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Distribution of the liquid aerosol produced from bursting bubbles in sea and distilled water

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ABSTRACT

Laser holographic techniques are used to observe and measure the liquid droplets (film drops) produced by air bubbles bursting in both seawater and distilled water. Film drop aerosol production is found not only to be a function of parent bubble size but also of the type of water (sea or distilled) in which the bubbles burst. For bubbles in the diameter range of about 4 mm–10 mm, fewer but bigger film drops are observed for the distilled water sample. Furthermore, the mass of the droplets produced per bubble is found to be greater for bubbles that burst in distilled water. In general, the film drop size range shifts towards larger sizes as the diameter of the parent bubble is increased. The largest drop sizes measured are 680 μm in distilled water and 500 μm in seawater for a 10 mm bubble. These values happen to be much greater than those hitherto reported.

1. Introduction

The liquid droplets produced by air bubbles that burst at the surface of the sea are known to play a major role in the formation of marine aerosols (Woodcock, 1952; Blanchard, 1963, 1983; Monahan, 1986). At the surface of polluted rivers and lakes, these droplets also serve as vehicles for the transfer of various chemical substances into the atmosphere (Blanchard and Syzdek, 1972; Blanchard and Hoffman, 1978; Wallace and Duce, 1978). In general, the bursting of an air bubble at a water surface produces two families of liquid droplets with physically distinct origins and characteristics which permit their classification as either film drops or jet drops. The film drops are created very early in the bubble breakup process from the shattering of the liquid film which constitutes the bubble cap, and are mostly too small to be visualized by the naked eye or ordinary photographic techniques. The jet drops, on the other hand, are produced much later from the disintegration by instability of the liquid jet that rises from the bottom of the collapsing bubble cavity, once the hemispherical dome disappears. They are relatively bigger in size (about $\frac{1}{10}$ the diameter of the parent bubble) and less numerous. In fact, their number never exceeds 8 or so for sufficiently small bubbles. Unlike the jet drops, the number of film drops produced per bursting bubble is still an unknown (increasing) function of the parent bubble size.

In 1986, Resch et al. (1986) using a holographic imaging technique succeeded in visualizing for the first time the various stages in the breakup of an air bubble at an air–water interface. In that paper, the authors were able to detect and measure all the film drops greater than about 10 μm in diameter (for bubble diameters of 4, 6, 8 and 10 mm) and presented tentative drop count histograms for a few bursting bubbles in each size range. However, in order to gain a broader insight into the role of the film drops in the sea-to-air exchange of particles, it is necessary to obtain statistically significant data on their size and mass distributions.

Such data will provide useful background information for a realistic modelling of the transport dynamics of the droplets in the marine atmosphere. On the other hand, film drop aerosol

Table 1. *Summary of results*

Type of water*	S.W.	D.W.	S.W.	D.W.	S.W.	D.W.	S.W.	D.W.
Bubble diameter (mm)	4	4	6	6	8	8	10	10
Number of bubbles	14	9	12	16	9	9	8	9
Total number of drops	224	168	378	261	224	172	397	237
Average number of drops per bubble	16.0	18.7	31.5	16.3	24.5	19.1	49.6	26.3
Average aggregate mass of drops per bubble (mg)	0.014	0.031	0.054	0.095	0.092	0.30	0.25	0.70
Maximum drop size detected (μm)	280	360	400	440	460	620	500	680
Average diameter of detected drops (μm)	98	110	113	181	124	256	150	302

* S.W. = seawater; D.W. = distilled water.

characteristics for bubbles that burst in distilled water may be used as comparative data in the study of the phenomenon over polluted rivers and lakes.

This paper compares film drop size and mass distributions for bubbles in seawater and distilled water, respectively. Droplet observation and measurement methods were based on optical holographic principles, and the data represents average values obtained from the bursting of between 8 and 16 bubbles for each specified bubble diameter (see Table 1).

2. Experimental method

A detailed description of the holographic imaging technique employed to detect and measure the film droplets produced by a single bursting bubble is given by Resch et al. (1986). The system consists principally of a pulsed YAG laser beam (which serves as a strong and coherent source of high-intensity light), and an opto-electronic circuit, which synchronizes the firing of the YAG laser with the rupture of the bubble film cap. The process is very much like flash photography, except that instead of a photographic film, a small glass plate coated on one side with a light-sensitive gelatine serves as the target (or holographic plate) on which the film drops are detected and recorded. In practice, upon development of the holographic plates, the film drops show up as dark spots. Such a plate, called an hologram, is shown in Fig. 1. Actually, the spots are the interference patterns formed on the holographic plate by the recombination of the laser waves that were diffracted by the drops and those waves that reached the recording plate without meeting any drops in their path. In order to appreciate the true sizes and spatial distribution of the droplets, the holograms must be "reconstructed", as has been done by Resch et al. (1986).

The holographic imaging technique is a powerful visualization method capable of providing reproducible information about the entire bubble breakup process, from only 150 μs after film cap rupture to the formation of the jet drops some 25 ms later. As a direct film drop measurement method (unlike other measuring techniques which relate detected drop nuclei to original drop size), optical holography has the advantage of being able to detect and size droplets in nucleus-free liquids as well. The technique is capable of providing informative data in the following bubble and drop size ranges.

(a) Bubbles of diameter equal to or greater than 3 mm; the opto-electronic synchronization system does not respond to bubbles of smaller diameter.

(b) Only those film drops that have diameters greater than about 10 μm are detected and measured. This value corresponds roughly to the diffraction limit of the holographic arrangement used, and is related to the sensitivity of the holographic plates.

In all the experiments, bubble rise distance to the surface of the water was about 3 cm. As it is difficult to generate bubbles having exactly integer mm diameters, bubble sizes quoted (in particular for bubbles larger than 6 mm in diameter) should be taken with a tolerance of about

Fig. 1. Typical appearance of film drop aerosol as recorded on a holographic plate (not to scale).

± 0.2 mm. Such big bubbles were generated by means of a common hypodermic syringe and sized with the aid of photographs taken of them before rupture. The production of the smaller bubbles was by means of capillary tubes for which the diameter of the bubble generated is directly proportional to the one-third power of the capillary tip diameter (Blanchard and Syzdek, 1977). The seawater sample was taken from the Mediterranean coast of France (where the salinity was 38.25‰) and passed through a 0.45 μm (Millipore) filter. The distilled water was laboratory-distilled tap water.

3. Results and discussion

The droplet size and mass distribution histograms for both seawater and distilled water are shown in Figs. 2 and 3, respectively. The mass distribution data are presented as the mass of film drops contained in each 20 μm size range (Fig. 3) and as the average aerosol mass per bubble burst (Fig. 4). A summary of the results is given in Table 1.

3.1. Film drop size range

Although no single measurement method so far reported has been successful in covering the entire film drop size range, previous estimates extend from a lower limit of less than 1 μm (Cipriano and Blanchard, 1981; Cipriano et al., 1983, 1987; Woolf et al., 1987; Blanchard and Syzdek, 1988) to more than 200 μm at the large end (Resch et al., 1986).

The present results indicate that the upper bound of the drop size range is a function of both parent bubble diameter and the liquid in which the bubbles burst. The larger the bubble, the bigger the maximum size of the film drops in the aerosol (see Table 1). A cumulative analysis of the drop size spectra reveals, however, that these

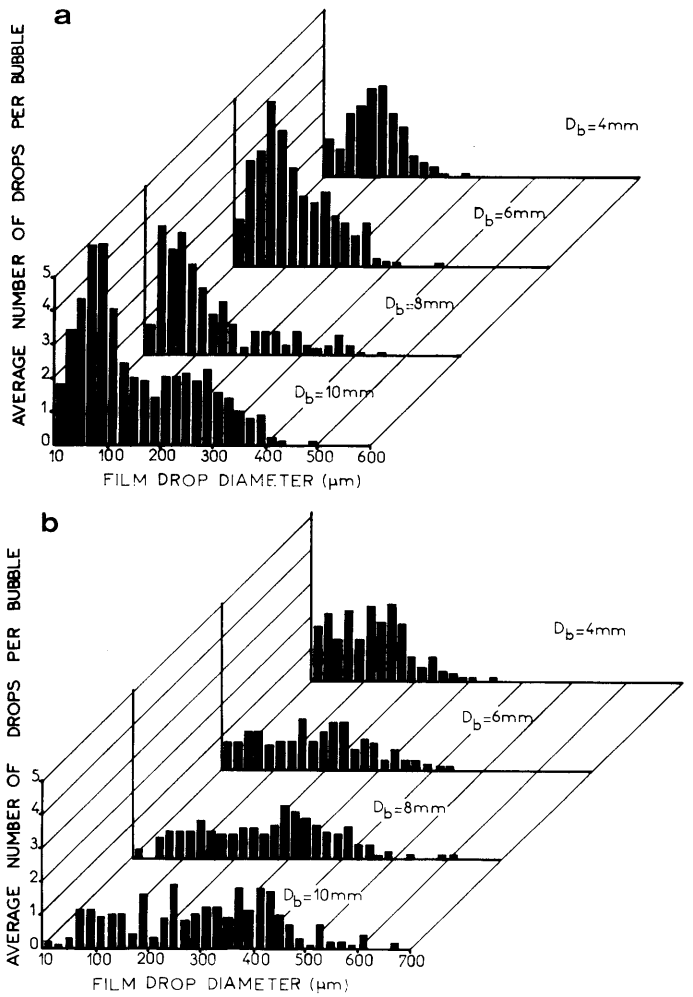

Fig. 2. Film drop size distribution histograms: (a) seawater (b) distilled water.

large drops constitute only a small fraction (<1%) of the total number of film drops in the aerosol.

Comparison of the maximum drop sizes for the two types of liquid show that, for a given bubble diameter, distilled water favours the production of larger drops. A possible explanation for this may be found, not necessarily in the amount of bubble film cap surface exposed above the free liquid level prior to breakup (Toba, 1959), but principally in the differences in bubble cap thickness. Bubbles in seawater have, in general, a longer surface life than those in distilled water (Blanchard, 1983) and therefore a thinner interfacial cap thickness at the moment rupture occurs. Thicker bubble caps, like large diameter bubbles, probably favour the creation of bigger film drops.

3.2. *Aerosol mass distribution*

As bubble caps in distilled water at the moment of rupture are thicker than those in seawater (bubbles, on average, have a shorter surface life in distilled water), one would expect to obtain a larger aerosol mass with bubbles in distilled water than in seawater. This was indeed the case.

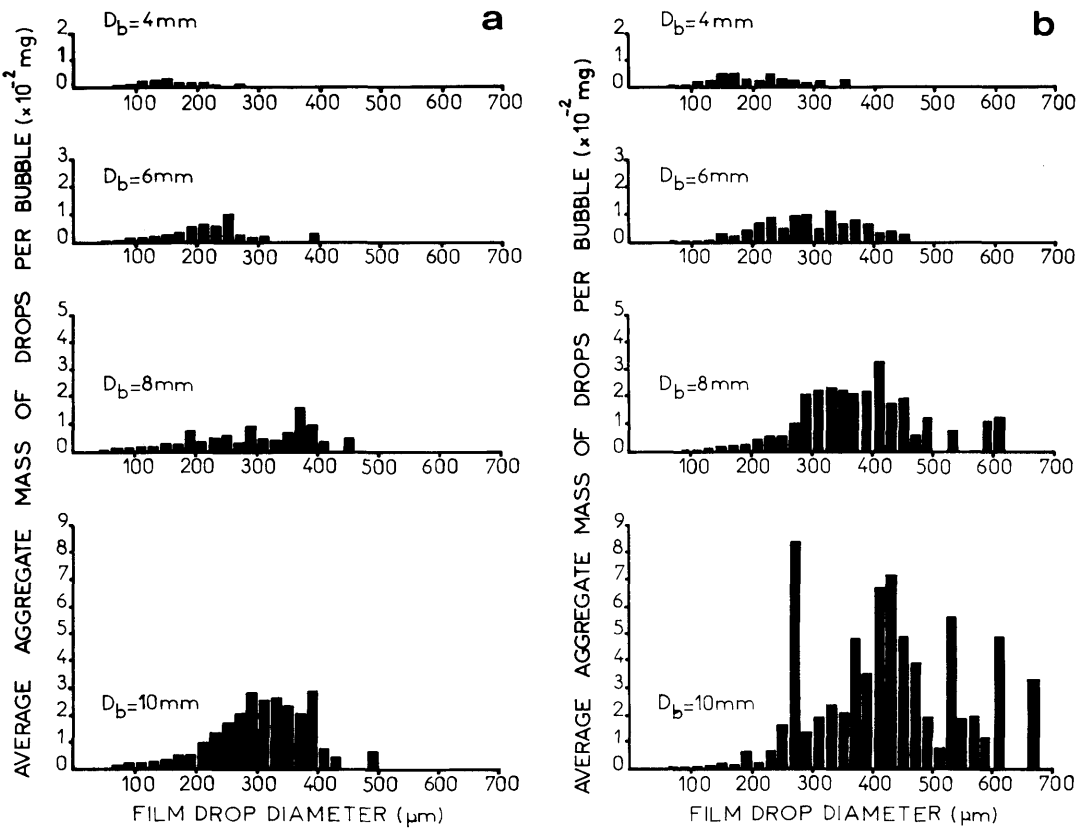

Fig. 3. Film drop mass distribution histograms: (a) seawater (b) distilled water.

Typical values of the average droplet mass per bubble, in the bubble diameter range of 4 mm–10 mm, varied between 0.014 and 0.25 mg for seawater and between 0.031 and 0.70 mg for distilled water

It is evident from the mass distribution histograms that the bulk of the aerosol mass per bubble was provided by droplets of diameter greater than 140 μm (Fig. 3). However, it is interesting to note from the size distribution histograms for seawater, that the number of film drops having diameters equal to or less than 140 μm accounted for the bulk of the total number of drops in the aerosol (Fig. 2a). This is not true for the distilled water samples.

3.3. Number of film drops produced per bubble

The number of drops detected per bursting bubble together with the corresponding standard deviations are shown in Fig. 5. The drop counts obtained, for bubbles in both seawater and distilled water, confirm that the number of drops produced increases with parent bubble diameter (Blanchard, 1963; Day, 1964; MacIntyre, 1974; Resch et al., 1986; Blanchard and Syzdek, 1988). The major significance of the present data, however, is the strong influence of the type of water in which the bubbles burst on the total number of film drops in the aerosol. Thus, with the exception of the 4 mm diameter bubbles, fewer but larger droplets were observed with the distilled water sample than in the seawater at all other bubble diameters (see Table 1). This can be explained by the shift toward larger drop diameters in the size distribution histograms for bubbles that burst in distilled water. One may also remark that whereas the film drop size distribution data for seawater appeared to always peak at drop diameters between 40 and 120 μm, the data for the distilled water showed a more

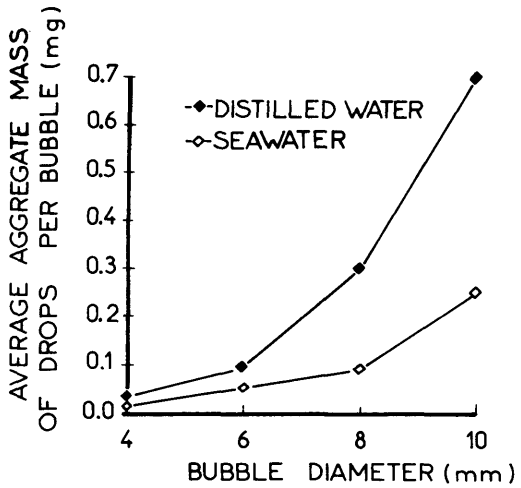

Fig. 4. Average droplet mass per bubble.

even distribution of the aerosol droplets over the entire size range covered by the present measurement method.

4. Conclusions

From the film drop aerosol data obtained in this work utilizing holographic imaging techniques, 3 major conclusions may be drawn.

(a) The number of film droplets produced in a bubble-generated aerosol, their sizes and mass distributions are all strongly influenced by the type of water in which the bubbles burst. Specifically, fewer but bigger drops are produced in distilled water than in seawater.

(b) Film drop sizes in the seawater aerosol rarely exceeded 500 μm in diameter, but are much larger than expected or hitherto reported.

(c) Although the proportion of the number of

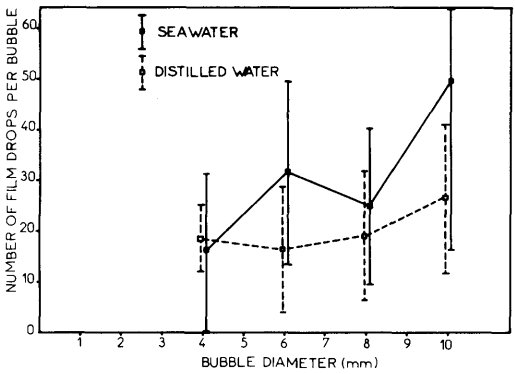

Fig. 5. Film drop count: number of drops with diameter greater than 10 μm produced per bubble.

the smaller drops in the aerosol is high, they account for a negligible fraction of the total aerosol mass. However, they may well play a more important role in the sea-to-air mass exchange process, since their smaller sizes render them less prone to gravitational fall-out.

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