Modeling of Droplet Evaporation from a Nebulizer in an Inductively Coupled Plasma

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Abstract.
The evaporation rate of sample droplets in an inductively coupled plasma is investigated through the development of two models using the direct simulation Monte Carlo technique. A standard continuum evaporation model is contrasted with a kinetic technique designed to obtain correct results over a large range of Knudsen numbers. The droplet evaporation rates predicted by the continuum desolvation model are found to be in agreement with those of previous studies. We present the first predicted spatial distribution of droplet concentrations and evaporation rates in an ICP flow.

INTRODUCTION

Atmospheric pressure argon inductively coupled plasmas (ICPs) are electrical discharges that have dramatically altered the practice of elemental and isotopic ratio analysis. [1,2] An analyte is most commonly introduced into an ICP for mass spectrometric, atomic emission, or atomic fluorescence analysis via a spray of dilute aqueous solution. Numerical modeling can assist in predicting the fate of sample droplets, which will lead to a better understanding of the amount of detectable analyte in the nebulizer spray. To this end, a two-phase flow computational model has been developed to predict the desolvation rate of droplets in the plasma. [3]

Analytical measurements of the droplet desolvation rate within the ICP are very limited due to experimental difficulties. Recent work addressed the issue of droplet desolvation in an ICP [4]. The effect of a specific distribution of particle sizes and velocities was not considered; thus, the model could not predict the behavior of the spray as a whole. In addition, our approach provides a more detailed description of the change in droplet velocities due to interactions with argon in the plasma and the two-dimensional size distribution of particles in the ICP.

The nebulizer chosen for this study is the direct injection high efficiency nebulizer (DIHEN)[5-7], a concentric micronebulizer for applications in which the sample is limited, expensive, or toxic. A fast flow of argon gas causes the liquid sample to disperse into droplets ranging from 0.5 to 25 μm in size with an average velocity of 12.4 m/s. These droplets are then injected directly into the base of the plasma.

The initial set of plasma conditions (temperatures, number densities, and flow velocities) was generated with the high frequency induction plasma (HiFI) model. [8,9] The energy loss by the plasma due to droplet evaporation is not taken into account by the HiFI code; however, the plasma temperatures will not be significantly affected assuming the limited volume of sample introduced. The plasma is approximately 5 cm in length, and ranges in temperature from 3,000 K within the central channel and outlying portions of the plasma to greater than 10,000 K, as shown in Fig. 1. The continuum solution is translated into discrete particles of argon with positions and velocities consistent with the temperature and pressure gradients as predicted with HiFI. The output of this program is then read into the DSMC simulation as a set of background gas flow parameters.

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14. ABSTRACT

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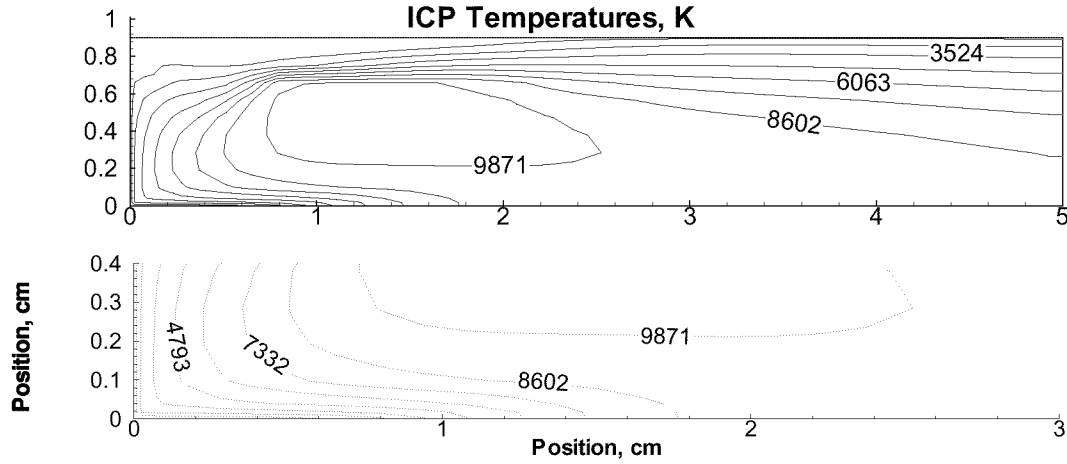Prescribed by ANSI Std Z39.18

FIGURE 1. ICP temperature gradients as predicted with the HiFI code. The lower contours show the portion of the plasma included in the DSMC simulation domain.

DSMC APPROACH AND NUMERICAL CHALLENGES

It is well known that the DSMC method is a reliable and efficient kinetic approach for modeling rarefied gas flows. [10] During the past several years, it has also been successfully applied to modeling of flows in the near-continuum regime. The numerical scope in this work, however, is quite different from the conventional application areas of the DSMC method. In this approach, the DSMC method is utilized in the simulation of droplet evaporation in a continuum regime. The selection of the DSMC method for this application is due to the fact that droplets exhibit particle, or microscopic, behavior that can be more reliably investigated with a kinetic approach. Since the DSMC method is the most promising kinetic approach in terms of different model implementations and modeling of two-and three-dimensional flows, it is used in this work to study droplet evaporation in a plasma.

There are several phenomena in this flow that have both interesting numerical and physical significance. The flow of interest is a two-phase flow. There are liquid droplets that may evaporate, coalesce, and condense, moving within a gas. This aspect raises many theoretical and practical questions connected with the necessity of an adequate description of droplet condensation/evaporation and interactions. Different flow regimes exist for the droplets and gas. While droplets are in the transitional or even free molecular regime with respect to each other, the surrounding gas is a continuum. Finally, the surrounding gas is at high temperatures, while the droplet surface temperature is relatively cool. This indicates that models developed to describe droplet/gas interactions, assuming a small difference in droplet surface and gas temperatures, are not likely to be applicable for this case. As a preliminary effort, this work examines droplet evaporation caused by a high-temperature argon gas using a DSMC scheme modified to include droplet-argon interactions.

The statistical modeling in low-density environments (SMILE) computational tool [11] based on the DSMC method is used in the computations. The SMILE tool is extended to simulate droplet behavior in the continuum regime. Several important features of the code enable us to improve the accuracy and efficiency of the simulation, such as an accurate collision scheme [12], species and spatial weights, and different grids for collisions and macroparameters, each adaptive to flow gradients.

The evaporation process has been modeled as follows. The ICP is maintained at a steady state, and it is reasonable to assume that the plasma properties are not affected with the introduction of a limited amount of sample. Hence, the argon plasma is modeled as a background argon gas having all the macroproperties (density, velocity, and temperature), of the calculated plasma. Using a background gas implies certain simplifications. Argon atoms are sampled once prior to the main simulation using equilibrium distributions with the location-dependent parameters obtained from HiFI. Using equilibrium distributions is reasonable since the plasma is in the continuum regime. These argon atoms are then fixed in the DSMC simulations such that they do not move over time and do not collide with each other. Therefore, the properties predicted with HiFI remain constant throughout the simulation. Argon-argon collisions are not modeled, and the positions of the argon atoms do not change, thus preserving the number densities and temperatures calculated with HiFI. No new

argon is introduced or removed from the simulation domain. Such an approach circumvents any potential computational difficulties connected with calculating argon collisions at atmospheric pressure.

The number of collisions calculated in a standard fashion is directly proportional to the total collision cross-sections of the particles in the gas and species number densities. The droplets generated by the DIHEN range from approximately 0.5 μm to 25 μm in diameter; therefore, the collision cross section of each droplet is correspondingly larger than a gas particle cross section. Since there is a high argon number density, one may conclude that to model each argon-droplet collision would be prohibitively large. To effectively model the collision process for droplets, a special procedure is implemented to reduce the number of droplet-argon collisions by a factor of approximately 1×10^{10}, but also increase the energy transfer during each collision by the same factor. This allows an equivalent amount of energy to be transferred during a single timestep, thus preserving both the calculation of droplet evaporation rate and the change in velocity of the droplets, while at the same time reducing the number of collisions calculated for each droplet.

The flow in the DIHEN is assumed to have an axial symmetry, and an axisymmetric SMILE code has therefore been used. To examine droplet evaporation in more detail, only a portion of the ICP was modeled adjacent to the nebulizer nozzle. The radius enclosed by the simulation is 4.0 mm from the center of the plasma, and extends 3.0 cm within the plasma. Although HiFI predicts temperatures and number densities for a plasma approximately 5 cm long with a 1 cm radius, all droplet evaporation occurs within the smaller computational domain, and thus any extension of the simulation domain beyond that point is unnecessary. The grid is uniform and consists of 40 cells in the y direction and 120 cells in the x direction. The time step is 1×10^{-6} s. The number of background argon molecules is about one million.

DISCUSSION OF EVAPORATION MODELS

Two droplet desolvation models are used to simulation particle evaporation and the effect of gas/droplet collisions on droplet temperature and diameters. The first one, a standard model for droplet evaporation, is widely used, especially in combustion applications [13-15]. Since a continuum regime is assumed, the model may not work properly in the limit of high Knudsen number. The second model uses a kinetic approach which yields equally accurate results for all Knudsen numbers [19,20]. The key assumption is that the droplet evaporation rate can be calculated by assuming small temperature and pressure changes from droplet/gas equilibria.

The continuum evaporation model is described as follows. The mass flux of water from the surface of a droplet is governed by the following equation [13]:

$$\dot{m}_v = 2\pi D \left(\frac{k}{c_p}\right)_g \ln(1 + B_M) \tag{1}$$

where $\dot{m}_v$ is the mass flux of water from the surface of the droplet, D is the diameter for the droplet, k is the thermal conductivity and c_p the heat capacity of the gas surrounding the droplet, and B_M is the mass transfer constant, based upon the mass fraction of water vapor at the droplet surface.

The droplet surface temperature is initially 298 K, and then increases as the droplet remains in the plasma. The rate of increase is dependent upon the amount of energy absorbed by the droplet over the course of a single timestep. The amount of energy absorbed is dependent in turn on the energy gained by the droplet through droplet-argon collisions and the energy lost to the surroundings via evaporation processes.

At some point, the evaporation rate of the droplet will become sufficiently high such that the net energy gain by the droplet during a single timestep approaches zero. At this point, no further energy is available to heat the droplet, and it will therefore reach a quasi-steady temperature. This representative temperature is known as the wet-bulb temperature of the droplet.

The above analysis yields a droplet evaporation model consistent with the D^2 law [13], which states that a plot of the square of an evaporating droplet's diameter versus time will be linear, with the slope equal to the evaporation constant for the droplet. For a droplet with a 10-μm initial diameter in a uniform 3,000 K environment, the above analysis indicates a wet-bulb temperature around 365 K, a droplet heating time of approximately 0.03 ms, and a total droplet evaporation time of 0.21 ms.

The limitations of the standard evaporation model are the assumption of a continuum regime and the requirement of a unitary Lewis number. The Lewis number is the ratio of thermal and mass diffusivities in the droplet; a value of 1 indicates that the energy and mass transport via diffusion are equally effective in the

boundary layer surrounding the droplet. This is not necessarily the case for every system. The Young-Carey model [18-20], a kinetic approach, does not share these two limitations.

Young [19,20] has proposed a general model to describe droplet evaporation occurring under nonequilibrium conditions:

$$j_w = \frac{p}{(2\pi m_w k T_\infty)^{1/2}} \left\{ L_{mm} \left(\frac{\Delta p_v}{p_{v,\infty}} \right) + L_{mq} \left(\frac{\Delta T}{T_\infty} \right) \right\} \tag{2}$$

The factors $\Delta p_v / p_{v,\infty}$ and $\Delta T / T_\infty$ represent the deviations from equilibrium associated with variations in pressure and temperature. Young derives the phenomenological coefficients, L_{mm} and L_{mq}, by analyzing the relative pressure and temperature jumps found at the droplet-continuum interfaces. A full analysis can be found in Young's work [19,20]; however, it is worth noting here that the primary differences between the Young-Carey model and the continuum model lies in the dependencies of the phenomenological coefficients on Sc, the Schmidt number; Pr, the Prandtl number; and Kn, the Knudsen number.

The gas-droplet surface Knudsen number is defined with the characteristic length taken to be the diameter of the evaporating droplet, as $Kn = \lambda/D$, and λ is the gas mean free path. The use of a Knudsen number dependent on D causes L_{mm} and L_{mq} to be proportional to D. Thus the evaporation rate is found to have a linear dependence of D^2 with time, which brings the kinetic approach into agreement with the continuum D^2 law.

RESULTS

Although the results of primary interest are the final points of evaporation for droplets generated by a DIHEN in an ICP, the simulation can also yield valuable information in combustion and atmospheric condensation problems. The DSMC/evaporation model is therefore tested under a variety of conditions, as described in Table 1.

TABLE 1. Initial conditions for simulations[2]

Case	Temperature	Droplet Distribution	Model	Figure
1	2000 K	10-μm	Continuum evaporation	2
2	ICP	10-μm	Continuum evaporation	3
3	2000 K	DIHEN	Continuum evaporation	4
4	ICP	DIHEN	Continuum evaporation	5
5	ICP	DIHEN	Continuum (with recondensation)	
6	2000 K	DIHEN	Kinetic evaporation (Young-Carey)	
7	ICP	DIHEN	Kinetic evaporation (Young-Carey)	

[2]All simulations use the DIHEN droplet velocity distribution.

The simplest of the cases considered consists of a uniform background gas at a temperature of 2,000 K with no net gas flow or macroscopic pressure gradients and an initial droplet diameter of 10 μm. This case will help to illustrate the trends in droplet evaporation due to the initial particle velocity distribution. Figure 2 shows the results of the simulation. Although a small number of droplets can reach distances of 8 mm from the nebulizer tip due to their higher velocities relative to the other droplets in the plasma, 90 percent of the droplets are completely desolvated within 4 mm. On average, a droplet will travel 2.5 mm, representing a residence time in the simulation domain of 0.20 ms.

Now consider simulation 2 (Table 1) with a constant droplet distribution but with a background gas consistent with an ICP. The pressure is close to atmospheric conditions; however, the temperature is much higher than 2,000 K. In addition, there is a large background gas flow in the positive x direction due to the 300 m/s nozzle exit velocity of the nebulizing gas (this high velocity is achieved due to the small DIHEN gas exit annulus, despite the relatively low volume flow of gas). The background flow increases the axial velocity of the droplets, leading to the result that some droplets survive to approximately 1.1 cm from the nozzle, more than 25 percent
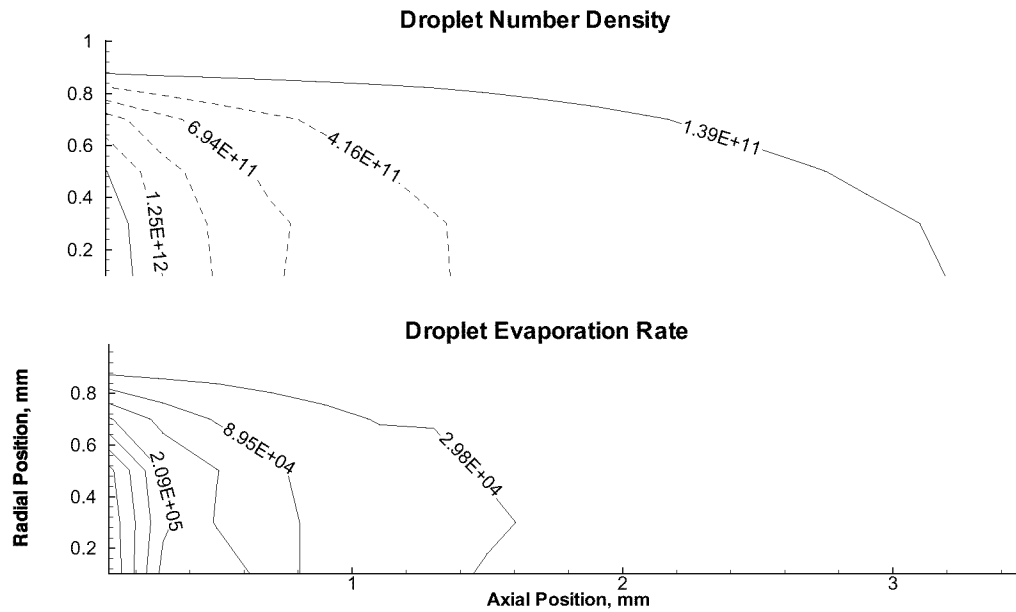
Droplet Number Density

Droplet Evaporation Rate

Radial Position, mm

Axial Position, mm

FIGURE 2. Droplet number density in m^{-3} and evaporation rate in m^{-3} $s^{-}1$ for a 10-μm uniform droplet distribution in a uniform 2000 K background gas.

farther than the droplets in the lower temperature, uniform gas. Any increase in evaporation rate due to higher temperatures leads to a shorter residence time of the droplet in the plasma; however, this is more than offset by the acceleration provided by the nebulizer gas. The average droplet will penetrate the plasma to 1.5 mm (indicating an average residence time of 0.12 ms, 40 percent shorter than in the low temperature simulation), while 90 percent will have evaporated within 6.0 mm. Figure 3 shows the results of the simulation.

The third condition under which the model was run is that of the uniform, lower temperature background gas with the inclusion of the DIHEN distribution of droplet sizes (Fig. 4). There is a large spread of droplet diameters, ranging from less than 0.5 μm to greater than 20 μm, randomly assigned to each droplet introduced into the simulation. Experimental data indicate little correlation between droplet size and droplet velocity, and thus the two parameters are decoupled.

The location at which the majority of droplets desolvate for this set of operating conditions is less defined. A sizable fraction of the droplets have a sufficiently low size such that evaporation occurs within several microseconds of entering the plasma. These droplets do not have sufficient time to significantly penetrate the plasma before desolvation occurs. However, the majority of the droplets are larger in size, and the simulation shows that 90 percent of the droplets evaporate within 2.3 mm of the nozzle exit.

Finally, the simulation was run using the background gas corresponding to the temperatures, pressures, and flows present in the ICP with the DIHEN distribution of droplets (case 4, see Fig. 5). As with the 10 μm droplet cases, the point at which 90 percent of the droplets have evaporated is farther from the exit nozzle of the DIHEN than for the case of uniform, but lower temperature, background gas. In this case, the 90 percent droplet evaporation point is located 3.1 mm into the simulation domain. However, larger and faster droplets can remain in the plasma up to 1.3 cm from the nebulizer tip.

Selected cases that used the standard evaporation model were repeated using the Young-Carey (kinetic) model of droplet evaporation. The kinetic evaporation model was used to calculate the evaporation time of droplets with the DIHEN size distribution in a 2,000 K uniform environment as well as a DIHEN droplet size distribution in the ICP. The point at which 90 percent of the droplets desolvate is located at 2.2 mm from the nebulizer tip for the ICP background gas temperatures and at 1.2 mm from the nebulizer tip for the 2000 K uniform background gas condition.

The evaporation time as obtained with the Young-Carey model differs significantly from that obtained using the continuum model. The Young-Carey treatment was derived for a case in which the deviation from an equilibrium situation is relatively small [19]. The simulations show that by lowering the background gas temperature to less than 1,000 K, the results of the kinetic evaporation model converge with those of the
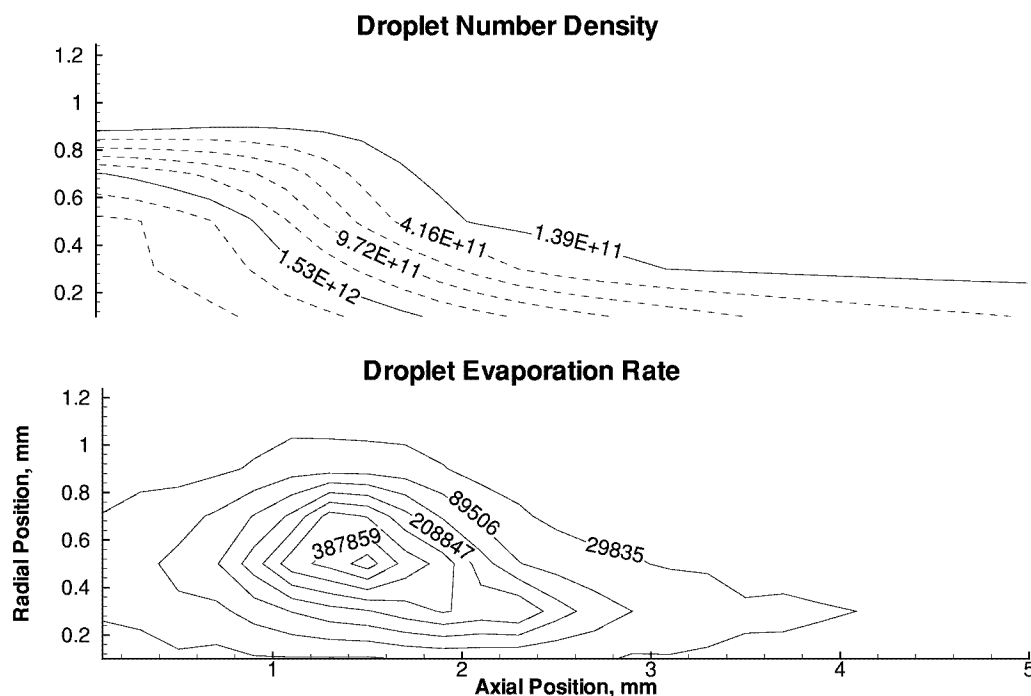
Droplet Number Density

Droplet Evaporation Rate

FIGURE 3. Droplet number density in m^{-3} and evaporation rate in m^{-3} s^{-1} for a 10-μm uniform droplet distribution in a background gas with temperature gradients identical to that of an ICP.

continuum evaporation model. This highlights the need for a kinetic-based evaporation model designed to give adequate solutions for droplet evaporation in situations that are far from equilibrium.

CONCLUSIONS

The study presented in this report demonstrated the ability to formulate and construct a general gas-particle evaporation model and simulation technique general to a wide range of Knudsen numbers and non-equilibrium flow conditions. Several numerical algorithms were developed to efficiently model high pressure gas-surface interactions and have been incorporated into the DSMC method. Two specific droplet evaporation models were considered and compared. The numerical scheme is sufficiently general to provide a framework for future extensions of the two phase flow modeling that will incorporate droplet condensation.

The results of the simulations show that the depth to which a droplet penetrates the plasma is, in general, less than 1 cm. The total time of evaporation agrees with that predicted in reference 6. However, the model in reference 6 did not include droplet transport and thus a full comparison is not possible. The plasma, however, is approximately 5 cm in length, and therefore desolvation processes can occupy 20 percent of the analyte's lifetime in the plasma. Considering that the analyte must then undergo the processes of evaporation to the gas phase, atomization, excitation, and ionization before it can be detected, a shorter time to desolvation would be preferred.

It can be seen that the high plasma temperatures lead to efficient desolvation; however, high gas velocities cause the analyte to penetrate the plasma to a higher degree. Although a lower gas velocity would lead to slower droplets, the nebulizer would be less efficient, producing larger droplets. Further studies of the design trade-offs between these two effects are necessary.

Finally, it can be seen that a more inclusive droplet evaporation model is required. Although the continuum model is likely to give accurate results, a particle-based method would be more comprehensive. Since the situation under study is dominated by continuum gas conditions, the continuum evaporation model can be used to give an acceptable desolvation rate. However, the influence of smaller droplets (higher Knudsen number) in terms of ICP/DIHEN performance and related plasma and combustion processes is still under investigation.
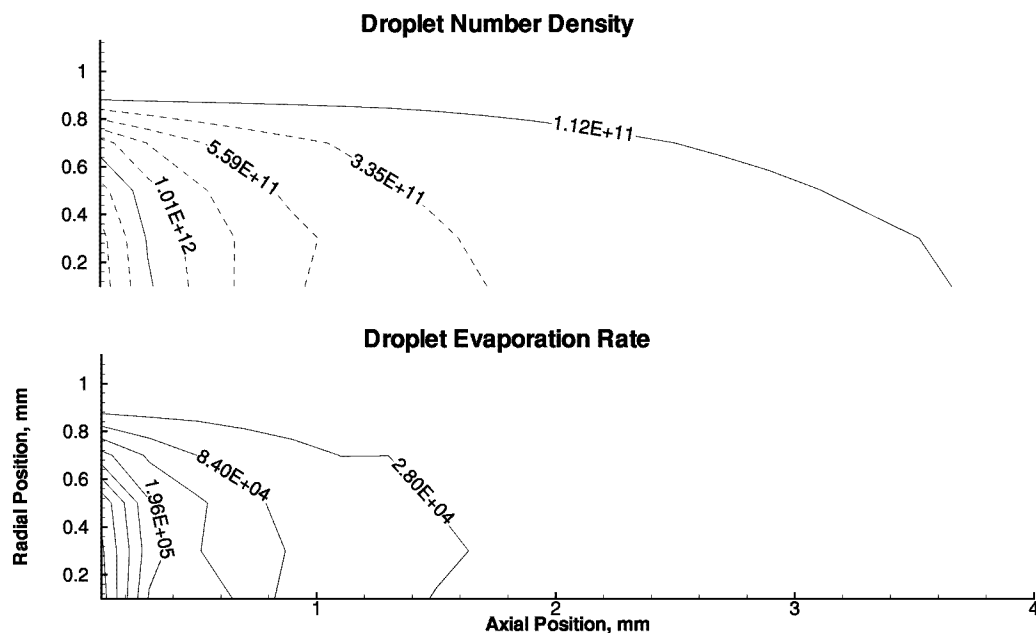
Droplet Number Density

Droplet Evaporation Rate

FIGURE 4. Droplet number density in m^{-3} and evaporation rate in m^{-3} $s^{-}1$ for a droplet size distribution representative of the DIHEN in a uniform 2000 K background gas.

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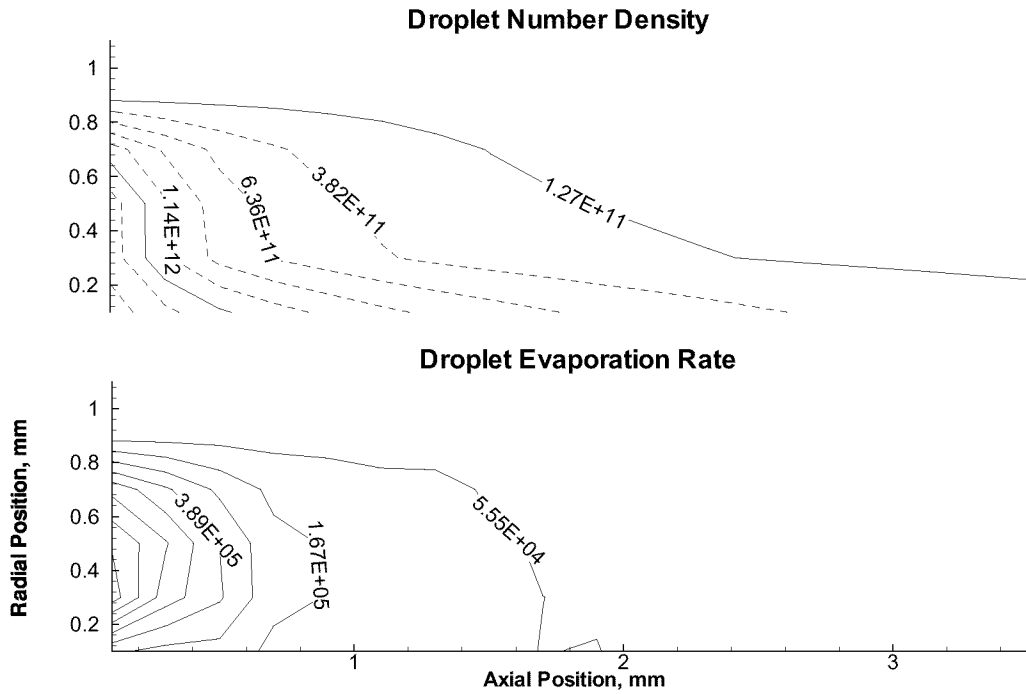
Droplet Number Density

Droplet Evaporation Rate

FIGURE 5. Droplet number density in m^{-3} and evaporation rate in $m^{-3}\,s^{-1}$ for a droplet size distribution representative of the DIHEN in a background gas with temperature gradients identical to that of an ICP.

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