude. The difference in the wind-speed dependence between the Wu (1993) model and those measurements of Smith et al. and Andrea's model should also be further investigated, as the production rate of spume drops in Wu's model was obtained through the scaling of laboratory measurements with the wind-friction velocity. Such a practice is also open to debate, as wave conditions, especially the occurrence and crest length of breaking waves in laboratory tanks and in the field, may not be capable of being scaled just by the wind-friction velocity.

4. Concluding remarks

Recent measurements of Smith et al. (1993) can be interpreted as reflecting two production mechanisms, each with a distinct spectral signatures: one component represents the production of film and jet drops by bursting bubbles and the other of spume drops by the wind tearing of wave crests. The former component is shown to match well with the measurements reported by de Leeuw (1986) and parameterized by Wu (1990b). We have greater confidence in the accuracy of our description of the component of spray production by bursting bubbles; in other words, either Wu's parameterization of de Leeuw's results or Smith et al.'s measurements and parameterization are acceptable for film and jet drops. As for spume drops, the production rate reflected in the measurements of Smith et al. is much smaller, by more than three orders of magnitude, than that suggested by Andreas (1992), but they have similar wind speed dependence. On the other hand, the measured production compares more favorably in magnitude with that suggested by Wu, at a wind speed around which spume drops constitute the principal component of sea spray, but their rates of increase of the spume-drop production with wind velocity are very much different. All of these lead us to think that we may be closer to reaching a parameterization of sea-spray production than originally thought. Further studies and refinements depend on the understanding and quantification of the influence of waves and wind, which is still unsettled.

REFERENCES

Andreas, E. L., 1992: Sea spray and the turbulent air–sea heat fluxes. *J. Geophys. Res.,* **97,** 11 429–11 441.

——, J. B. Edson, E. C. Monahan, M. P. Rouault, and S. D. Smith, 1995: The spray contribution to net evaporation from the sea: A review of recent progress. *Bound.-Layer Meteor.,* **72,** 3–52.

de Leeuw, G., 1986: Vertical profiles of giant particles close above the sea surface. *Tellus,* **38B,** 51–61.

Fitzgerald, J. W., 1975: Approximation formulas for the equilibrium size of an aerosol particle as a function of its dry size and composition and the ambient relative humidity. *J. Appl. Meteor.,* **14,** 1044–1049.

Gathman, S. G., 1983: Optical properties of the marine aerosol as predicted by the Navy aerosol model. *Opt. Eng.,* **22,** 57–62.

Miller, M. A., 1987: An investigation of aerosol generation in the marine planetary boundary layer. M. S. thesis, Dept. of Meteorology, The Pennsylvania State University, 142 pp.

Monahan, E. C., C. W. Fairall, K. L. Davidson, and P. J. Boyle, 1983: Observed inter-relations between 10 m winds, ocean whitecaps and marine aersols. *Quart. J. Roy. Meteor. Soc.,* **109,** 379–392.

Preobrazhenskii, L. Yu., 1973: Estimation of content of spray drops in the near-water layer of the atmosphere. *Fluid Mech. Sov. Res.,* **2,** 59–100.

Slingo, A., 1990: Sensitivity of the earth's radiation budget to changes in low clouds. *Nature,* **343,** 49–51.

Slinn, S. A., and W. G. N. Slinn, 1981: Modeling of atmospheric particulate deposition of natural waters. *Atmospheric Pollutants in Natural Waters,* S. J. Eisenreich, Ed., Ann Arbor Sci., 23–53.

Smith, M. H., P. M. Park, and I. E. Consterdine, 1993: Marine aerosol concentrations and estimated fluxes over the sea. *Quart. J. Roy. Meteor. Soc.,* **119,** 809–824.

Vrins, E., and P. Hofschreuder, 1983: Sampling total suspended particulate matter. *J. Aerosol. Sci.,* **14,** 318–322.

Wu, Jin, 1973: Spray in the atmospheric surface layer: Laboratory study. *J. Geophys. Res.,* **78,** 511–519.

——, 1981: On critical roughness Reynolds numbers of the atmospheric surface layer. *J. Geophys. Res.,* **86,** 6661–6665.

——, 1990a: Vertical distribution of spray droplets near the sea surface—Influences of jet-drop ejection and surface tearing. *J. Geophys. Res.,* **95,** 9775–9778.

——, 1990b: On parameterization of sea spray. *J. Geophys. Res.,* **95,** 18 269–18 279.

——, 1993: Production of spume drops by the wind tearing of wave crests: The search for quantification. *J. Geophys. Res.,* **98,** 18 221–18 227.

——, J. J. Murray, and R. J. Lai, 1984: Production and distributions of sea spray. *J. Geophys. Res.,* **89,** 8163–8169.