The potential role of sea spray droplets in facilitating air-sea gas transfer

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Abstract. For over 30 years, air-sea interaction specialists have been evaluating and parameterizing the role of whitecap bubbles in air-sea gas exchange. To our knowledge, no one, however, has studied the mirror image process of whether sea spray droplets can facilitate air-sea gas exchange. We are therefore using theory, data analysis, and numerical modeling to quantify the role of spray on air-sea gas transfer. In this, our first formal work on this subject, we seek the rate-limiting step in spray-mediated gas transfer by evaluating the three time scales that govern the exchange: \( \tau_{\text{air}} \), which quantifies the rate of transfer between the atmospheric gas reservoir and the surface of the droplet; \( \tau_{\text{int}} \), which quantifies the exchange rate across the air-droplet interface; and \( \tau_{\text{aq}} \), which quantifies gas mixing within the aqueous solution droplet.

1. Introduction

Early in the study of air-sea gas transfer—roughly 35 years ago—ocean scientists began considering bubble-mediated transfer as a way to explain the nonlinear increase in the gas transfer velocity that becomes apparent when the 10-m wind speed \( (U_{10}) \) reaches about 10 m s\(^{-1}\) (Thorpe 1982; Merlivat and Memery 1983; Monahan and Spillane 1984). After all, stage-B whitecaps, which are the source of most ocean bubbles, reach a fractional ocean coverage of 1% at \( U_{10} = 10 \text{ m s}^{-1} \), and that coverage increases faster than the cube of the wind speed (Monahan and O’Muircheartaigh 1980). In the intervening years, many others took up the study of bubble-mediated gas transfer (e.g., Woolf and Thorpe 1991; Wallace and Wirick 1992; Keeling 1993; Woolf 1993, 2005; Asher et al. 1996; Monahan 2002; Vlahos and Monahan 2009; Vlahos et al. 2011).

Surprisingly few, however, have considered what appears to be the mirror-image process of bubble-mediated air-sea gas transfer—spray-mediated transfer. We therefore begin here a formal treatment of how and whether sea spray might influence the rate of air-sea gas transfer. We will generally follow the approach that Andreas and colleagues used for assessing how sea spray affects air-sea heat and moisture transfer (Andreas 1992, 2010; Andreas and DeCosmo 2002; Andreas et al. 1995, 2015). Microphysical modeling of the temperature and radius evolution of individual spray droplets underlies most of this work. In turn, we will begin our study of spray-mediated gas transfer by first studying the microphysical, thermodynamic, and gas transfer properties of individual spray droplets. We quantify these processes with time scales that ultimately reveal where the rate-limiting step is for spray-mediated gas transfer and, consequently, inform our decision on how next to proceed.
Figure 1. Gas transfer processes near the air-sea interface. The figure depicts three routes for gas transfer: interfacial exchange, which follows the sea-air difference in the partial pressure of the gas, $\Delta p$; bubble-mediated exchange; and spray-mediated exchange, in which droplet size is a crucial parameter.

Figure 1 is a schematic overview of air-sea gas transfer. The air and sea are always exchanging gases across the interface as a consequence of Henry’s law equilibrium and the differences in partial pressures of the gases in the sea and in the near-surface air ($\Delta p$). This interfacial exchange goes approximately linearly with wind speed (Jähne et al. 1987; Woolf 2005). The copious bubbles that begin to form once whitcapping begins when the 10-m wind speed reaches about 4 m s$^{-1}$ provide additional contact between air and sea and affect the gas transfer rate. Because whitecap coverage increases approximately with the cube of the wind speed, bubble-mediated gas transfer is presumed to go roughly as the cube of the wind speed (Monahan and Spillane 1984; Monahan 2002; Woolf 2005).

Also coincident with the onset of whitcapping is sea spray production. As with bubbles, spray is intimately tied to wave breaking; the number of spray droplets produced therefore also increases as roughly the third power of wind speed. In most marine environments, spray droplets cool rapidly to a temperature lower than the air thermometer. They also evaporate slowly, thereby losing fresh water and becoming more saline.

Figure 1 identifies two classes of spray droplets: small and large. The distinction is relative, not absolute, as we will explain later. Small droplets remain suspended long enough to come to radius (evaporative) equilibrium with the environment; large droplets fall back into the sea before reaching radius equilibrium—these are re-entrant spray droplets.

The small droplets generally transport gas from the ocean to the atmosphere because they lose fresh water, become more saline, and therefore become an increasingly inhospitable environment for dissolved gases. If the near-surface relative humidity is 75% or greater, the efflorescence point for saline droplets, these droplets reach equilibrium as aqueous solution droplets. In an environment with
relative humidity less than 75%, these small droplets ultimately lose all their water and all their gas to the atmosphere and leave behind just a dry salt crystal.

While large droplets carry most of the mass in the spray, they cannot participate fully in air-sea gas transfer because of limits on their lifetime. They return to the ocean before they become hypersaline and, thus, can exchange gas either to or from the ocean depending, mostly, on the sign of $\Delta p$. The time scales that govern the rates for these various processes are our subject here.

2. Theoretical background

2.1. Spray droplet evolution

In the early stages of evolution, both droplet temperature ($T$) and radius ($r$) approximately follow exponential relations:

$$
\frac{T(t) - T_{eq}}{T_w - T_{eq}} = \exp(-t/\tau_{T}),
$$

$$
\frac{r(t) - r_{eq}}{r_0 - r_{eq}} = \exp(-t/\tau_{r}).
$$

Here, $t$ is the time since the droplet formed; $T_w$, the sea surface temperature (the initial droplet temperature); $r_0$, the initial droplet radius; $T_{eq}$ and $r_{eq}$, the droplet’s equilibrium temperature and radius; and $\tau_T$ and $\tau_r$, the corresponding e-folding times to reach these equilibrium values. Later figures show values of $\tau_T$ and $\tau_r$. The microphysical quantities $T_{eq}$, $r_{eq}$, $\tau_T$, and $\tau_r$ all depend on the initial droplet radius, temperature, and salinity and on environmental conditions like air temperature, relative humidity, and barometric pressure.

2.2. Gas time scales

In the context of the time scales $\tau_T$ and $\tau_r$, over which spray droplets themselves evolve, we turn next to consider the time scales that govern how gas molecules interact with water droplets. The cloud physics literature—namely, the books by Seinfeld and Pandis (2006), Pruppacher and Klett (2010), and Lamb and Verlinde (2011) — have provided the background for this work.

Figure 2 schematically shows how an individual spray droplet may exchange an arbitrary gas with an atmospheric reservoir. Although spray-mediated transfer can involve both invasion and evasion, we suspect evasion will be the primary direction for spray-mediated air-sea gas transfer.

The Henry’s law coefficient ($K_H$) is crucial in all studies of gas transfer across an air-water interface. We define $K_H$ for spray droplet geometry according to the relation

$$
C_d(r_0) = K_H p_g(r_0).
$$

Here, $p_g(r_0)$ is the partial pressure (or fugacity) of arbitrary gas $g$ at the external surface of a droplet, and $C_d(r_0)$ is the concentration of the gas just inside the droplet (see figure 2). Our preferred units for $p_g$ and $C_d$ are atm and mol L$^{-1}$, respectively. Hence, $K_H$ has units of mol L$^{-1}$ atm$^{-1}$.

To put all gas quantities in (3) in terms of a concentration, we use the ideal gas law to convert $p_g$ to the concentration of the gas in air ($C_a$). Equation (3) thus becomes

$$
C_d(r_0) = K_H R T C_a(r_0),
$$

where $C_a$ also has units of mol L$^{-1}$. Also in (4), $R$ ($= 8.20574 \times 10^{-2}$ atm L mol$^{-1}$ K$^{-1}$) is the universal gas constant, and $T$ is the kelvin temperature. Notice, the product $K_H R T$ is dimensionless. Equation (4) explains the apparent discontinuity in concentration in figure 2 at the surface of the spray droplet.
All three of our primary sources — Seinfeld and Pandis (2006, pp. 556–557), Pruppacher and Klett (2010, p. 764), and Lamb and Verlinde (2011, p. 508) — estimate the time scale $\tau_{aq}$ that quantifies mixing inside the aqueous solution droplet (figure 2) as

\[ \tau_{aq} = \frac{r_0^2}{\pi^2 D_{g,sw}}, \]  

where $D_{g,sw}$ is the molecular diffusivity of gas $g$ in seawater.

Using an approach outlined by Pruppacher and Klett (2010, p. 761), we derive estimates for $\tau_{air}$ and $\tau_{int}$ of

\[ \tau_{air,PK} = \frac{r_0^3 K_H RT_d}{3 D_g (r_0 + \Delta_g)} \]  

and

\[ \tau_{int,PK} = \frac{r_0 K_H RT_d}{3 \alpha} \left( \frac{2 \pi M_g}{RT_a} \right)^{1/2}. \]

In these, $D_g$ is the molecular diffusivity of the gas in air; $M_g$ the molecular weight of the gas; $T_a$ the air temperature; and $T_d$ the droplet temperature (assumed uniform in this derivation). If the droplet is...
small enough, the air and gas may no longer behave as continuous fluids; the \( \Delta \) in (6), the gas “jump” length, which is approximately the mean free path of the gas in air (\( \lambda_g \); see figure 2), determines what “small enough” is. Gas molecules cross this jump length as dictated by the kinetic theory of gases (e.g., Bohren and Albrecht 1998, pp. 188, 234–235). Thus, in (7), we recognize \( \left( \frac{2\pi M_g}{RT_a} \right)^{1/2} \) as \( 4/\bar{v}_g \), where \( \bar{v}_g \) is the average speed of an ideal gas molecule according to the Maxwell–Boltzmann speed distribution (Bohren and Albrecht 1998, pp. 60–64).

Finally, in (7), \( \alpha \) is the mass accommodation coefficient of the gas: the ratio of the number of gas molecules that stick to a droplet over the number that strike the droplet. The values of \( \alpha \) for our six gases are still emerging, particularly in the case of sea spray where the evaporative flux will require consideration of significant changes in the efflux rates as the drop shrinks. In a comparable microphysical model for heat and water vapor exchange with a spray droplet, Andreas (1989, 1990) used \( \alpha = 0.036 \) for water vapor (Pruppacher and Klett 2010, table 13-1). Based on Davis (2006), values of unity are appropriate for condensation coefficients at relatively low temperatures such that re-emission of the gas does not occur, but the emerging accommodation coefficient (what leaves the droplet) is the most appropriate scaler for a spray model. It is important to note that values of unity are based on the assumption that the Knudsen number is sufficiently large, though this may not hold in a shrinking droplet (Davis 2006). Also, values based on isolated droplets tend to overestimate evaporation rates by at least 65% (Devarakonda et al. 1998) avoiding attenuation due to droplet interactions. In the absence of compelling evidence for a definitive choice, we conservatively apply both 0.036 and 1.0 across all gases and evaluate the sensitivity of this parameter in the interpretation of our results.

Although Lamb and Verlinde (2011, p. 507) derive the same estimate as (7) for the interfacial time scale (i.e., \( \tau_{int,LV} = \tau_{int,PK} \)), they ignore the jump layer and, thereby, derive as the time scale in air [cf. (6)]

\[
\tau_{air,LV} = \frac{r_0^2 \ K_H \ RT_d}{3 D_g}. \tag{8}
\]

In contrast with (6) and (8), Seinfeld and Pandis (2006, pp. 538, 549–551, 580–582) assume that the gas concentration at the external surface of the spray droplet is constant with time and thereby estimate the gas time scale in air as

\[
\tau_{air,SP} = \frac{r_0^2}{4 D_g}. \tag{9}
\]

But they do a much more interesting derivation of the interfacial time scale, \( \tau_{int} \).

On allowing the gas concentration in the droplet to vary with radial position, Seinfeld and Pandis (2006, pp. 551–554) derive

\[
\tau_{int,SP} = \frac{r_0^2}{\beta_1^2 D_g,sw}, \tag{10}
\]

where \( \beta_1 \) is the first positive root of

\[
\beta_4 \cos(\beta_4) + L = 0. \tag{11}
\]

In (11),

\[
L = \frac{r_0 H}{D_g,sw} - 1, \tag{12}
\]

where
For large values of \( L \), nominally above 100, \( \beta_i \) from (11) is \( \pi \) (e.g., Carslaw and Jaeger 1996, p. 492) and

\[
\lim_{L \to \infty} \tau_{\text{int,SP}} = \frac{r_0^2}{\alpha^2 D_{g,sw}}. \tag{14}
\]

We will see this case often in our subsequent calculations because \( L \) is large when \( r_0 \) is large, \( \alpha \) is large, or the gas is not very soluble (i.e., small \( K_{ij} \)). Of our six gases, all except carbon dioxide would be termed only weakly soluble. Notice also that (5) and (14) are identical.

Equations (5) and (10) follow from the assumption that the fluid within the droplets is motionless; gas molecules thus would move only through molecular diffusion, \( D_{g,sw} \). Ample evidence exists, however, that the fluid, even in small droplets, develops circulations when a shear stress occurs at the droplet’s surface (Clift et al. 1978, pp. 36–38, 127–129; Pruppacher and Klett 2010, pp. 386–393). Consequently, mixing of the gas within a droplet and across its interface is surely much faster than \( \tau_{aq} \) and \( \tau_{\text{int,SP}} \) suggest. We thus consider (5) and (10) as upper bounds for these time scales.

### 2.3. Droplet residence time

A droplet’s residence time in air provides the context for all of these time scales. In short, a spray droplet can exchange heat, water vapor, and gases with air only during its (often brief) lifetime between creation and re-entry into the ocean.

Because spume droplets, which are created by the wind’s tearing droplets right off the wave crests, accomplish most of the spray-mediated heat and moisture exchange, Andreas and colleagues (Andreas 1992; Andreas and DeCosmo 2002; Andreas et al. 2015) based a residence time scale on the behavior of these droplets:

\[
\tau_f = \frac{H_{1/3}/2}{u_f(r_0)}. \tag{15}
\]

Here, \( u_f(r_0) \) is the terminal fall velocity of droplets of radius \( r_0 \). Also, \( H_{1/3} \) is the significant wave height; consequently, \( H_{1/3}/2 (= A_{1/3}) \) is the significant wave amplitude, the height above mean sea level where many spray, and most spume, droplets originate. Equation (15), in essence, models the residence time in air of a droplet undergoing ballistic flight.

### 3. Spray droplet time scales

Figure 3 compares these several time scales over a range of radii for one set of environmental conditions and for two gases: helium and carbon dioxide. In both plots, the \( \tau_f \), \( \tau_g \), and \( \tau_r \) curves are the same because these times do not depend on the gas.

We selected helium (\( M_g = 4 \text{ g mol}^{-1} \)) and carbon dioxide (\( M_g = 44 \text{ g mol}^{-1} \)) for display in figure 3 because their molecular masses span the range from smallest to largest in our set of six gases and thereby demonstrate how the size of the gas molecule influences \( \tau_{aq}, \tau_{\text{int}}, \) and \( \tau_{sp} \).

This is how we interpret figure 3. Any spray-mediated transfer must occur in a time less than the droplet’s residence time, \( \tau_f \). This time scale decreases as the radius increases because \( u_f \) increases with radius. The \( \tau_f \) curves in figure 3 are for a 10-m wind speed of 12 m s\(^{-1}\); the \( \tau_f \) curve will rise for increasing wind speed (because \( H_{1/3} \) increases), allowing more spray-mediated exchange to occur. The \( \tau_f \) curve in figure 3, as an example, is above \( \tau_r \) for radii up to about 30 \( \mu \text{m} \). Droplets of 30 \( \mu \text{m} \) and smaller thus can reach radius equilibrium and become more saline before they fall back into the ocean. Droplets larger than 30 \( \mu \text{m} \), where \( \tau_f < \tau_r \), on the other hand, fall back into the water before losing...
much water and, consequently, return to the ocean with approximately their original salinity. A droplet’s temperature evolution is much faster. Figure 3 shows that droplets up to about 200 µm in radius have time to reach an equilibrium temperature of $T_{eq}$ at this wind speed before they fall back into the sea.

Our several estimates of gas time scales are comparable to and even shorter than $\tau_f$. At first sight, we might say mixing of gas molecules within a spray droplet—parameterized as $\tau_{aq}$—is the slow step in spray-mediated gas transfer because, at least in these two figures, it is the largest of the gas time scales. (In figure 3, $\tau_{aq} = \tau_{int,SP}$. More on this shortly.) But remember, $\tau_{aq}$ is a maximum value that does not recognize any fluid motion in a spray droplet that would certainly enhance gas mixing by at least an order of magnitude. Consequently, we suspect that the true $\tau_{aq}$ will be at least ten times shorter than depicted.

For the air time scales, $\tau_{air}$, in figure 3, we show only $\tau_{air,PK}$ (6), because it is near the Lamb and Verlinde (2011) scale (8) and because the Seinfeld and Pandis (2006) scale (9) is based (unrealistically) only on molecular diffusion of gas molecules to and from a spray droplet. Notice in figure 3 that $\tau_{air,PK}$ is orders of magnitude less than $\tau_f$, which we consider to represent very fast exchange. $\tau_{air,PK}$ does, however, increase by roughly three orders of magnitude between helium and carbon dioxide as a consequence of how increasing molecular weight slows molecular velocities in air.

The final time scale shown in figure 3 quantifies exchange across a droplet’s interface, and we represent this exchange with both $\tau_{int,PK}$ (7), and $\tau_{int,SP}$, (10). For helium and carbon dioxide and our choice of the accommodation coefficient, 0.036, $\tau_{int,SP}$ has the same calculated value as $\tau_{aq}$—compare (5) and (14). For an accommodation coefficient of 1 the $\tau_{int,SP}$ becomes 2 orders of magnitude smaller. And because $\tau_{int,SP}$ is formulated in terms of the molecular diffusivity in the spray droplet, $D_{g,sw}$, we expect it to be lower, too, if we acknowledge the likelihood of fluid motion within spray droplets. The second interfacial time scale, $\tau_{int,PK}$, (7), is orders of magnitude less than the Seinfeld and Pandis (2006) prediction, $\tau_{int,SP}$. A mass accommodation coefficient of unity further reduces the $\tau_{int,PK}$ by a factor of 30. And although $\tau_{int,PK}$ does increase with the increasing molecular weight of the gas molecule, it always represents extremely fast gas transfer over the chosen mass accommodation coefficient range.

4. Conclusions

Gas molecules transit between the deep interior of a sea spray droplet and an atmospheric reservoir in five distinct steps. 1) The molecules must diffuse between the deep interior of the droplet and its interior surface (time scale $\tau_{aq}$), 2) must cross the air-droplet interface (time scale $\tau_{int}$), and 3) the molecules must move between the ambient atmosphere and the exterior surface of the droplet (time scale $\tau_{air}$). Within spray droplets, 4) some gas molecules will dissociate, and 5) some will undergo chemical reactions.

Of the six gases we are currently considering, all must complete the first three steps for spray-mediated transfer to occur; but only carbon dioxide participates in the additional fourth and fifth steps—dissociation and chemical reactions. Hence, for helium, neon, argon, oxygen, and nitrogen, the time scales we have evaluated are sufficient for modeling their role in spray-mediated transfer. In future work, we will consider how the chemical behavior of carbon dioxide influences the rate at which it undergoes spray-mediated exchange.

We found several opinions as to how to parameterize the time scales for transfer in the air, $\tau_{air}$, and transfer across the air-droplet interface, $\tau_{int}$. For $\tau_{air}$, we view the time scale based on Pruppacher and Klett’s (2010) approach ($\tau_{air,PK}$) as most appropriate because it is similar to Lamb and Verlinde’s (2011) result and more physical that Seinfeld and Pandis’s (2006). In contrast, the interfacial time scale from Seinfeld and Pandis ($\tau_{int,SP}$) seems more appropriate than Pruppacher and Klett’s because it more realistically treats gas diffusion within spray droplets. Meanwhile, all our main sources agree on the aqueous time scale, $\tau_{aq}$, that parameterizes diffusive mixing within droplets.
Figure 3. For droplets of radius $r_0$, the figure shows the time scales for temperature ($\tau_T$) and radius ($\tau_r$) evolution, the droplet residence time ($\tau_f$), and the time scales that govern transfer of helium (left panel) and carbon dioxide (right panel) molecules to or from a spray droplet in air ($\tau_{\text{air}}$), across the air-droplet interface ($\tau_{\text{int,PK}}$) and within the droplet ($\tau_{\text{aq}}$). The interfacial time scale derives from both Pruppacher and Klett (2010; $\tau_{\text{int,PK}}$), equation (7), and Seinfeld and Pandis (2006; $\tau_{\text{int,SP}}$), equation (10). The time scale in air derives from Pruppacher and Klett ($\tau_{\text{air,PK}}$), equation (6); the aqueous mixing time scale ($\tau_{\text{aq}}$) is (5). Conditions here are for a 10-m wind speed of 12 m s$^{-1}$, air temperature and water temperature of 20°C, relative humidity of 85%, and sea surface salinity of 34 psu; the mass accommodation coefficient is set at $\alpha = 0.036$. For this value, $\beta_1$ in the Seinfeld and Pandis $\tau_{\text{int,SP}}$ is $\pi$, and $\tau_{\text{int,SP}}$ is the same as $\tau_{\text{aq}}$.

A caveat regarding $\tau_{\text{int,SP}}$ and $\tau_{\text{aq}}$ is that mixing within spray droplets likely also results from fluid motions, not just from molecular processes quantified by the molecular diffusivity of a gas in seawater, $D_{g,sw}$. In light of this physics, both exchange across the droplet interface and within a droplet could be one or more orders of magnitude faster than estimated from $\tau_{\text{int,SP}}$ and $\tau_{\text{aq}}$.

As a consequence, $\tau_{\text{air}}$ and our revised estimates of $\tau_{\text{int,SP}}$ (regardless of mass accommodation coefficient values 0.036 or 1.0) and $\tau_{\text{aq}}$ are all shorter than the microphysical time scales $\tau_T$ and $\tau_r$ that quantify a spray droplet’s physical evolution. Hence, to estimate spray-mediated gas transfer, we can reasonably assume that the gas in a spray droplet is in instantaneous equilibrium with the atmospheric reservoir. The gas concentration in the droplet will therefore evolve as the radius, temperature, and salinity of the droplet evolve. Thus our next step is to track simultaneously this gas concentration for a spectrum of droplets as they are injected into the near-surface marine atmosphere and evolve according to the Andreas (1989, 1990) microphysical model.

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