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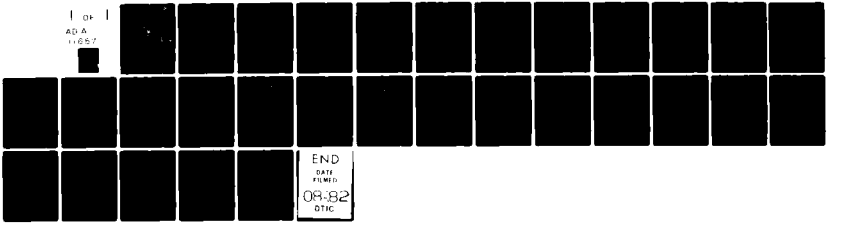

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NAVAL POSTGRADUATE SCHOOL

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GENERATION RATE OF MARINE AEROSOLS AS DETERMINED FROM A BOUNDARY LAYER MODEL

by

C. W. Fairall, K. L. Davidson,
and G. E. Schacher

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- The rate of generation of aerosols by bubbles bursting at the sea surface is not well known. Unfortunately, the rate is a very difficult parameter to measure. The rate can be inferred from a boundary layer model that includes determining evolutions of aerosol spectra from parameterizations of generation, transport and removal processes. Using

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this technique, data from CEWCOM-78 have been analyzed to produce the aerosol surface flux volume spectrum from 0.8 to 15 μm radius at a wind speed of 9 m/sec. Using this flux spectrum and equilibrium aerosol spectra from JASIN, flux spectra are calculated for wind speeds from 6 to 18 m/sec.

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1. INTRODUCTION

This is one of a series of reports dealing with modeling of the marine atmospheric boundary layer. The basics of the Naval Postgraduate School modeling approach are given in Fairall (1981). That report also describes the current status of modeling the pertinent physical processes. The utility of the model for tactical use and initial model validation are given in Davidson et al (1982). This report is in a somewhat different vein. The other reports deal with use of the model to describe or predict the evolution of boundary layer properties. Here we use the model "in reverse" to determine an important physical parameter, the rate of generation of marine aerosols.

There are two components to aerosols in the marine boundary layer: 1) continental (background) and 2) locally generated sea-spray droplets (Barnhardt and Streete, 1970). The sea-spray droplets are generated primarily by the bursting of bubbles at the sea surface (Blanchard and Woodcock, 1957). The bubbles are produced by biological activity, chemical reaction and breaking waves (whitecaps). Thus, the sea surface is a continuous source of sea-salt aerosols in the marine boundary layer. At wind speeds greater than about 3 meters per second, whitecaps are the primary contributor to the bursting bubbles. These surface produced aerosols can be characterized by a surface flux spectrum, $F_s(r)$, which represents the volume of aerosol per particle radius interval produced per square centimeter of ocean surface each second as a function of aerosol particle radius. This quantity is a function of wind speed.

The continuous production of sea-salt aerosols is balanced by several removal mechanisms. One obvious mechanism is the loss of particles as they fall back to the surface. This settling under gravity is called "Stokes fall-out" and is characterized by the Stokes velocity. The particles are transported vertically by turbulence in the marine boundary layer and maintained at

a nearly uniform mixing ratio throughout the mixed layer. The growth of the height of the layer constitutes another loss mechanism called "entrainment" (Deardorff, 1976). The final loss mechanism is "rainout" which occurs when the particles become condensation nuclei in the formation of clouds.

Given the surface flux spectrum and a parameterization of the removal mechanisms, one could predict evolutions of the aerosol density spectrum (Fairall et al, 1982). Unfortunately, the surface flux spectrum is not known. However, we can reverse the process—that is, use parameterizations of the removal processes and evolutions of the aerosol spectrum--to obtain estimates of the surface flux spectrum. This paper describes the calculation of $F_s(r)$ from data taken by the NPS Environmental Physics Group during the Cooperative Experiment for West Coast Oceanography and Meteorology (CEWCOM-78) and the Joint Air-Sea Interaction Experiment (JASIN). Details about the experiments, equipment, measurements, and analysis are given by Fairall (1981).

2. AEROSOL SPECTRUM

The first step to analyzing the evolution of the aerosol spectrum is to remove those variations due to changes in the ambient relative humidity. This is done by transforming each spectrum to a reference relative humidity (RH = 80% or a saturation ratio S = 0.8). We make the usual assumption that the particle radius at saturation S is given by (Fitzgerald, 1975)

$$r_S = r \cdot g(S) \tag{1}$$

where r is the radius at S = 0.8 and $g(S) = 0.81 \exp [0.066S/(1.058-S)]$. If we have a measured aerosol number density spectrum, $n'(r_S)$, then the transformed spectrum at standard saturation, n(r), is:

$$dn/dr = n(r) = n'(r \cdot g(S)) \cdot g(S) \tag{2}$$

Since we prefer to work with the volume spectrum $V(r) = 4/3\pi r^3 n(r)$, the corresponding relationship is:

$$dV/dr = V(r) = V'(r \cdot g(S))/g^2(S) \tag{3}$$

We now must separate the measured spectrum into its two basic components:

1) The background continental aerosol of nonlocal origin, V_c, which is present above and within the marine layer; and
2) The sea-salt aerosol locally generated at the sea surface, V_s, which is well mixed throughout the marine layer (h is the height of the mixed layer). Thus, we assume:

$$V(r) = V_c(r) + V_s(r) \qquad Z < h \tag{4a}$$

$$V(r) = V_c(r) \qquad Z > h \tag{4b}$$

The continental aerosol component is represented with a Junge type distribution,

$$V_c(r) = A/r \tag{5}$$

where A is the continental coefficient. Since there is very little sea-salt volume production for $r < 0.3$ μm, we can use the small size range of the

spectrum to calculate the continental coefficient ($A = rV(r)$). This is documented in Fig. 1 where ensemble averages of volume spectra indicated that $V(r)$ is essentially independent of wind speed at $r = 0.1$ μm. At low wind speeds, the $r = 0.1$ μm and $r = 0.3$ μm ensemble average spectra are described quite accurately by Eqn (5). The claim that A is an accurate index of continental influence is nicely validated by comparison with atmospheric Radon activity (Larsen et al, 1979) from CEWCOM-78 (Fig. 2). Thus, the locally generated sea-salt component can be calculated using:

$$V_s(r) = V(r) - A/r \tag{6}$$

The remainder of this report will deal exclusively with the volume spectra transformed to standard humidity, $V_s(r)$. We write this as V_s to simplify the notation--with the understanding that V_s refers to the spectrum at RH = 80%, with radius, r, an implied variable.

The evolution of the aerosol spectrum in the well mixed marine boundary is described by the following equation (Fairall et al, 1982),

$$hdV_s/dt = F_s - (W_e + W_s)\, V_s \tag{7}$$

(which is presented here without further discussion)

where W_e is the entrainment velocity and W_s is the Stokes velocity (Wu, 1979)

$$W_s = 1.57 \times 10^{-2}\ (1 + 1.2(0.81/g(S))^3)r^2\, g^2(S) \tag{8a}$$

$$W_e = dh/dt - \bar{W} \tag{8b}$$

where r is in μm, W_s is in cm/sec and $\bar{W}$ is the mean vertical air motion due to synoptic scale weather (subsidence). In Eqn (7) we have left out the cloud formation removal mechanism because it acts on a much longer time scale. One assumption implicit in Eqn (7) is that the convective mixing velocity, W_* (Kaimal et al, 1976), is much larger than W_s. Since W_* is typically 1 m/sec while W_s is on the order of 0.1 m/sec for the largest particles ($r = 15$ μm) considered here, the assumption is reasonable.

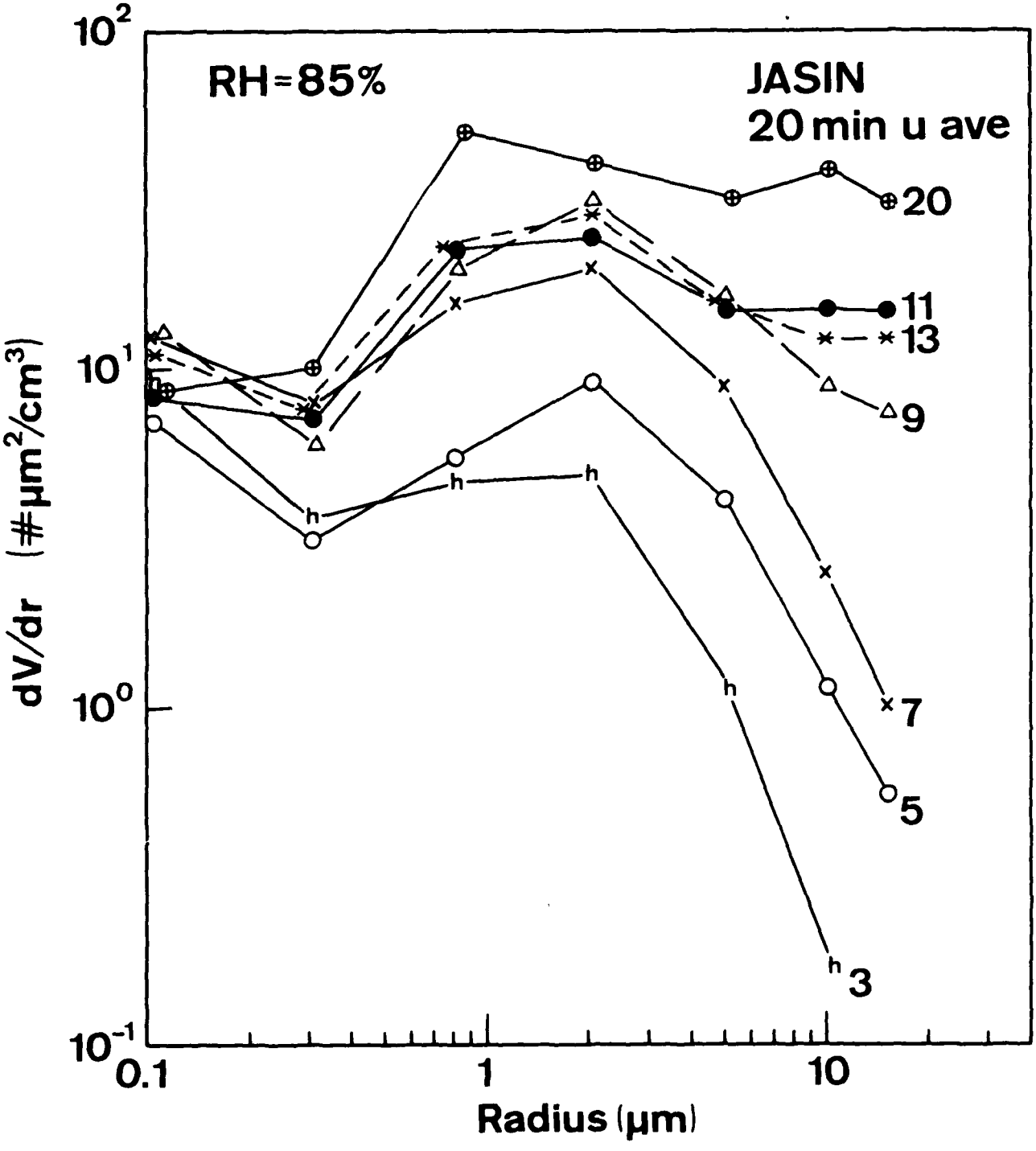

Figure 1. Ensemble average total aerosol volume spectra from JASIN. The number to the right of the spectrum is the wind speed category in m/sec.

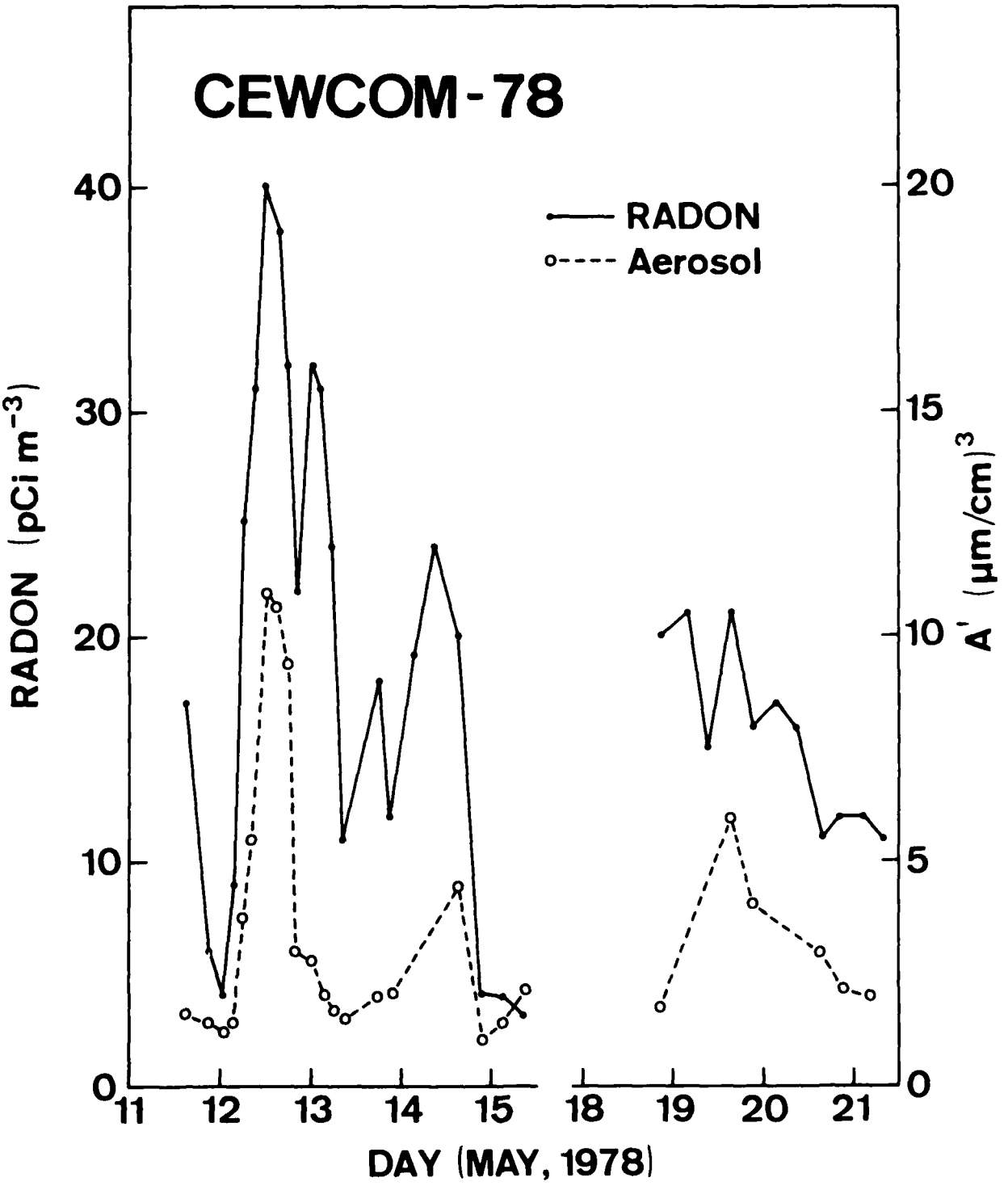

Figure 2. Time series of atmospheric ^{222}Rn activity (solid line, Larsen, Kasemir and Bressan, 1979) and continental aerosol coefficient, A (dashed line).

3. FLUX SPECTRUM ANALYSIS

The CEWCOM-78 data have been used to evaluate the surface flux term of Eqn (7). Since the other terms of the expression were measured, one simply calculates F_s using a rearrangement of the terms. Thus the final expression for the flux calculation is,

$$F_s = h dV_s/dt + (W_e + W_s)V_s \qquad (9)$$

A period was chosen from CEWCOM-78 where all data were available, the synoptic conditions were fairly stable, the wind speed was reasonably constant and a good mixed layer was present (Table I). There was a slowly decreasing continental influence indicated by steady NW winds (A in Table I and Fig. 2).

Application of Eqn (9) requires a value for the entrainment velocity. If the subsidence velocity is known, then W_e can be calculated from dh/dt using Eqn (8b). Since it is difficult to obtain $\bar{W}$ to 0.1 cm/s accuracy, it was decided to estimate W_e empirically from the evolution of the mixed-layer temperature and water vapor density. This was accomplished by simulating the analysis period with a dynamic mixed-layer model. After initializing the model with the atmospheric parameters at the beginning of the period, the entrainment velocity was adjusted to yield best agreement with the temperature and water vapor density at the end of the period. The mean W_e for the period was 0.35 cm/s. This implies $\bar{W} = -0.3$ cm/s for the first 8 hours and $\bar{W} = 0$ for the last 12 hours.

The scatter and uncertainty of the aerosol and mixed layer depth measurements introduces certain trade-offs between averaging times and statistical validity. For short averaging times, the random scatter of the individual terms is considerable, leading to large variations in the time derivative terms. By increasing the averaging time to 4 hours, the F_s "signal-to-noise" becomes more reasonable. A time series of the individual terms of Eqn (9) is

TABLE I. CEWCOM-78 meteorological and aerosol data for the time period analyzed for this report. The aerosol volume spectra density dV/dr is given at each radius r, in μm. The height of the boundary layer was determined from an acoustic sounder.

DATE	TIME	U (m/s)	RH (%)	h (m)	A	r=.3	r=.8	r=2	r=5 ($\mu m^2/cm^3$)	r=10	r=15
5/20	1130	7.4	89.4	420	7.8	15.1	12.5	9.79	2.26	0.801	0.437
5/20	1230	7.7	87.6	430	5.2	9.15	12.5	11.9	2.08	0.644	0.327
5/20	1330	8.2	86.4	430	4.5	8.39	11.9	12.2	2.91	1.11	0.628
5/20	1430	8.1	82.1	415	4.7	8.54	10.6	11.4	2.24	0.780	0.420
5/20	1530	8.3	80.9	437	4.0	7.96	12.0	11.2	1.26	0.296	0.127
5/20	1630	8.2	81.1	437	4.0	7.85	11.9	13.6	2.47	0.795	0.411
5/20	1730	9.0	82.5	443	3.6	7.26	12.9	13.4	2.55	0.854	0.455
5/20	1830	10.0	83.8	430	2.6	6.54	14.0	13.2	3.30	1.37	0.819
5/20	1930	10.3	84.3	420	2.4	6.07	13.8	13.2	3.71	1.73	1.11
5/20	2030	9.8	86.8	360	2.1	6.54	15.9	14.2	4.02	1.63	0.958
5/20	2130	10.1	87.7	377	1.9	6.07	14.9	13.2	3.34	1.22	0.678
5/20	2230	9.5	87.6	397	2.7	6.82	13.6	12.8	3.52	1.42	0.833
5/20	2330	9.2	87.6	400	2.7	6.15	12.9	12.7	3.46	1.46	0.891
5/21	30	9.6	87.3	420	3.0	6.14	13.1	11.8	2.58	0.993	0.569
5/21	130	9.2	88.3	490	2.0	5.69	13.5	12.9	3.33	1.26	0.710
5/21	230	8.3	87.4	520	1.7	5.29	13.3	13.4	3.44	1.39	0.818
5/21	330	8.2	86.1	560	2.7	6.11	11.8	10.9	2.18	0.731	0.390
5/21	430	8.5	83.1	597	2.2	5.55	11.6	9.71	1.78	0.659	0.369
5/21	530	8.2	81.3	620	2.0	5.33	10.9	9.75	1.85	0.732	0.431
5/21	630	7.9	79.7	680	1.8	5.01	11.1	10.7	1.96	0.678	0.365

shown in Fig. 3 for two different particle sizes. The average surface production flux spectrum is shown in Fig. 4. This flux spectrum applies to the average surface conditions for the entire 20 hour period (average wind speed was about 9 m/sec). The error bars represent the uncertainty in the mean estimate.

The relative contributions of entrainment, gravitational fallout and surface production are nicely illustrated by defining an equivalent surface production vertical velocity, $W_F = F_s/V_s$ so that in equilibrium ($dV_s/dt = 0$)

$$W_F = W_e + W_s \tag{10}$$

For the average conditions found in the analysis period, the entrainment and Stokes terms were roughly equal at a particle radius of 5 μm (Fig. 5).

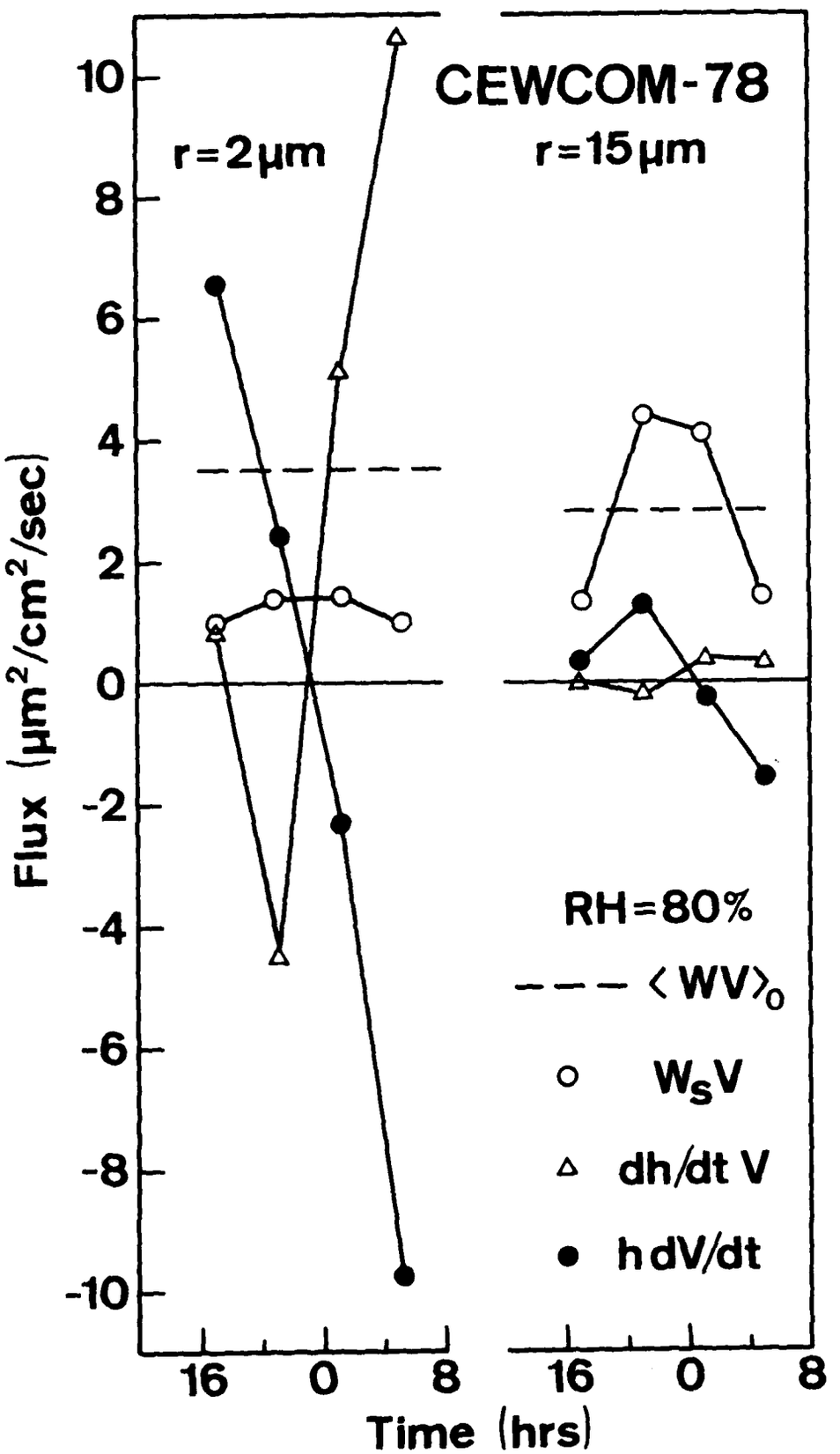

Figure 3. Time series of the terms of Eqn (9) for 2 μm radius and 15 μm radius particles. The dashed line is the average contribution of the surface production term and should be the sum of the other three terms.

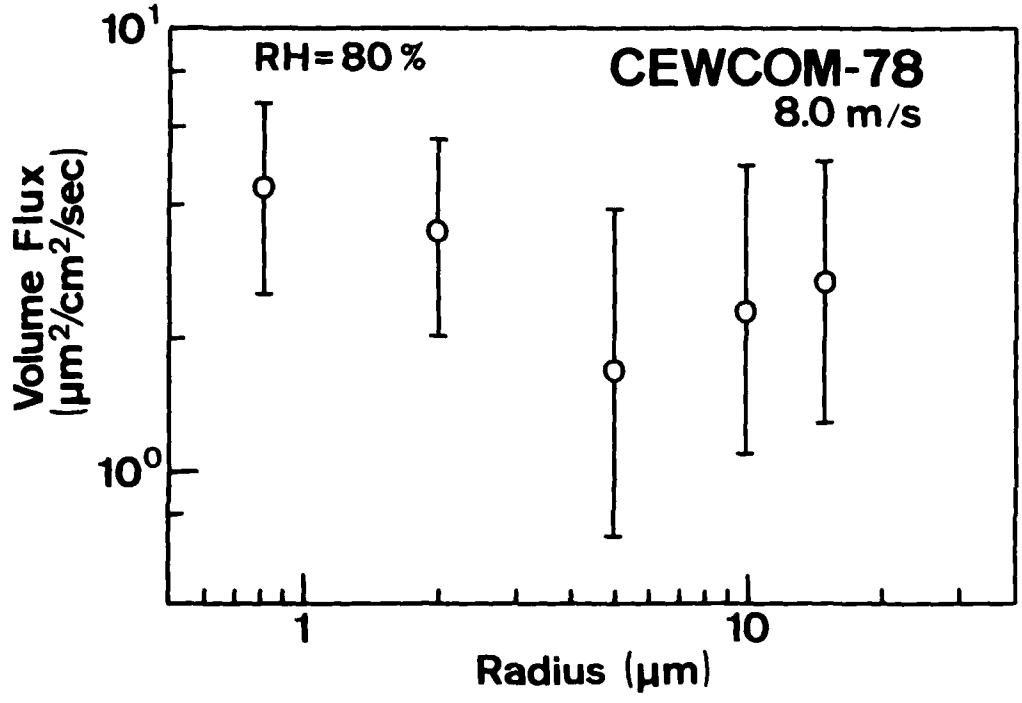

Figure 4. The ensemble average surface flux spectrum, $F_s(r)$, for the CEWCOM-78 analysis period (average wind speed, 9 m/sec).

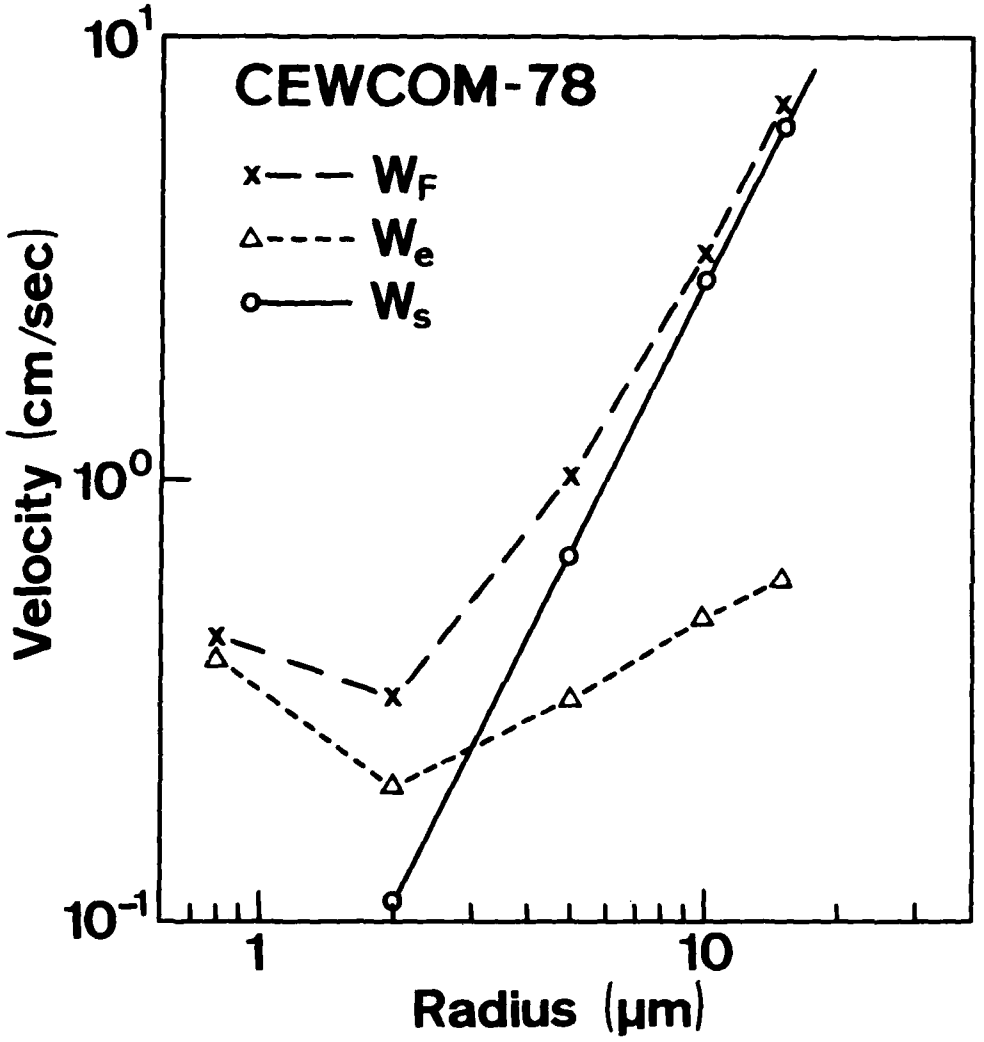

Figure 5. Ensemble average contribution of entrainment (W_e), Stokes fallout (W_s) and surface flux $W_F = F_s/V_s$ assuming a state of dynamic equilibrium.

4. FLUX SPECTRUM AND WIND SPEED

Because of the reasonably constant wind speeds during the 20 hour CEWCOM-78 period, we were able to improve the statistical certainty for the F_s calculation by combining all the data from the period, assuming that the flux would be a reasonable representation for U = 9 m/sec wind speed. Thus, we now have the surface flux spectrum at a single wind speed. In order to estimate the flux at other wind speeds, we note that the right hand side of Eqn (10) is nearly independent of wind speed for equilibrium conditions. Therefore, the flux at one wind speed can be related to the flux at other wind speeds if the equilibrium volume spectra are known:

$$F_s(U_1) = F_s(U_2)\ V_s(U_1)/V_s(U_2) \qquad (11)$$

We have available from JASIN a large set of ensemble averages of aerosol volume spectra at different wind speeds (Fig. 6 and Table II). It is a simple matter to apply this data to Eqn (11) using the CEWCOM-78 aerosol flux and equilibrium spectrum to generate the surface volume flux spectra as a function of wind speed (Fig. 7 and Table III). At a wind speed of 6.0 m/sec, the flux can be integrated in radius space to yield a total sea-salt dry mass production rate of 4.8 mg/m^2/day which compares well with the 4.4 mg/m^2/day estimated by Blanchard (1963) and 4.2 mg/m^2/day estimated by Kritz and Rancher (1980).

TABLE II. Equilibrium sea-salt aerosol spectra in $\mu m^2/cm^3$ as a function of radius (μm) and wind speed (m/sec).

r → / ↓ u	0.8	2	5	10	15
6	2.8	3.4	3	0.3	0.05
9	10	10	5	1	0.35
11	18	25	13	6.4	4.2
13	20	30	20	15	15
15	24	33	22	22	22
18	28	35	28	28	28
9*	9	11	2	0.8	0.4

TABLE III. Surface sea-salt aerosol flux, $F_s(r)$, ($\mu m^2/cm^2/sec$) as a function of particle radius (μm) and wind speed (m/sec).

r → / ↓ u	0.8	2	5	10	15
6	1.3	1.1	2.5	1.0	0.33
9	4.5	3.1	4.2	3.3	2.3
11	8.2	7.7	11	21	27
13	9.1	9.2	17	49	48
15	11	10	19	72	140
18	17	11	24	92	180
9*	4.1	3.4	1.7	2.3	2.6

* CEWCOM-78 Analysis Period

5. NON-EQUILIBRIUM

For time periods of a few hours, the aerosol spectrum may not be in a state of dynamic equilibrium. If we rewrite Eqn (7) in the following form:

$$dV_s/dt + V_s/\tau_p = F_s/h$$

where the time constant, τ_p, is

$$\tau_p = h/(W_e + W_s) \tag{13}$$

then we see the analogy of aerosols and a capacitor charged by an applied "voltage", F_s, through a "resistance", $(W_e + W_s)^{-1}$. In this analogy, the "capacitance" is h.

The response time of the aerosol density is a strong function of particle radius because $W_s \sim r^2$. Values of τ_p for S = 0.8 and h = 400 m are given in Table IV.

TABLE IV. The equilibrium time constant for aerosols at S = 0.8, h = 400 and W_e = 0.4 cm/sec.

r,μm	0.5	1	5	10	15
τ_p, hours	28	22	11	3.5	0.5

The boundary layer mixing time, τ_m, which represents the time required for changes in aerosol density to be evenly distributed throughout the marine layer is:

$$\tau_m = h/W_* \tag{14}$$

For the CEWCOM-78 analysis period (h = 400 m and W_* = 0.6 m/sec) we find τ_m = 0.16 hours. Therefore, short term variations in mixing volume (dh/dt) will lead to changes in the aerosol density because the production response time is much slower than the mixing time. Thus, for time periods on the order of one hour, changes in the aerosol density (hdV_s/dt) will be highly correlated with the mixing volume term (V_s dh/dt). This effect, which is particularly noticeable for smaller particles (see Fig. 3 and Table III), is shown in Fig. 8.

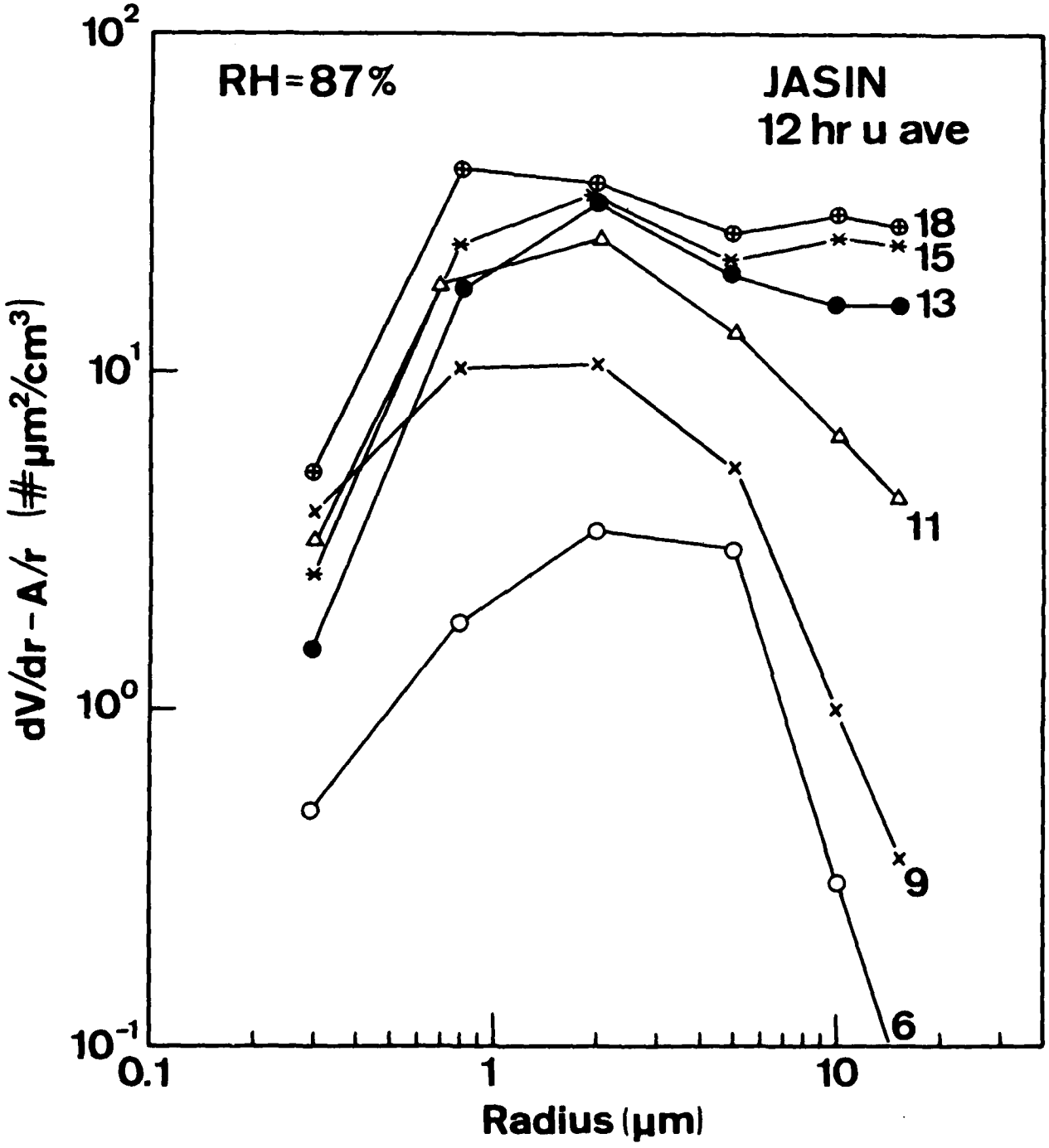

Figure 6. Ensemble average equilibrium sea salt volume spectra from JASIN. The number to the right of the spectrum is the wind speed category in m/sec.

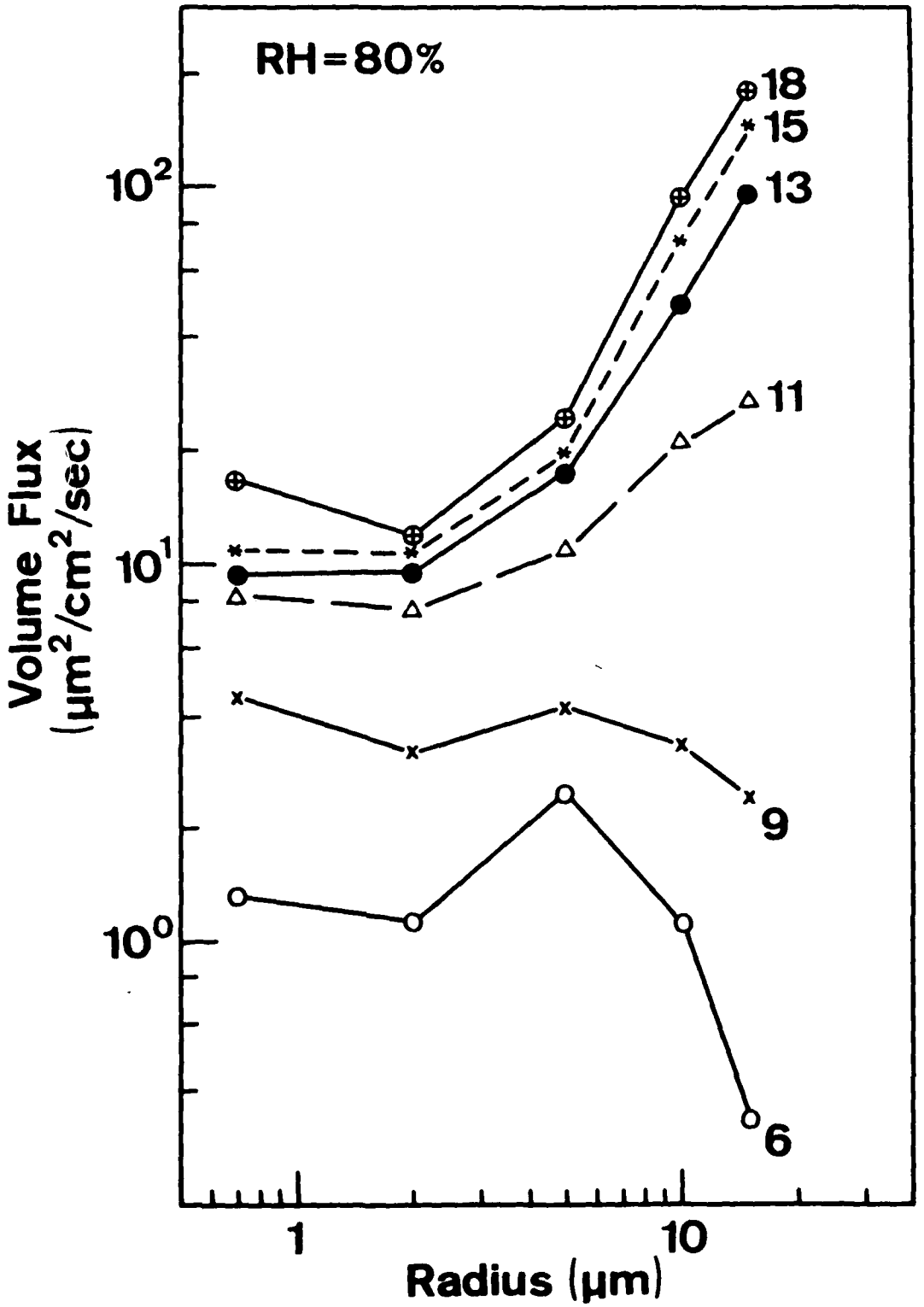

Figure 7. Ensemble average surface flux spectra deduced from Eqn (11) and Table II. The number to the right of the spectrum is the wind speed in m/sec.

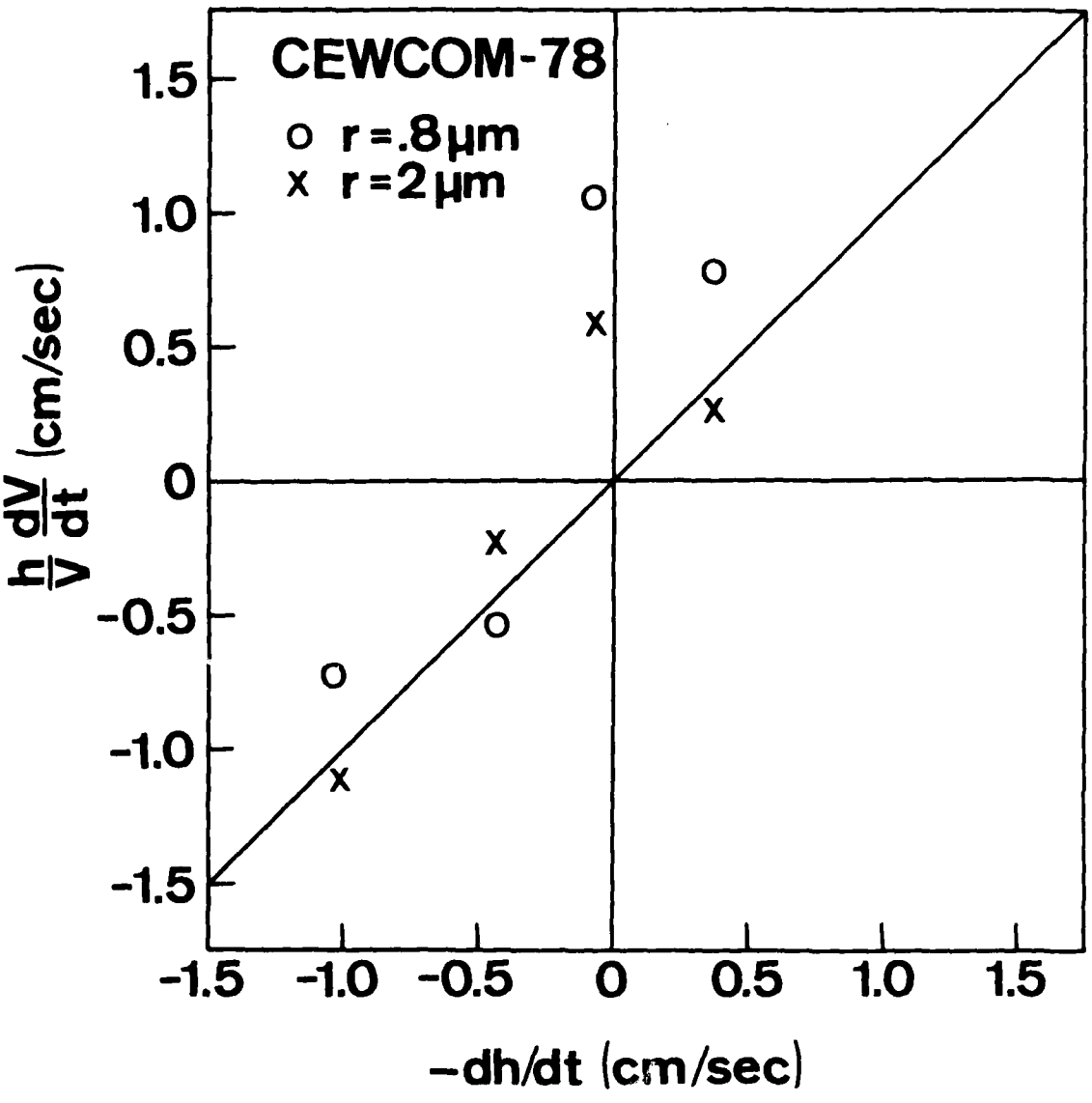

Figure 8. Changes in aerosol spectral density (dV_s/dt) versus changes in mixing volume (-dh/dt). This graph illustrates the dominance of these terms in Eqn (9) for short time periods.

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