An Eulerian model for sea spray transport and evaporation

Fabrice Veron¹ and Luc Mieussens²

¹School of Marine Science and Policy, University of Delaware, Newark DE, USA ²Bordeaux INP, Univ. Bordeaux, CNRS, IMB, UMR 5251, F-33400 Talence,France

(Received xx; revised xx; accepted xx)

Reliable estimates of the fluxes of momentum, heat, and moisture at the air-sea interface are essential for accurate long term climate projections, as well as the prediction of short term weather events such as tropical cyclones. In recent years, it has been suggested that these estimates need to incorporate an accurate description of the transport of sea spray within the atmospheric boundary layer and the drop-induced fluxes of momentum, heat, and moisture, so that the resulting effects on the atmospheric flow can be evaluated. In this paper we propose a model based on a theoretical and mathematical framework inspired from kinetic gas theory. This approach reconciles the Lagrangian nature of the spray transport with the Eulerian description of the atmosphere. In turn, this enables a relatively straightforward inclusion of the spray fluxes and the resulting spray effects on the atmospheric flow. A comprehensive dimensional analysis has led us to identify the spray effects that are most likely to influence the speed, temperature, and moisture of the airflow. We also provide an example application to illustrate the capabilities of the model in specific environmental conditions. Finally, suggestions for future work are offered.

1. Introduction

The fluxes of momentum, heat, and moisture at the air-sea interface are crucial ingredients in the overall energy and mass balances of both the oceans and the atmosphere. As such, these air-sea fluxes are important boundary conditions for atmospheric and oceanic models that attempt to capture the physics and evolution of weather and climate. These fluxes can be relatively well estimated in moderate wind speed conditions using so-called bulk parameterizations, which are extensions of the classical turbulent boundary layer flow theory applied to the air-sea interface (e.g. Fairall *et al.* 1996). However, the presence of surface waves and, in particular, breaking events give rise to phenomena such as bubble injections and turbulence generation on the waterside, and airflow separation and spray generation on the airside. In turn, these phenomena cause the flows to depart from the classical boundary layer descriptions (e.g. Thorpe 1993; Melville 1994; Anis & Moum 1995; Melville 1996; Terray et al. 1996; Veron & Melville 2001; Hristov et al. 1998; Hare et al. 1997; Adrian 1991; Edson & Fairall 1998; Donelan et al. 2004). Consequently, in conditions when surface wave breaking is pervasive, be it because of intense winds or the presence of coastlines, air-sea fluxes are not fully resolved. In fact, in the past decade, measurements at high wind speeds confirm that simple air-sea flux parameterizations are still lacking for conditions in which breaking is ubiquitous (Drennan *et al.* 2007; Zhang et al. 2008; Bell et al. 2012).

The knowledge gap in high winds can be, in part, attributed to our incomplete understanding of the role of sea spray. Indeed, in strongly forced conditions, it is well known that significant amounts of sea spray are generated at the surface of the ocean and get subsequently transported into the marine atmospheric boundary layer. In turn, these spray drops are believed to affect the multiple air-sea fluxes. However, there are still significant uncertainties as to the amount of momentum, heat, and moisture that the drops exchange with the atmosphere while suspended (Andreas 1990; Veron 2015). In this paper, we develop a model that accounts for the direct effects of the spray on the atmosphere.

Sea spray is composed of seawater drops ejected from the sea surface, generally due to breaking waves and breaking related phenomena such as bubble entrainment and whitecaps. Spray is mainly formed through two pathways. The first occurs when bubbles entrained by breaking waves rise to the surface and burst (see Lewis & Schwartz 2004; de Leeuw et al. 2011). When the bubble surface shatters, small film drops with radii less than $O(1) \ \mu m$ are formed (Spiel 1998; Fuentes *et al.* 2010). The subsequent collapse of the cavity that is leftover leads to the ejection of so-called jet drops with radii of order $O(1-40) \ \mu m$ (Wu 2002; Walls et al. 2015; Wang et al. 2017). The second process occurs when the wind is sufficiently strong to tear off water from the top of waves or from splashing from plunging breaking waves. Early measurements by Koga (1981) showed that globules or filaments, produced on the front face of the waves, breakup under the shearing effect of the wind and generate a distribution of large drops (Marmottant & Villermaux 2004; Mueller & Veron 2009b). These are referred to as spume drops (Andreas et al. 1995; Veron 2015). This filament breakup process is likely accompanied by a bag breakup phenomenon whereby small water canopies are inflated by the wind and shatter (Veron et al. 2012; Troitskaya et al. 2018). The splash production of drops from plunging breaking waves has only been recently investigated (Erinin et al. 2019). In general, spume and splash drops have radii larger than $O(20) \ \mu m$.

Once ejected from the ocean surface, these drops are transported and dispersed in the atmospheric boundary layer where they interact with the turbulent airflow and exchange momentum, heat, and moisture with the atmosphere (e.g. Fairall *et al.* 1994; Edson *et al.* 1996; Andreas & DeCosmo 1999; Andreas 2002; Andreas *et al.* 2008; Mueller & Veron 2014b). It is through these exchanges that sea spray is believed to play a critical role in the overall air-sea heat and moisture fluxes which fuels the intensity of tropical cyclones (Bao *et al.* 2000; Andreas & Emanuel 2001; Andreas 2004; Andreas *et al.* 2008, 2010; Liu *et al.* 2010; Bianco *et al.* 2011; Rosenfeld *et al.* 2012).

In order to quantify the spray effects on the air-sea fluxes, both an accurate quantification of the source of spray, and a suitable knowledge of the thermodynamics and transport of aqueous drops in air, are required. With regards to the thermodynamics, seminal works by Pruppacher & Klett (1978) and Andreas (1990) have guided numerous studies, (e.g. Rouault et al. 1991; Andreas 1992; Edson & Fairall 1994; Andreas et al. 1995; Edson et al. 1996; Mestayer et al. 1996; Makin 1998; Andreas & Emanuel 2001; Van Eijk et al. 2001; Lewis & Schwartz 2004; Barenblatt et al. 2005; Mueller & Veron 2010b,a; Bao et al. 2011; Mueller & Veron 2014a, b). Likewise, in order to assess the momentum exchanges between drops and the airflow, comprehensive knowledge of the drop dynamics at ejection and during its flight through the turbulent atmosphere is needed. Nowadays, a drop's thermodynamics and its (Lagrangian) dynamics are quite well understood and modeled. Thus, a complete assessment of the spray-mediated air-sea fluxes relies on the proper estimate of the amount of spray present in, and ejected into, the airflow. This is typically quantified using the so-called sea spray generation function, or spray source function, i.e. the number distribution of spray drops generated at the ocean surface (per unit surface area and unit time). However, this source function has proven difficult to directly measure, especially for the larger drops which may reside within a wave height of the surface and thus cannot be easily detected by typical fixed height measurements. Consequently, despite recent laboratory measurements (Veron *et al.* 2012; Ortiz-Suslow *et al.* 2016; Troitskaya *et al.* 2018), theoretical estimates and parameterizations of the spray generation function are not well constrained, particularly for field applications and large drop radii. Yet, these large and heavy drops may have the largest potential for exchanging momentum with the wind (Andreas 1998; Fairall *et al.* 2009). Small film drops with radii smaller than $O(1) \mu m$ eventually partake in the global marine aerosol flux but are expected to have a minimal influence on the overall heat and momentum air-sea fluxes (Andreas 1990). Accordingly, in this paper, we consider spray drops with radii in the range $O(1) \mu m$ to O(1) mm.

Overall, the currently incomplete quantification of the spray source function (or equivalently the spray concentration and velocity distributions) is a substantial limiting factor in properly assessing the impact of sea spray on the total air-sea fluxes of heat, moisture, and momentum. In this paper, however, we do not focus on the details of the spray generation process, or the spray generation function and its shortcomings. The generation function serves as a boundary condition for the model proposed therein, and the choice of which generation function is best suited for a particular set of conditions is left to the user.

Still, based on spray concentration functions commonly used (e.g. Fairall *et al.* 1994; Andreas 2002; Andreas *et al.* 2010) it is possible to estimate, even if roughly, a number of useful parameters. For example, one can estimate that even in wind speed conditions reaching approximately 50 ms^{-1} , the total spray volume fraction (i.e. the volume occupied by the liquid drops in a unit volume of air) is on the order of $O(10^{-5})$ †. In these conditions, drop collisions and coalescence are expected to be extremely improbable. However, because mass and heat capacity of water drops are larger than that of the air, estimates of the spray mass and heat content ratios show that they can reach up to O(1)% and O(10)% respectively. This simply means that while the spray drops occupy a negligible volume, they carry a significant amount of the total mass and heat contained in the air-spray mixture. Thus, one should expect that the spray could indeed play a substantial role in the total air-sea momentum, heat, and moisture fluxes.

As hinted above, an additional major challenge associated with modeling spraymediated exchanges is the Lagrangian nature of transport of the drops through the atmosphere, whereas the spray concentration function and the airflow are traditionally described and modeled in an Eulerian frame of reference (Edson & Fairall 1994; Edson *et al.* 1996; Mueller & Veron 2009*a*, 2010*a*; Shpund *et al.* 2011, 2012; Richter & Sullivan 2013*b*,*a*; Helgans & Richter 2016; Richter & Sullivan 2014; Peng & Richter 2017, 2019).

In this paper, we present a theoretical framework, originally developed in the context of kinetic gas theory, with which we reconcile the Eulerian and Lagrangian approaches. This kinetic ("discrete mechanics") technique is also, in part, motivated and supported by the spray volume concentration estimate above which indicates that in most conditions, the spray drop concentration is so-called "thin". We believe that the model presented here will prove useful in simulating the transport of spray drops and the exchanges of momentum, heat, and moisture between the drops and the atmosphere.

 $[\]dagger$ Note that the spray generation function of Fairall *et al.* (1994) or Andreas (2002) have not been validated at such high wind speeds; however, they are generally regarded as reliable and typically serve as a reference. As such, the order of magnitude estimate presented here still provides informative overview.

2. Sea spray evolution equations

To be able to describe the influence of the sea spray on the atmosphere, both the motion and the thermodynamics of single drops need to be examined. This is inherently a Lagrangian approach where we assume that each spray drop is made of a single fluid with density ρ_p , and is spherical with radius r_p and mass $m_p = \rho_p \frac{4}{3}\pi r_p^3$. Each drop is then uniquely defined by its position $\boldsymbol{x}_p \in \mathbb{R}^3$; its velocity $\boldsymbol{v}_p \in \mathbb{R}^3$, its radius $r_p \in [0, +\infty[$, its mass $m_p \in [0, +\infty[$, and its temperature $T_p \in [0, +\infty[$. We note that the drop mass and radius are independent variables because sea spray drops are made of sea water but evaporation only transfers pure water (water vapour) between the spray drop and the air. Consequently, at a particular time, given only the drop radius but ignoring its time history, one cannot state precisely the drop mass. For instance, a small drop could be newly formed and have the density of sea water ρ_{sw} , or it could be a large drop that has substantially evaporated, and thus would be saltier with a density superior to that of sea water.

2.1. Motion of individual spray drops

The motion of individual spray drops is given by:

$$\frac{d\boldsymbol{x}_p(t)}{dt} = \boldsymbol{v}_p(t) \tag{2.1}$$

$$m_p(t)\frac{d\boldsymbol{v}_p(t)}{dt} = \boldsymbol{\mathcal{F}}(t).$$
(2.2)

The first equation relates the location of the drop $x_p(t)$ to its velocity $v_p(t)$. The second equation is Newton's Second law where \mathcal{F} accounts for gravity and air frictional effects

$$\boldsymbol{\mathcal{F}} = m_p \boldsymbol{g} + \boldsymbol{F} \tag{2.3}$$

but does not include higher order effects such as the Basset history and the added mass terms, or Faxen, Saffman, and Magnus effects. These equations follow individual drops and are therefore *Lagrangian* in nature. Gravity is noted g and ρ is the density of the (moist) air. The drag force exerted by the fluid on the drop is described with a general drag law

$$\boldsymbol{F} = \frac{1}{2} \rho C_{Dp} A \left(\boldsymbol{u} - \boldsymbol{v}_p \right) \left| \boldsymbol{v}_s \right|$$
(2.4)

where \boldsymbol{u} is the velocity of the air (at the location of the drop as if it were absent), C_{Dp} is the drag coefficient of the drop and $A = \pi r_p^2$ its cross sectional area. This drag coefficient generally depends on the drop Reynolds number $Re_p = \frac{2r_p|\boldsymbol{v}_s|}{\nu}$, with $\boldsymbol{v}_s = \boldsymbol{v}_p - \boldsymbol{u}$, usually referred to as the slip velocity, and $\nu = \mu/\rho$ the kinematic viscosity of air. It is convenient to express the drag coefficient as $C_{Dp} = \frac{24}{Re_p} \boldsymbol{\Xi}$, where $\boldsymbol{\Xi}$ is an empirical correction factor which accounts for departures from the Stokes flow as Re_p increases and the flow around drops becomes turbulent (e.g. Clift & Gauvin 1970).

Equivalently, one can express the viscous drag force in a more compact form

$$\boldsymbol{F} = \frac{m_p}{\tau_D} \left(\boldsymbol{u} - \boldsymbol{v}_p \right) \tag{2.5}$$

where τ_D is the drop response time to the drag force. In this case, just like the drag coefficient, τ_D can be expressed as $\tau_D = \frac{\tau_p}{\Xi}$ where $\tau_p = \frac{2r_p^2\rho_p}{9\mu}$ is the drop Stokes time scale. Naturally, in Stokes flow with $Re_p \ll 1$, $\Xi \to 1$, $\tau_D \to \tau_p$, $C_{Dp} \to \frac{24}{Re_p}$ and the viscous drag $\mathbf{F} \to \frac{m_p}{\tau_p} (\mathbf{u} - \mathbf{v}_p) = 6\pi r_p \mu (\mathbf{u} - \mathbf{v}_p)$. Non-continuum effects can also be accounted for in the correction factor Ξ through the use of a Cunningham slip coefficient

(Davies 1945), but these non-continuum effects do not have a significant influence on the drag of drops with radii larger than approximately 0.3-0.5 μm .

2.2. Thermodynamics of individual spray drops

In addition to the drop position and momentum, it is clear from equation (2.2) that we need to determine the mass of the drop and thus its thermodynamical state. As sea spray drops evaporate, their mass change according to (Pruppacher & Klett (1996), their equation 13.9):

$$\frac{dm_p(t)}{dt} = 4\pi r_p D_v^* \left(\rho_v - \rho_{v,p}\right),$$
(2.6)

where ρ_v is the water vapour density, and D_v^* the molecular diffusivity for water vapour modified to include non-continuum effects. While the formulation of Pruppacher & Klett (1996) was originally derived for stationary drops in a motionless atmosphere, we also include ventilation effects using a correction coefficient based on the drop Reynolds number (see below). In this model, the mass transfer results from the difference between the ambient vapour density and that directly adjacent to the surface of the drop, noted $\rho_{v,p}$. To estimate $\rho_{v,p}$ properly, it is generally necessary to account for the effects of drop curvature (surface tension), and salinity. With these effects explicitly written out, the evolution equation for the mass of a saline sea spray drop takes the following form (Pruppacher & Klett (1996) equation 13.26):

$$\frac{dm_p(t)}{dt} = \frac{4\pi r_p D_v^* M_w p_v^{sat} (T_{air})}{RT_{air}} \times \left[Q_{RH} - \frac{T_{air}}{T_p} \exp\left(\frac{L_v M_w}{R} \left(\frac{1}{T_{air}} - \frac{1}{T_p}\right) + \frac{2M_w \Gamma_p}{R\rho_w r_p T_p} - \frac{I\Phi_s m_s (M_w/M_s)}{m_p - m_s} \right) \right] \\
= \mathcal{M}(t)$$
(2.7)

where it is noted that the rate of variation \mathcal{M} depends on both the fields of the carrier fluid and on the Lagrangian state of the drop, i.e. $\mathcal{M} = \mathcal{M}(\rho, \boldsymbol{u}, T_{air}; \boldsymbol{v}_p, r_p, m_p, T_p)$. In the equation above, Q_{RH} is the fractional relative humidity; R is the universal gas constant; M_w and M_s are the molecular weight of water and salt, respectively; $p_v^{sat}(T_{air})$ is the saturation water vapour pressure at the air temperature T_{air} ; T_p is drop temperature assumed uniform inside the drop; L_v is the latent heat of vaporization; Γ_p is the surface tension for a flat surface at the interface of the drop; m_s is the mass of salt in the drop; I is the number of ions salt dissociates into (I=2), and Φ_s is the osmotic coefficient. In equation (2.7), the term between brackets [] simply represents the difference between the ambient humidity Q_{RH} surrounding the drop and the humidity at the surface of the drop which results from three distinct effects that are reflected in the three terms inside the *exp*. The first term accounts for the drop temperature which differs from the ambient air temperature. The second term reflects curvature and surface tension effects. The last term accounts for salinity effects in the spray drop.

Noting that the mass of salt inside a spray drop remain constant (i.e. evaporation or condensation transfers only pure water), at a given time,

$$m_p(t) = \frac{4}{3}\pi\rho_{sw}r_{p0}^3 + \frac{4}{3}\pi\rho_w\left(r_p(t)^3 - r_{p0}^3\right),$$
(2.8)

where ρ_{sw} is the density of sea water, ρ_w is the density of (pure) water, and r_{p0} is the radius of a drop at formation (t = 0). In the equation above, the first term is the mass of the drop at formation and the second term represents the mass lost due to the evaporation of pure water. Incidentally, this computation shows that the mass of salt in the drop (required in (2.7)) is $m_s = m_p(t) - \frac{4}{3}\pi r_p(t)^3 \rho_w$. Thus, assuming that ρ_w does not change substantially in time (because of temperature changes for example),

$$\frac{dm_p(t)}{dt} = 4\pi r_p^2 \rho_w \frac{dr_p(t)}{dt}
= 4\pi r_p^2 \rho_w \mathcal{R}(t)
= \mathcal{M}(t),$$
(2.9)

which readily gives the evolution equation for the drop radius.

Similarly, the temporal evolution of the drop temperature T_p is given in Pruppacher & Klett (1996) (their equation 13.65):

$$m_p c_{ps} \frac{dT_p(t)}{dt} = 4\pi r_p k_a^* \left(T_{air} - T_p \right) + L_v \frac{dm_p(t)}{dt}$$
(2.10)

which yields

$$\frac{dT_p(t)}{dt} = \frac{4\pi r_p}{m_p c_{ps}} \left(k_a^* \left(T_{air} - T_p \right) + L_v D_v^* \left(\rho_v - \rho_{v,p} \right) \right) = \mathcal{T}(t)$$
(2.11)

where the rate of variation $\mathcal{T} = \mathcal{T}(\rho, \boldsymbol{u}, T_{air}; \boldsymbol{v}_p, r_p, m_p, T_p)$ also depends on the macroscopic fields of the carrier fluid and on the Lagrangian variables of the drop. In the expression for \mathcal{T} , we neglect high order effects related to temporal and spatial gradients in the air temperature (Michaelides & Feng 1994). In the equation above, c_{ps} is the specific heat of salty water, and k_a^* the modified thermal conductivity of air. The two terms in $\mathcal{T}(t)$ represent the changes to the drop temperature from the sensible and latent heat flux exchanges.

The modified diffusivity for water vapour, D_v^* , and the modified thermal conductivity of air, k_a^* , include ventilation and noncontinuum effects and are respectively:

$$D_{v}^{*} = \frac{f_{v}D_{v}}{\frac{r_{p}}{r_{p}+\Delta_{v}} + \frac{D_{v}}{r_{p}\alpha_{c}}\left(\frac{2\pi M_{w}}{RT_{air}}\right)^{1/2}},$$
(2.12)

$$k_a^* = \frac{f_h k_a}{\frac{r_p}{r_p + \Delta_T} + \frac{k_a}{r_p \alpha_T \rho_a c_{p,a}} \left(\frac{2\pi M_a}{RT_{air}}\right)^{1/2}},$$
(2.13)

where D_v and k_a are the molecular diffusivity for water vapour and thermal conductivity of air, respectively. Also, M_a is the molecular weight of dry air; $c_{p,a}$ and ρ_a are respectively the specific heat (at constant pressure) and density of dry air, with constants $\Delta_v =$ 8×10^{-8} , $\alpha_c = 0.06$, $\Delta_T = 2.16 \times 10^{-7}$, and $\alpha_T = 0.7$. We also include ventilation coefficients $f_v = f_h = 1 + \sqrt{Re_p}/4$, which are corrections to the heat and water vapour diffusivity for large instantaneous drop Reynolds numbers, i.e. $Re_p >> 1$ (Edson & Fairall 1994). These are analogous to the mean Sherwood and Nusselt numbers corrected using Ranz-Marshall correlations (Ranz *et al.* 1952).

In short, in this Lagrangian approach, the system that governs the evolution of sea spray drops is

$$\frac{d\boldsymbol{x}_p}{dt} = \boldsymbol{v}_p, \qquad \frac{d\boldsymbol{v}_p}{dt} = \frac{1}{m_p}\boldsymbol{\mathcal{F}},
\frac{dr_p}{dt} = \boldsymbol{\mathcal{R}}, \qquad \frac{dm_p}{dt} = \boldsymbol{\mathcal{M}}, \qquad \frac{dT_p}{dt} = \boldsymbol{\mathcal{T}}.$$
(2.14)

Figure 1 shows the time evolution of a water drop's temperature (blue), radius (red),

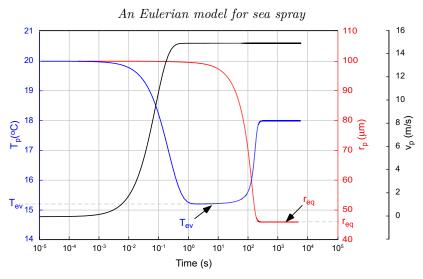


FIGURE 1. Time evolution of a water drop's temperature (blue), radius (red), and velocity (black). The drop is placed at time t = 0 in air with a wind speed of $U_{10} = 15$ m s⁻¹, a temperature of $T_{air} = 18^{\circ}C$ and a relative humidity of 75%. The drop has an initial radius of $r_p = 100\mu m$ and initial temperature of $T_p = 20^{\circ}C$.

and velocity (black) when placed at time t = 0 in air with a wind speed of $U_{10} = 15$ m s⁻¹, a temperature of $T_{air} = 18^{\circ}C$ and a relative humidity of 75%. The drop is initially at rest and with an initial radius of $r_p = 100\mu m$ and initial temperature of $T_p = 20^{\circ}C$. From figure 1, it is apparent that the drop responds quickly to the drag forces imposed on it. It also transfers sensible heat to the air and rapidly cools down. The latent heat transfer between the air and the drop is slower than both momentum and sensible heat transfers. Indeed, the radius of the drop starts to diminish because of evaporation only after having been exposed to the ambient atmospheric conditions for several seconds. The separation between latent and sensible heat transfers from a spray drop were first noted by Andreas (1990) who subsequently explored the implication of this phenomenon for air-sea spray-mediated heat fluxes (e.g. Andreas 1990, 1992; Andreas *et al.* 1995; Andreas & DeCosmo 1999; Andreas & Emanuel 2001). As such, the results of figure 1 are not new but they will inform results in this paper.

2.3. Drop conservation and kinetic equation

Let us now consider a set of drops described by the distribution function $f(t, \boldsymbol{x}, \boldsymbol{v}, r, m, T)$, which is the number density of drops at time t in the phase space $\mathbb{R}^3_{\boldsymbol{x}} \times \mathbb{R}^3_{\boldsymbol{v}} \times [0, +\infty[_r \times [0, +\infty[_m \times [0, +\infty[_T. \text{ It is the number of drops per } d\boldsymbol{x}, \text{ per } d\boldsymbol{v}, \text{ per } dr, \text{ per } dm, \text{ per } dT$ and has units of $[(Number of drops) (m)^{-3} (ms^{-1})^{-3} (m^{-1}) (kg^{-1}) (K^{-1})]$. In other words, $f(t, \boldsymbol{x}, \boldsymbol{v}, r, m, T) d\boldsymbol{x} d\boldsymbol{v} dr dm dT$ is the number of drops that at time t are at position $\boldsymbol{x} \pm d\boldsymbol{x}$ (where $d\boldsymbol{x} = dx dy dz$) with velocity $\boldsymbol{v} \pm d\boldsymbol{v}$, with a radius $r \pm dr$, with mass $m \pm dm$, and at temperature $T \pm dT$.

The governing (conservation) equation for this distribution function is

$$\frac{\partial}{\partial t}f + \boldsymbol{\nabla} \cdot (\boldsymbol{v}f) + \boldsymbol{\nabla}_{\boldsymbol{v}} \cdot \left(\frac{\boldsymbol{\mathcal{F}}}{m}f\right) + \frac{\partial}{\partial r}\left(\mathcal{R}f\right) + \frac{\partial}{\partial m}\left(\mathcal{M}f\right) + \frac{\partial}{\partial T}\left(\mathcal{T}f\right) = 0 \qquad (2.15)$$

This is known as the Williams-Boltzmann equation (Marchiso & Fox 2013) and can be understood as a conservation law for the number of drops in the multi-dimensional phase space that represents physical space \boldsymbol{x} , but also velocity space \boldsymbol{v} , radius r, mass m, and temperature T domains. Indeed, the first two terms of this equation are recognized as the classical conservation law involving the flux divergence. Likewise, ∇_{v} denotes differentiation with respect to the velocity vector components. Note that the Eulerian rates of variation of velocity, radius, mass, and temperature used in (2.15) are defined through their Lagrangian versions (2.2), (2.7), (2.11), (2.9), in which the Lagrangian variables $\boldsymbol{x}_p, \boldsymbol{v}_p, r_p, m_p, T_p$ are replaced by the Eulerian kinetic variables $\boldsymbol{x}, \boldsymbol{v}, r, m$, and T. If the drops do not interact, this change of variables is straightforward. In the case of spray drops, there is a weak interaction because the drops modify the airflow, which in turn can influence the drop transport (see section 3). In that case, equation (2.15) is nonetheless recovered using so called mean-field techniques (see Desvillettes *et al.* (2008) for a rigorous derivation of a simple example). For particles with strong interactions, such as charged particles submitted to a long range interaction force induced by all the other particles, the derivation of (2.15) can be substantially more complex and the Eulerian rates of variation require some averaging process. This is not necessary here in the case of sea spray.

Note also that collisions, breakup, coalescence, and nucleation of drops are normally taken into account by source (and sink) terms in the right-hand side of (2.15). However, since the volume fraction of sea spray is expected to be very small (see the introduction), collision and coalescence are expected to be negligible. Nucleation processes are important in cloud formation but are neglected for sea spray applications. Finally, breakup is expected to be a substantial mechanism of spray formation per se, particularly for large spume drops (e.g Mueller & Veron 2009b), but presumably only for short times after the initial ejection of the spray. Furthermore, the details of generation of large spume drops at the ocean surface remains poorly resolved. Thus, in this analysis, we consider the spray distribution after the spray is formed and once the drops are no longer fragmenting in the airflow after their ejection from the interface. Overall, sources and sinks terms are expected to provide minimal contributions to the overall flux balances and can be neglected.

The source of drops can therefore be naturally taken into account by boundary conditions. Indeed, equation (2.15) is set in a moving domain where one boundary is the free sea surface. At this boundary, where drops are injected into the airflow, the distribution function of incoming drops has to be defined. To that end, let \boldsymbol{x} be a point on this free surface, $\boldsymbol{u}_s(t, \boldsymbol{x})$ be the velocity of the free surface at this point, and $\boldsymbol{n}(t, \boldsymbol{x})$ the unit vector normal to this free surface, directed toward the air flow. Then, the source of drops coming from \boldsymbol{x} is simply $f(t, \boldsymbol{x}, \boldsymbol{v}, r, m, T)$ for every \boldsymbol{v} such that $(\boldsymbol{v} - \boldsymbol{u}_s(t, \boldsymbol{x})) \cdot \boldsymbol{n}(t, \boldsymbol{x}) > 0$ (i.e. drops whose velocities are directed toward the air flow). This value, denoted by f_{gen} thus needs to be known.

Fortunately, because of the interests in assessing spray impacts on Earth's climate and weather, some efforts have been spent in attempting to evaluate the air-sea flux of spray. In practice, so-called sea spray generation functions, $S(t, \boldsymbol{x}, r)$, are commonly utilized. They give the number of drops generated at the surface per unit surface area, per unit time, and per radius increment. We note however that $S(t, \boldsymbol{x}, r)$ is not trivial to obtain and nearly all spray generation estimates derive from measurements of drop concentration, $N(t, \boldsymbol{x}, r)$, at some elevation above the surface. $N(t, \boldsymbol{x}, r)$ is then the number of drops per unit volume of air, per radius increment, and is a function of space, time, and drop radius. The difference between the flux and the concentration is typically estimated assuming steady state resulting from a balance between gravitational deposition and turbulent suspension. Under these assumptions, the source flux $S(\boldsymbol{x}, r)$ can then be inferred from concentration measurements (e.g. Andreas *et al.* 2010; Veron 2015) and $S(\boldsymbol{x}, r) = v_o(r)N(\boldsymbol{x}, r)$ where $v_o(r)$ is a vertical suspension (or deposition) velocity (Slinn & Slinn 1980; Andreas *et al.* 2010). However, suspension velocities are likely to be a function of near-surface turbulence and the details of the drop formation process (Mueller & Veron 2009b; Veron *et al.* 2012; Troitskaya *et al.* 2018).

For the purpose of the model, we assume that the (known) radius dependent ejection velocity is $v_{gen}(r)^{\dagger}$. We also note that the drops generated at the surface have the same density as sea water, and therefore their mass m_{gen} is given by $m_{gen}(r) = \frac{4}{3}\pi r^3 \rho_{sw}$. Likewise, we can also assume that all the drops are ejected with the same sea temperature $T_{sw}(t, \boldsymbol{x})$. Then, dimensional arguments show that the number of drops generated at the surface per unit surface area, per unit time, per velocity, radius, mass, and temperature increment is

$$(\boldsymbol{v} - \boldsymbol{u}_s(t, \boldsymbol{x})) \cdot \boldsymbol{n}(t, \boldsymbol{x}) f_{gen}(t, \boldsymbol{x}, \boldsymbol{v}, r, m, T) = S \delta_{\boldsymbol{v} - \boldsymbol{v}_{gen}(r)} \delta_{m - m_{gen}(r)} \delta_{T - T_{sw}(t, \boldsymbol{x})}, \quad (2.16)$$

where $(\boldsymbol{v} - \boldsymbol{u}_s(t, \boldsymbol{x})) \cdot \boldsymbol{n}(t, \boldsymbol{x}) > 0$, and where the " δ " symbols denote Dirac delta functions centered in $\boldsymbol{v}_{gen}(r)$, $m_{gen}(r)$ and $T_{sw}(t, \boldsymbol{x})$, for the velocity, mass, and temperature, respectively. Once the S and $\boldsymbol{v}_{gen}(r)$ are known, this relation fully defines the boundary value f_{gen} for equation (2.15).

Finally, note that the Williams-Boltzmann equation (2.15) needs to be solved concurrently with the equations for the atmospheric airflow, i.e. the (*Eulerian*) conservation of mass, momentum, and energy respectively:

$$\frac{\partial}{\partial t}\rho + \boldsymbol{\nabla} \cdot (\rho \boldsymbol{u}) = 0, \qquad (2.17)$$

$$\frac{\partial}{\partial t}(\rho \boldsymbol{u}) + \boldsymbol{\nabla} \cdot (\rho \boldsymbol{u} \otimes \boldsymbol{u}) = \boldsymbol{\nabla} \cdot \boldsymbol{\tau} + \rho \boldsymbol{g}, \qquad (2.18)$$

$$\frac{\partial}{\partial t}E + \boldsymbol{\nabla} \cdot (E\boldsymbol{u}) = \boldsymbol{\nabla} \cdot \boldsymbol{Q} + \boldsymbol{\nabla} \cdot (\boldsymbol{\tau}\boldsymbol{u}) + \rho \boldsymbol{g} \cdot \boldsymbol{u}, \qquad (2.19)$$

where ρ is the air density, \boldsymbol{u} the air velocity, $\boldsymbol{\tau}$ atmospheric stress including pressure, E the total energy density of the air and \boldsymbol{Q} the heat flux.

3. Airflow including spray effects

The equations (2.17), (2.18), and (2.19) reflect conservation of mass, momentum, and energy for the airflow, but do not include the effects of the spray drops on the atmosphere. Using the kinetic description outlined above, feedback from the drops on the atmosphere is relatively straightforward to account for.

3.1. Conservation of mass

For example, since $\mathcal{M}dt$ is the mass lost (gained) from a drop during a small time interval dt, the mass gained (lost) by the fluid during dt, from all the drops at position $x \pm dx$ is

$$- d\boldsymbol{x} dt \int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\infty} \int_{\mathbb{R}^{3}_{\boldsymbol{v}}}^{\infty} \mathcal{M}f(t, \boldsymbol{x}, \boldsymbol{v}, r, m, T) d\boldsymbol{v} dr dm dT.$$
(3.1)

Note that the domain considered here now comprises all possible values in the entire phase space $\mathbb{R}^3_{\boldsymbol{v}} \times [0, \infty]_r \times [0, \infty]_m \times [0, \infty]_T$. To lighten the notation, we denote

$$\int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\infty} \int_{\mathbb{R}^{3}_{\boldsymbol{v}}} \mathcal{A}(t, \boldsymbol{x}, \boldsymbol{v}, r, m, T) d\boldsymbol{v} dr dm dT = \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \mathcal{A} d\boldsymbol{\mathfrak{X}}$$
(3.2)

[†] Note that the ejection velocity can take a more general form in order to include, for example, distributions of ejection angles. Here, v_o can be understood as the vertical component of v_{gen} .

where $\mathfrak{X} = (\boldsymbol{v}, r, m, T)$, and, $\boldsymbol{\Omega}_{\mathfrak{X}} = \mathbb{R}^3_{\boldsymbol{v}} \times [0, +\infty[_r \times [0, +\infty[_m \times [0, +\infty[_T, \infty[_m \times [0, +\infty[_T, \infty[_m \times [0, +\infty[_T, \infty[_m \times [0, +\infty[_m \times [0, \pm (0, +\infty[_m \times [0, +\infty[_m$

So, conservation of mass in the air, accounting for the mass exchange between the air and the drops, reads:

$$\frac{\partial}{\partial t}\rho + \boldsymbol{\nabla} \cdot (\rho \boldsymbol{u}) = -\pi_m \tag{3.3}$$

with

$$\pi_m = \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \mathcal{M} f d\boldsymbol{\mathfrak{X}}, \tag{3.4}$$

the mass production term due to spray drops. In other words, the water vapour transferred from (evaporation) or to (condensation) the spray drops will change the density of the air. In fact, while equations (2.17), (2.18), and (2.19) were pertinent for dry air, it is now necessary to consider moist air, i.e. the (binary) mixture of dry air and water vapour. As such, ρ in equation (3.3) is the density of moist air and $\rho = \rho_v + \rho_a$, the sum of the water vapour density and the density of dry air. In the remainder of the paper, unless explicitly noted as "dry", the air is considered to be moist air.

Furthermore, we need an equation for the water vapour density which is used to estimate the rate of variation of both the mass and the temperature of the drops (see equations (2.6), and (2.11)). Water vapour advects with the same velocity as the moist air, plus a small molecular motion v_v induced by the gradients in water vapour. The equation for the water vapour density is then

$$\frac{\partial}{\partial t}\rho_v + \boldsymbol{\nabla} \cdot \left(\rho_v \left(\boldsymbol{u} + \boldsymbol{v}_v\right)\right) = -\pi_m.$$
(3.5)

The diffusion from molecular motion can further be modeled with a Fick's diffusion law $\boldsymbol{v}_v = \frac{-1}{q} D_v \boldsymbol{\nabla} q$ with diffusivity D_v , and where $q = \rho_v / \rho$ is the specific humidity of air. Thus, the conservation of water vapour reads:

$$\frac{\partial}{\partial t}\rho_v + \boldsymbol{\nabla} \cdot (\rho_v \boldsymbol{u}) = \boldsymbol{\nabla} \cdot (\rho D_v \boldsymbol{\nabla} q) - \pi_m.$$
(3.6)

For completeness, equations (3.6) and (3.3) yield the mass conservation equation for dry air:

$$\frac{\partial}{\partial t}\rho_a + \boldsymbol{\nabla} \cdot (\rho_a \boldsymbol{u}) = \boldsymbol{\nabla} \cdot (\rho D_v \boldsymbol{\nabla} (1-q)).$$
(3.7)

where $\boldsymbol{v}_a = \frac{-1}{(1-q)} D_v \boldsymbol{\nabla} (1-q)$. Incidentally, $\rho_a \boldsymbol{v}_a + \rho_v \boldsymbol{v}_v = 0$ since we consider here a binary mixture with $\rho = \rho_v + \rho_a$.

3.2. Conservation of momentum

Equivalently, if \mathbf{F} is the force from the fluid on the drop, the force on the fluid from all drops at position $\mathbf{x} \pm d\mathbf{x}$ is $-d\mathbf{x} \int_{\Omega_{\mathcal{X}}} \mathbf{F} f d\mathbf{\mathfrak{X}}$. The change of momentum of the air is also due to an increase (decrease) in mass of water vapour coming from evaporating (condensing) drops. The corresponding rate of variation of momentum is $-d\mathbf{x} \int_{\Omega_{\mathcal{X}}} \mathbf{v} \mathcal{M} f d\mathbf{\mathfrak{X}}$. Then conservation of momentum in the air, accounting for the momentum exchange between the air and the drops, reads:

$$\frac{\partial}{\partial t}(\rho \boldsymbol{u}) + \boldsymbol{\nabla} \cdot (\rho \boldsymbol{u} \otimes \boldsymbol{u}) = \boldsymbol{\nabla} \cdot \boldsymbol{\tau} + \rho \boldsymbol{g} - \boldsymbol{\pi}_{\boldsymbol{u}}$$
(3.8)

where

$$\boldsymbol{\pi}_{\boldsymbol{u}} = \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \boldsymbol{F} f d\boldsymbol{\mathfrak{X}} + \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \boldsymbol{v} \mathcal{M} f d\boldsymbol{\mathfrak{X}}$$
(3.9)

is the momentum production term due to the drops. The corresponding equation for the air velocity is

$$\rho\left(\frac{\partial \boldsymbol{u}}{\partial t} + (\boldsymbol{u} \cdot \boldsymbol{\nabla}) \,\boldsymbol{u}\right) + \boldsymbol{\nabla} p = \boldsymbol{\nabla} \cdot \boldsymbol{\sigma} + \rho \boldsymbol{g} - \boldsymbol{\pi}_{\boldsymbol{u}} + \boldsymbol{\pi}_{m} \boldsymbol{u},$$

$$= \boldsymbol{\nabla} \cdot \boldsymbol{\sigma} + \rho \boldsymbol{g} - \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \boldsymbol{F} f d\boldsymbol{\mathfrak{X}} - \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} (\boldsymbol{v} - \boldsymbol{u}) \mathcal{M} f d\boldsymbol{\mathfrak{X}}$$
(3.10)

where the stress tensor has been decomposed into its atmospheric pressure and viscous components as $\boldsymbol{\tau} = -p\boldsymbol{I} + \boldsymbol{\sigma}$. Also, the viscous stress tensor is easily obtained from the gradient velocity tensor and its transpose using $\boldsymbol{\sigma} = \mu \left(\boldsymbol{\nabla} \boldsymbol{u} + (\boldsymbol{\nabla} \boldsymbol{u})^T - \left(\frac{2}{3} \boldsymbol{\nabla} \cdot \boldsymbol{u}\right) \boldsymbol{I} \right)$ In developing equations (3.8) and (3.10), strictly speaking, one needs to first consider separate momentum equations for dry air and water vapour. These can easily be combined by neglecting the terms involving molecular diffusive motions for dry air and water vapour (see equations (3.6) and (3.7) above) of order $O(\boldsymbol{v}_a^2)$ or $O(\boldsymbol{v}_v^2)$.

3.3. Conservation of energy

Finally, we seek to estimate the energy exchanged between the drops and the air, which we'll note π_e . The energy of a single drop is the sum of its internal and kinetic energy:

$$E_p(t) = m_p(t)e_p(t) + \frac{1}{2}m_p(t)\boldsymbol{v}_p^2(t)$$
(3.11)

where e_p denotes the drop specific internal energy which is a function of $T_p(t)$. So, the rate of change of a spray drop energy is

$$\frac{d}{dt}E_p(t) = \left(e_p(t) + \frac{1}{2}\boldsymbol{v}_p^2(t)\right)\frac{d}{dt}m_p(t) + m_p(t)\left(\frac{d}{dt}e_p(t) + \boldsymbol{v}_p(t)\cdot\frac{d\boldsymbol{v}_p(t)}{dt}\right)$$

$$= \left(e_p(t) + \frac{1}{2}\boldsymbol{v}_p^2\right)\mathcal{M} + m_p\frac{d}{dt}e_p(t) + \boldsymbol{v}_p\cdot\boldsymbol{\mathcal{F}}$$

$$= \mathcal{E}(t).$$
(3.12)

However, the gravity is a force which is external to the closed air/drops system, and its work should not be accounted for in the energy exchange between the drops and the air. Consequently, to define the rate of variation of energy of the drop, the work of \mathcal{F} is replaced by the work of the drag force \mathbf{F} in the previous relation. Furthermore, the specific internal energy of the drop is simply the specific internal energy of salty water at the temperature of the drop, so $e_p(t) = e_{sw}(T_p(t))$. Then, from the definition of c_{ps} , we obtain $\frac{d}{dt}e_p(t) = c_{ps}\frac{dT_p(t)}{dt} = c_{ps}\mathcal{T}$. In fact, equation (2.10) can now be understood as the rate of change of a drop's internal energy. Finally,

$$\mathcal{E}(t) = e_{sw}(T_p)\mathcal{M} + m_p c_{ps}\mathcal{T} + \boldsymbol{v}_p \cdot \boldsymbol{F} + \frac{1}{2}\mathcal{M}\boldsymbol{v}_p^2.$$
(3.13)

In the equation above, one can see that changes in internal energy of a drop may arise from both a change of mass and a change of temperature, while the change of kinetic energy results from the work of friction forces and a change in drop mass. As with the mass and momentum conservation equations above, the energy production term from all the drops is

$$\pi_e = \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \mathcal{E} f d\boldsymbol{\mathfrak{X}} \tag{3.14}$$

in which \mathcal{E} has to be understood as an Eulerian quantity derived from its Lagrangian definition (3.13). In other words, equation (3.14) reads:

$$\pi_e = \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} e_{sw}(T) \mathcal{M} f d\boldsymbol{\mathfrak{X}} + \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} mc_{ps} \mathcal{T} f d\boldsymbol{\mathfrak{X}} + \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \boldsymbol{v} \cdot \boldsymbol{F} f d\boldsymbol{\mathfrak{X}} + \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \frac{1}{2} \boldsymbol{v}^2 \mathcal{M} f d\boldsymbol{\mathfrak{X}}. \quad (3.15)$$

We can now develop the energy conservation for the air, accounting for the input from the spray. However, in contrast with the derivation of the momentum equation above, we chose here to keep the moist air energy density E explicitly separated into the energy density for dry air E_a , and that of the water vapour, E_v . With spray effects included, equation (2.19) reads:

$$\frac{\partial}{\partial t} (E_a + E_v) + \nabla \cdot ((E_a + E_v) \boldsymbol{u}) + \nabla \cdot (E_a \boldsymbol{v}_a + E_v \boldsymbol{v}_v)
= \nabla \cdot \boldsymbol{Q} + \nabla \cdot (\boldsymbol{\sigma} \boldsymbol{u}) - \nabla \cdot ((\boldsymbol{u} + \boldsymbol{v}_a) p_a + (\boldsymbol{u} + \boldsymbol{v}_v) p_v)
+ \rho \boldsymbol{g} \cdot \boldsymbol{u} - \pi_e$$
(3.16)

where p_a is the partial pressure for dry air, and p_v the water vapour partial pressure, and where terms of order $O(\boldsymbol{v}_a^2)$ or $O(\boldsymbol{v}_v^2)$ are once more neglected. The separation between water vapour and dry air is useful here because we wish to keep the rate of work done by the respective partial pressures. We note here that neglecting $\boldsymbol{v}_v p_v$ and $\boldsymbol{v}_a p_a$ would allow us to simply use $p = p_v + p_a$ and incorporate the pressure in the stress tensor of the (moist) air as in equation (3.10). However, it is convenient to keep the partial pressures explicitly out of the stress tensor so that expressions for the enthalpy of the dry air and water vapour can be uncovered. For example, as done above for the drop, the energy density of the water vapour can be decomposed into internal and kinetic energy:

$$E_v = \rho_v e_v + \frac{1}{2} \rho_v |\boldsymbol{u}|^2 \tag{3.17}$$

where e_v is the specific internal energy for water vapour, and where the contribution of the diffusion velocity to the kinetic energy can be neglected. By definition, the specific enthalpy for the water vapour is $h_v = e_v + p_v/\rho_v$, and thus

$$E_v + p_v = \rho_v h_v + \frac{1}{2} \rho_v |\boldsymbol{u}|^2.$$
(3.18)

Similarly,

$$E_a + p_a = \rho_a h_a + \frac{1}{2} \rho_a |\mathbf{u}|^2.$$
(3.19)

in which $h_a = e_a + p_a/\rho_a$ is the specific enthalpy of dry air, and e_a the specific internal energy for dry air. Therefore, equation (3.16) can be written as:

$$\frac{\partial}{\partial t} (\rho_a e_a + \rho_v e_v) + \nabla \cdot ((\rho_a h_a + \rho_v h_v) \boldsymbol{u})
+ \nabla \cdot (\rho_a h_a \boldsymbol{v}_a + \rho_v h_v \boldsymbol{v}_v)
+ \frac{\partial}{\partial t} \left(\frac{1}{2}\rho |\boldsymbol{u}|^2\right) + \nabla \cdot \left(\frac{1}{2}\rho |\boldsymbol{u}|^2 \boldsymbol{u}\right)
= \nabla \cdot \boldsymbol{Q} + \nabla \cdot (\boldsymbol{\sigma} \boldsymbol{u}) + \rho \boldsymbol{g} \cdot \boldsymbol{u} - \pi_e$$
(3.20)

Using equations (3.6), (3.8), and (3.20), we find the equation governing e, the specific

internal energy of moist air:

$$\frac{\partial}{\partial t} (\rho e) + \nabla \cdot (\rho e \boldsymbol{u}) = \nabla \cdot \boldsymbol{Q} + \boldsymbol{\sigma} : \nabla \boldsymbol{u} - p \nabla \cdot \boldsymbol{u} \\ - \nabla \cdot \boldsymbol{J} \\ + \boldsymbol{\pi}_{\boldsymbol{u}} \cdot \boldsymbol{u} - \frac{1}{2} |\boldsymbol{u}|^2 \boldsymbol{\pi}_m - \boldsymbol{\pi}_e$$
(3.21)

where $\rho e = \rho_a e_a + \rho_v e_v$, and

$$\boldsymbol{J} = \rho_a h_a \boldsymbol{v}_a + \rho_v h_v \boldsymbol{v}_v
= -\rho D_v \left(h_a \boldsymbol{\nabla} (1-q) + h_v \boldsymbol{\nabla} q \right)$$
(3.22)

is a flux of enthalpy due to small molecular motions.

For modeling purposes, it is generally practical to look at the temperature of the carrier flow T_{air} . With tedious but simple algebra, the energy conservation equation (3.21) can be written as

$$\rho c_{v} \left(\frac{\partial T_{air}}{\partial t} + \boldsymbol{u} \cdot \boldsymbol{\nabla} T_{air} \right) = \boldsymbol{\nabla} \cdot (\kappa \boldsymbol{\nabla} T_{air}) + \boldsymbol{\sigma} : \boldsymbol{\nabla} \boldsymbol{u} - p \boldsymbol{\nabla} \cdot \boldsymbol{u} - \boldsymbol{\nabla} \cdot \boldsymbol{J} + e_{a} \boldsymbol{\nabla} \cdot (\rho_{a} \boldsymbol{v}_{a}) + e_{v} \boldsymbol{\nabla} \cdot (\rho_{v} \boldsymbol{v}_{v}) + \pi_{\boldsymbol{u}} \cdot \boldsymbol{u} - \frac{1}{2} |\boldsymbol{u}|^{2} \pi_{m} - \pi_{e} + e_{v} \pi_{m}$$
(3.23)

where we have taken the heat flux as given by the Fourier law $\mathbf{Q} = \kappa \nabla T_{air}$ and where c_v is the specific heat of air at constant volume, derived from the corresponding specific heat of water vapour and dry air using:

$$c_v = qc_{v,v} + (1-q)c_{v,a} \tag{3.24}$$

Also, in equation (3.23), e_v and e_a are taken at the air temperature T_{air} .

Expanding the spray production terms leads to:

$$\rho c_{v} \left(\frac{\partial T_{air}}{\partial t} + \boldsymbol{u} \cdot \boldsymbol{\nabla} T_{air} \right) = \boldsymbol{\nabla} \cdot (\kappa \boldsymbol{\nabla} T_{air}) + \boldsymbol{\sigma} : \boldsymbol{\nabla} \boldsymbol{u} - p \boldsymbol{\nabla} \cdot \boldsymbol{u} - \boldsymbol{\nabla} \cdot \boldsymbol{J} + e_{a} \boldsymbol{\nabla} \cdot (\rho_{a} \boldsymbol{v}_{a}) + e_{v} \boldsymbol{\nabla} \cdot (\rho_{v} \boldsymbol{v}_{v}) - \int_{\boldsymbol{\Omega}_{\mathcal{X}}} (\boldsymbol{v} - \boldsymbol{u}) \cdot \boldsymbol{F} f d\boldsymbol{\mathfrak{X}} - \int_{\boldsymbol{\Omega}_{\mathcal{X}}} \frac{1}{2} |\boldsymbol{v} - \boldsymbol{u}|^{2} \mathcal{M} f d\boldsymbol{\mathfrak{X}} - \int_{\boldsymbol{\Omega}_{\mathcal{X}}} m c_{ps} \mathcal{T} f d\boldsymbol{\mathfrak{X}} + \int_{\boldsymbol{\Omega}_{\mathcal{X}}} (e_{v}(T_{air}) - e_{sw}(T)) \mathcal{M} f d\boldsymbol{\mathfrak{X}}$$

$$(3.25)$$

In the equation above, the spray feedback terms are on the right hand side and evidently contain f and its integrals. The first three feedback term represent the heat gained by the air due to the work of the frictional drag force exerted by the drops on the air, a loss of kinetic energy of the drops (because of a change of mass as the drop evaporate), and the exchange of sensible heat between the air and the drops due to the variation of the temperature of the drops. The last feedback term represents a transfer in internal energy from the drops to water vapour as drops evaporate.

Also, we develop the term resulting from molecular diffusion,

$$\mathcal{D} = -\nabla \cdot \boldsymbol{J} + e_a \nabla \cdot (\rho_a \boldsymbol{v}_a) + e_v \nabla \cdot (\rho_v \boldsymbol{v}_v)$$

= $RT_{air} \left(\frac{1}{M_w} - \frac{1}{M_a} \right) \nabla \cdot (\rho D_v \nabla q)$
+ $(c_{p,v} - c_{p,a}) \rho D_v \nabla q \cdot \nabla T_{air}$ (3.26)

where $c_{p,v}$ and $c_{p,a}$ are respectively the specific heat at constant pressure for water vapour and dry air. Finally,

$$e_{sw}(T) = c_{ps}(T - T_{ref}) + e_{sw}(T_{ref})$$
 and,
 $e_v(T_{air}) = c_{v,v}(T_{air} - T_{ref}) - \frac{R}{M_w}T_{ref} + L_v$ (3.27)

where T_{ref} is a reference temperature, generally taken as $T_{ref} = 0^{\circ}C$, such that the latent heat of vaporization, L_v , is the enthalpy difference between water vapour and liquid water at T_{ref} . To simplify the notation, we write the last feedback term in (3.25) as:

$$\Upsilon = \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \left(e_v(T_{air}) - e_{sw}(T) \right) \mathcal{M} f d\boldsymbol{\mathfrak{X}}$$
(3.28)

4. Dimensional analysis

In this section, we attempt to simplify the system given by William-Boltzman equation (equation (2.15)), with the atmospheric flow conservation equations which all include the contributions from the spray (equations (3.3), (3.10), and (3.25)).

4.1. Characteristic quantities and dimensionless variables

Here, we take r_o as a characteristic radius of a population of drops. Their corresponding vertical settling velocity is denoted by v_o . The space scale x_o is a typical transport scale for these drops which we choose as the significant wave height (Andreas 2004). Thus, the time scale $t_o = x_o/v_o$ roughly corresponds to a typical average time for a drop to fall from the significant wave height to the mean water level. And reas (2004) considers t_o as an adequate proxy for the drop time of flight, but we emphasize here that the details of the ejection mechanism such as the initial velocity and angle of ejection of the drops are not accounted for. There is some evidence that large drops ejected near the wave crest may fall back near the crest and not at the mean water level (Mueller & Veron 2014a). And the ejection velocity, particularly for small, bubble-generated drops, can be significant (Blanchard 1989; Spiel 1995, 1997, 1998). Nevertheless, for the purpose of the non-dimensional scaling, t_o is a suitable estimate for the drop time of flight. The characteristic temperature of the drops is naturally taken as the sea temperature and the characteristic mass drop is therefore $m_o = \rho_{sw} \frac{4}{3} \pi r_o^3$. Finally, the corresponding characteristic values $\mathcal{F}_o, \mathcal{R}_o, \mathcal{M}_o, \text{ and } \mathcal{T}_o$ of the the rates of variation $\mathcal{F}, \mathcal{R}, \mathcal{M}, \mathcal{T}$ of the drop momentum, radius, mass and temperature, respectively, are obtained by using formulas (2.3), (2.7), (2.11), and (2.9). Because sea spray is made of spray drops that are disperse in size (ranging here from $O(1) \mu m$ to O(1) mm), defining the characteristic value f_o of the distribution function f is delicate. However, we shall see later that f_o appears in all spray feedback terms and thus naturally falls out of a term-by-term dimensional comparison.

For the air flow, we take the same space and time scales. However, since air flow and spray drops can have very different velocities, especially for large spume drops with An Eulerian model for sea spray

substantial mass and inertia, we use a velocity scale u_o for the air flow as a typical wind speed, possibly different from v_o . An obvious choice for u_o is the mean wind speed measured at 10m height above the surface, U_{10} . The characteristic air mass density is ρ_o , and its characteristic temperature is T_o which is the same as the characteristic sea temperature. Derived characteristic values of viscosity μ_o and heat transfer κ_o coefficients can be easily obtained.

In the following, the corresponding dimensionless quantities will be denoted with the superscript ', and defined by $[.]' = [.]/[.]_o$; for instance $t' = t/t_o$ for the time variable.

4.2. Dimensionless Williams-Boltzmann equation

The Williams-Boltzmann equation (2.15) can be non-dimensionalized to

$$\frac{\partial}{\partial t'}f' + \nabla' \cdot (f'\boldsymbol{v}') + \frac{t_o}{\tau_v} \nabla_{\boldsymbol{v}'} \cdot \left(\frac{f'}{m'}\mathcal{F}'\right)
+ \frac{t_o}{\tau_R} \frac{\partial}{\partial r'} \left(\mathcal{R}'f'\right) + \frac{t_o}{\tau_m} \frac{\partial}{\partial m'} \left(\mathcal{M}'f'\right) + \frac{t_o}{\tau_T} \frac{\partial}{\partial T'} \left(\mathcal{T}'f'\right) = 0,$$
(4.1)

where the relaxation times τ_v , τ_R , τ_m , and τ_T are characteristic times for the variation of the momentum, radius, mass, and temperature of the drops, defined by

$$\tau_v = \frac{m_o v_o}{\mathcal{F}_o}, \qquad \tau_R = \frac{r_o}{\mathcal{R}_o}, \qquad \tau_m = \frac{m_o}{\mathcal{M}_o}, \qquad \tau_T = \frac{T_o}{\mathcal{T}_o}.$$
(4.2)

We note that the momentum term can further be decomposed into its gravitational and drag components such that

$$\frac{t_o}{\tau_v} \boldsymbol{\nabla}_{\boldsymbol{v}'} \cdot \left(\frac{f'}{m'} \mathcal{F}'\right) = \boldsymbol{\nabla}_{\boldsymbol{v}'} \cdot \left(\frac{t_o}{\tau_g} \boldsymbol{g}' f' + \frac{t_o}{\tau_{Do}} \frac{1}{\tau'_D} \left(\frac{1}{\eta} \boldsymbol{u}' - \boldsymbol{v}'\right) f'\right)$$
(4.3)

where $\eta = v_o/u_o$ is the ratio of the characteristic drop and air flow velocities, and where the dimensionless gravity vector is $\mathbf{g}' = \mathbf{g}/g = -e_z$, with $g = |\mathbf{g}|$ the gravity magnitude. Then, $\tau_g = v_o/g$ is the characteristic drop time scale due to gravity and $\tau_{Do} = 2r_o^2 \rho_{sw}/(9\mu_o \Xi_o)$ is the characteristic drop response time to the drag force, i.e $\tau_D = \tau_{Do} \tau'_D$ with $\tau'_D = r'^2/(\mu' \Xi')$.

Figure 2 shows the characteristic times of variation of the drag force τ_{Do} , radius τ_R , mass τ_m , and temperature τ_T , of the drop as a function of the characteristic radius r_0 . These timescales are plotted here for specific ambient conditions, but they do vary (albeit weakly), in particular with relative humidity (see Andreas 1990). They are, however, strong functions of the drop radius. Indeed, τ_{Do} is proportional to the drag on the drop, i.e. its cross sectional area, and τ_R , τ_m , and τ_T are all controlled by molecular exchanges through the drops surface area. Thus, all these time scales are proportional to r_o^2 . From figure 2, one can easily see that small drops transfer sensible heat faster than larger ones do. Also, $\tau_T \ll \tau_R$ and the temperature of a drop will change and adapt to ambient atmospheric conditions much faster (approximately 1000 times) than its radius will.

We note here that in the absence of wind or other sources of airflow fluctuations and turbulence, the terminal fall velocity of a drop, $v_t(r_o)$, is such that $v_t/g = \tau_{Do}$, and $v_t(r_o)$ is a function of the drop radius only. However, when the airflow is turbulent, the terminal velocity v_t and the settling (or deposition) velocity v_o , of small drops in particular, can be substantially different. Indeed, small drops are affected by the turbulence in the airflow such that v_t does not adequately represent the actual velocity at which these drops effectively settle (e.g. Slinn & Slinn 1980; Andreas *et al.* 2010). Here, we adopt a formulation found in Smith *et al.* (1993) and originally proposed by Carruthers &

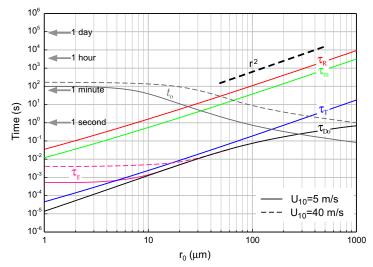


FIGURE 2. Characteristic times of variation of the drag force τ_{Do} , radius τ_R , mass τ_m , and temperature τ_T , of the drop as a function of the characteristic radius r_0 and for water temperature of $T_{air} = 18^{\circ}C$, $T_p = 20^{\circ}C$, and a 75% relative humidity. Also, the characteristic times τ_g and t_o are shown for wind speeds of $U_{10} = 5 \text{ m s}^{-1}$ with a significant wave height of $x_o = 0.5 \text{ m}$ (solid lines), and $U_{10} = 40 \text{ m s}^{-1}$ with a significant wave height of $x_o = 6.4 \text{ m}$ (dashed lines).

Choularton (1986)

$$v_o = \frac{v_t}{1 - exp\left(\frac{v_t}{C_D U_{10}}\right)},\tag{4.4}$$

where $C_D \sim O(10^{-3})$ is the air-sea aerodynamic drag coefficient. Thus τ_g is function of both the drop radius and the wind speed. It is plotted on figure 2 for $U_{10} = 5 \text{ m s}^{-1}$ with a significant wave height estimated at $x_o = 0.5 \text{ m}$ (solid line), and for $U_{10} = 40 \text{ m}$ s⁻¹ with a significant wave height estimated at $x_o = 6.4 \text{ m}$ (dashed line).

It is worthwhile to compare the relaxation times with t_o , the atmospheric residence time of spray drops. For convenience, we have plotted t_o on figure 2, and we note that, t_o also depends on the drop settling velocity and thus on the wind speed. For the remainder of the analysis, it is useful to introduce the following dimensionless numbers

$$\alpha_g = \frac{t_o}{\tau_g}, \quad \alpha_D = \frac{t_o}{\tau_{Do}}, \quad \alpha_R = \frac{t_o}{\tau_R}, \quad \alpha_m = \frac{t_o}{\tau_m}, \quad \alpha_T = \frac{t_o}{\tau_T}, \tag{4.5}$$

such that the nondimensionalized Williams-Boltzmann equation can be written as:

$$\frac{\partial}{\partial t'}f' + \nabla' \cdot (f'\boldsymbol{v}')
+ \nabla_{\boldsymbol{v}'} \cdot \left(\alpha_g \boldsymbol{g}' f' + \alpha_D \frac{1}{\tau'_D} \left(\frac{1}{\eta} \boldsymbol{u}' - \boldsymbol{v}'\right) f'\right)
+ \alpha_R \frac{\partial}{\partial r'} (\mathcal{R}' f')
+ \alpha_m \frac{\partial}{\partial m'} (\mathcal{M}' f')
+ \alpha_T \frac{\partial}{\partial T'} (\mathcal{T}' f') = 0.$$
(4.6)

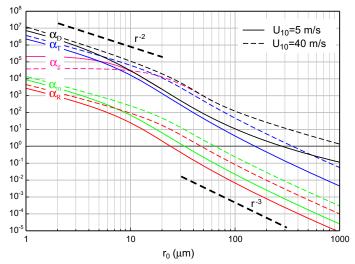


FIGURE 3. Non-dimensional relaxation times for the drag force on the drop α_D , the drop temperature α_T , the drop mass α_m , and the drop radius α_R . Solid lines indicate $U_{10} = 5 \text{ m s}^{-1}$ with a significant wave height estimated at $x_o = 0.5 \text{ m}$, and dashed lines indicate $U_{10} = 40 \text{ m} \text{ s}^{-1}$ with a significant wave height estimated at $x_o = 6.4 \text{ m}$.

Figure 3 shows these non-dimensional *alphas* which are the ratios of the drops residence time t_o , to the relaxation times τ_v , τ_R , τ_m , and τ_T . They are plotted as a function of r_o and for a wind speed of $U_{10} = 5 \text{ m s}^{-1}$ (solid lines) and $U_{10} = 40 \text{ m s}^{-1}$ (dashed lines). When these ratios are above 1, this simply indicates that the drops remain suspended in the airflow long enough to exchange their momentum, sensible heat, or water (vapour) mass with the surrounding atmosphere. For example, from figure 3, we deduce that at $U_{10} = 5 \text{ m s}^{-1}$, drops with $r_o < 150 \ \mu\text{m}$ will exchange momentum and sensible heat with the atmosphere, while only drops with $r_o < 30 \ \mu\text{m}$ will have the opportunity to exchange latent heat with the air, thereby transferring water vapour to the atmosphere.

4.3. Dimensionless equations for the airflow including feedbacks

In this section, we non-dimensionalize the equations for the airflow including spray feedback effects.

4.3.1. Conservation of mass

The dimensionless mass conservation equation for the air is

$$\frac{\partial}{\partial t'}\rho' + \frac{1}{\eta}\nabla' \cdot (\rho'\boldsymbol{u}') = -\alpha_m \chi \int_{\boldsymbol{\Omega}_{\mathfrak{X}'}} \mathcal{M}' f' d\boldsymbol{\mathfrak{X}'}, \qquad (4.7)$$

where $\chi = m_o f_o v_o^3 r_o T_o m_o / \rho_o$. Physically, χ is a mass fraction, i.e. the mass of the drops represented by f_o , divided by the mass of a characteristic volume of air.

4.3.2. Airflow velocity

In non-dimensional variables, equation (3.10) reduces to

$$\rho'\left(\frac{\partial \boldsymbol{u}'}{\partial t'} + \frac{1}{\eta}(\boldsymbol{u}'\cdot\boldsymbol{\nabla}')\boldsymbol{u}'\right) + \frac{1}{\eta}\frac{1}{\gamma\mathrm{Ma}^2}\boldsymbol{\nabla}'p' = \frac{1}{\eta}\frac{1}{\mathrm{Re}}\boldsymbol{\nabla}'\cdot\boldsymbol{\sigma}' + \frac{1}{\eta}\frac{1}{\mathrm{Fr}^2}\rho'\boldsymbol{g}' + \frac{1}{\eta}\frac{1}{\mathrm{Fr}^2}\rho'\boldsymbol{g}' - \alpha_D\chi\int_{\boldsymbol{\Omega}_{\mathfrak{X}'}}\frac{m'}{\tau'_D}(\boldsymbol{u}'-\eta\boldsymbol{v}')f'd\boldsymbol{\mathfrak{X}'} - \alpha_m\chi\int_{\boldsymbol{\Omega}_{\mathfrak{X}'}}(\eta\boldsymbol{v}'-\boldsymbol{u}')\mathcal{M}'f'd\boldsymbol{\mathfrak{X}'},$$

$$(4.8)$$

where the nondimensional number in front of the pressure gradient contains the Mach number of the air flow defined by $\operatorname{Ma} = u_o/c_o$ with $c_o = \sqrt{\gamma p_o/\rho_o}$ the sound speed in air, $\gamma = c_p/c_v$ the ratio of specific heats of air, and $p_o = \rho_o RT_o$ the characteristic value for the atmospheric pressure. The nondimensional number in front of the shear stress is the inverse of the Reynolds number of the air flow $\operatorname{Re} = \rho_o u_o x_o/\mu_o$, where $\mu_o = \mu(T_o)$ is a characteristic value for the viscosity of air. The non-dimensional number in the gravity term contains the Froude number $\operatorname{Fr} = u_o/\sqrt{x_og}$. The other dimensionless numbers have been defined in previous sections.

It is possible to substantially simplify the equation above. Indeed, we can see from figure 3 that $\alpha_m \ll \alpha_D$ for all drop radii. Therefore, among the spray feedback terms, only that related to the drag force on the fluid from the drops remains.

4.3.3. Airflow temperature

The same procedure leads to the nondimensional form of the temperature equation (equation (3.25)):

$$\rho' \left(\frac{\partial T'_{air}}{\partial t'} + \frac{u'}{\eta} \cdot \nabla' T'_{air} \right)$$

$$= \frac{1}{\eta} \frac{\gamma}{\text{Pe}_{\text{T}}} \nabla' \cdot (\kappa' \nabla' T'_{air})$$

$$+ \frac{1}{\eta} \gamma(\gamma - 1) \frac{\text{Ma}^2}{\text{Re}} \sigma' : \nabla' u'$$

$$- \frac{1}{\eta} (\gamma - 1) p' \nabla' \cdot u'$$

$$+ \mathcal{D}'$$

$$+ \gamma(\gamma - 1) \text{Ma}^2 \alpha_D \chi \int_{\Omega_{\mathfrak{X}'}} \frac{m'}{\tau'_D} |u' - \eta v'|^2 f' d\mathfrak{X}',$$

$$- \gamma(\gamma - 1) \text{Ma}^2 \alpha_m \chi \int_{\Omega_{\mathfrak{X}}} \left(\frac{1}{2} |\eta v' - u'|^2 \right) \mathcal{M}' f' d\mathfrak{X}'$$

$$- \frac{c_{ps}}{c_v} \alpha_T \chi \int_{\Omega_{\mathfrak{X}'}} m' \mathcal{T}' f' d\mathfrak{X}'$$

$$+ \Upsilon'$$

$$(4.9)$$

where the dimensionless number in front of the heat flux contains the Peclet number for heat $\text{Pe}_{\text{T}} = c_p x_o u_o \rho_o / \kappa_o$, with c_p the specific heat of air at constant pressure.

The non-dimensional diffusion term reads:

$$\mathcal{D}' = \frac{1}{\eta} \frac{1}{\operatorname{Pe}_v} \frac{R}{c_v} q_o T'_{air} \left(\frac{1}{M_w} - \frac{1}{M_a} \right) \boldsymbol{\nabla}' \cdot (\rho' \boldsymbol{\nabla}' q') + \frac{1}{\eta} \frac{1}{\operatorname{Pe}_v} q_o \frac{c_{p,v} - c_{p,a}}{c_v} \rho' \boldsymbol{\nabla}' q' \cdot \boldsymbol{\nabla}' T'_{air}$$
(4.10)

where Pe_v is the Peclet number for mass transfer of water vapour $\text{Pe}_v = x_o u_o/D_v$ and q_o is a characteristic specific humidity. Finally, the non-dimensional internal energy term:

$$\Upsilon' = \frac{c_{v,v}}{c_v} \alpha_m \chi \int_{\Omega_{\mathfrak{X}}} \left(T'_{air} - \frac{T_{ref}}{T_o} \right) \mathcal{M}' f' d\mathfrak{X}'
+ \frac{L_v}{T_o c_v} \alpha_m \chi \int_{\Omega_{\mathfrak{X}}} \mathcal{M}' f' d\mathfrak{X}'
- \frac{RT_{ref}}{M_w T_o c_v} \alpha_m \chi \int_{\Omega_{\mathfrak{X}}} \mathcal{M}' f' d\mathfrak{X}'
- \frac{c_{ps}}{c_v} \alpha_m \chi \int_{\Omega_{\mathfrak{X}}} \left(T' - \frac{T_{ref}}{T_o} \right) \mathcal{M}' f' d\mathfrak{X}'
- \frac{e_{sw}(T_{ref})}{T_o} \alpha_m \chi \int_{\Omega_{\mathfrak{X}}} \mathcal{M}' f' d\mathfrak{X}'$$
(4.11)

Again, using figure 3, it is possible to see that several feedback terms can be neglected. With the typical values for c_{ps} , c_v , c_p , $c_{p,v}$, $c_{p,a}$, γ , and considering a low Mach number (see below) and values of T and T_o between 0 and about $40^{\circ}C$, only two feedback terms remain. They are the spray feedback due to sensible heat variations from the drop (second to last term in (4.9)) and the latent heat transfer from the drops to the water vapour (second term on the right-hand side of (4.11)). We also keep the molecular diffusion term $(\mathcal{D}' \text{ in } (4.9))$.

5. Discussion

5.1. Spray feedback estimates

Evidently, while individual drops may have sufficient time to transfer a particular property to the atmosphere, the spray induced feedback on the airflow, if any, depends on the aggregated drop effects, and thus on the drop size distribution and its integrals. Below, we present an example application of the model in which we examine the spray effects on the airflow. However, a comprehensive study of the influence of spray on the atmosphere over a wide range of environmental conditions is beyond the scope of this paper. Here, we simply illustrate the capability of the model with an example which corresponds to realistic marine weather conditions, yet not as extreme as storm events during which sea spray is known to be ubiquitous, and for which the present model is certainly expected to work well.

To this end, we consider the simplified equations obtained from the dimensional analysis in the section above. In dimensional form, they are:

$$\frac{\partial}{\partial t}\rho + \boldsymbol{\nabla} \cdot (\rho \boldsymbol{u}) = -\int_{\boldsymbol{\Omega}_{\boldsymbol{\mathfrak{X}}}} \mathcal{M} f d\boldsymbol{\mathfrak{X}}, \qquad (5.1)$$

$$\frac{\partial}{\partial t}q + \boldsymbol{u} \cdot \boldsymbol{\nabla}q = -\frac{1-q}{\rho} \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \mathcal{M}fd\boldsymbol{\mathfrak{X}} + \boldsymbol{\nabla} \cdot (\rho D_{v}\boldsymbol{\nabla}q), \tag{5.2}$$

$$\rho\left(\frac{\partial \boldsymbol{u}}{\partial t} + (\boldsymbol{u} \cdot \boldsymbol{\nabla})\boldsymbol{u}\right) + \boldsymbol{\nabla}p = \boldsymbol{\nabla} \cdot \boldsymbol{\sigma} + \rho \boldsymbol{g} - \int_{\boldsymbol{\Omega}_{\mathfrak{X}}} \boldsymbol{F} f d\boldsymbol{\mathfrak{X}}, \tag{5.3}$$

$$c_{v}\rho\left(\frac{\partial T_{air}}{\partial t} + \boldsymbol{u}\cdot\boldsymbol{\nabla}T_{air}\right) = \boldsymbol{\nabla}\cdot(\kappa\boldsymbol{\nabla}T_{air}) + \boldsymbol{\sigma}:\boldsymbol{\nabla}\boldsymbol{u} - p\boldsymbol{\nabla}\cdot\boldsymbol{u} + \mathcal{D}$$
$$-\int_{\boldsymbol{\Omega}_{\mathfrak{X}}} mc_{ps}\mathcal{T}fd\boldsymbol{\mathfrak{X}}$$
$$+\int_{\boldsymbol{\Omega}_{\mathfrak{X}}} L_{v}\mathcal{M}fd\boldsymbol{\mathfrak{X}}$$
(5.4)

As expected, for all the properties of the air that we are interested in (density, humidity, velocity, temperature), the feedback terms are integrals of the drop-induced rate of change of these quantities $(\mathcal{M}, \mathbf{F}, \mathcal{T})$, weighed by the number (concentration) distribution of the spray drops. The time rate of change of the drop velocity, mass, and temperature, are known from the microphysical equations (2.2), (2.7), and (2.11), although they are given in a Lagrangian frame of reference (see below). Equally importantly, the distribution function $f(t, \boldsymbol{x}, \boldsymbol{v}, r, m, T)$ now needs to be estimated.

Because spray is thought to have substantial impacts on short term weather events such as tropical storms, the airborne concentrations of drops and marine aerosols have long been of interest and several data sets have been acquired over the past decades. These data generally report steady and homogeneous, radius-dependent, drop concentration functions, noted N(r), which represent the number of drops per unit volume of air dxand per radius increment dr. To illustrate the model presented in this paper, we choose a somewhat reduced and idealized case in which we consider an initial drop size distribution which is instantly introduced in the airflow at t = 0. Here, we assume that this population of drops contains newly formed, motionless, sea water drops. In other words the initial drop distribution is:

$$f(t = 0, \boldsymbol{v}, r, m, T) = f_0(\boldsymbol{v}, r, m, T) = \delta_{\boldsymbol{v}-0} N(r) \delta_{m-m_{sw}(r)} \delta_{T-T_{sw}},$$
(5.5)

with $m_{sw}(r) = \frac{4}{3}\pi r^3 \rho_{sw}$ the mass of a drop of sea water. For the purpose of this discussion, we estimate the spray concentration N(r) from the source distribution S(r) using $N(r) = \frac{S(r)}{v_o(r)}$, and use the source distribution outlined in Andreas *et al.* (2010), which merges the functions proposed by Fairall *et al.* (1994) and Monahan *et al.* (1986).

In this example, we chose $U_{10} = 15 \text{ m s}^{-1}$ and limit our analysis to spray drops with radii from $r = 1 \ \mu\text{m}$ to $r = 1 \ \text{mm}$. Furthermore, two-way coupling is not accounted for and the atmospheric conditions imposed on the drops are kept fixed. In other words, we consider that the drop evolve in steady atmospheric conditions and the forcing imposed on them is fixed, but we do estimate the change that the drops would otherwise induce on the atmosphere. Thus, drop velocity, mass, and temperature evolve according to the Lagrangian equations (2.2), (2.7), and (2.11), and the drop distribution evolves following equation (2.15). For the latter, our approach, which we detail below, is based on a semianalytic method which uses the distribution N(r) and the corresponding solution to Williams-Boltzmann equation.

Following equations (5.1) to (5.4), at time t, if we ignore the effect of molecular diffusion of vapour in dry air, the spray-induced effects for mass, humidity, velocity, latent, and sensible heat, are given by:

$$\mathbb{M}_{S}(t) = -\int_{0}^{t} \int_{\Omega_{\mathfrak{X}}} \mathcal{M}(\mathfrak{X}) f(\tilde{t}, \mathfrak{X}) d\mathfrak{X} d\tilde{t}, \qquad \mathbb{H}_{S}(t) = \frac{1-q}{\rho} \mathbb{M}_{S}(t), \tag{5.6}$$

$$\mathbb{U}_{S}(t) = -\frac{1}{\rho} \int_{0}^{t} \int_{\Omega_{\mathfrak{X}}} \boldsymbol{F}(\mathfrak{X}) f(\tilde{t}, \mathfrak{X}) d\mathfrak{X} d\tilde{t},$$
(5.7)

$$\mathbb{T}_{S,L}(t) = -\frac{L_v}{c_v \rho} \mathbb{M}_S(t), \qquad \mathbb{T}_{S,S}(t) = -\frac{1}{c_v \rho} \int_0^t \int_{\Omega_{\mathfrak{X}}} mc_{ps} \mathcal{T}(\mathfrak{X}) f(\tilde{t}, \mathfrak{X}) d\mathfrak{X} d\tilde{t}, \qquad (5.8)$$

where $f(t, \mathfrak{X})$ is the spatially homogeneous solution to equation (2.15) in which t is a dummy time variable used for the integration and with f_0 as initial condition (see above). All feedback quantities involve integrals of the form:

$$\int_{\Omega_{\mathfrak{X}}} \phi(\mathfrak{X}) f(t,\mathfrak{X}) d\mathfrak{X}, \tag{5.9}$$

which can be calculated as follows. We first consolidate the Lagrangian equations (2.2), (2.7), (2.9), (2.11) as a differential system,

$$\frac{dX(t)}{dt} = V(X(t)), \qquad X(0) = \mathfrak{X}_0, \tag{5.10}$$

where $X(t) = (\mathbf{v}(t), r(t), m(t), T(t))$. Using a change in variable, which for all times t transforms $\mathfrak{X}_0 = (\mathbf{x}_0, \mathbf{v}_0, r_0, m_0, T_0)$ to $\mathfrak{X} = X(t)$, a known result from transport theory then states that:

$$\int_{\Omega_{\mathfrak{X}}} \phi(\mathfrak{X}) f(t,\mathfrak{X}) d\mathfrak{X} = \int_{\Omega_{\mathfrak{X}_0}} \phi(X(t)) f_0(\mathfrak{X}_0) d\mathfrak{X}_0.$$
(5.11)

This formula then allows us solve the feedback equations above, so long as we can solve for the Lagrangian equations (2.2), (2.7), (2.9), (2.11). Because the Lagrangian equations for the spray are unnecessarily complicated for this simple example, we use the reduced relaxation model given in Andreas (1989). It also has the added benefit of simple analytical solutions.

Therefore, we approximate equations (2.7) and (2.11) with:

$$\frac{dr(t)}{dt} = \frac{r_{eq}(r_0) - r(t)}{\tau_R(r_0)},\tag{5.12}$$

and

$$\frac{dT(t)}{dt} = \frac{T_{ev}(r_0) - T(t)}{\tau_T(r_0)},$$
(5.13)

where τ_R and τ_T are shown in figure 2, $r_{eq}(r_0) = \alpha r_0$ is the radius at equilibrium (α depends only on ambient atmospheric conditions), and T_{ev} the so-called evaporating temperature. The use of this simplified relaxation model is justified by examining the results in figure 1, which shows that the temporal evolution (and thus the time rate of change) of a drop's temperature, velocity, and radius, can indeed be approximated by an exponential. Here, we consider air with a wind speed of $U_{10} = 15 \text{ m s}^{-1}$, a temperature of $T_{air} = 18^{\circ}C$ and a relative humidity of 75%, which leads to $\alpha \approx 0.46$. The spray

Lagrangian equations then yield

$$u(t) = U_{10} - U_{10}e^{-\int_0^t \frac{ds}{\tau_D(r(s))}},$$
(5.14)

$$r(t) = \left(\alpha + (1 - \alpha)e^{-t/\tau_R(r_0)}\right)r_0,$$
(5.15)

$$m(t) = m_0 + \frac{4}{3}\pi \left(\left(\alpha + (1 - \alpha)e^{-t/\tau_R(r_0)} \right)^3 - 1 \right) r_0^3 \rho_w,$$
(5.16)

$$T(t) = T_{ev}(r_0) + (T_0 - T_{ev}(r_0))e^{-t/\tau_T(r_0)},$$
(5.17)

where, for simplicity, we only consider the horizontal component of the velocity u(t). The corresponding Lagrangian rates of changes are

$$F(t) = \frac{m(t)}{\tau_D(r(t))} (U_{10} - u(t)), \qquad (5.18)$$

$$\mathcal{R}(t) = \frac{\alpha r_0 - r(t)}{\tau_R(r_0)},\tag{5.19}$$

$$\mathcal{M}(t) = 4\pi r(t)^2 \rho_w \mathcal{R}(t), \qquad (5.20)$$

$$\mathcal{T}(t) = \frac{T_{ev}(r_0) - T(t)}{\tau_T(r_0)}.$$
(5.21)

where equation (5.11) naturally leads to a relatively simple estimate of the airside variations in density, humidity, temperature, and speed, which are induced by the introduction in the airflow of a population of sea-water spray drops at time t = 0.

5.1.1. Estimation of the feedback on the humidity of the air

From equation (5.1), the water mass (per unit volume. i.e. the density) transferred to the air by the evaporating spray is

$$\mathbb{M}_{S}(t) = -\int_{0}^{t} \int_{\Omega_{\mathfrak{X}_{0}}} \mathcal{M}(\tilde{t}) f_{0}(\mathfrak{X}_{0}) d\mathfrak{X}_{0} d\tilde{t}, \qquad (5.22)$$

in which $\mathcal{M}(\tilde{t})$ is given by equation (5.20) above. Using the known value for f_0 and integrating in time yields

$$\mathbb{M}_{S}(t) = \int_{0}^{+\infty} \frac{4}{3} \pi r_{0}^{3} \rho_{w} \left(1 - \alpha^{3} - \phi \left(\frac{t}{\tau_{R}(r_{0})} \right) \right) N(r_{0}) \, dr_{0}, \tag{5.23}$$

where $\phi(\zeta) = (1 - \alpha)(3\alpha^2 e^{-\zeta} + 3\alpha(1 - \alpha)e^{-2\zeta} + (1 - \alpha)^2 e^{-3\zeta}).$

Note that for large times, \mathbb{M}_S tends toward

$$(1 - \alpha^3) \int_0^{+\infty} \frac{4}{3} \pi r_0^3 \rho_w N(r_0) \, dr_0, \qquad (5.24)$$

which simply represent the initial pure water mass, less the maximum water mass that can potentially evaporate, $\int_0^{+\infty} \frac{4}{3}\pi(\alpha r_0)^3\rho_w N(r_0) dr_0$. In fact, this limit readily gives a quick upper limit estimate of the potential total water vapour that can be transferred to the atmosphere.

In figure 4, we plot the corresponding relative humidity increase as a function of time, assuming that the spray drop distribution is introduced in the airflow at t = 0. Similarly to the flow conditions of figure 1, the air has a steady wind speed of $U_{10} = 15 \text{ m s}^{-1}$, a temperature of $T_{air} = 18^{\circ}C$, and a relative humidity of 75%. The drops all have an initial temperature of $T_p = 20^{\circ}C$. Within $O(10^3)$ s, the drop distribution has transferred the bulk of its water vapour to the atmosphere. During that time, the air relative humidity

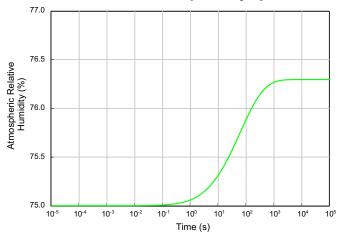


FIGURE 4. Relative humidity due to the spray drops with a size distribution N(r). The distribution is assumed to be introduced at t = 0 in air with a steady wind speed of $U_{10} = 15$ m s⁻¹, a temperature of $T_{air} = 18^{\circ}C$, and a relative humidity of 75%. The drops all have an initial temperature of $T_p = 20^{\circ}C$.

has increased from 75% to approximately 76.3%. We note here that figure 4 shows the aggregated results for the whole size distribution N(r). In figure 2, the relaxation time τ_m for this distribution ranges from $O(10^{-2})$ s to $O(10^4)$ s and we therefore expect the small drops to evaporate much faster than the large drops. But the time over which atmospheric humidity is increased which is depicted in figure 4, is due to the whole size dependent distribution N(r), along with the corresponding size dependent relaxation time $\tau_m(r)$. In fact, it appears that the distribution N(r) does not possess a sufficient amount of large drops of sizes O(1) mm to drive the relative humidity increase over long times, and the relative humidity stabilizes at times shorter than $O(10^4)$ s, time that would be required for the largest drops to evaporate. In other words, the larger drops in the distribution do not contribute much to the change in humidity; if they did, the atmospheric humidity would still be changing until times of order $O(10^4)$ s.

This is somewhat fortuitous because the model above also assumes that all the drops of the distribution, including the largest ones, remain suspended in the air. And it is likely that large spume drop do not stay suspended long enough, at this wind speed, to fully evaporate (see figures 2 and 3). Furthermore, two-way coupling was not accounted for in this idealized example and the atmospheric conditions are considered steady. In reality, ambient relative humidity changes as the spray evaporates. Of course, the fully unsteady solutions can be obtained by solving equation (2.15) which gives the spray size distribution, concurrently with equations (5.1) or (5.2), (5.3) and (5.4) which give the atmospheric condition including the (instantaneous) spray effects. In that sense, the result presented above is a slight overestimate of the the feedback effects from the drop distribution on the atmospheric relative humidity. F. Veron & L. Mieussens

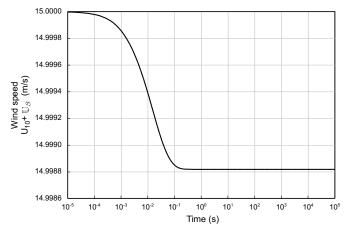


FIGURE 5. Decrease of the air velocity due to the drag force exerted by the drops given by the distribution N(r). Initial ambient conditions are the same as those shown in figure 4

5.1.2. Estimation of the feedback on the air velocity

Similarly, from equation (5.11), at time t, the variation in wind speed due to the spray distribution is

$$\mathbb{U}_{S}(t) = -\frac{1}{\rho} \int_{0}^{t} \int_{\Omega_{\mathfrak{X}_{0}}} F(\tilde{t}) f_{0}(\mathfrak{X}_{0}) d\mathfrak{X}_{0} d\tilde{t}
= -\frac{1}{\rho} \int_{0}^{t} \int_{\Omega_{\mathfrak{X}_{0}}} \frac{\frac{4}{3} \pi r(\tilde{t})^{3} \rho_{w} + m_{sw}(r_{0})}{\tau_{D}(r(\tilde{t}))} U_{10} e^{-\int_{0}^{\tilde{t}} \frac{ds}{\tau_{D}(r(\tilde{t}))}} N(r_{0}) dr_{0} d\tilde{t}.$$
(5.25)

Neglecting high Reynolds number corrections on the drag force, thus assuming that $\tau_D(r) = \tau_p(r) = \frac{2r^2 \rho_{sw}}{9\mu}$ (see section 2.1), and noting that $\frac{\tau_D}{\tau_R} \approx O(10^{-3}) \ll 1$ (see figure 2), lead to a simpler analytical estimate

$$\mathbb{U}_{S}(t) = -\frac{1}{\rho} U_{10} \alpha^{2} \left(1 + \frac{\rho_{w}}{\rho_{sw}} \right) \int_{0}^{+\infty} m_{sw}(r_{0}) \left(1 - e^{-\frac{t}{\alpha^{2} \tau_{D}(r_{0})}} \right) N(r_{0}) dr_{0}.$$
(5.26)

Note that for large times, \mathbb{U}_S tends toward:

$$-\frac{1}{\rho}U_{10}\alpha^{2}\left(1+\frac{\rho_{w}}{\rho_{sw}}\right)\int_{0}^{+\infty}m_{sw}(r_{0})N(r_{0})\,dr_{0},\tag{5.27}$$

where $\int_0^{+\infty} m_{sw}(r_0) N(r_0) dr_0$ is sea spray mass fraction. Again, this gives a quick way to estimate the potential dynamic effect of a given spray concentration $N(r_0)$.

Figure 5 shows the variation in wind speed due to the spray distribution. For the exchange of momentum, figure 2 indicates that the relaxation time is sufficiently short for most of the drop sizes to fully exchange momentum with the atmosphere. In fact, figure 5 shows that the whole distribution has completed its momentum exchange in less that O(1) s. Yet, the momentum extracted from the wind to accelerate the spray drops (which are assumed to start at rest) is negligible, at least in the example taken here. This simply indicates that the spray mass loading given by $N(r_0)$ at this wind speed is too small to have a dynamic effect on the carrier fluid. Larger mass loading evidently can have a substantial impact on the spray-atmosphere momentum exchange (see for example the recent work of Richter & Sullivan (2013b,a); Richter (2015); Richter *et al.* (2016)).

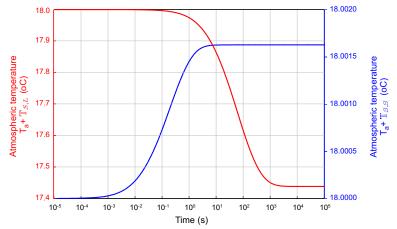


FIGURE 6. Variation of the air temperature due to the sensible (blue) and latent (red) heat flux from the distribution of sea spray drops given by N(r). Initial ambient conditions are the same as those shown in figure 4

5.1.3. Estimation of the feedback on the air temperature

From equation (5.4), the rate of change of the air temperature due to the evaporation of sea spray is the sum of the two terms corresponding to the spray latent and sensible heat transfers. Together, equations (5.8) and (5.23) give the spray induced latent heat $\mathbb{T}_{S,L}(t)$. Likewise, equations (5.8) and (5.11) give the component corresponding to the spray sensible heat

$$\begin{aligned} \mathbb{T}_{S,S}(t) &= -\frac{1}{c_v \rho} \int_0^t \int_{\Omega_{\mathfrak{X}_0}} m(\tilde{t}) c_{ps} \mathcal{T}(\tilde{t}) f_0(\mathfrak{X}_0) d\mathfrak{X}_0 d\tilde{t} \\ &= \frac{1}{c_v \rho} \int_0^{+\infty} \left(m_{sw}(r_0) (1 - e^{-t/\tau_T(r_0)}) \right. \\ &\quad \left. + \frac{4}{3} \pi r_0^3 \rho_w \psi \left(\frac{t}{\tau_T(r_0)}, \frac{\tau_T(r_0)}{\tau_R(r_0)} \right) \right) c_{ps}(T_{sw} - T_{ev}(r_0)) N(r_0) \, dr_0, \end{aligned}$$

$$(5.28)$$

with

$$\psi(x,y) = (\alpha^3 - 1)(1 - e^{-x}) + 3\alpha^2(\alpha - 1)\frac{1}{1+y}(1 - e^{-x(1+y)}) + 3\alpha(\alpha - 1)^2\frac{1}{1+2y}(1 - e^{-x(1+2y)}) + (1 - \alpha)^3\frac{1}{1+3y}(1 - e^{-x(1+3y)}).$$
(5.29)

Noting that $\frac{\tau_T}{\tau_R} \approx O(10^{-3}) \ll 1$ (see figure 2), the expression above can be substantially simplified:

$$\mathbb{T}_{S,S}(t) = \frac{1}{c_v \rho} \int_0^{+\infty} \left(m_{sw}(r_0) - 6\alpha^2 (1-\alpha) \frac{4}{3} \pi r_0^3 \rho_w \right) (1 - e^{-t/\tau_T(r_0)}) c_{ps}(T_{sw} - T_{ev}(r_0)) N(r_0) \, dr_0.$$
(5.30)

Figure 6 shows the resulting variation in the air temperature induced respectively by the latent (red curve) and sensible (blue curve) heat exchanges from the spray. Again, just as with the spray effects on the humidity, the relaxation time for the latent heat flux is such that the spray contribution may be slightly overestimated when accounting for the

F. Veron & L. Mieussens

entire size distribution including a few large drops. The temperature change induced by the spray sensible flux is likely to be more accurate, from a time-scale point of view, but the results show that it is a negligible part of the total spray-induced air temperature change. Furthermore, figure 1 shows that the drops eventually reach and equilibrium temperature T_{eq} which is generally larger than the evaporation temperature T_{ev} assumed in the relaxation model of equation (5.13). Thus the atmospheric temperature change induced by the sensible flux from the spray is likely to be even smaller than that estimated here. Overall, figure 6 shows that the bulk to the spray effects on the air temperature comes from the latent heat exchange from the drop distribution. As such, just like the effects on the relative humidity, it also takes $O(10^3)$ s to fully establish. In the example taken here, the atmospheric temperature is decreased by approximately $0.5^{\circ}C$.

5.2. Perspectives

Lastly, we offer here a discussion of possible future work and potential avenues for improvements to the preliminary model presented above.

5.2.1. Incompressible approximation

While the approach presented here can be applied to a range of particle-laden flows, in the case of sea spray, the air can generally be considered as nearly incompressible. Indeed, the Mach number ranges from 0.01 for a wind speed of 5 m s⁻¹ to approximately 0.2 in extreme winds of 80 m $\,\mathrm{s}^{-1}$. In this case, it is customary to replace the Navier-Stokes equations by the corresponding incompressible equations. Formally, these equations are obtained by assuming a uniform and steady density for the air, in which case the conservation equation of the mass is replaced by $\nabla \cdot \boldsymbol{u} = 0$; this new relationship is then included in the momentum equation. Furthermore, if the variations of temperature are sufficiently low (of the order of a few percent in Kelvin, or less than a dozen degrees Celsius, which is generally the case for sea spray), one can take into account the natural convection by using the well-known Boussinesq approximation in which the gravity term ρg is replaced with $\frac{T-T_o}{T_o}\rho_o g$. In our case, terms accounting for feedback effects from the spray complicate the traditional Boussinesq approximation. Therefore, for readers interested in pursuing the analysis beyond what is presented here, we recommend proceeding with the so called "low Mach" asymptotic analysis (Feireisl & Novotný 2009) where equations are written as a series expansion using the Mach number as the small parameter.

5.2.2. Turbulent flows

The airflow over the wavy ocean surface is turbulent and the fluctuations in the air velocity thereby induce fluctuations in the velocity of the drops. When the Navier-Stokes equations for the airflow are solved with a sufficient resolution, the present model accounts for these fluctuations. However, solving Navier-Stokes equations with domains sufficiently large to cover spray transport generally requires numerical resolutions which are not sufficient to capture the turbulence of the air, which is then typically modeled using LES or RANS approaches. In this case, the diffusion induced by the turbulence is parametrized without specifically describing the fluctuations of the velocity field. Equivalently, our model can be modified to account for the turbulent fluctuations of the air and drop velocities. In the Lagrangian description, fluctuations for the drop velocities can be modeled by a turbulence-induced forcing thus leading to a stochastic differential equation (analogous to a Langevin's equation) for v_p . In the Eulerian framework, a corresponding (and still deterministic) Williams-Boltzmann equation can be obtained through the Feynman-Kac representation theorem (see for example Domelevo & Villedieu (2007)).

5.2.3. Numerical simulations

The Eulerian model presented here to study spray and spray effects on the atmosphere is consistent with the typical Eulerian approach for solving large-scale atmospheric flows. From a numerical point of view, this simply means that similar numerical methods can be used for the spray, the carrier atmospheric flow, and the feedback/coupling terms. Meanwhile, using a Lagrangian model to describe the spray and an Eulerian model for the carrier fluid is an approach that is frequently used (e.g. Edson & Fairall 1994; Edson *et al.* 1996; Mueller & Veron 2014*a*,*b*; Richter & Sullivan 2013*b*,*a*; Helgans & Richter 2016; Richter & Sullivan 2014; Peng & Richter 2019; Richter *et al.* 2019), but which presents a number of challenges such as numerical stability problems and a computing cost that can quickly become prohibitive especially for unsteady flows.

Practically, an Eulerian numerical simulation will not directly solve the complete Williams-Boltzmann (equation (2.15)) because the size of the phase space is generally far too large (at least 9 variables in a three-dimensional physical space). Thus, so called "fluid models" obtained from the kinetic equation are generally favored. These models can be obtained from conservation equations for moments of the distribution function $f(t, \boldsymbol{x}, \boldsymbol{v}, r, m, T)$, with the addition of an appropriate closure hypothesis. This approach is used successfully in combustion (Desjardins *et al.* 2008; Marchiso & Fox 2013), and several types of models exist. In particular, the "Quadrature-Based Moment Method" approach seems quite suitable for the spray problem presented here. For example, we briefly describe below the single-node closure in which the distribution $f(t, \boldsymbol{x}, \boldsymbol{v}, r, m, T)$ is reduced to $n(t, \boldsymbol{x}, r, m)\delta_{\boldsymbol{v}-\boldsymbol{U}(t, \boldsymbol{x}, r, m)}\delta_{T-\Theta(t, \boldsymbol{x}, r, m)}$. This simply means that at any point in space, drops of radii r and mass m all have the same speed \boldsymbol{U} and the same temperature Θ , while these parameters can change in time and space. With these simplifications, it is straightforward to obtain three evolution equations for n, \boldsymbol{U} and Θ . Indeed, integrating (2.15) multiplied by 1, \boldsymbol{v} , and T, yields:

$$\frac{\partial n}{\partial t} + \boldsymbol{\nabla} \cdot (n\boldsymbol{U}) + \frac{\partial}{\partial r}(\bar{\mathcal{R}}n) + \frac{\partial}{\partial m}(\bar{\mathcal{M}}n) = 0, \qquad (5.31)$$

$$\frac{\partial n\boldsymbol{U}}{\partial t} + \boldsymbol{\nabla} \cdot (n\boldsymbol{U} \otimes \boldsymbol{U}) + \frac{\partial}{\partial r}(\bar{\mathcal{R}}n\boldsymbol{U}) + \frac{\partial}{\partial m}(\bar{\mathcal{M}}n\boldsymbol{U}) = n\boldsymbol{g} + \frac{1}{\tau_D}n(\boldsymbol{u} - \boldsymbol{U}), \qquad (5.32)$$

$$\frac{\partial n\Theta}{\partial t} + \boldsymbol{\nabla} \cdot (n\boldsymbol{U}\Theta) + \frac{\partial}{\partial r}(\bar{\mathcal{R}}n\Theta) + \frac{\partial}{\partial m}(\bar{\mathcal{M}}n\Theta) = \bar{\mathcal{T}}n, \qquad (5.33)$$

where $\overline{\mathcal{R}}$, $\overline{\mathcal{M}}$, and $\overline{\mathcal{T}}$ are the rate of variations of the drops in which the kinetic variables \boldsymbol{v} and T are replaced by the values \boldsymbol{U} and Θ of the closure. This system can be used instead of the complete Williams-Boltzmann equation, which is no longer necessary.

The feedback terms are also easily obtained from n, U, and Θ . Thus, the reduced model consists of semi-macroscopic equations with n, U, and Θ above and the equations for the air flow. It is then possible to further reduce the model by eliminating the variables r and m. To that end, two types of approaches exist. The first is based on a direct integration and leads to the loss of the polydispersed nature of the problem; this obviously seems inappropriate in the case of sea spray. The second is to discretize the variables r and m, as in the "sampling" and "sectional" or "multi-fluid" approaches; this seems better suited for the problem at hand (see Marchiso & Fox (2013)).

For realistic applications, this one node closure is not sufficient, and several higher order closure have been recently developed to obtain more accurate models, see Fox *et al.* (2018) and the references therein.

6. Conclusions

In this paper, we have presented a novel model to estimate sea spray feedback effects on the atmospheric flow. Evidently, the spray effects depend on the exchange rates of momentum, heat, and mass, from the population of drops that make up sea spray. The spray mediated exchanges are therefore generally obtained from the equation of motion and the thermodynamics of individual drops as they are transported in the atmospheric boundary layer. These are naturally given from a Lagrangian point of view. In fact, Lagrangian drop transport models are valuable tools to estimate spray residence times in the air. However, the atmospheric flow is better described in a conventional Eulerian frame of reference with the usual mass, momentum, and energy conservation equations, i.e. the standard Navier-Stokes set. Evaluating spray effects on the atmosphere thus generally involves coupling the Lagrangian spray transport with the Eulerian atmospheric flow. This is quite impractical and usually computationally expensive. In this paper, we present a model which utilizes the kinetic theory framework and thus reconciles the Eulerian and Lagrangian methods. This kinetic approach affords substantial advantages as it leads to a coupled Eulerian spray-atmosphere model which can be solved to estimate spray feedback effects.

We further performed a dimensional analysis and isolated the spray induced dominant terms in the equations for the atmospheric flow. Subsequently, we presented, for illustration purpose, an example estimate of the spray feedback effects in which we have used a commonly accepted sea spray drop size distribution forcing function. A complete investigation of the spray effects over a wide range of atmospheric wind speed, temperature, and humidity is beyond the scope of this paper. Nevertheless, the spray induced feedback on the atmosphere which results from this simple estimate, and for these particular atmospheric conditions, shows that the spray latent flux, but not the sensible flux, drives an atmospheric temperature change. The spray evaporation also has a measurable effect on the relative humidity in the airflow. The spray effects on the momentum of the airflow are negligible in the condition studied. We expect spray momentum effects to occur only at substantial spray mass loading.

While the model developed here is robust, the largest source of error and uncertainty lies with the difficulty of defining a reliable spray size distribution. Clearly, experiments are needed in order to obtain, over a wide range of environmental conditions, measured spray size distributions, or spray generation functions which are generally needed to assess air-sea fluxes.

Acknowledgements

This work was supported in part on grant OCE-1829660 from the US National Science Foundation.

REFERENCES

- ADRIAN, R J 1991 Particle-imaging techniques for experimental fluid mechanics. Annu. Rev. Fluid Mech. 23 (1), 261–304.
- ANDREAS, E L 1989 Thermal and size evolution of sea spray droplets. CRREL Report 89-11.
- ANDREAS, E L 1990 Time constants for the evolution of sea spray droplets. Tellus B 42 (5), 481-497.
- ANDREAS, E L 1992 Sea spray and the turbulent air-sea heat fluxes. J. Geophys. Res. 97 (C7), 11429–11441.
- ANDREAS, E L 1998 A new sea spray generation function for wind speeds up to 32m s^{-1} . J. Phys. Oceanogr. 28 (11), 2175–2184.
- ANDREAS, E L 2002 A review of the sea spray generation function for the open ocean. In Atmosphere-Ocean Interactions (ed. W A Perrie), , vol. 1, pp. 1–46. Southhampton, UK: WIT Press.
- ANDREAS, E L 2004 Spray stress revisited. J. Phys. Oceanogr. 34 (6), 1429–1440.
- ANDREAS, E L & DECOSMO, J 1999 Sea spray production and influence on air-sea heat and moisture fluxes over the open ocean. In Air-Sea Exchange: Physics, Chemistry and Dynamics (ed. G L Geernaert), pp. 327–362. Kluwer.
- ANDREAS, E L, EDSON, J B, MONAHAN, E C, ROUAULT, M P & SMITH, S D 1995 The spray contribution to net evaporation from the sea: a review of recent progress. *Bound.-Layer Meteor.* 72 (1-2), 3–52.
- ANDREAS, E L & EMANUEL, K A 2001 Effects of sea spray on tropical cyclone intensity. J. Atmos. Sci. 58 (24), 3741–3751.
- ANDREAS, EDGAR L., JONES, KATHLEEN F. & FAIRALL, CHRISTOPHER W. 2010 Production velocity of sea spray droplets. J. Geophys. Res. 115, C12065.
- ANDREAS, E L, PERSSON, P O G & HARE, J E 2008 A bulk turbulent air-sea flux algorithm for high-wind, spray conditions. J. Phys. Oceanogr. 38 (7), 1581–1596.
- ANIS, A & MOUM, J N 1995 Surface wave-turbulence interactions: Scaling $\epsilon(z)$ near the sea surface. J. Phys. Oceanogr. 25, 346–366.
- BAO, J W, FAIRALL, C W, MICHELSON, S A & L, BIANCO 2011 Parameterizations of sea-spray impact on the air-sea momentum and heat fluxes. *Mon. Wea. Rev.* **139**, 3781–3797.
- BAO, J W, WILCZAK, J M, CHOI, J-K & KANTHA, L H 2000 Numerical simulations of airsea interaction under high wind conditions using a coupled model: A study of hurricane development. Mon. Wea. Rev. 128, 2190–2210.
- BARENBLATT, G I, CHORIN, A J & PROSTOKISHIN, V M 2005 A note concerning the lighthill "sandwhich model" of tropical cyclones. *Proc. Natl. Acad. Sci.* **102**, 11148–11150.
- BELL, MICHAEL M., MONTGOMERY, MICHAEL T. & EMANUEL, KERRY A. 2012 Air-sea enthalpy and momentum exchange at major hurricane wind speeds observed during cblast. J. Atmos. Sci. 69 (3), 3197–3222.
- BIANCO, L, BAO, J W, FAIRALL, C W & MICHELSON, S A 2011 Impact of sea-spray on the atmospheric surface layer. *Bound.-Layer Meteor.* .
- BLANCHARD, D C 1989 The size and height to which jet drops are ejected from bursting bubbles in sea water. J. Geophys. Res. **94** (C8), 10999–11002.
- CARRUTHERS, DJ & CHOULARTON, TW 1986 The microstructure of hill cap clouds. Quarterly Journal of the Royal Meteorological Society 112 (471), 113–129.
- CLIFT, R & GAUVIN, W H 1970 The motion of particles in turbulent gas streams. Proc. Chemeca'70 1.
- DAVIES, C N 1945 Definitive equations for the fluid resistance of spheres. Proc. Phys. Soc. London 57 (Pt.4), 259–270.
- DESJARDINS, O., FOX, R.O. & VILLEDIEU, P. 2008 A quadrature-based moment method for dilute fluid-particle flows. Journal of Computational Physics 227 (4), 2514 – 2539.
- DESVILLETTES, L, GOLSE, F & RICCI, V 2008 The mean-field limit for solid particles in a Navier-Stokes flow. J. Stat. Phys. 131 (5), 941–967.
- DOMELEVO, K & VILLEDIEU, P 2007 A hierarchy of models for turbulent dispersed two-phase flows derived from a kinetic equation for the joint particle-gas pdf. *Commun. Math. Sci.* **5** (2), 331–353.
- DONELAN, M A, HAUS, B K, REUL, N, PLANT, W J, STIASSNIE, M, GRABER, H C, BROWN,

O B & SALTZMAN, E S 2004 On the limiting aerodynamic roughness of the ocean in very strong winds. *Geophys. Res. Lett.* **31** (18).

- DRENNAN, W M, ZHANG, J A, FRENCH, J R, MCCORMICK, C & BLACK, P G 2007 Turbulent fluxes in the hurricane boundary layer. part ii: Latent heat flux. J. Atmos. Sci. 64 (4), 1103–1115.
- EDSON, J.B. & FAIRALL, C.W. 1998 Similarity relationships in the marine surface layer. J. Atmos. Sci. 55, 2311–2328.
- EDSON, J B, ANQUETIN, S, MESTAYER, P G & SINI, J F 1996 Spray droplet modeling. 2. an interactive eulerian-lagrangian model of evaporating spray droplets. J. Geophys. Res. 101, 1279–1293.
- EDSON, J B & FAIRALL, C W 1994 Spray droplet modeling. 1. lagrangian model simulation of the turbulent transport of evaporating droplets. J. Geophys. Res. 99 (C12), 25295–25311.
- ERININ, MA, WANG, SD, LIU, R, TOWLE, D, LIU, X & DUNCAN, JH 2019 Spray generation by a plunging breaker. *Geophysical Research Letters*.
- FAIRALL, C W, BANNER, M L, PEIRSON, W L, ASHER, W & MORISON, R P 2009 Investigation of the physical scaling of sea spray spume droplet production. J. Geophys. Res. 114 (C10).
- FAIRALL, C W, BRADLEY, E F, ROGERS, D P, EDSON, J B & YOUNG, G S 1996 Bulk parameterization of air-sea fluxes for tropical ocean-global atmosphere coupled-ocean atmosphere response experiment. J. Geophys. Res. 101, 3747–3764.
- FAIRALL, C W, KEPERT, J D & HOLLAND, G J 1994 The effect of sea spray on surface energy transports over the ocean. *Global Atmos. Ocean Syst.* 2, 121–142.
- FEIREISL, EDUARD & NOVOTNÝ, ANTONÍN 2009 Singular limits in thermodynamics of viscous fluids. Birkhäuser Verlag, Basel.
- FOX, RODNEY O., LAURENT, FRÉDÉRIQUE & VIÉ, AYMERIC 2018 Conditional hyperbolic quadrature method of moments for kinetic equations. *Journal of Computational Physics* 365, 269 – 293.
- FUENTES, E., COE, H., GREEN, D., DE LEEUW, G. & MCFIGGANS, G. 2010 Laboratorygenerated primary marine aerosol via bubble-bursting and atomization. Atmos. Meas. Techn. 3 (1), 141–162.
- HARE, J E, HARA, T, EDSON, J B & WILCZACK, J M 1997 A similarity analysis of the structure of airflow over surface waves. J. Phys. Oceanogr. 27, 1018–1037.
- HELGANS, BRIAN & RICHTER, DAVID H. 2016 Turbulent latent and sensible heat flux in the presence of evaporative droplets. *International Journal of Multiphase Flow* **78**, 1–11.
- HRISTOV, TIHOMIR, FRIEHE, CARL & MILLER, SCOTT 1998 Wave-coherent fields in air flow over ocean waves: Identification of cooperative behavior buried in turbulence. *Physical review letters* 81 (23), 5245.
- KOGA, M 1981 Direct production of droplets from breaking wind-waves, its observation by a multi-colored overlapping exposure photographing technique. *Tellus* **33**, 552–563.
- DE LEEUW, G, ANDREAS, E L, ANGUELOVA, M D, FAIRALL, C W, LEWIS, E R, O'DOWD, C, SHULTZ, M & SCHWARTZ, S E 2011 Production flux of sea spray aerosol. *Reviews in Geophysics* 49.
- LEWIS, E R & SCHWARTZ, S E 2004 Sea Salt Aerosol Production: Mechanisms, Methods, Measurements, and Models. Amer. Geophys. Union, Washington.
- LIU, B, LIU, H, XIE, L, GUAN, C & ZHAO, D 2010 A coupled atmosphere-wave-ocean modeling system: Simulation of the intensity of an idealized tropical cyclone. *Mon. Wea. Rev.* 139 (1), 132–152.
- MAKIN, V K 1998 Air-sea exchange of heat in the presence of wind waves and spray. J. Geophys. Res. 103, 1137–1152.
- MARCHISO, E & FOX, RO 2013 Computational Models for Polydisperse Particulate and Multiphase Systems. Cambridge University Press.
- MARMOTTANT, P H & VILLERMAUX, E 2004 On spray formation. J. Fluid Mech. 498, 73-111.
- MELVILLE, W K 1994 Energy dissipation by breaking waves. J. Phys. Oceanogr. 24, 2041–2049.
- MELVILLE, W K 1996 The role of surface-wave breaking in air-sea interaction. Annu. Rev. Fluid Mech. 28, 279–321.
- MESTAYER, P G, VAN EIJK, A M J, DE LEEUW, G & TRANCHANT, B S 1996 Numerical simulation of the dynamics of sea spray over the waves. J. Geophys. Res. 101, 20771– 20797.

- MICHAELIDES, EFSTATHIOS E & FENG, ZHIGANG 1994 Heat transfer from a rigid sphere in a nonuniform flow and temperature field. *International journal of heat and mass transfer* **37** (14), 2069–2076.
- MONAHAN, E.C., SPIEL, D.E. & DAVIDSON, K.L. 1986 A model of marine aerosol generation via whitecaps and wave disruption. In *Oceanic Whitecaps* (ed. EdwardC. Monahan & GearóidMac Niocaill), *Oceanographic Sciences Library*, vol. 2, pp. 167–174. Springer Netherlands.
- MUELLER, J & VERON, F 2010a A lagrangian stochastic model for sea-spray evaporation in the atmospheric marine boundary layer. *Bound.-Layer Meteor.* **137**, 135–152.
- MUELLER, J A & VERON, F 2009*a* A lagrangian stochastic model for heavy particle dispersion in the atmospheric marine boundary layer. *Bound.-Layer Meteor.* **130** (2), 229–247.
- MUELLER, J A & VERON, F 2009b A sea state-dependent spume generation function. J. Phys. Oceanogr. **39**, 2363–2372.
- MUELLER, J A & VERON, F 2010b Bulk formulation of the heat and water vapor fluxes at the air-sea interface, including nonmolecular contributions. J. Atmos. Sci. 67, 234–247.
- MUELLER, J A & VERON, F 2014a Impact of sea spray on air-sea fluxes part 1: Results from stochastic simulations of sea spray drops over the ocean. J. Phys. Oceanogr. p. In Revision.
- MUELLER, J A & VERON, F 2014b Impact of sea spray on air-sea fluxes part 2: Feedback effects. *J. Phys. Oceanogr.* p. In Revision.
- ORTIZ-SUSLOW, DAVID G, HAUS, BRIAN K, MEHTA, SANCHIT & LAXAGUE, NATHAN JM 2016 Sea spray generation in very high winds. *Journal of the Atmospheric Sciences* 73 (10), 3975–3995.
- PENG, TIANZE & RICHTER, DAVID 2017 Influence of evaporating droplets in the turbulent marine atmospheric boundary layer. *Boundary-layer meteorology* **165** (3), 497–518.
- PENG, TIANZE & RICHTER, DAVID 2019 Sea spray and its feedback effects: Assessing bulk algorithms of air–sea heat fluxes via direct numerical simulations. *Journal of Physical Oceanography* **49** (6), 1403–1421.
- PRUPPACHER, HR & KLETT, JD 1996 *Microphysics of Clouds and Precipitation*, , vol. 18. Springer Science & Business Media.
- PRUPPACHER, H R & KLETT, J D 1978 Microphysics of Clouds and Precipitation. D. Riedel.
- RANZ, WE, MARSHALL, W R_ & OTHERS 1952 Evaporation from drops. Chem. eng. prog 48 (3), 141–146.
- RICHTER, DAVID H. 2015 Turbulence modification by inertial particles and its influence on the spectral energy budget in planar Couette flow. *Physics of Fluids* 27.
- RICHTER, DAVID H, DEMPSEY, ANNE E & SULLIVAN, PETER P 2019 Turbulent transport of spray droplets in the vicinity of moving surface waves. *Journal of Physical Oceanography* (2019).
- RICHTER, DAVID H., GARCIA, OMAR & ASTEPHEN, CHRISTOPHER 2016 Particle stresses in dilute, polydisperse, two-way coupled turbulent flows. *Physical Review E* **93**, 013111.
- RICHTER, DAVID H. & SULLIVAN, PETER P. 2013a Momentum transfer in a turbulent, particleladen Couette flow. *Phys. Fluids* **25**, 053304.
- RICHTER, DAVID H. & SULLIVAN, PETER P. 2013b Sea surface drag and the role of spray. *Geophys. Res. Lett.* **40** (3), 656–660.
- RICHTER, DAVID H. & SULLIVAN, PETER P. 2014 Modification of near-wall coherent structures by inertial particles. *Physics of Fluids* **26**, 103304.
- ROSENFELD, D, WOODLEY, W L, KHAIN, A, COTTON, W R, CARRIO, G, GINIS, I & GOLDEN, J H 2012 Aerosol effects on microstructure and intensity of tropical cyclones. Bull. Amer. Meteor. Soc 93, 987–1001.
- ROUAULT, M P, MESTAYER, P G & SCHIESTEL, R 1991 A model of evaporating spray droplet dispersion. J. Geophys. Res. 96, 7181–7200.
- SHPUND, J, PINSKY, M & KHAIN, A 2011 Microphysical structure of the marine boundary layer under strong wind and spray formation as seen from simulations using a 2d explicit microphysical model. part i: The impact of large eddies. J. Atmos. Sci. 68 (10), 2366–2384.
- SHPUND, J, ZHANG, J A, PINSKY, M & KHAIN, A 2012 Microphysical structure of the marine boundary layer under strong wind and spray formation as seen from simulations using a 2d explicit microphysical model. part ii: The role of sea spray. J. Atmos. Sci. 69 (12), 3501–3514.

- SLINN, S A & SLINN, W G N 1980 Prediction for particle deposition on natural waters. Atmos. Env. 14 (9), 1013–1016.
- SMITH, M H, PARK, P M & CONSTERDINE, I E 1993 Marine aerosol concentration and estimated fluxes over the sea. Quart. J. Roy. Meteor. Soc. 119, 809–824.
- SPIEL, D E 1995 On the births of jet drops from bubbles bursting on water surfaces. J. Geophys. Res. 100 (C3), 4995–5006.
- SPIEL, DONALD E. 1997 More on the births of jet drops from bubbles bursting on seawater surfaces. J. Geophys. Res. 102 (C3), 5815–5821.
- SPIEL, D E 1998 On the births of film drops from bubbles bursting on seawater surfaces. J. Geophys. Res. 103 (C11), 24907–24918.
- TERRAY, E A, DONELAN, M A, AGRAWAL, Y C, DRENNAN, W M, KAHAMA, K K, WILLIAMS III, A J, HWANG, P A & KITAIGORODSKII, S A 1996 Estimates of kinetic energy dissipation under breaking waves. J. Phys. Oceanogr. 26, 792–807.
- THORPE, S A 1993 Energy loss by breaking waves. J. Phys. Oceanogr. 23, 2498–2502.
- TROITSKAYA, YU, KANDAUROV, A, ERMAKOVA, O, KOZLOV, D, SERGEEV, D & ZILITINKEVICH, SERGEI 2018 The "bag breakup" spume droplet generation mechanism at high winds. part i: Spray generation function. *Journal of Physical Oceanography* **48** (9), 2167–2188.
- VAN EIJK, A M J, TRANCHANT, B S & MESTAYER, P G 2001 Seacluse: Numerical simulation of evaporating sea spray droplets. J. Geophys. Res. 106, 2573–2588.
- VERON, F 2015 Ocean spray. Annu. Rev. Fluid Mech. 47, 507-538.
- VERON, F, HOPKINS, C, HARRISON, E & MUELLER, J 2012 Sea spray spume droplet production in high wind speeds. *Geophys. Res. Lett.* **39** (16).
- VERON, F & MELVILLE, W K 2001 Experiments on the stability and transition of wind-driven water surfaces. J. Fluid Mech. 446, 25–65.
- WALLS, PETER LL, HENAUX, LOUIS & BIRD, JAMES C 2015 Jet drops from bursting bubbles: How gravity and viscosity couple to inhibit droplet production. *Physical Review E* **92** (2), 021002.
- WANG, XIAOFEI, DEANE, GRANT B, MOORE, KATHRYN A, RYDER, OLIVIA S, STOKES, M DALE, BEALL, CHARLOTTE M, COLLINS, DOUGLAS B, SANTANDER, MITCHELL V, BURROWS, SUSANNAH M, SULTANA, CAMILLE M & OTHERS 2017 The role of jet and film drops in controlling the mixing state of submicron sea spray aerosol particles. Proceedings of the National Academy of Sciences 114 (27), 6978–6983.
- WU, J 2002 Jet drops produced by bubbles bursting at the surface of seawater. J. Phys. Oceanogr. 32, 3286–3290.
- ZHANG, J A, BLACK, P G, FRENCH, J R & DRENNAN, W M 2008 First direct measurements of enthalpy flux in the hurricane boundary layer: The cblast results. *Geophys. Res. Lett.* 35 (14).