Evaporation of Nonequilibrium Raindrops as a Fog Formation Mechanism

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ABSTRACT

To gain insights into the poorly understood phenomenon of precipitation fog, this study assesses the evaporation of freely falling drops departing from equilibrium as a possible contributing factor to fog formation in rainy conditions. The study is based on simulations performed with a microphysical column model describing the evolution of the temperature and mass of evaporating raindrops within a Lagrangian reference frame. Equilibrium defines a state where the latent heat loss of an evaporating drop is balanced by the sensible heat flux from the ambient air, hence defining a steady-state drop temperature. Model results show that the assumption of equilibrium leads to small but significant errors in calculated precipitation evaporation rates for drops falling in continuously varying ambient near-saturated or saturated conditions. Departure from equilibrium depends on the magnitude of the vertical gradients of the ambient temperature and moisture as well as the drop-size-dependent terminal velocity. Contrasting patterns of behavior occur depending on the stratification of the atmosphere. Raindrops falling in inversion layers remain warmer than the equilibrium temperature and lead to enhanced moistening, with supersaturation achieved when evaporation proceeds in saturated inversions. Dehydration occurs in layers with temperature and water vapor increasing with height due to the vapor flux from the environment to the colder drops. These contrasts are not represented when equilibrium is assumed. The role of nonequilibrium raindrop evaporation in fog occurrences is further emphasized with simulations of a case study characterized by fog forming under light rain falling in a developing frontal inversion. Good agreement is obtained between fog water content observations and simulations representing only the effects of rainfall evaporation. This study demonstrates the need to take into account the nonequilibrium state of falling raindrops for a proper representation of an important mechanism contributing to precipitation fog occurrences.

1. Introduction

It has long been recognized that low cloud ceilings and fog are integral parts of precipitating large-scale low pressure systems (George 1940a,b,c; Dolezel 1944; Goldman 1951; Byers 1959; Petterssen, 1969; Westcott 2004; Croft and Burton 2006). However, relatively few studies have been dedicated to the understanding of such cloud systems in perturbed weather conditions. With respect to fog, examples of its common occurrence in precipitation (referred to as precipitation fog) have been described by Tardif and Rasmussen (2007) for the New York City region. Although some instances of precipitation fog have been reported in regions of extratropical cyclones characterized by transitions in precipitation type (Stewart 1992; Stewart and Yiu 1993; Stewart et al. 1995), it has been more widely described as occurring in liquid precipitation associated with warm fronts (George 1940a,b,c; Byers 1959; Petterssen 1969). Goldman (1951) discusses such scenarios by citing factors influencing the evolution of low cloud ceilings as the evaporation of rainfall, advection of warm humid air, and turbulent mixing. Empirical rules were formulated to forecast ceiling evolution based on these principles and
results were found to correlate somewhat with the observations. The roles of various physical mechanisms were however not investigated in detail. Tardif and Rasmussen (2008) performed a detailed characterization of precipitation fog events in the New York City region and found that foggy conditions are often associated with cloud bases lowering to the surface as light liquid precipitation falls into sharp low-level temperature inversions. The available evidence, therefore, suggests that fog formation in warm precipitation involves processes taking place within temperature inversions, such as the turbulent mixing of air parcels with contrasting temperatures and/or raindrops falling from warm layers aloft and evaporating into the colder air near the surface. This study focuses on the latter mechanism.

The common explanation for the formation of fog by the evaporation of precipitation has been presented by Petterssen (1969). For raindrops of temperature $T_r$ warmer than the temperature of ambient air $T$, evaporation vanishes when $e = e_{ws}(T_r) > e_{ws}(T)$ (see Table 1 for the list of symbols and their definitions). This simplistic model is illustrated in Fig. 1, in which the evolving environmental conditions follow the path defined by $A \rightarrow B \rightarrow C$ as the evaporation of precipitating hydrometeors moistens the environment. Since saturation is reached at $B$, the final point $C$ (vapor pressure equal to the saturation vapor pressure at the surface of the precipitation particles) defines a state characterized by supersaturation and thus possible activation of cloud condensation nuclei (CCN) and fog formation. This simple explanation however neglects the evolution of the temperature of the environment and of the precipitation particles relative to the sensible and latent heat exchanges. Furthermore, the notion of drops remaining warmer than the ambient air over a considerable length of time during their freefall is in clear contradiction with the widely accepted hypothesis of equilibrium. Equilibrium defines a state where the latent heat loss of an evaporating drop is balanced by the sensible heat flux from the ambient air, hence defining

![Diagram illustrating the creation of supersaturation by the evaporation of warm rain into a colder environment as discussed by Petterssen (1969) and Jiusto (1981). The solid line represents the saturation curve. Raindrops of temperature $T_r$ initially fall into a subsaturated atmosphere at temperature $T$ (state described by point A). The atmosphere is moistened by the evaporation of the raindrops until saturation is reached (path from point A to point B). Because $T_r > T$, raindrops continue evaporating until the humidity in the atmosphere is increased to a value satisfying $e(T) = e_{ws}(T_r)$ (path from point B to point C). The final state of the atmosphere (point C) is characterized by $e(T) > e_{ws}(T)$ (supersaturation).]
a steady-state drop temperature. As will be shown in the next section, this steady-state “equilibrium temperature” is equal to the ambient air temperature in saturated conditions so drops no longer evaporate. If in fact equilibrium would be the rule rather than the exception, this would rule out precipitation evaporation as a mechanism directly responsible for the creation of supersaturation and fog formation. As will be shown herein, a mechanism contributing to fog formation during precipitation is not represented when the equilibrium assumption is used in the calculation of raindrop evaporation.

Few studies have addressed this problem. Dolezel (1944) attempted a description of the evolution of the ambient relative humidity and fog formation associated with evaporating raindrops falling into subcloud layers of various lapse rates. However, Dolezel’s model was based on a simplistic formulation of the calculation of the falling temperature and the resulting supersaturation. Tests performed with the model used in this study reveal that Dolezel’s approach leads to appreciable errors in the calculated temperature of the falling drops and the resulting supersaturation. Best (1952) also studied differences between the ambient and drop temperatures in near-saturated conditions but restricted the discussion to cases of drops falling in warmer layers, while Caplan (1966) had similar goals but used a model based on simplifying assumptions affecting the accuracy of the results. More recently, Donaldson and Stewart (1993) performed numerical experiments by considering the role of melting–freezing precipitation particles. Such particles remain at a constant temperature of 0°C and thus can remain noticeably warmer or colder than the environment. This was found to be an enhancing factor in the generation of supersaturation. However, important supersaturation was not produced in their simulations when liquid hydrometeors were considered. Such results, combined with clear evidence that precipitation fog events are most often associated with liquid precipitation rather than with melting or freezing particles in some regions (Tardif and Rasmussen 2008), provide motivation for a study focusing on microphysical processes associated with fog formation in rainy conditions.

A one-dimensional microphysical model solving for drop temperature and mass, coupled with the evolution of the ambient temperature and moisture conditions, is developed to investigate the drops’ departure from equilibrium and its impacts on the evolution of ambient relative humidity conditions. The evaporation of raindrops warmer than the ambient air of the temperature inversions and its role in the creation of supersaturation and fog formation is emphasized.

This paper is organized as follows. Key theoretical aspects of raindrop evaporation are reviewed in section 2, along with a definition of their equilibrium state. The numerical model used in this study is described in section 3. The departure from equilibrium of raindrops falling in layers of contrasting vertical gradients is quantitatively assessed in section 4 using numerical model simulations, while section 5 discusses the creation of supersaturation by the evaporation of nonequilibrium raindrops falling in inversions. Section 6 describes an observed precipitation fog event involving liquid precipitation and a developing low-level inversion. Simulation results are presented that illustrate the role of nonequilibrium raindrop evaporation in fog formation for this event. A summary and conclusions are outlined in section 7.

2. Evaporating raindrops and their equilibrium temperature

A comprehensive description of the state of an evaporating raindrop involves several processes such as sensible heat fluxes in the gas and liquid media and water vapor fluxes in the surrounding air (Sedunov 1974; Pruppacher and Klett 1997). Simplifications are, however, applied to render the problem tractable. For instance, Chang and Davis (1974) found that evaporation is only weakly controlled by heat conduction inside the drop. Evaporation can therefore be described with sufficient accuracy by assuming a drop having a uniform temperature equal to that of its surface (Watts and Farhi 1975). Raindrops may be considered as perfect spheres, neglecting the shape distortion due to surface tension and drag force. Straka and Gilmore (2006) showed that the effects of drop distortion on evaporation generally remain small, particularly in light rain conditions, which are of special interest in this study.

The heat balance of an evaporating sphere of pure water within a vapor field in the continuum regime (appropriate for precipitation-size drops) is expressed as (Rogers and Yau 1989)

$$\rho_c c_w \frac{4}{3} \pi r^3 \frac{dT}{dt} = L \frac{dm}{dt} + \frac{dh}{dt}.$$  \hspace{1cm} (1)

The sum of the latent heat ($L \frac{dm}{dt}$) and sensible heat ($\frac{dh}{dt}$) flux terms on the right-hand side is the net energy flux to or from the drop. Heat fluxes to or from a spherical drop at rest exposed to fresh ambient conditions are well described by analytical solutions of the diffusion equation. Mass and heat fluxes between a drop and its environment are described by transient analytical solutions with associated adjustment time scales toward steady state (see Pruppacher and Klett 1997). These transient solutions, however, do not accurately describe the state of freely falling (ventilated) drops, where ambient
air is constantly renewed on their forward faces and modified air is rejected from their back sides. To describe this situation, Kinzer and Gunn (1951) and Abraham (1968) developed models of mass and heat transfer involving transient interactions between packets of fresh environmental air with the spherical surface of drops. Expressions of mass and heat fluxes for ventilated drops involve steady-state water vapor density and temperature gradients enhanced by the so-called ventilation coefficients $F_v$ and $F_h$ for vapor and heat, respectively:

$$\frac{dm}{dt} = -F_v 4\pi r \rho_{v,\text{sat}} (T_r - \rho_v)$$ \hspace{1cm} (2a)

$$\frac{dh}{dt} = -F_h 4\pi \kappa a (T_r - T)$$ \hspace{1cm} (2b)

The rapid adjustment to a steady equilibrium temperature for drops exposed to a subsaturated isothermal environment is a well-known concept. A drop reaches equilibrium when the latent heat flux is balanced by the sensible heat flux so that the net energy flux to the hydrometeor vanishes and its temperature no longer evolves over time [see Eq. (1)]. An expression for the equilibrium temperature ($T_e$) is derived using Eqs. (2a) and (2b), taking $L \frac{dm}{dt} = -\frac{dh}{dt}$, assuming $F_v = F_h$, setting $T_r = T_c$, and solving for $T_e$:

$$T_e = T - \frac{LD_v}{\kappa_a} [\rho_{v,\text{sat}} (T_c) - \rho_v].$$ \hspace{1cm} (3)

This equation can be solved numerically using an iterative method. It is clearly seen that the equilibrium temperature is colder than the environment in a sub-saturated atmosphere and becomes $T_e = T$ at saturation. Note that $T_e$ is not the wet-bulb temperature in spite of the similarity in their expressions.

Kinzer and Gunn (1951) derived the following expression for the relaxation time scale to $T_e$ by assuming equality of the ventilation coefficients for heat and vapor ($F_v = F_h = \bar{f}$):

$$\tau = \frac{\rho_w c_w r^2}{3\bar{f} \left[ \kappa_a + LD_v \left( \frac{d\rho_v}{dT} \right)_\text{sat} \right]}.$$ \hspace{1cm} (4)

This relaxation time typically ranges from a fraction of a second for cloud and small drizzle drops to a few seconds for the larger drizzle and raindrops.

Studies focusing on evaporating raindrops (Abraham 1962; Syono and Takeda 1962; Beard and Pruppacher 1971; Pruppacher and Rasmussen 1979; Srivastava and Coen 1992) or involving rainfall evaporation (Srivastava 1985; Clough and Franks 1991; Brown 1993; Hu and Srivastava 1995, among others) generally consider some estimate of the equilibrium temperature for the precipitating hydrometeors, such as the wet-bulb temperature or other more detailed approximations. This equilibrium assumption is commonly justified by the fact that the drop temperature has a short relaxation time toward its equilibrium state as determined by Eq. (4) and verified by laboratory experiments (Kinzer and Gunn 1951; Iwai and Kato 1998). The validity of the equilibrium assumption may be challenged however when drops are falling along continuously varying ambient temperature and moisture conditions as will be described herein. Changes in the ambient conditions surrounding raindrops on time scales shorter than their relaxation time scale prevent drops from reaching a steady equilibrium state. A numerical model has been designed to quantitatively investigate this phenomenon.

3. Numerical model

Similar to Best (1952) and Caplan (1966), a model simulating the evolution of the temperature and mass of drops as they fall through vertical temperature and humidity gradients has been developed. A more complete formulation is used compared to prior studies, by relaxing the quasi-steady assumption of Caplan (1966), and using more sophisticated formulations of the ventilation coefficients. A complete time dependency of the mass and temperature of the raindrops is considered, and the evolution of the ambient temperature and moisture in response to the drops evaporation is fully taken into account to examine time-dependent conditions.

The model is based on Eqs. (1) and (2), describing the evolutionary patterns of drop temperature and mass within a Lagrangian reference frame as drops fall through strata of the atmosphere with varying temperatures, moisture levels, and pressures. The ventilation coefficients are expressed following Beard and Pruppacher (1971) and Pruppacher and Rasmussen (1979):

$$\bar{f} = 1.0 + 0.108 (N_{Sc,v}^{1/3} N_{Re}^{1/2})^2,$$

for $N_{Sc,v}^{1/3} N_{Re}^{1/2} < 1.4$, and

$$\bar{f} = 0.78 + 0.308 (N_{Sc,v}^{1/3} N_{Re}^{1/2}),$$

for $1.4 \leq N_{Sc,v}^{1/3} N_{Re}^{1/2} \leq 51.4$. \hspace{1cm} (5a, 5b)

The Schmidt and Reynolds numbers are expressed as

$$N_{Sc,v} = \frac{\mu_a}{\rho D_v}$$ \hspace{1cm} (6)
\[ N_{Re} = 2 \rho \frac{V_t}{\mu}, \] (7)

with the dynamic viscosity of air taken as a function of air temperature (Rogers and Yau 1989):

\[ \mu_a = 1.72 \times 10^{-5} \left( \frac{393.0}{T + 120.0} \right) \times \left( \frac{T}{273.15} \right) \text{ (units in kg m}^{-1}\text{s}^{-1}). \] (8)

The diffusivity of water vapor in air in the vicinity of drops is formulated as in Hall and Pruppacher (1976),

\[ D_v = 0.211 \times 10^{-4} \left( \frac{1013.25}{p} \right) \times \left( \frac{T}{273.15} \right)^{1.94} \text{ (units in m}^2\text{s}^{-1}), \] (9)

while the thermal conductivity is taken as (Beard and Pruppacher 1971)

\[ \kappa_a = 4.1868 \times 10^{-3} (5.69 + 0.017T) \] (units in J m\(^{-1}\) s\(^{-1}\) K\(^{-1}\)). \] (10)

Precipitation particles are introduced at the top of the model and fall at their terminal velocity calculated using the exponential relationship of Ferrier (1994):

\[ V_{t,00} = 4854d e^{-195d} \text{ (units in m} \text{s}^{-1}), \] (11)

where \( d \) is the diameter of the drops (in meters). The terminal velocity aloft (at pressure \( p \) and temperature \( T \)) is obtained by applying a correction for the variation of air density with altitude as in Dotzek and Beheng (2001), without taking into account the effects of the updraft vertical velocity:

\[ V_i(p, T) = V_{t,00} \left( \frac{\rho_{00}}{\rho} \right)^{\alpha}, \] (12)

where \( \rho_{00} \) is the reference air density at the ground and \( \alpha \) is set to 0.4 (Foote and du Toit 1969).

The model may be run using a monodisperse drop size distribution to provide a clearer identification of the results' dependence on drop size, while a bin version incorporates a more realistic representation of the combined effects of raindrops distributed following a prescribed size distribution. The equations described above are applied for every bin “i,” with packets of \( n_i \) drops of size \( r_i \) launched periodically from the top of the model. Bins of uniform width of 0.5 mm spanning drop sizes up to 5 mm in diameter are used. The frequency with which drops are introduced at the top of the model and the various \( n_i \) are prescribed following the Marshall–Palmer exponential size distribution (Marshall and Palmer 1948) (Fig. 2) corresponding to a chosen rain rate. The introduction of drops of individual sizes is performed sequentially, starting with the larger drops to take into account their larger terminal velocities. A very fine vertical grid spacing of 0.5 m is used. The accuracy of the model at representing the transient evolution of the drop temperature has been validated by Tardif (2007) by reproducing experimental laboratory conditions and results reported by Kinzer and Gunn (1951) and Iwai and Kato (1998).

Changes to the ambient temperature \(dT\) and water vapor content \(dp_v\) at a given model level due to drops in bin \( i \) are calculated through the combined influence of the sensible and latent heat fluxes from the \( n_i \) drops of size \( r_i \) as they fall though the model layer while exposed to the local ambient conditions over a time interval \( dt_i \) equal to \( dz/V_{t,i} \), where \( dz \) is the grid spacing and \( V_{t,i} \) is the corresponding terminal velocity of the drops. The total changes in ambient conditions may then be expressed as

\[ dT = \frac{1}{\rho c_p} \sum_i n_i \frac{T}{4 \pi r_i^2} \int_k \kappa_a (T_{v,i} - T) \, dt_i \] \hspace{1cm} (13a)

\[ dp_v = \sum_i n_i \frac{T}{4 \pi r_i^2} \int_k \left( \rho_{v,sat}(T_{v,i}) - \rho_v \right) \, dt_i. \] \hspace{1cm} (13b)

The coupled system of equations is solved using a fully implicit Crank–Nicholson numerical scheme (Richtmyer and Morton 1967), with a time step corresponding to the
The design of the model includes simplifying assumptions. The interactions among drops (e.g., collision–coalescence) are neglected. Individual drops at the same vertical level are assumed to be exposed to identical ambient conditions at any given time. The separation between drops is assumed to be large enough to neglect direct drop-to-drop interactions. Another simplification consists of neglecting the curvature and solute effects in the calculation of saturation vapor pressure at the surface of the drops. Precipitation-sized drops are large enough to neglect the curvature effect (Pruppacher and Klett 1997) and are assumed to be composed of pure water. The saturation vapor pressure at the surface of the drops is calculated as a function of temperature using the relation proposed by Buck (1981). The contribution from radiative heat loss to the drop energy balance (Roach 1976) is neglected. This contribution should be taken into account for droplets located near cloud top underneath a clear atmosphere where the divergence of the radiative fluxes is important (Rasmussen et al. 2002). However, radiative cooling is negligible in the cloud interior or in the subcloud layer underneath overcast cloud conditions typically accompanying stratiform precipitation. It is assumed that raindrops fall in a horizontally homogeneous atmosphere at rest, where possible circulations induced by the evaporation of precipitation are neglected.

4. Departure from equilibrium and approach to saturation

Idealized numerical experiments are first presented, assessing the departure from equilibrium for raindrops of various sizes falling in contrasting atmospheric layers. The top of the model (base of the precipitating cloud) is set at an altitude of 1 km, where drops are assumed at equilibrium under saturated ambient conditions at a temperature of 15°C. Drops encounter various ambient temperature and water vapor density profiles during their downward trajectory. Various temperature lapse rates are specified in a subcloud layer with a constant relative humidity (RH) profile of 95%, or with a constant water vapor density profile. Other profiles are characterized by relative humidity gradients in an isothermal layer and in a dry-adiabatic layer. The atmospheric pressure profile is obtained by assuming a hydrostatic atmosphere and a pressure at the surface of 1013.25 hPa. The difference between the drop and the equilibrium temperature \( T \) as the drops reach the ground is shown in Fig. 3. Results show departures up to a few tenths of a degree depending on the strength of the gradients and the drop sizes. Drops falling into warmer layers remain colder than the equilibrium temperature (Figs. 3a, 3b, and 3d) while drops falling into colder and/or dryer layers remain warmer than the equilibrium temperature (Figs. 3a and 3c). The largest departure from equilibrium is obtained for the larger drops falling in the strongest inversions.

The impacts of nonequilibrium drops on the evolution of ambient conditions toward saturation are investigated next. To illustrate the concept more clearly, the model is integrated over time by only considering raindrops of 1-mm diameter with a number concentration corresponding to rainfall rate of 2.5 mm h\(^{-1}\) (concentration equal to 271 m\(^{-3}\) with a bin width of 1 mm). Various initial conditions are defined with temperature profiles ranging from isothermal (ISO) to dry-adiabatic (DryAD) conditions, and an inversion with a \(+10\) K km\(^{-1}\) lapse rate (INV). An initial constant relative humidity profile of 95% is used. For comparison, other simulations were performed where an equilibrium temperature is imposed on the drops. The temporal evolution of the near-surface supersaturation \( S \) is shown in Fig. 4. The reader is reminded that, since a single drop size is considered here, ambient conditions are influenced by only a fraction of the number of drops that would normally be observed for the chosen rain rate, hence the long time scale characterizing the evolution of the ambient conditions. Both the equilibrium and nonequilibrium simulations approach saturation at approximately the same rate. Small differences are nevertheless noticed when examining the evolving conditions near saturation (inset in Fig. 4). Assuming a drop temperature at equilibrium leads to an underestimation of 0.07% in \( S \) when drops fall through a temperature inversion and an overestimation when they fall through a dry-adiabatic layer. No differences are found for isothermal and constant RH conditions as equilibrium is reached and maintained by drops falling in vertically constant ambient conditions.

Contrasting behavior in the evolution of the environmental temperature and humidity are shown for drops falling in nonisothermal conditions (Fig. 5). Atmospheric cooling is observed in the DryAD layer from the sensible heat exchange with the cold drops, but is accompanied by a depletion of the ambient water vapor beginning halfway into the simulation. The dehydration occurs when the water vapor density difference between the surface of the colder drops and the air becomes negative \( [\rho_{\text{sat}}(T) - \rho_{w}] < 0 \), inducing a flux of vapor from the environment to the colder drops. This drying partially offsets the cooling and contributes to a slower increase in \( S \). In contrast, enhanced moistening is taking place in the INV simulation with cooling taking place during the first half of the simulation, transitioning to warming during the second half. This transition to warming
occurs as evaporating drops become warmer than the air temperature.

When an equilibrium temperature is imposed on the raindrops, warming of the air is absent in the INV simulation and cooling is underestimated by 0.15°C at the end of the DryAD simulation (Fig. 5a). Also, the dehydration simulated in the DryAD layer is not represented if equilibrium is imposed, leading to an overestimation of 0.18 g kg⁻¹ in the specific humidity at the end of the simulation (Fig. 5b). In the INV case, an underestimation of 0.08 g kg⁻¹ is obtained as a result of assuming equilibrium. Similar to the ISO simulation (in which drops remain at equilibrium), DryAD and INV simulations performed with the imposed equilibrium are characterized by vanishing temperature and water vapor density contrasts between drops and their environment as saturation is approached.

5. Supersaturation from rainfall evaporation

The previous results show that nonequilibrium effects are small, but suggest that they become significant when a threshold phenomenon such as reaching and exceeding saturation is of interest, as in the case of fog. To further examine the importance of nonequilibrium effects and associated precipitation evaporation as a supersaturation-producing mechanism, results from a series of similar simulations are presented in which the initial conditions have been set at saturation.

a. Drops falling in contrasting stratification

Simulations were performed using three contrasting temperature profiles, corresponding to the pseudo-adiabatic (PseudoAD) and isothermal (ISO) lapse rates, as well as an inversion layer with a +10 K km⁻¹ lapse rate (INV). All other parameters are the same as in the previous simulations.

The evolution of the temperature contrasts between the surfaces of the raindrops and the ambient air and the resulting changes in the ambient near-surface temperature, specific humidity and corresponding supersaturation (S) with respect to their initial values, are shown in Figs. 6a–d, respectively. Results show that a saturated isothermal layer (drops at equilibrium) induces no evaporation or condensation, nor changes in ambient temperature, and thus maintains steady saturation. In contrast, dehydration of the atmosphere is associated with the condensation on colder-than-ambient drops.
falling through pseudo-adiabatic conditions, leading to a decreasing supersaturation with time. This occurs in spite of the cooling from the sensible heat exchange with the cold drops. Warmer-than-ambient drops falling through a temperature inversion continuously evaporate causing the temperature and specific humidity to increase with time. The trajectory describing the evolution of the ambient water vapor and temperature conditions with respect to saturation is shown in the \((T, e)\) plane in Fig. 7. As suggested earlier, Petterssen’s (1969) simple conceptual model (Fig. 1) does not adequately describe the effects of rainfall into inversions by neglecting the evolution of ambient temperature. Rather, supersaturation is achieved by a rate of water vapor increase that is slightly greater than the effect of increasing temperature (the slope of the trajectory is greater than the slope of the saturation curve).

As shown earlier, drops in equilibrium, such as those falling in isothermal layers, do not exert any forcing on the ambient temperature and moisture (and therefore supersaturation) when conditions are at saturation. On the other hand, drops falling into temperature and humidity gradients under saturated conditions lead to either a reduction or increase in relative humidity due to the slower thermal relaxation time of the drops compared to changes in the ambient conditions. Therefore, the evaporation of rainfall is not a direct fog-forcing mechanism in layers where the temperature is decreasing with height, in that it does not generate supersaturated conditions responsible for the activation of cloud condensation nuclei (CCN). CCN activation in such layers must be related to other processes such as advection, turbulent mixing, or adiabatic cooling of the moistened air. In contrast, the addition of water vapor from the evaporation of warm raindrops in inversions leads to supersaturated conditions and therefore can be considered as a direct fog-inducing mechanism. This shows the importance of considering nonequilibrium effects when interested in processes near saturation or when transitions from subsaturated to supersaturated conditions are of importance, as is the case for fog and low cloud ceilings forming in precipitation.

b. Rainfall through inversions: Sensitivity experiments

The results presented up to this point provide evidence that evaporation of nonequilibrium raindrops falling through temperature inversions can lead to supersaturated conditions. In this section, the sensitivity of supersaturation to the size of the raindrops, the character of the inversions (magnitude of the temperature lapse rate), and the rainfall intensity is examined. The metric used to assess the potential for fog formation by rainfall evaporation is the average supersaturation

![Figure 4: Temporal evolution of supersaturation near the ground surface from simulations performed with initial conditions at RH = 95% and with three different lapse rates (dry adiabatic, isothermal, and an inversion with a lapse rate of \(+10^\circ\text{C km}^{-1}\)). Results from the full simulations are shown by the black lines, while simulations performed by imposing equilibrium are shown as gray lines.](image-url)

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tendency at the lowest model level during the first hour of simulation.

1) DROP SIZE AND INVERSION STRENGTH

Figure 8a shows the rate of increase of supersaturation resulting from the evaporation of raindrops with diameters ranging from 0.5 to 3.5 mm falling through inversions with lapse rates ranging from 0.1 to 15 K km$^{-1}$. A drop size distribution (DSD) corresponding to a rain rate of 2.5 mm h$^{-1}$ (light rain) is imposed at the top of the model using the corresponding Marshall–Palmer (MP) relationship (see Fig. 2). Although deviations from the MP distribution have been reported in the literature, the MP distribution represents a reasonable approximation to DSDs in frontal precipitation, particularly for the millimeter-size drops. The magnitude of the supersaturation tendency increases with the strength of the inversion. Given the specified DSD, this trend with respect to size is maximized for 1.5-mm-diameter drops, with an absolute maximum rate of increase of 0.057% h$^{-1}$ achieved for a +15 K km$^{-1}$ inversion. This is compared to 0.033% and 0.014% h$^{-1}$ when the lapse rates are +10 and +5 K km$^{-1}$, respectively. These rates are reduced by 50% when 2.25-mm drops are considered. Even though large drops maintain larger departures from equilibrium and an associated greater potential for evaporation, a combination of an exponential decrease in number concentration and larger terminal velocity giving larger drops less time to interact with the environment leads to a lesser contribution to the increase of $S$. The production of supersaturation is thus sensitive to the convolution between the effects of drop size and drop concentration, leading to a maximum contribution from millimeter-sized drops in light rainfall conditions.

2) PRECIPITATION RATE AND DROP SIZE DISTRIBUTION

To assess the sensitivity of supersaturation production to precipitation rate, a similar series of simulations was performed with the number concentration of raindrops drawn from an MP distribution corresponding to a rain rate of 0.3 mm h$^{-1}$ (Fig. 8b). The highest rate of increase in $S$ is 0.005% h$^{-1}$ and is obtained for drops of 1-mm diameter falling through the strongest inversion considered.

An additional series of simulations was performed with conditions corresponding to a rain rate of 0.3 mm h$^{-1}$ as before, but this time with the concentration of raindrops drawn from the Joss-drizzle DSD (Mätzler 2002). This distribution is characterized by a greater concentration of small drops (diameter $< 0.75$ mm) and smaller concentration in the larger drop range compared to the MP distribution (Fig. 2). The results show a general decrease in the production rate of supersaturation of 30%–36% with the Joss-drizzle distribution for the various inversion strengths (Fig. 9). The size of the drops producing the largest tendencies is shifted to 0.5 from 1.0 mm compared to the MP distribution. The highest rate of increase is 0.003% h$^{-1}$ for drops of 0.5-mm diameter falling in the 15 K km$^{-1}$ inversion. This decrease is the result of the drop concentration being smaller by 50% for the 1-mm drops, and smaller by at least an order of magnitude for drops 1.75 mm in diameter and larger.

These idealized sensitivity experiments indicate that the attainable supersaturation from rainfall evaporation into inversions is highly sensitive to the characteristics of the inversions themselves and the microphysical structure of the rainfall. This sensitivity of supersaturation to the shape of the DSD suggests the indirect role of microphysical processes other than evaporation, such as coalescence and breakup. An investigation of the role of
these processes in shaping the DSD and their associated impacts on supersaturation is outside the scope of this work, but results obtained suggest that it should be the subject of further study.

6. An observed precipitation fog event

To further confirm the role of nonequilibrium raindrop evaporation as a mechanism contributing to fog formation, an episode characterized by low cloud ceilings and fog formation observed during precipitation is investigated using observations and a model simulation. The event occurred in the New York, New York, area on 6 and 7 February 2004 as a midlatitude cyclone propagated over the northeastern United States and eastern Canada (Fig. 10). Data from various instruments (listed in Table 2) deployed at the Brookhaven National Laboratory (BNL) (see Fig. 10 for location) and routine radio soundings from the nearby Upton National Weather Service station (OKX) are used for the investigation.

Figure 11 shows the temporal evolution of the cloud ceiling height, near-surface horizontal visibility, and precipitation rate (Figs. 11a–c, respectively) observed on 6 and 7 February. A low cloud ceiling was first detected an hour after precipitation onset and roughly 12 h prior to fog. An average near-surface horizontal visibility of 2 km was observed under persisting low ceilings, even during the period of most intense rainfall.

Fig. 6. Temporal evolution of the (a) difference between the temperature of raindrops ($T_r$) and the air temperature ($T_a$), (b) normalized temporal evolution (difference with value at the initial time) of air temperature, (c) specific humidity, and (d) of ambient supersaturation at the lowest model level in simulations performed with a pseudo-adiabatic lapse rate, isothermal profile, and an inversion characterized by temperature increasing at a rate of 10 K km$^{-1}$.

Fig. 7. Evolution of the near-surface ambient water vapor pressure against the evolving temperature (solid line with arrow) in response to evaporating raindrops of 1 mm falling in an inversion with a lapse rate of +10 K km$^{-1}$. Conditions corresponding to saturation are shown by the dashed line.
Visibility experienced a downward trend to reach values below the 1-km fog threshold around 2200 UTC as rainfall intensity decreased below 2 mm h\(^{-1}\). Foggy conditions persisted until 0815 UTC on 7 February, a period characterized by intermittent precipitation.

During the period leading to fog formation and early evolution (from 2000 UTC on 6 February to 0400 UTC 7 February), the mean sea level pressure pattern induced a light southerly onshore flow throughout the region’s coastal areas (Fig. 10), with relatively warm air flowing over the cold coastal water. This scenario might suggest initial fog formation at sea, with the fog subsequently advected onshore. However, observations from buoys offshore did not indicate saturation until fog formed inland. Fog also formed as precipitation was observed at stations located farther inland (e.g., in central New Jersey) under calm wind conditions. Therefore, evidence points to scenarios other than an advection fog event.

Radio soundings provide evidence of important changes in the vertical structure of the lower atmosphere during the period between low cloud and fog formation (Fig. 12). Considerable warming occurred below 750 hPa, with a rate maximized at about 950 hPa leading to a strong surface-based inversion (Fig. 12b). The central role of differential temperature advection in the creation of this inversion is confirmed by the high temporal resolution observations on a 90-m tower, showing an increase of 4°C between 2000 and 2100 UTC at 85 m compared to an increase of 1°C at the surface (Fig. 13). Fog formation occurred during the latter stages of the development of this inversion as warming rates decreased. The vapor content of the lower atmosphere did not keep up with the warming in the upper part of the inversion, resulting in a decrease in dewpoint depression above 150 m (Fig. 14). Moisture advection and rainfall evaporation therefore did not provide the necessary water vapor to maintain near-saturated conditions aloft during this period, except in the lowest and strongest parts of the inversion. This suggests that a sensitive balance between the precipitation evaporation and differential advection of the temperature and moisture has to be in place in order for fog to form ahead of the warm front. The low-level inversion persisted until about 0400 UTC, at which time rapid cooling was observed with a shift to a northerly flow bringing cold-air advection. The latter stages of the
fog event were thus influenced by processes other than rainfall evaporation in an inversion and are therefore outside the scope of this study.

The role of nonequilibrium raindrop evaporation on the formation of fog is investigated using a simulation of the column precipitation evaporation model described in section 3, where the liquid water mixing ratio (LWMR) is calculated from supersaturation through an iterative bulk condensation scheme as in Bergot and Guédalia (1994). This approach is somewhat simplistic due to the

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FIG. 10. Surface weather observations and corresponding sea level pressures (hPa) and frontal analyses from the National Oceanic and Atmospheric Administration’s (NOAA) Hydrometeorological Prediction Center at 0000 UTC 7 Feb 2004. The black dot near the center of the figure indicates the location of the instrumented site at the BNL.
underlying assumption that liquid fog is at saturation. However, uncertainties in simulated LWMR inherent to this simplicity cannot be satisfactorily resolved without comprehensive information about the ambient aerosol particles, as is the case here. The gravitational settling flux of the condensed water is parameterized as in Brown and Roach (1976) with a gravitational settling velocity derived from fog monitor (Droplet Measurement Technologies model FM-100) data (Tardif 2007). The top of the model is set at 2300 m, corresponding to the base of Fig. 11. Observed evolution of (a) cloud base (30-s ceilometer returns, black rectangles) and its associated ceiling height (dots), (b) near-surface horizontal visibility (fog threshold marked by the light gray line), and (c) precipitation rate from the GEONOR Inc. rain gauge, on 6 and 7 Feb 2004. The hatched area in (c) indicates snow, light gray indicates a period of mixed precipitation, and the gray-shaded area indicates the presence of liquid precipitation. The upward- and downward-pointing arrows in (a) indicate sunrise and sunset, respectively.
the precipitating cloud determined from the 0000 UTC 7 February sounding (Fig. 14). Precipitation particles are introduced at the top of the model with their temperature set at equilibrium, with an evolving precipitation rate adjusted to match the precipitation accumulation observed at the surface. Rainfall rate–dependent Marshall–Palmer raindrop size distributions are assumed. The simulation extends out to 0400 UTC 7 February.

As discussed above, horizontal advection played an important role in the evolution of the temperature and humidity profiles. Time-dependent horizontal advections are estimated using the BNL tower and sounding observations, complemented with high temporal resolution retrievals from a profiling microwave radiometer (MWR) (Ware et al. 2003). A comparison between the MWR temperature retrievals and sounding profiles (Figs. 15a and 15b), along with the temporal evolution of temperature observed at the upper tower level (85 m) and lowest retrieval level (100 m) (Fig. 15c), indicate that the transition toward a stable lower atmosphere was well captured by radiometric retrievals. Advection estimates, used as lateral boundary conditions, are determined during

Fig. 12. Skew $T$–log $p$ plots of soundings from OKX at (a) 1200 UTC 6 Feb and (b) 0000 UTC 7 Feb 2004. The thick solid line shows the air temperature while the thinner black solid line shows the dewpoint temperature.

Fig. 13. Observed profiles of potential temperature from the BNL tower during the period of cloud lowering and fog formation on 6 Feb 2004.

Fig. 14. Observed profiles of air temperature (solid line) and dewpoint temperature (dashed line) over the lowest 4 km of the atmosphere from the OKX sounding at 0000 UTC 7 Feb 2004. Light dotted–dashed lines represent the reference dry-adiabatic profiles, while light dashed lines represent the pseudo-adiabatic profiles.
iterative integrations of the column rainfall model as described in the appendix. The simulation is initialized at 2000 UTC, shortly before the initiation of differential temperature advection, creating the inversion. The initial temperature and humidity profiles were determined from a combination of tower observations and MWR retrievals.

The time–height cross section of the air temperature derived from tower and soundings observations and MWR retrievals, and liquid water mixing ratio (LWMR) profiles resulting from supersaturation generated by the evaporation of nonequilibrium raindrops, are shown in Figs. 16a and 16b, respectively. Condensation first takes place aloft in the lower part of the layer with the strongest vertical temperature gradient. The maximum production of water subsequently propagates downward following the base of the inversion during its migration toward the surface (marked by the solid line in Fig. 16a). As the temperature becomes steady, the top of the fog layer remains slightly below the top of the surface-based inversion located slightly below 150 m. With the atmosphere aloft no longer warming after 2300 UTC, rainfall evaporation moistens the subsaturated layer and leads to the creation of supersaturated conditions and condensation shortly after 0100 UTC 7 February, as warm raindrops continue evaporating as they fall through the upper part of the inversion.

The comparison of LWMR estimates derived from FM-100 spectrometer measurements and simulated LWMR from the evaporation of nonequilibrium raindrops is shown in Fig. 17. The measured LWMR was characterized by values generally below 0.05 g kg$^{-1}$, with periods of enhanced LWMR. Upon closer inspection of the measurements, circumstantial evidence suggests the contamination of fog droplet measurements by the breakup of raindrops in the interior of the horn.
in the FM-100 and the subsequent intake of drop fragments into the instrument. Some periods of enhanced LWMR have been found to correspond to shifts in the wind direction leading to a flow directed into the inlet of the instrument as rain was falling. Fluctuations in LWMR could not be explained through physically based arguments, such as a correspondence with periods of enhanced turbulent mixing generating higher levels of supersaturation and enhanced droplet growth rates (Gerber 1991; Korolev and Isaac 2000). Correlation with fluctuations in other measured quantities (e.g., temperature) could not be established. Although circumstantial in nature, the evidence presented suggests that the highlighted data points in Fig. 17 should be considered suspicious. The appearance and early increase of LWMR are well represented by the model simulation, as well as LWMR values measured after 0000 UTC during the periods when the FM-100 measurements are not contaminated by splashing raindrops. Given the simplifying assumptions used to define the conditions of the simulation (e.g., steady temperature after 2300 UTC), it can be concluded that the simulated fog water forming as a result of precipitation evaporation into the inversion is in good agreement with the FM-100 measurements deemed reliable. Although turbulent mixing in the saturated inversion cannot be ruled out as a possible contributing factor without a more detailed study, these results provide compelling evidence of the role of nonequilibrium raindrop evaporation in the formation of fog within the lower part of a frontal inversion.

7. Summary and conclusions

The various scenarios and associated complex processes and interactions leading to fog contribute to the difficulty in accurately forecasting its formation. Progress in this area can be obtained through a better understanding of the physical processes contributing to fog formation. In this study, a common yet relatively overlooked fog type (precipitation fog) has been investigated. It has been postulated that the evaporation of raindrops remaining out of equilibrium could play a significant role in the formation of this fog type. The investigation provides a quantitative assessment of the departure from equilibrium for raindrops falling through a vertically varying ambient temperature and humidity conditions, based on simulations of a Lagrangian numerical model depicting the evolution of raindrop temperature and evaporation taking place in light rain conditions. Simulation results have been presented providing evidence that equilibrium is not reached for raindrops falling in atmospheric layers characterized by nonzero temperature and/or humidity vertical gradients. The sensitivity of the temporal evolution of the ambient relative humidity due to the nonequilibrium raindrops can be taken to be negligible when considering conditions away from saturation. However, in spite of the raindrops’ small departure from equilibrium (few hundredths to a few tenths of a degree), the nonequilibrium effect becomes significant as saturation is approached and reached. As suggested by the approximate solutions of Best (1952) and Caplan (1966), the more detailed simulation results presented herein confirm that, contrary to common belief, dehydration of the atmosphere may occur during rainfall through a flux of vapor from the environment to the drops as saturation is approached and reached. Another consequence of nonequilibrium is an enhanced moistening and warming of the low-level
air from drops becoming warmer than the environment when falling in an inversion layer. These features are not represented when equilibrium is assumed.

More importantly, simulations performed with initial conditions at saturation have shown that raindrops falling in saturated inversions remain warmer than the ambient air (out of equilibrium) and lead to supersaturation ($S$) as their evaporation provides an influx of water vapor to a saturated environment. The supersaturation production rate is a function of the rainfall rate and the magnitude of the vertical gradients. A marked dependence on the convolution between drop size and concentration has been illustrated. The increase in the potential for the generation of supersaturation for larger raindrops, combined with the typical exponential decay of the drop concentration with increasing sizes often encountered in natural rain conditions for a given rain rate, leads to a maximum contribution to the generation of $S$ from drops of 0.5–1.5-mm diameter. The concept has also been tested on a real case study characterized by fog formation during light liquid precipitation falling through a developing surface-based frontal inversion. Good agreement of the simulation results and observations was found.

Other processes likely contribute to fog formation as precipitation occurs. Results presented herein suggest that the direct influence of nonequilibrium raindrops is less likely in scenarios characterized by elevated inversions and/or the presence of drizzle, scenarios that are quite common, as shown by Tardif and Rasmussen (2008). Scenarios characterized by a moistened atmospheric boundary layer flowing over colder surfaces and/or the occurrences of upslope flow and/or the presence of shear-induced turbulent mixing are also favorable for the creation of supersaturation and fog. The variety and complexity of factors playing a role in the likelihood of fog formation in perturbed weather conditions is thus considerable. Further investigations should therefore focus on assessing the relative contribution of these processes. The present study has nevertheless clearly demonstrated the role of evaporating nonequilibrium raindrops in promoting supersaturation and hence fog formation in specific scenarios.

Finally, it should be noted that the various microphysical parameterization schemes commonly used in numerical models (Kessler 1969; Schlesinger et al. 1988; Feingold 1993; Ferrier 1994; Gregory 1995; Reisner et al. 1998; Cohard and Pinty 2000; Thompson et al. 2004; Milbrandt and Yau 2005; Posselt and Lohmann 2008) all neglect nonequilibrium effects as the various formulations lead to vanishing evaporation rates as saturation is approached, regardless of the vertical distribution of the ambient conditions encountered by the falling raindrops. Thus, fog and cloud formation due to nonequilibrium raindrop evaporation cannot be properly represented in simulations based on these parameterizations. A suitable parameterization scheme including the nonequilibrium effects on drop temperature should therefore be developed to enhance our capability to forecast a potentially hazardous phenomenon.

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APPENDIX

Lateral Boundary Conditions Used in the Column Precipitation Evaporation Model

Estimates of temperature and humidity tendencies, representing horizontal advections used as lateral boundary conditions in the column simulation of rainfall evaporation, are derived from a combination of measurements from radiosondes, an instrumented tower, and a profiling microwave radiometer (MWR). Hourly estimates of horizontal advection for a given height are derived from the observed or retrieved profiles calibrated against sounding data as

$$\text{Adv} X(\Delta t) = \left( \frac{\Delta X_{s}^{12\text{hrs}}}{\Delta X_{\text{Obs}}^{12\text{hrs}}} \right) \left( \frac{\Delta X_{\text{Obs}}^{\Delta t}}{\Delta t} \right) - \frac{\Delta X_{\text{precip}}^{\Delta t}}{\Delta t},$$ (A1)

where $\text{Adv} X(\Delta t)$ is the advection tendency of the variable $X$ (temperature or water vapor mixing ratio) over the time interval $\Delta t$, $\Delta X_{s}^{12\text{hrs}}$ is the temperature difference between the two soundings 12 h apart, $\Delta X_{\text{Obs}}^{12\text{hrs}}$ is the corresponding difference in local observations (MWR retrievals or tower measurements), $\Delta X_{\text{Obs}}^{\Delta t}$ is the difference between observations at time $t + \Delta t$ and a reference time $t$, and $\Delta X_{\text{precip}}^{\Delta t}$ is the change due to precipitation evaporation calculated with the precipitation evaporation model [see (13a) and (13b)] over the same time interval. This method takes advantage of the higher temporal resolution of MWR retrievals to better resolve changes in conditions aloft while maintaining consistency with the more reliable but scarce sounding data. Due to possible interactions between the precipitation sources and/or sinks of the heat and vapor and advection estimates, the precipitation evaporation model is integrated in an iterative manner using the advection estimates described above until convergence of the hourly estimates is obtained.

This procedure is applied to represent the evolution of the temperature throughout the column, using MWR retrievals above 100 m and tower observations below. For humidity, less accurate results may be obtained due to the sensitivity of the MWR brightness temperature measurements in its water vapor channels related to the backscattering of microwave radiation by raindrops, or due to a lack of accuracy of the in situ relative humidity sensors on the tower. A simple linear tendency derived from soundings is used above 100 m instead of MWR output to estimate $\Delta X_{\text{Obs}}^{\Delta t}$ in Eq. (A1), leading to a gradual transition from the saturated conditions at 1200 UTC to the subsaturated conditions between 150 and 2300 m at 0000 UTC shown in Fig. 14. Closer to the surface, sounding and tower profiles suggest that saturation was maintained in the lowest 150 m throughout the 12-h period between soundings. Advection tendencies near the surface were therefore estimated on the basis of maintaining saturation as warming was taking place, but constrained not to create supersaturated conditions.

With this methodology, evolving profiles representing observed conditions are obtained, including contributions to cooling and moistening from precipitation evaporation and allowing supersaturation to develop due to the presence of nonequilibrium raindrops falling into the inversion.

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