

Effects of Salinity and Surface Tension on Microbubble-Mediated Sea-to-Air Transfer of Surfactants

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A series of experiments were conducted in artificial seawater to study the effects of salinity and surface tension on the bubble-mediated sea-to-air transfer of surfactant materials. The bubble transport of a surfactant tracer (¹⁴C-labeled oleic acid) from seawater to the air-water interface and, subsequently, to a 10-cm height above the water surface by bubble bursting was measured. The change in surface tension induced by bubbling was also determined. We have found that the amount of oleic acid transported to the water surface by bubble scavenging increases as the salinity increases. In consequence, so does the quantity of oleic acid ejected into the air. The ratio of the quantity of oleic acid ejected to a height of 10 cm above the water surface to the amount transported to the water surface was constructed. This ratio was found to increase with decreasing surface tension, indicating that the energetics of the bubble bursting process are strongly influenced by the surface tension encountered during bursting.

INTRODUCTION

A significant quantity of surface active material transported to the atmosphere from the sea is mediated by bubbles produced by wave-breaking events. These bubbles, ascending through the water column, scavenge and concentrate the indigenous surface active materials. Upon reaching the water surface, the bubbles burst and eject a portion of the adsorbed surfactant material together with some seawater into the air. This scavenging and bursting process is thought to be a major source of organic materials, bacteria, and trace metals [Blanchard, 1989] and of condensation nuclei [Cipriano *et al.*, 1987] in the atmospheric boundary layer above oceanic areas.

The transport of surfactant materials from the ocean to the atmosphere by the bubble scavenging and bursting process has received considerable attention over the past 2 decades. Blanchard [1989] has reviewed a number of studies which have examined the bursting of single bubbles in seawater. These studies have revealed that the top (first) jet droplet produced by a bursting bubble is approximately one-tenth the bubble diameter and that the ejection height reached by this droplet is dependent on the size of the producing bubble, first increasing and then decreasing with bubble size with a maximum attained height of nearly 20 cm for 2-mm bubbles. Blanchard [1989] has also reviewed a number of studies which have examined the scavenging of bacteria by single rising bubbles. These investigations have found that the quantity of bacteria scavenged is dependent on the type of bacteria studied. Scavenging increases to a defined limit with increasing bubble rise distance and, for a given rise distance, is largest for bubbles of approximately 500 μm in diameter. From a series of laboratory experiments with bubble plumes, Hoffman and Duce [1976] have identified several factors affecting the organic carbon (OC) content of marine aerosols resulting from bursting bubbles. They found that the OC to sodium ratio of marine aerosols is dependent on the bubble rise distance, the quantity and nature of organic material in

their water samples, and the relative abundance of surfactant in the mixture. Recently, our group [Tseng *et al.*, 1992] has reported on laboratory experiments with bubble plumes and a variety of surfactants in artificial seawater. We have found that the quantity of surfactant transported to the surface microlayer by bubbling decreases as bubble size increases from approximately 200 to 750 μm in diameter. We have also found that the quantity of surfactant ejected to a height 10 cm above the water surface by bubble bursting is linearly proportional to the quantity of surfactant transported to the surface microlayer by bubbling and that the constant of proportionality decreases as bubble size increases. The fact that Tseng *et al.* [1992] find surfactant materials ejected to a height of 10 cm by bubbles less than 750 μm in diameter is in opposition to the results provided by Blanchard [1989] which indicate that bubbles larger than 750 μm in diameter are necessary to obtain jet droplet ejection heights of this magnitude. However, the flow hydrodynamics and bubble bursting processes of single bubbles and bubble plumes are quite different. D. Blanchard (private communication, 1990) has suggested that perhaps bubble coalescence in the thin foam layer generated by a bubble plume is occurring prior to bursting and that this coalescence accounts for the observed discrepancy. Another possibility is some synergistic effect (for example, reduced aerodynamic drag due to drafting) provided by the continuous production of jet droplets from an upwelling bubble plume. This issue has yet to be resolved.

The purpose of the present study was to examine variations in two factors, salinity and surface tension, which might influence the quantity of surfactant materials ejected from seawater to the atmosphere by bursting bubbles. Variations of these two factors are known to occur at the outfalls of major rivers and estuaries. Two series of experimental results are reported here. One series examined the quantity of surfactant transported to the water surface and, subsequently, to the air by bubble scavenging and bursting; the other series measured the surface tension of seawater under various bubbling and seawater conditions.

Using ¹⁴C-labeled oleic acid as a surfactant tracer, we find that as the salinity increases from 0 ppt to 35 ppt, substantially more oleic acid is transported from the water column to the water surface and, in turn, into the air. The ratio of the amount of oleic acid ejected to an altitude of 10 cm above the water surface to the amount transported to the water surface was constructed. We find that this ratio increases with decreasing surface tension. This result indicates that the bubble bursting process is strongly influenced by the surface tension encountered during bursting.

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FACILITY, EXPERIMENTS, AND EXPERIMENTAL CONDITIONS

Experiments were conducted in an acrylic test tank (Figure 1) having internal dimensions of 15.8 x 10.0 cm in cross section and 32.5 cm in height. Three "seawater" conditions were used in experiments: doubly deionized-distilled water, artificial seawater prepared by mixing 35 ppt of Instant Ocean (Aquarium Systems Inc., Mentor, Ohio) into doubly deionized-distilled water, and "clean" (i.e., surfactant depleted) 35-ppt artificial seawater. The water was filtered through Teflon micropore filters, and 3.5 L was added to the tank. This resulted in a water level 10 cm below the top of the chamber. Air bubbles were produced by flowing purified dry air through two porous glass frits which were mounted in plastic frames 2.5 cm above the bottom of the tank. Four different mean airflow rates of 0.5, 1.0, 2.0, and 4.0 mL/min per cm² of water surface area (158 cm²) were maintained. The bubbles were observed to have a lognormal size distribution with a mean diameter of 203 μ m and a standard deviation of 61 μ m regardless of the water type. (Scott [1975a] and others have observed larger bubbles in fresh water than in salt water due to increased bubble coalescence. We have observed similar results within the water column only when frits of higher porosity were utilized.)

Oleic acid was used as an additive surface active material and tracer. Previous studies by our group [Skop *et al.*, 1991; Tseng *et al.*, 1992] have shown that oleic acid is readily scavenged by rising bubbles and is ejected along with other products of the bubble-bursting process, thereby making it a reasonable model to examine the effects of salinity and surface tension on bubble scavenging and bursting. In experiments which measured the surface tension of seawater, unlabeled oleic acid was employed. In experiments measuring surfactant transport to the water surface and air, ¹⁴C-labeled oleic acid was used. In the latter case, radiolabeled oleic acid was constituted with unlabeled oleic acid and dissolved in methanol to yield a concentration of 20 μ g/mL having an activity of approximately 1.0 μ Ci/mL. After filling the tank with 3.5 L of water, 3.5 mL of the oleic acid solution was injected into the center of the water column and mixed thoroughly with a magnetic stirring bar. This resulted in an initial oleic acid concentration of 20 μ g/L.

The material transported to the water surface and ejected to a height of 10 cm into the air by bursting bubbles was collected by

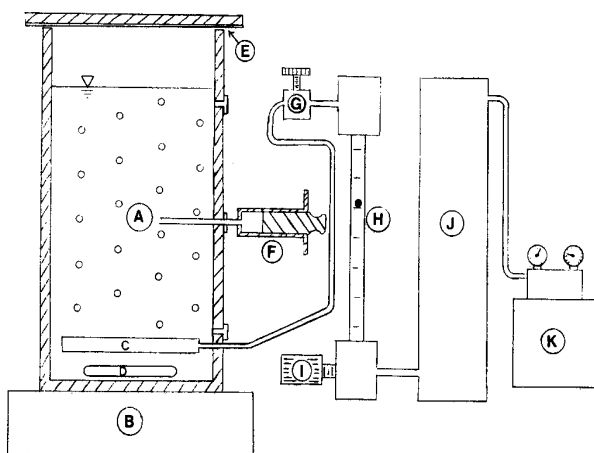


Fig. 1. Bubble test tank apparatus: A, artificial seawater with ¹⁴C-labeled surfactant; B, magnetic stirrer; C, porous bubbling frit; D, magnetic stirring bar; E, cover and ejection collection filter; F, sampling syringe; G, airflow splitter and pressure dump valve; H, rotameter; I, airflow control valve; J, air drying and filtering container; K, air pump.

3M CHA grade chromatography paper. The stirring bar mixing process itself was found to cause a slight degree of surface microlayer enrichment. This mixing-enriched microlayer was removed by chromatography paper prior to the initiation of bubbling. A sample blank caused by bulk water absorption was also inherent to each sampling. This blank value was determined experimentally by sampling the surface immediately after the mixing-enriched microlayer was removed. The blank value was then subtracted from all subsequent bubbling enrichment data. It should be noted that the blank value ($\sim 0.001 \mu\text{g}/\text{cm}^2$) was quite small in comparison to the values resulting from bubble transport microlayer enrichment. The mean pickup efficiency of surface microlayer samplings for oleic acid was predetermined to be 88%. This number was later incorporated into surface microlayer sample determinations to obtain a corrected surface microlayer concentration. A scintillation counter was used to quantitate the radioactivity of each collection filter. This count was then converted from disintegrations per minute to micrograms by the absolute calibration value derived from the radiolabeled oleic acid/methanol mixture. The experimental technique described above has been used previously in studies of microbubble scavenging and sea-to-air surfactant transport. Further details are given by Skop *et al.* [1991] and Tseng *et al.* [1992].

The surface tension of each seawater type, both before and after bubbling, was determined by calibrated spreading oils. This method, originally proposed by Adam [1937] and described more recently by Garrett and Duce [1980], uses a series of buoyant calibrated oils to determine surface tension. Table 1 gives the spreading pressure for 17 oils provided by the U.S. Naval Research Laboratory (NRL) and used in our experiments. During these studies, we found that the surface tension of seawater under certain bubbling conditions was lower than 44.5 dyn/cm, the lowest spreading pressure value of the NRL oils. Therefore a number of other chemicals with lower spreading pressures were also utilized (Table 1). In measuring the surface tension, toothpicks were dipped in each oil, touched with the adhering droplet to the water surface, and discarded after use. This procedure was repeated with several oils. The surface tension of the water was then bracketed between the calibrated values of any two oils in the set, one of which would spread on the water surface and the other of which would not. With rigorous surface cleaning (absorption of the surface layer seven times with Kim-Wipes after each surface tension determination), the use of these oils in the test tank was found to have no significant effect on subsequent surface tension measurements following later babbings.

The protocol followed was to measure the surface tension of the undisturbed water surface, clean; bubble at 0.5 mL/(min-cm²) for 30 s, measure, clean; bubble at 1.0 mL/(min-cm²) for 30 s, measure, clean; bubble at 2.0 mL/(min-cm²) for 30 s, measure, clean; bubble at 4.0 mL/(min-cm²) for 30 s, measure, clean; and then repeat the sequence, but in descending order. The surface tension determinations in the ascending and descending sequences were invariably identical or within 2 to 3 dyn/cm of each other which, from Table 1, is the limit of resolution of the calibrated spreading oils. More accurate and less intrusive techniques for surface tension determinations (i.e., a ring tensiometer) were not used because of the relative confinement of the test tank necessitated by the use of radiolabeled materials.

We found from surface tension measurements that Instant Ocean salt contained a significant quantity of surface active contaminants. The surface tension of the 35-ppt artificial seawater solution was found to be much lower than that of the distilled water following the bubbling process (results presented in a later

TABLE 1. Spreading Pressures of NRL Oils and Non-NRL Materials

Substance	Spreading Pressure
<i>NRL Oils</i>	
1	73.0
2	72.6
3	72.1
4	71.6
5	71.0
6	70.5
7	70.0
8	69.3
9	68.7
10	66.6
11	64.6
12	62.0
13	60.8
14	56.7
15	53.4
16	48.1
17	44.5
<i>Non-NRL Materials</i>	
18 (benzyl alcohol)	39.0
19 (o-xylene)	30.1
20 (toluene)	28.5
21 (n-butanol)	24.6
22 (ethanol)	22.8
23 (isopropyl alcohol)	21.8
24 (n-hexane)	18.5

section). Therefore a new technique was developed and used to remove surface active contaminants from artificial seawater. In this technique, the salt water was first mixed for 1 hour with 35 g of activated charcoal and subsequently purified by passing it through a 12-g bed of fresh activated charcoal. A similar technique has been described by Scott [1975b]. After filtering through two Teflon micropore filters, the water was placed in the test tank. The water was then further purified of residual organic surfactant by bubbling at an airflow rate of 4.0 mL/(min-cm²) through two porous glass frits mounted on the tank bottom. A collection filter was mounted approximately 1 to 2 cm above the water surface to collect and remove surface active contaminants ejected by bursting bubbles. This bubbling process was continued for 1 hour with air collection filters changed every 15 min. As evidenced by surface tension measurements, this technique resulted in relatively "clean" 35 ppt artificial seawater (i.e., the surface tensions determined following subsequent bubbleings were within 3 dyn/cm of those determined for deionized-distilled water).

In order to investigate separately the effects of salinity and surface tension on bubble-mediated sea-to-air transfer, we have conducted three sets of experiments using different water conditions. They are (1) deionized-distilled water with 0 ppt salinity, (2) cleaned artificial seawater with 35 ppt salinity, and (3) uncleaned artificial seawater with 35 ppt salinity. Experiments 1 and 2 produce similar surface tension values but have markedly different salinities. Experiments 2 and 3 have the same salinity but produce markedly different surface tensions.

RESULTS AND DISCUSSIONS

For simplification, the bubble transport of surfactant material

from the water column to the water surface microlayer and to the atmosphere at 10-cm altitude will be denoted "surface transport" and "air transport," respectively. The results presented below are all based on mean values of two or more experiments. The standard deviations of surface transport and air transport values after 120 s of bubbling, representing four individual 30-s accumulations, were generally less than 20% and 40% of their mean values, respectively. The standard deviation of the surface pressure (defined as the difference between the surface tension before and after bubbling) after 30 s of bubbling at 4.0 mL/(min-cm²) was generally less than 10% of its mean value.

Shown in Figure 2 are the surface pressures of the different seawater types as a function of airflow rate after 30 s of bubbling. The surface pressures for distilled water and 35-ppt cleaned and uncleaned artificial seawaters without oleic acid are also plotted. The surface pressure curves of both distilled water and 35-ppt cleaned artificial seawater without oleic acid are similar and have much lower surface pressure values than the other four curves. This finding indicates that the deionized-distilled water and the 35-ppt artificial seawater prepared by our purification technique are relatively "contamination free" compared with uncleaned artificial seawater.

The surface pressure curves are seen to increase with airflow rate but then reach a saturation value or increase less rapidly at higher airflow rates. With the addition of oleic acid, the surface pressure for 35-ppt uncleaned artificial seawater is higher than that of 35-ppt cleaned artificial seawater which is, in turn, somewhat higher than that of distilled water. The differences in surface pressure at each airflow rate, derived by $\Pi_{\text{oleic}} - \Pi$, where Π_{oleic} and Π are the surface pressure of seawater with and without oleic acid, respectively, were also calculated. We have found that the increase of surface pressure for 35-ppt uncleaned artificial seawater due to the addition of oleic acid is generally less than about 6 dyn/cm. On the other hand, the increase of surface pressure for

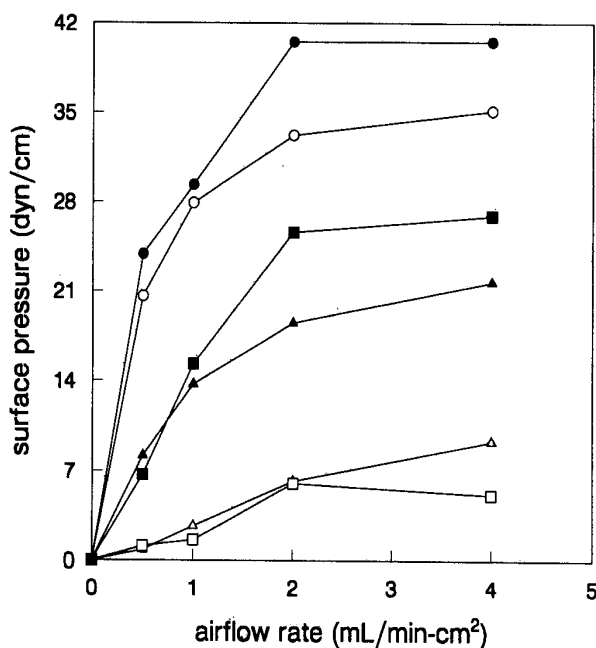


Fig. 2. Variations of surface pressure with airflow rate after 30 s of bubbling for different types of water. The water types are deionized-distilled water (open triangles), 35-ppt artificial seawater (open circles), 35-ppt clean artificial seawater (open squares), deionized-distilled water with oleic acid (solid triangles), 35-ppt artificial seawater with oleic acid (solid circles), and 35-ppt clean artificial seawater with oleic acid (solid squares).

distilled water and 35-ppt cleaned artificial seawater due to the addition of oleic acid is generally between 6 and 20 dyn/cm. This finding indicates that oleic acid, in the quantity used here (20 $\mu\text{g/L}$), acts primarily as a tracer in uncleaned artificial seawater but serves as a significant surface active material in "contamination free" water.

The surface transport of oleic acid for all three sets of experiments is plotted in Figure 3 as a function of bubbling time. The results shown here are cumulative from each bubbling interval of 30 s at an airflow rate of 4.0 $\text{mL}/(\text{min}\cdot\text{cm}^2)$. The surface transport values of oleic acid in all cases indicate a decline in transport rate with increasing air bubbling time, a phenomenon due primarily to the depletion of surfactant from the water column as previously reported [Skop *et al.*, 1991]. We observe that the quantity of surface transport for 35-ppt cleaned artificial seawater is somewhat higher than that for 35-ppt uncleaned artificial seawater. The surface transport for the deionized-distilled water is much smaller than for the other two cases studied.

We can now make some inferences regarding the effects of salinity on the microbubble-mediated scavenging process and on the resulting surface pressure of seawater. The observation of increased surface transport with 35-ppt salinity versus 0-ppt salinity can be explained by noting that the increased ionic content of seawater tends to reduce the solubility of the hydrophobic tail of the oleic acid molecule. Hence the molecule has an increased affinity for the bubble-water interface and is scavenged, or "salted out," more readily than in distilled water [Davies and Rideal, 1963]. The observation that the surface pressures of deionized-distilled water and 35-ppt cleaned artificial seawater are not in great difference (see Figure 2) while the surface transport of oleic acid for the former is half that of the latter can be explained by reference to findings made by D'Arrigo [1984]. He notes that increased salinity tends to disrupt hydrogen bonding between surfactant molecules on the water surface. Hence the surface film monolayer has a greatly expanded film pressure versus area relationship for seawater as compared with distilled water. That is, the same number of surfactant molecules per unit area should

produce less surface pressure on seawater than on distilled water or, equivalently, more surfactant molecules per unit area are needed on seawater than on distilled water to produce the same surface pressure. As was noted previously, the surface pressure for the 35-ppt uncleaned artificial seawater is dominated by non-oleic acid contaminants rather than by the amount of oleic acid transported to the water surface.

The air transport of oleic acid to a 10-cm altitude is shown in Figure 4 as a function of bubbling time. We note that the air transport for 35-ppt cleaned seawater is less than half that of 35-ppt uncleaned seawater even though both have nearly the same surface transport. The air transport for deionized-distilled water is smaller than the other two cases, as was the situation for surface transport.

As mentioned previously, our group [Tseng *et al.*, 1992] has found that bubble transport of surfactant to the air (10-cm altitude) is linearly proportional to the quantity of surfactant transported to the sea surface microlayer. The constant of proportionality appears, in the absence of significant surface tension changes, to be independent of the type of material but is a function of the airflow rate and bubble size. (Our experiments used uncleaned artificial seawater. Hence the surface tension was dominated by extraneous contaminants rather than by the surfactants studied.) This conclusion is again confirmed here by results, plotted in Figure 5, for artificial seawaters of various salinities and contamination at an airflow rate of 4.0 $\text{mL}/(\text{min}\cdot\text{cm}^2)$. The constants of proportionality for these three cases, using least squares fitting to the equation

$$\text{air transport} = C \times \text{surface transport},$$

are summarized in Table 2. Also summarized in Table 2 for each case are the surface pressures after 30 s of bubbling at 4.0 $\text{mL}/(\text{min}\cdot\text{cm}^2)$.

We can now draw some conclusions concerning the effects of surface tension and salinity on the microbubble-mediated sea-to-air transfer of surfactants. Comparing the air transport to surface transport ratios of oleic acid for 35-ppt uncleaned and cleaned artificial seawaters, we see that the former is approximately 3 times the latter. The surface pressure of the 35-ppt uncleaned seawater is also substantially larger than the surface pressure of the 35-ppt cleaned seawater. These results indicate that the surface tension encountered during the bubble-bursting process strongly influences this process.

We can think of two possible explanations for this finding. Assume, as the first possible explanation, that the amount of surfactant carried by a jet droplet is proportional to the amount of surfactant scavenged from solution by a rising bubble (or equivalently, that the proportion of the bubble microlayer skimmed off a collapsing bubble in the droplet formation process is constant). Our results then say that the percentage of this surfactant material reaching an altitude of 10 cm increases as the surface tension decreases. This in turn says, since the droplets are in free flight, that the distribution of initial upward velocities of the droplets formed during bursting increases in magnitude as the surface tension encountered during bursting decreases.

Assume next, as the second possible explanation, that the distribution of initial upward velocities of the droplets formed during bursting is independent of the surface tension encountered during bursting or perhaps even decreases slightly in magnitude as the surface tension encountered during bursting decreases. This assumption in turn requires that the amount of surfactant material carried by a jet droplet increases as the surface tension decreases

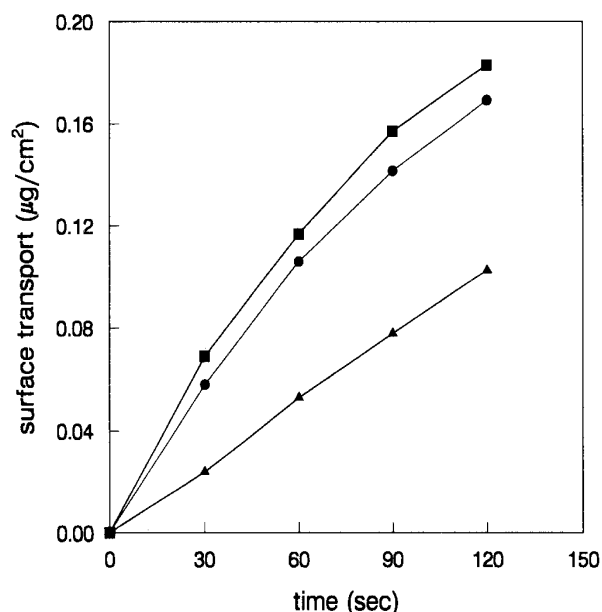


Fig. 3. Cumulative surface transport of oleic acid versus time. The water types are deionized-distilled water (triangles), 35-ppt artificial seawater (circles), and 35-ppt clean artificial seawater (squares). The results are for an airflow rate of 4.0 $\text{mL}/(\text{min}\cdot\text{cm}^2)$.

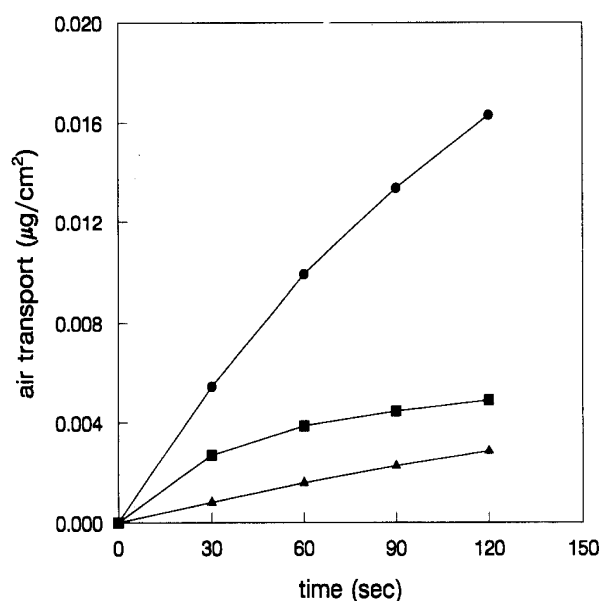


Fig. 4. Cumulative air transport of oleic acid versus time. The water types are deionized-distilled water (triangles), 35-ppt artificial seawater (circles), and 35-ppt clean artificial seawater (squares). The results are for an airflow rate of 4.0 mL/(min-cm²).

TABLE 2. Surface Pressures after Bubbling and Constants of Proportionality Between Air Transport (10-cm Altitude) and Surface Transport

Seawater Type	Π_{oleic} dyn/cm	$C \times 10^2$
Distilled	21.7	2.86
35-ppt cleaned	26.9	2.92
35-ppt uncleaned	40.5	9.53

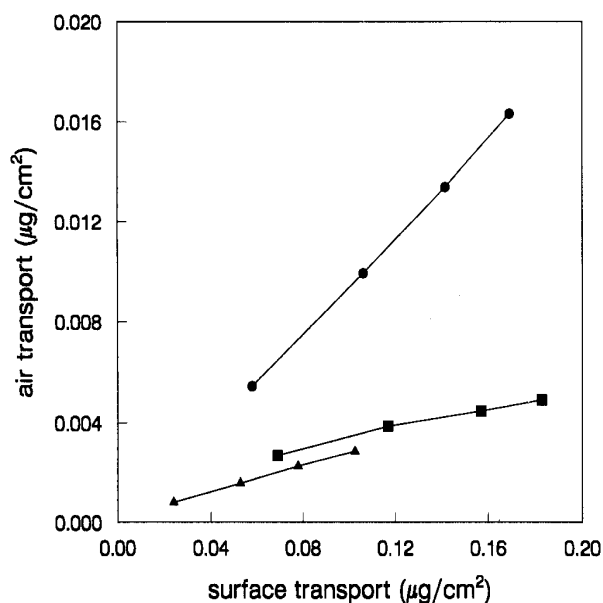


Fig. 5. Variation of cumulative air transport with cumulative surface transport for oleic acid. The water types are deionized-distilled water (triangles), 35-ppt artificial seawater (circles), and 35-ppt clean artificial seawater (squares). The results are for an airflow rate of 4.0 mL/(min-cm²).

(or equivalently, that the proportion of the bubble microlayer skimmed off a collapsing bubble in the droplet formation process increases as the surface tension decreases).

The latter mechanism is supported by the numerical calculations of Peng [1992] and the experimental observations of Blanchard [1990]. Peng studied the effects of surface tension on the dynamics of a jet droplet formed by the collapse of an initial surface depression. He found that the height achieved by the jet droplet, while dependent on the size of the initial surface depression, was only very weakly dependent on the surface tension. Blanchard studied the behavior of jet droplets generated by the impingement of bubbles on a preexisting monolayer of oleic acid. He found that the droplets produced tended to be larger than the droplets produced by the impingement of identical bubbles on a clean interface. This was the result of droplet coalescence in some cases or because of the production of a larger (but much slower) droplet in other cases. It should be noted that the studies of Peng [1992] and Blanchard [1990] dealt only with single bubbles. Hence our conclusion that the primary effect of a decrease in surface tension is to increase the proportion of the microlayer skimmed off a collapsing bubble in the droplet formation process must be regarded as tentative.

Comparing the air transport to surface transport ratios of 35-ppt cleaned artificial seawater and deionized-distilled water, we find the former to be just slightly higher than the latter, while the surface pressure associated with 35-ppt cleaned artificial seawater is 20% higher than the surface pressure associated with distilled water. In this case, however, it is likely that the absence of salinity in the distilled water plays an offsetting role to the increased surface tension in the distilled water in modifying the bubble bursting process. We have observed visually that surface bubbles in the distilled water tend to coalesce with each other before bursting, a phenomenon not readily apparent in the 35-ppt seawaters. Peltzer and Griffin [1987] have noticed an identical tendency in their studies of the effects of salinity on the decay of foam columns. Such coalescence, assuming conservation of total microlayer surfactant material, would tend to result in a higher concentration of surfactant in the microlayer of the coalesced bubble than in the microlayers of the coalescing bubbles. (For example, consider two bubbles having identical volumes V and identical surface microlayer concentrations Γ , in grams per square centimeters, of surfactant material. Assuming conservation of total microlayer surfactant material, the coalesced bubble of volume $2V$ would then have a surface microlayer concentration of 1.26Γ .)

Acknowledgments. The support of this research was provided by the Office of Naval Research under contracts N-00014-89-J-1354 and N-00014-91-J-1171 and by the U.S. Naval Research Laboratory under contracts N-00014-89-J-2008 and N-00014-91-K-2013. We gratefully acknowledge the help of William Barger of the Naval Research Laboratory who generously sent us a set of calibrated spreading oils and Chen Peng who assisted us in conducting parts of these experiments. Ruo-Shan Tseng also wishes to acknowledge the support provided by a Rosenstiel Postdoctoral Fellowship of the University of Miami. The comments of the reviewers concerning the original version of this manuscript were most helpful and insightful and their efforts are appreciated.

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(Received May 11, 1992;
revised January 13, 1993;
accepted January 14, 1993.)