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Entropy **2009**, *11*, 713-747; doi:10.3390/e11040713

OPEN ACCESS
entropy
ISSN 1099-4300
www.mdpi.com/journal/entropy

Review

The Maximum Entropy Formalism and the Prediction of Liquid Spray Drop-Size Distribution

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Received: 27 August 2009 / Accepted: 26 October 2009 / Published: 2 November 2009

Abstract: The efficiency of any application involving a liquid spray is known to be highly dependent on the spray characteristics, and mainly, on the drop-diameter distribution. There is therefore a crucial need of models allowing the prediction of this distribution. However, atomization processes are partially known and so far a universal model is not available. For almost thirty years, models based on the Maximum Entropy Formalism have been proposed to fulfill this task. This paper presents a review of these models emphasizing their similarities and differences, and discusses expectations of the use of this formalism to model spray drop-size distribution.

Keywords: maximum entropy formalism; liquid sprays; drop-size distribution; drop-velocity distribution

PACS Codes: 47.55-t, 47.55.D, 89.70.Cf

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1. Introduction

A liquid spray is defined as a flow of individual droplets evolving in a surrounding gaseous medium. Each droplet has its own diameter and velocity. Many industrial processes or domestic applications involve liquids as sprays rather than as continuous flows (field treatment in agriculture, drug delivery in medical therapy, mixture preparation for combustion purposes, coating processes, fire extinction, atmosphere cleaning, powder fabrication…[1,2]). Whatever the process or the application involving a liquid spray, its efficiency depends on the spray characteristics, among which droplet-size distribution and velocity distribution are the most important ones. Therefore, models to predict these characteristics are very much required in order to control the spray production and improve the efficiency of applications involving a spray.

The common process to produce liquid sprays consists in ejecting a liquid flow into a gaseous environment that can be at rest or in motion. As soon as the liquid flow issues from the nozzle, deformations appear on the liquid interface. These deformations grow in space and time until the liquid flow cannot sustain them anymore and disintegrates into a cloud of droplets. This flow deformation and disintegration is designated as the atomization process. Two major factors control atomization processes, namely, the presence of initial disturbances on the liquid-gas interface and mechanisms that allow some of these disturbances to grow. The characteristics of the resulting spray depend on both factors. Theoretical analyses have been carried out on the initial distortion and disintegration of liquid streams [3,4]. These approaches are based on the determination of unstable waves that grow on the liquid-gas interface and therefore dominate its breakup. Coupled with a breakup scheme, a theoretical mean drop-diameter can be deduced from the characteristics of the dominant wave. Despite these approaches have reported important information in many situations, they can predict a limited number of spray characteristics and are applicable in a restricted domain. For instance, high-energy atomization processes are still untouched by theoretical approaches. On the other hand, experimental investigations conducted on liquid atomization processes reveal the complexity of this phenomenon that involves liquid and gas turbulence, interfacial instability, liquid cavitation, gas and liquid interaction… [5]. Thus, liquid atomization processes are still partially understood which explains the absence of a universal model that would allow the prediction of liquid spray characteristics.

Babinski and Sojka [6] classify the models to predict spray drop-size distributions in three groups, i.e., the empirical method, the discrete probability method and the maximum entropy method. The empirical method consists of determining a mathematical form that fits the experimental data collected for a wide range of operating conditions. This technique has produced many empirical mathematical spray drop-size distributions. Among others, we can quote the log-normal, upper-limit, root-normal, Rosin-Rammler, Nukiyama-Tanasawa and log-hyperbolic distributions [1,6-8]. *A priori*, it is not possible to know which of these distributions will be better to reproduce a given situation. Of course, their ability to fit spray drop-size distributions is directly a function of the number of parameters these distributions contain. This number varies from two to four and can reach eight for empirical joint diameter-velocity distributions (see [8] for instance).

The discrete probability method postulates that polydispersion in liquid sprays is due to initial condition fluctuations [6]. A deterministic model as those mentioned above [3,4] is used to predict a specific diameter for a given set of initial conditions. The polydispersion of the drop-size is obtained by describing the fluctuating initial conditions by a continuous probability density function (input pdf). The dispersion methods essentially define a transform from this input pdf to the drop-size distribution pdf using the breakup model as a transform function. This approach is very attractive but it remains difficult to apply as the problem of the determination of the drop-diameter distribution is replaced by the one of the determination of the initial condition fluctuation distribution. Furthermore, for many atomization processes and particularly those involving high energy, it is believed that the origin of polydispersion is not solely related to fluctuations of initial conditions.

For almost thirty years, models based on the Maximum Entropy Formalism (MEF) have been developed to predict liquid spray drop-diameter and velocity distributions. MEF is a method of inference allowing the determination of a probability density function from a limited amount of information. This information specifies properties that the distribution must satisfy and comes from a partial knowledge of the distribution sought. Among the distributions that satisfy the available information, the MEF states that the most likely or least biased solution is the one whose statistical entropy is a maximum. MEF is often identified with statistical thermodynamics but that identification is too limiting. It has found numerous applications in engineering and sciences. Two points motivate the application of this formalism to formulate liquid spray drop-diameter and velocity distribution models. First, liquid atomization is a non deterministic process and may not be modeled by conventional deterministic methods. Second, liquid spray drop-diameter and velocity distributions are mathematically represented by probability distributions. Application of MEF to determine liquid spray characteristics can address two questions, namely, can we predict drop-size and velocity distributions if partial information (obtained either from a model or from an experiment) is available, and, what is the minimum information required and what is its form to predict a relevant drop-size and velocity distributions?

This paper present a review of the models based on the MEF to predict drop-size distributions. Section 2 recalls some mathematical definitions associated to liquid spray drop-size distributions. Section 3 summarizes the MEF as it is used in the formulation of drop-size distribution models. The main section of this paper (Section 4) describes the different approaches based on the application of the MEF and the paper ends with a summary and conclusions.

2. Mathematical Representation of Spray Drop-Size Distribution

Paloposki [9] provided an accurate and complete mathematical definition of spray drop-size distribution differencing the temporal and spatial descriptions. As a first approach, the introduction of mathematical concepts to describe spray drop-size distribution does not require this differentiation. On the other hand, it becomes essential when dealing with diagnostics dedicated to the measurement of this spray characteristic. The definitions presented in this section are commonly encountered in the literature [1,2].

Several types of drop-size distribution can be defined [10]. Among them, the number-based and volume-based drop-diameter distributions are those that are commonly used in the literature. Let us consider a spray containing N_T droplets. Each droplet is assumed spherical and therefore fully characterized by a diameter D. (The assumption of sphericity is maintained throughout the paper except otherwise mentioned.) The droplet diameter ranges in the interval $[D_{min}; D_{max}]$ where D_{min} and D_{max} are the minimum and maximum drop diameter, respectively. This interval is divided into n_c classes $[D_i - \Delta D_i/2; D_i + \Delta D_i/2]$ where D_i and ΔD_i are the median diameter and the width of class i, respectively. The droplets are distributed in this class series according to their diameters and it is assumed that all droplets belonging to a given class i have the same diameter equal to the class median diameter D_i. This assumption is reasonable provided that $\Delta D_i << D_i$, which is usually the case. By introducing N_i, the number of droplets in class i, it becomes:

$$\sum_{i=1}^{n_c} N_i = N_T \tag{1}$$

Similarly, the total volume of liquid V_T contained in the spray and the liquid volume V_i contained in each class are introduced. The following equation can be written:

$$\sum_{i=1}^{n_c} V_i = V_T \tag{2}$$

The spray drop-size distribution is characterized by the number-fraction distribution P_{ni} and the volume-fraction distribution P_{vi} defined by:

$$\begin{cases} P_{ni} = \dfrac{N_i}{N_T} \\ P_{vi} = \dfrac{V_i}{V_T} \end{cases} \tag{3}$$

Considering that the drop diameter is a continuous variable, continuous number-based drop-size distribution $f_n(D)$ and volume-based drop-size distribution $f_v(D)$ are introduced:

$$\begin{cases} P_{ni} = \displaystyle\int_{D_i-\Delta D_i/2}^{D_i+\Delta D_i/2} f_n(D)dD \\ P_{vi} = \displaystyle\int_{D_i-\Delta D_i/2}^{D_i+\Delta D_i/2} f_v(D)dD \end{cases} \tag{4}$$

Equations 1 to 3 indicate that the sums ΣP_{ni} and ΣP_{vi} for i varying from 1 to n_c are both equal to 1. Consequently, according to Equation 4, the distributions f_n and f_v are both normalized. However, it is important to mention here that, despite the fact that the distributions P_{ni} and P_{vi} show similar mathematical properties, only the distribution P_{ni} is a probability distribution (it expresses the probability of finding a drop of a given diameter). By no means P_{vi} can be associated to a probability: it expresses a volume-fraction. In consequence, strictly speaking, only f_n is a probability density function. To illustrate the difference between the number-fraction and the volume-fraction distributions, one can estimate the change produced by adding one supplementary droplet with diameter D_k (class k) in the set of N_T droplets. Now the total number of droplets is $N_T + 1$, and, for $i \neq k$, $P_{ni} = N_i/(N_T + 1)$ and $P_{vi} = V_i/(V_T + \pi D_k^3/6)$. We see that the modification of the number-fraction distribution is independent of any characteristic of the added droplet whereas the modification of the volume-based distribution depends on the diameter of this droplet. In the number-based representation, each droplet is considered as an event and has the same weight whereas in the volume-based representation, the contribution of each droplet is not equivalent but is weighted by its volume.

A mean drop-diameter series D_{kl} standardized by Mugele and Evans [11] is commonly used to characterize liquid spray drop-size distributions. This series is defined by:

$$D_{kl} = \left(\frac{\int_0^\infty f_n(D) D^k dD}{\int_0^\infty f_n(D) D^l dD} \right)^{\frac{1}{(k-l)}} = \left(\frac{\int_0^\infty f_v(D) D^{k-3} dD}{\int_0^\infty f_v(D) D^{l-3} dD} \right)^{\frac{1}{(k-l)}} \quad (5)$$

From a mathematical point of view, the indexes k and l can take any real values. The most encountered mean diameters are listed in Table 1. As pointed out by Sowa [10], the mean diameter D_{10} corresponds to the arithmetic mean of the number-based distribution f_n and D_{43} to the arithmetic mean of the volume-based distribution f_v. Often encountered in the literature, the Sauter mean diameter D_{32} corresponds to the diameter of the drop that has the same volume to surface area ratio as the entire spray.

A given spray can be indifferently described by the number-based or by the volume-based drop-size distribution. Therefore, these two distributions must be related to each other. Making use of Equation 5, it can be demonstrated that:

$$f_v(D) = \left(\frac{D}{D_{30}} \right)^3 f_n(D) \quad (6)$$

Table 1. Commonly encountered mean drop-diameters.

k	l	Designation	Relation
1	0	Number mean-diameter	$N_T D_{10} = \sum_{i=1}^{n_c} N_i D_i$
2	0	Surface mean-diameter	$N_T \pi D_{20}{}^2 = \sum_{i=1}^{n_c} N_i \pi D_i{}^2$
3	0	Mass mean-diameter	$\frac{N_T \pi D_{30}{}^3}{6} = \sum_{i=1}^{n_c} \frac{N_i \pi D_i{}^3}{6}$
3	2	Sauter mean-diameter	$\frac{D_{32}}{6} = \sum_{i=1}^{n_c} \frac{N_i \pi D_i{}^3}{6} \Big/ \sum_{i=1}^{n_c} N_i \pi D_i{}^2$
4	3	De Brouckere	$D_{43} = \int_0^\infty f_v(D) dD$

The distributions defined in this section can be generalized. Using the characteristic length (the diameter D) or surface (πD^2) of each drop, the definitions of the length-based and surface-based distributions are straightforward [10]. These distributions are rarely used in the literature. Furthermore, as we will see in section 4, liquid spray drops might be also described by a joint size-velocity distribution, each droplet of a spray having their own velocity. In this case, the droplets are distributed in a two dimensional space according to their diameter D_i and velocity U_j and a two-dimensional probability distribution P_{ij} expressing the probability of finding a droplet with diameter D_i and velocity U_j, is introduced. This probability distribution satisfies:

$$\sum_i \sum_j P_{ij} = 1 \tag{7}$$

and is associated to a normalized continuous distribution $f(D,U)$.

3. The Maximum Entropy Formalism (MEF)

The objective of this section is to present the mathematical aspect of the MEF as used in the literature for the prediction of the spray drop-size and velocity distributions. This presentation is derived from Sellens and Brzustowski [12]. For more information concerning the origin of this formalism and its applications, refer to Kapur [13] where an extensive list of references is available.

MEF is a tool of statistical inference that allows the determination of a probability distribution from a limited amount of information. This formalism states that the most likely (or least biased) probability distribution P_i that satisfies a set of constraints that expresses known characteristics of the distribution sought is the one whose Shannon's entropy S is maximum. This entropy is defined by:

$$S = -k \sum_i P_i \, ln(P_i) \tag{8}$$

where k is a constant. The constraints, expressing the available information concerning the distribution sought, can be given the following form:

$$\sum_i P_i g_{r,i} = \langle g_r \rangle \qquad\qquad r = 1,\ldots,m \tag{9}$$

where the quantities $\langle g_r \rangle$ are known moments of the distribution and constitute the available information. An additional constraint arises from the normalization of the probability distribution, i.e.,:

$$\sum_i P_i = 1 \tag{10}$$

There are numerous probability distributions that satisfy a given set of constraints (Equations 9 and 10), but there is only one whose entropy S is maximum. Using the method of Lagrangian multipliers, it can be demonstrated that this probability distribution is given by:

$$P_i = exp\left(-\lambda_0 - \sum_{r=1}^{m} \lambda_r g_{r,i} \right) \tag{11}$$

where the Lagrangian multipliers λ_0, λ_1,…, λ_r must be determined. The normalization constraint (Equation 10) relates the multipliers as follows:

$$exp(\lambda_0) = \sum_i exp\left(-\sum_{r=1}^{m} \lambda_r g_{r,i} \right) \tag{12}$$

and the other constraints (Equation 9) gives the following m relationships:

$$\langle g_r \rangle exp(\lambda_0) = \sum_i g_{r,i} \, exp\left(-\sum_{r=1}^{m} \lambda_r g_{r,i} \right) \tag{13}$$

The resolution of Equations 12 and 13 allows the Lagrangian multipliers to be determined as a function of the information, i.e., the *m* moments $\langle g_r \rangle$. By assuming a uniform discretization of the solution space, an equivalent continuous formulation can be derived. Let's introduce ψ, the solution space that contains all the permissible states, a probability density function *f* is defined by:

$$P_i = \int_{\Psi_i - \Delta\Psi_i/2}^{\Psi_i + \Delta\Psi_i/2} f d\Psi \tag{14}$$

The set of constraints (9) and (10) is now replaced by:

$$\begin{cases} \int_{\Psi} f d\Psi = 1 \\ \int_{\Psi} f g_r d\Psi = \langle g_r \rangle \qquad r = 1,\ldots,m \end{cases} \tag{15}$$

and the statistical entropy of the probability density function is:

$$S = -k \int_{\Psi} f \, ln(f) d\Psi \tag{16}$$

The PDF that maximizes *S* (Equation 16) subject to the set of constraints (Equation 15) is:

$$f = exp\left(-\lambda_0 - \sum_{r=1}^{m} \lambda_r g_r \right) \tag{17}$$

Note that if no information is introduced in the formalism, i.e., system of Equation 15 reduces to the normalization constraint, Equation 17 returns a constant distribution ($f = \exp(-\lambda_0)$). This solution illustrates that when nothing is known about the reachable states, the most reasonable suggestion consists in attributing the same probability of occurrence to each state.

4. Application of MEF to Determine Liquid Spray Drop-Size Distribution

The first attempt to predict the liquid spray drop-size distribution on the basis of the MEF is due to Sellens and Brzustowski [12,14]. They considered the breakup of a liquid sheet (thickness *t*, velocity U_s) in the vicinity of the breakup region. The constraints expressed conservation laws that are expected to be satisfied in any physical process, namely, conservation of mass, momentum and energy. These constraints required to account for the size and the velocity of each drop. Thus, the solution is defined by drop diameter *D* and velocity *U* so that an element of the solution space is $d\Psi = dDdU$. The parameters are non-dimensionalized by the spray mass mean-diameter D_{30} and the liquid sheet velocity U_s. (Non-dimensionalized parameters are noted $\delta = D/D_{30}$ for the diameter, $\tau = t/D_{30}$ for the sheet thickness and $u = U/U_s$ for the velocity. These notations will be kept throughout the paper otherwise mentioned.) As far as the energy constraint is concerned, Sellens and Brzustowski pointed out the necessity of writing two separate constraints for the kinetic energy and the surface energy in order to account for irreversibility of certain energy transformations. Indeed, in an atomization process, kinetic energy is readily transformed into surface energy but the reverse transformation is not possible. Sellens and Brzustowski based their formalism on the following list of constraints:

- Normalization:

$$\iint_{\Psi} f d\delta du = 1 \tag{18}$$

- Mass conservation:

$$\iint_{\Psi} f\delta^3 d\delta du = 1 + S_m \tag{19}$$

- Momentum conservation:

$$\iint_{\Psi} f\delta^3 u d\delta du = 1 + S_{mv} \tag{20}$$

- Kinetic energy conservation:

$$\iint_{\Psi} f\delta^3 u^2 d\delta du = 1 + S_{ke} \tag{21}$$

- Surface energy conservation:

$$\iint_{\Psi} f\delta^2 d\delta du = \frac{1}{3\tau} + S_s \tag{22}$$

where the source terms S_m, S_{mv}, S_{ke} and S_s allow accounting for losses of mass, momentum, kinetic energy or surface energy during the breakup process, respectively. For instance, a high evaporation rate would be represented by a negative mass source term whereas condensation of the vapor in the ambient medium would be taken into account with a positive mass source term. It must be noted that in their development, Sellens and Brzustowski related the initial total liquid mass M and the total number of droplets N_T by $M = N_T\rho_L\pi D_{30}^3/6$ (where ρ_L is the liquid density) which implicitly imposes $S_m = 0$. This small inconsistency was corrected in their next publications. Following Equation 17, the joint diameter-velocity distribution derived by Sellens and Brzustowski has the form:

$$f = exp\left(-\lambda_0 - \lambda_1\delta^2 - \lambda_2\delta^3 - \lambda_3\delta^3 u - \lambda_4\delta^3 u^2\right) \tag{23}$$

By integrating this function over the velocity space, i.e., from $u = 0$ to a maximum permitted velocity u_m, Sellens and Brzustowski derived the following number-based drop-diameter distribution $f_n(\delta)$:

$$f_n(\delta) = \sqrt{\frac{\pi}{4\lambda_4\delta^3}} \times \left(erf\left(u_m\sqrt{\lambda_4\delta^3} + \frac{\lambda_3}{2}\sqrt{\frac{\delta^3}{\lambda_4}} \right) - erf\left(\frac{\lambda_3}{2}\sqrt{\frac{\delta^3}{\lambda_4}} \right) \right) \times exp\left(-\lambda_0 - \lambda_1\delta^2 - \lambda_2\delta^3 - \lambda_3\delta^3 u - \lambda_4\delta^3 u^2\right) \tag{24}$$

Considering a loss-free atomization process (all term sources equal to 0), Sellens and Brzustowski [12] demonstrated that the number-based drop-diameter distribution given by Equation 24 is very similar in shape to the Rosin-Rammler distribution which is a two-parameter empirical drop-diameter distribution extensively used to represent spray drop-size distributions [2,9]. However, the number-based maximum entropy distribution reveals a drawback: the probability associated with the smaller drop sizes is much larger than is expected, i.e., it doesn't decrease towards 0 when $D = 0$. Then, the left tail of the distribution contradicts almost all experimental observations reported in the literature. As far as the drop-velocity distribution is concerned, Sellens and Brzustowski [14] pointed out the necessity of a non-zero momentum source term in order to produce an initial velocity distribution. The assumption of a uniform velocity in the liquid sheet would lead to a uniform velocity in the droplets if $S_{mv} = 0$. With $S_{mv} \neq 0$, each drop category reports a Gaussian velocity distribution. Coupling the initial droplet velocity distribution with a simple air drag model Sellens and Brzustowski investigated the downstream velocity distribution behavior [14]. For all drop categories, the velocity variance rapidly decreases towards 0 whatever the initial velocity distribution. Therefore, in real sprays, a mean velocity should be sufficient to characterize any drop category. Furthermore, this shows that spray drop-velocity distribution is strongly dependent on the nature of the gaseous environment rather than on the initial velocity distribution.

Parallel to Sellens and Brzustowski investigation, Li and Tankin [15,16] developed their own approach. This approach has been extensively used as we will see in the following. In their first contribution, Li and Tankin [15] derived a drop-diameter distribution on the basis of a single constraint expressing the conservation of mass. Contrary to Sellens and Brzustowski [12,14], the parameter of the solution space was the volume V of the drop instead of the diameter D, i.e., $d\Psi = dV$. The probability distribution P_i of finding a droplet with a volume V_i they obtained is:

$$P_i = exp\left(-\lambda_0 - \lambda_1 V_i\right) \tag{25}$$

Then, by performing a change of variable from V to D with $V = \pi D^3/6$ and noting that:

$$P_i = \int_{\text{class } i} f_n(V) dV = \int_{\text{class } i} f_n(D) dD \tag{26}$$

they obtained the following number-based drop-diameter distribution:

$$f_n(D) = 3\alpha D^2 \, exp\left(-\alpha D^3\right) \tag{27}$$

where the parameter α is a function of the parameters introduced in the mass conservation law, i.e., the liquid density, the liquid mass flow rate and the number of drops produced per unit time. This parameter can be related to any mean drop diameter, in particular:

$$\alpha = \frac{1}{\left(D_{32}\Gamma\left(\frac{5}{3}\right)\right)^3} \tag{28}$$

where Γ is the Gamma function. The enthusiastic aspect of the diameter distribution given by Equation 27 is that it is a form of the Nukiyama-Tanasawa distribution which is another well-known empirical drop-diameter distribution [1,9,17]. Li and Tankin [15] compared their solution under the volume-based form (using Equation 6) with experimental drop-size distributions drawn from the literature. The parameter α was calculated from Equation 28 where the mean diameter D_{32} was the one reported by the experiment, i.e., the mathematical distribution was forced to have the same Sauter mean diameter as the one of the measured distribution. The authors pointed out a satisfactory agreement (good prediction of the peak diameter and slight underestimation of the distribution width). Furthermore, contrary to Sellens and Brzustowski's solution (Equation 24), Li and Tankin's solution shows the expected trend at small drop-diameter and reaches 0 when $D = 0$. This particular behavior of

Li and Tankin's solution is appreciable and comes from the change of variable described above and that imposes $f_n(D) \propto D^2$. However, as it will be discussed later in this paper, this change of variable violates the MEF.

In their second contribution, Li and Tankin [16] extended the previous work to derive a joint drop diameter-velocity distribution. They wrote a set of constraints on the basis of the conservation of mass, momentum and energy during the process. The control volume in their formulation extended from the nozzle exit of the injector down to the plane where the droplets are formed. Therefore, the constraints they used express that the mass, momentum and energy of the droplets just downstream of their formation are the same respectively as those of the continuous liquid bulk at the nozzle exit plus any source terms which exist in the region between the nozzle exit and the plane of the droplet formation. This aspect constitutes a fundamental difference with Sellens and Brzustowski [12,14] approach that focuses on the breakup region only. A second important difference in Li and Tankin [16] formulation concerns the writing of the energy constraint: instead of writing two separate constraints for the kinetic energy and the surface energy they proposed a total energy constraint expressing the conservation of the sum of the directed kinetic energy and surface energy. Conducting their development in the volume-velocity solution space ($d\Psi = dVdU$), and using the mean drop volume V_m ($= \pi D_{30}^3/6$) and the mean velocity U_0 of the liquid flow issuing from the nozzle to define the dimensionless parameters v ($= V/V_m$) and u ($= U/U_0$), their combined total energy constraint had the form:

$$\iint_{\Psi} f\left(vu^2 + B' Kv\right) dv du = 1 + S_e \tag{29}$$

where $B' = 2\sigma/\rho_L U_0^2$, σ is the liquid-gas surface tension coefficient and K is the drop surface area to volume ratio. The mass and momentum conservation constraints they used are similar to those of Sellens and Brzustowski (Equations 19 and 20), which, as noticed above, implicitly supposes that $S_m = 0$. Following the same mathematical manipulation as in their first approach, Li and Tankin [16] obtained the following joint drop diameter-velocity distribution:

$$f = 3\delta^2 exp\left(-\lambda_0 - \lambda_1\delta^3 - \lambda_2\delta^3 u - \lambda_3\left(\delta^3 u^2 + B\delta^2\right)\right) \tag{30}$$

where:

$$B = \frac{12}{We} \qquad\qquad We = \frac{\rho_L {U_0}^2 D_{30}}{\sigma} \tag{31}$$

We is a liquid Weber number, dimensionless number often encountered in liquid atomization. It can be demonstrated that Equation 30 converges to the previous solution (Equation 27) when $We \rightarrow \infty$. The distributions obtained by Sellens and Brzustowski (Equation 23) and by Li and Tankin (Equation 30) show two differences: 1 – Li and Tankin's distribution (Equation 30) is proportional to δ^2: as mentioned above this is the signature of the solution space modification during the mathematical development, 2 – Li and Tankin's distribution (Equation 30) is explicitly a function of the liquid-gas surface tension through the Weber number: this is presented by the authors as an advantage as surface tension is an important parameter in liquid atomization processes. Furthermore, Li and Tankin [16] claimed that their solution did not require any non-zero momentum source term to predict velocity distribution but such evidence was not shown in their paper. Li *et al*. [18] reported numerical applications of the joint drop diameter-velocity distribution given by Equation 30 investigating the influence of the source terms S_e and S_{mv}. For each drop category, the velocity distribution was a truncated Gaussian distribution. Furthermore, these calculations revealed an important and complicated influence of the source terms and of the Weber number on the resulting distribution characteristics. This clearly indicates that the prior determination of the source terms required for these approaches to be fully predicting constitutes a tricky and crucial step.

The question of whether the drop-diameter distributions obtained from the application of the MEF show a better ability to represent real spray drop-size distribution has been addressed by Bhatia and Durst [19]. They first compared the number-based diameter distribution due to Li and Tankin [15] (Equation 27) with the Rosin-Rammler, log-normal and log-hyperbolic distributions. These three empirical distributions depend on 2, 2 and 4 parameters, respectively. These distributions were compared with the experimental distributions used by Li and Tankin [15] as well as local distributions of water sprays produced by a solid-cone nozzle and measured with a Phase Doppler technique (PDT). A description of working of this instrument is available in [20]. This diagnostic performs a local measurement (being delimited by the intersection of two laser beams the measuring volume is far less than the volume of the entire spray) and reports the diameter and velocity of the drops that pass at the location of measurement. The PDT is a temporal sampling instrument: such as single-particle counters, it registers signals proportional to temporal frequency (count/s). Contrary to Li and Tankin [15], Bhatia and Durst compared the number-based drop-diameter distributions. Whereas, for all situations, the log-normal and log-hyperbolic distributions showed a good agreement with the measured distributions, they noticed a poor ability of the Li and Tankin's solution (Equation 27) and of the Rosin-Rammler distribution to represent number-based drop-size distribution. It must be noted here that Li and Tankin's solution tested in this work (Equation 27) is a one parameter distribution and therefore has less ability to fit drop-size distribution. Furthermore, one should mention that the empirical Rosin-Rammler distribution has been derived to fit volume-based drop-size distribution and that the use of the corresponding number-based distribution is not recommended.

Bhatia and Durst [19] also performed a similar work on joint diameter-velocity distributions comparing Sellens and Brzustowski's distribution (Equation 23), Li and Tankin's distribution (Equation 30) and the empirical two-dimensional hyperbolic-distribution. For each case, the parameters of the distributions were numerically determined in order to provide the best fit. These comparisons showed a much better ability of the hyperbolic-distribution to represent the measured distributions than the two maximum entropy distributions. In particular, the maximum entropy number-based diameter distributions $f_n(\delta)$ obtained by integrating the two-dimensional distributions over the velocity space, were in strong disagreement with the experimental observations. Bhatia and Durst [19] concluded that the two-dimensional coupled distributions due to Sellens and Brzustowski [12,14] or to Li and Tankin [16] are not appropriate to approximate drop-size distributions in sprays. One should note here that the two-dimensional hyperbolic distribution depends on eight parameters and has therefore a better fitting capability than the tested maximum entropy distributions that depend on four parameters only. Furthermore, one should emphasize that the lack of agreement between Sellens and Brzustowski's number-based drop-diameter distribution (Equation 24) and measurements was amplified by the non-physical behavior of the mathematical distribution at the space origin: as mentioned earlier, this distribution doesn't reach zero when $D = 0$. Sellens eliminated this drawback in the following investigation.

Sellens [21] completed the approach he developed with Brzustowski by including a better description of the liquid sheet before the breakup stage and by reconsidering the set of constraints. As reported by many experimental observations, flat liquid sheets are subject to a growing undulation before breaking up. This undulation is a Kelvin-Helmholtz instability resulting from the interaction between the liquid and gas and is often modeled as a sinusoid with a given wavelength and a growing amplitude (see [4]). Since Sellens focused the MEF application in the breakup region, the description of the initial state had to account for the undulated nature of the liquid sheet. To achieve this, Sellens introduced two components of velocity in his formulation; one along the mean motion of the liquid sheet U_x, and one perpendicular to the sheet U_y. Thus, the solution space had three dimensions and $d\Psi = dD dU_x dU_y$. Basing the parameter non-dimensionalization on the mean streamwise sheet velocity U_s and the mass mean diameter D_{30}, the set of constraints required by the problem was the following:

- Normalization:

$$\iiint_{\Psi} f d\delta du_x du_y = 1 \tag{32}$$

- Mass conservation:

$$\iiint_{\Psi} f\delta^3 d\delta du_x du_y = 1 \tag{33}$$

- x Momentum conservation:

$$\iiint_{\Psi} f\delta^3 u_x d\delta du_x du_y = u_{s\,x} + S_{mv\,x} \tag{34}$$

- y Momentum conservation:

$$\iiint_{\Psi} f\delta^3 u_y d\delta du_x du_y = u_{s\,y} + S_{mv\,y} \tag{35}$$

- Kinetic energy conservation:

$$\iiint_{\psi} f\delta^3 \left(u_x{}^2 + u_y{}^2\right) d\delta du_x du_y = \overline{u_{s\,x}{}^2 + u_{s\,y}{}^2} + S_{ke} \tag{36}$$

- Surface energy conservation:

$$\iiint_{\psi} f\delta^2 d\delta du_x du_y = \frac{1}{3\tau} + S_s = \delta_{20}{}^2 = \frac{1}{\delta_{32}} \tag{37}$$

where the index *s* indicates a liquid sheet characteristic. Note that two momentum constraints, one for each direction, were written. Contrary to the previous formulation (Equation 19) we see that the mass source term does not appear explicitly in the mass constraint (Equation 33). This is because the reference mass used for the non-dimensionalization process, i.e., $\rho_L \pi D_{30}{}^3/6$, is a characteristic of the droplets (the final stage) not of the liquid sheet (the initial stage). The right hand side of Equation 33 is actually $\delta_{30}{}^3 = 1$. This new formulation is more appropriate than the previous one. Note also that, as for the mass conservation, the surface energy conservation (Equation 37) might be expressed as a function of a mean drop-diameter of the spray.

Sellens pointed out the necessity of adding a supplementary constraint because of the low contribution of the small drop diameter to the moments expressed by Equations 32–37. Indeed, the lowest order moment of drop size is the second order (Equation 37) and the set of constraints has very little effect on the nature of the distribution of small drops. Therefore, the resulting probability associated with the smaller drop sizes is much larger than is generally observed, i.e., when $D = 0$, $f_n(D)$ is not equal to zero. To solve this problem, Sellens introduced a supplementary constraint that limits the liquid surface area to volume ratio of the entire spray. This new constraint, called the partition constraint, is given by:

$$\iiint_{\psi} f\delta^{-1} d\delta du_x du_y = K_p \tag{38}$$

where K_p expresses the strength of the partition constraint. In agreement with Equation 17, the set of constraints Equations 32–38 led to the following solution:

$$f = exp\left(-\lambda_0 - \lambda_1\delta^2 - \lambda_2\delta^3 - \lambda_3\delta^3 u_x - \lambda_4\delta^3 u_y - \lambda_5\delta^3\left(u_x^{\ 2} + u_y^{\ 2}\right) - \lambda_6\delta^{-1}\right) \tag{39}$$

This solution involves two supplementary parameters compared to Equation 23 which correspond to the number of supplementary constraints added in the new formulation. The difficulty in applying this model lies in the prior determination of the right hand sides of the constraints. As a first approach Sellens proposed evaluating these quantities from experiments. The experimental work considered water sprays produced by swirl atomizers. Swirl atomizer produces a thin conical liquid sheet whose disintegration is structured by the development of a sinusoidal Kelvin-Helmholtz instability. Analyzing liquid sheet visualizations allowed the velocity components u_{sx} and u_{sy} to be determined. The sprays were analyzed with a PDT. The measurements were performed at some distance from the breakup plane and at one location only for each operation condition. The application of the model for comparison purpose made use of the mean diameters D_{30} and D_{32} reported by the measurements. The parameter S_{ke} and K_p were determined in order to fit the experimental data and the two momentum source terms (S_{mx} and S_{my} in Equations 34 and 35) were taken equal to 0. Thus, enough information was introduced in the formulation to allow the 6 parameters in Equation 39 to be determined. As far as the drop-size distributions $f_n(\delta)$ were concerned, the agreements were acceptable: the right tails were well reproduced, the peak diameters were slightly underestimated and the calculated left tail slightly stiffer. Agreements were also observed for the droplet mean velocity and velocity variance except for the small drops for which both the mean and the variance were overestimated by the model. This was explained by the fact that, at the measurement location, small droplets had been significantly influenced by drag forces. The model doesn’t take this behavior into account. The agreement reported for the drop-diameter distribution might be surprising since measurements were local and did not considered the all spray produced by the liquid sheet as described by the model. This could be due to a perfect spatial homogeneity of the drop-size distribution, but, as reported by many experimental works performed on swirl atomizer sprays, we know that this is not the case. The agreement reported by Sellens is likely to be due to the fact that most of the information required by the model was derived from experiments. In other words, the maximum entropy distribution was forced to have identical characteristics as the measured distribution. To the least, this demonstrates that the maximum entropy distribution obtained by Sellens [21] has a good propensity to represent spray drop-size distributions. Finally, Sellens [21] investigated the influence of some source terms on the maximum entropy distribution. He first noted that the introduction of a more realistic undulating sheet model avoided to impose arbitrary non-zero momentum source term to obtain realistic velocity distributions as it was the case for Sellens and Brzutowski's formulation. Second, he found that the kinetic energy source term mainly affects the droplet mean velocity and has a negligible influence on the drop-diameter distribution and that the mean diameter δ_{32} (surface energy source term) and the partition coefficient influence mainly the drop-diameter distribution with a negligible impact on the mean velocity. This indicates that each constraint has a targeted influence on one component of the solution space contrary to what Li *et al*. [18] reported concerning Li and Tankin's solution (Equation 30). This aspect of Sellens' solution will be exploited later by the author.

Li *et al*. [22] and Li and Tankin [23] compared their maximum entropy two-dimensional distribution (Equation 30) with measured distributions. Water sprays were produced by a swirl atomizer. Visualizations of the hollow cone were analyzed in order to locate the liquid sheet breakup plane and the more appropriate location to perform drop diameter and velocity measurements. The breakup plane located at 7.5 mm from the nozzle and the spray characteristics were measured at 10 mm from the nozzle, distance at which all droplets were formed. A two-component PDT was used: this equipment performs a local measurement of the drop diameter as well as two velocity components. Contrary to Bhatia and Durst [19] and Sellens [21], Li *et al*. performed measurements as a function of the radial position of the measuring volume. After having checked the axisymmetry of the mean axial and mean tangential velocities and of the Sauter mean diameter, a global integrated joint size-axial velocity distribution was constructed from the individual point measurements, weighting each local measurement by their time of collection and the ratio of their optical probe area to the ring area represented at that location. The set of constraints of Li and Tankin's model (Equations 18–20, 29) requires some information to allow the maximum entropy distribution (Equation 30) to be determined. The liquid sheet mean axial velocity at the nozzle exit U_0 is determined from the experiment as well as the mass median diameter D_{30}. Thus, the number We is determined. The mass source term in Equation 19 is taken equal to zero. The surface area increases between the nozzle exit and the drop formation. On the other hand the kinetic energy decreases between the nozzle exit and breakup. Thus, Li *et al*. [22] estimated the energy source term S_e of the combined energy constraint (Equation 29) to zero. The momentum source term S_{mv} was estimated using a drag model where the conical sheet is unfolded into a triangular shape. This model yielded a value for S_{mv} of –0.017. The number-based drop-diameter distribution $f_n(\delta)$ obtained from the integration of the joint drop diameter-velocity distribution (Equation 30) over the velocity space agreed reasonably well with measurements: the peak diameter was well estimated as well as the whole distribution width. However, although the predicted mean velocity was in agreement with the measurements for large drop-diameter, it was considerably overestimated for small diameter drops. Similar to the results found by Sellens [21], this behavior was due to a lack of a drag model in the prediction of the drop-size and velocity distribution. Li *et al*. [22] completed their MEF model with a drag model to incorporate the influence of the air on the droplets between the plane of droplet production (7.5 mm) and the plane of measurement (10 mm). This drag model that considered each droplet individualy considerably improved the results. Therefore, contrary to Bhatia and Durst [19], this application demonstrated the ability of Li and Tankin's solution to represent spray drop-diameter and velocity distribution. Li and Tankin [23] emphasized that the pejorative results reported by Bhatia and Durst [19] as far as their maximum entropy distribution was concerned was due to the use of local drop-diameter and velocity distribution measurements and to the absence of any drag model to compensate the fact that measurements are never performed were the droplets are produced.

Chin *et al*. [24] reconsidered Li and Tankin's approach [16] to predict the volume and velocity distribution ($d\Psi = dVdU$) of the drops produced by a cylindrical liquid jet subject to a Rayleigh breakup process. This well-known process has been described in many references (see [1,2] for instance). The Rayleigh breakup process is a capillary instability that occurs on a small velocity liquid column. The characteristic features of this process are the growth on a sinusoidal perturbation on the liquid column and the production of droplets with roughly the same diameter of the order of twice the initial jet diameter. According to the velocity of the jet, much smaller droplets (called satellite drops) may be produced between two main droplets. Following Li and Tankin [16], the control volume extended from the nozzle exit down to a plane where all droplets were formed. However, Chin *et al*. [24] reconsidered the used of a single energy constraint in Li and Tankin's approach by noting that when a single constraint is used no information is provided to how the total energy source is distributed between the kinetic energy and the surface energy. Thus, any combination of constant total energy source will result in the same probability density function. In consequence, as recommended by Sellens [21], Chin *et al*. [24] used separate constraints for kinetic energy and surface energy. Using the jet velocity U_{jet} at the nozzle exit and the mass mean drop-diameter D_{30} to calculate the dimensionless parameters, the normalization, mass conservation, momentum conservation and kinetic energy conservation constraints were similar to Equations 18–21, respectively (without the explicit appearance of the source term S_m as in Equation 33) and the surface energy conservation was written as:

$$B\iint_{\Psi} f\delta^2 d\delta du = \frac{2B}{3\delta_{jet}} + S_s \tag{40}$$

where B is given by Equation 31. Then, following Li and Tankin's mathematical manipulations [16], the final diameter-velocity distribution they obtained was:

$$f = 3\delta^2 \, exp\left(-\lambda_0 - \lambda_1\delta^3 - \lambda_2\delta^3 u - \lambda_3\delta^3 u^2 - \lambda_4 B\delta^2\right) \tag{41}$$

As in Equations 27 and 30, $f \propto \delta^2$ indicates that the change of variable expressed by Equation 26 has been performed. This solution is identical to Equation 30 except that $\delta^3 u^2$ and $B\delta^2$ depend on their own parameter. Chin *et al*. [24] derived analytical expressions for each source term included in the constraints (except for S_m that was taken equal to zero) assuming that all droplets produced by a Rayleigh instability had the same diameter D_{30}. They obtained:

$$S_{mv} = u_m - 1 \qquad S_{ke} = \left(u_m^{\,2} + u_{rms}^{\,2}\right) - 1 \qquad S_s = \frac{B}{\delta_{32}} - \frac{2B}{3\delta_{jet}} \tag{42}$$

where u_m and u_{rms} are the mean drop velocity and the root mean square droplet velocity fluctuation, respectively. Performing PDT measurements and using the measured values for D_{30}, D_{32}, u_m and u_{rsm} as input information for the model, the authors qualified the agreement between the estimations and the measurements as reasonably good: they underlined discrepancies between the peak heights of the estimated distributions and of the experimental data. They also demonstrated that the use of a combined energy constraint, which leads to the solution given by Equation 30, reported a prediction in strong disagreement with the number-based drop-size distribution $f_n(\delta)$.

Based on Sellens'observations [21], Ahmadi and Sellens [25] emphasized that the constraints on momentum and kinetic energy (Equations 20 and 21) in the spray carry only velocity information, i.e., they have a negligible influence on the number-based drop-size distribution $f_n(\delta)$. It is therefore possible to consider drop size and velocity separately. They developed a simplified MEF model to predict the number-based drop-diameter distribution ($d\Psi = dD$) using the normalization, the mass conservation, the surface energy conservation and the partition constraints (Equations 18, 19, 22 and 38), which, in dimensional form, can be rewritten as:

$$\begin{cases} \int_0^\infty f_n(D)dD = 1 \\ \int_0^\infty f_n(D)D^3dD = D_{30}{}^3 \\ \int_0^\infty f_n(D)D^2dD = \dfrac{D_{30}{}^3}{D_{32}} \\ \int_0^\infty f_n(D)D^{-1}dD = \dfrac{1}{D_{-10}} \end{cases} \tag{43}$$

leading to the following number-based drop-diameter distribution:

$$f_n(D) = exp\left(-\lambda_0 - \lambda_1 D^2 - \lambda_2 D^3 - \lambda_3 D^{-1}\right) \tag{44}$$

Using the mean diameters D_{30}, D_{32} and D_{-10} from the experiments, they reported good agreement with experimental distributions of water sprays produced by a swirl atomizer they measured with a PDT as well as those provided by Li and Tankin [15] and Bhatia and Durst [19]. In each situation the peak diameter and distribution width were well estimated. Ahmadi and Sellens [25] furthermore observed that the agreement obtained with this three-parameter distributions (Equation 44) is as good as the one obtained with the four-parameter log-hyperbolic distribution. They concluded that three moments are required as input information to predict a spray drop-diameter distribution, namely, D_{-10}, D_{30} and D_{32} and that the advantage of this model compared to empirical distributions such as log-hyperbolic distribution for instance, is that the parameters in the MEF distribution are directly linked to a physical model. Finally, since Ahmadi and Sellens [25] succeeded in representing experimental data—and in particular those published by Bhatia and Durst [19]—with their maximum entropy distribution as well as with the one due to Li and Tankin [16], they concluded that Bhatia and Durst misapplied the distribution function.

The investigations due to Sellens' group [12,14,21,25] and to Li and Tankin's group [15,16,24] have motivated developments and applications of models based on the MEF to predict liquid spray drop-diameter distributions. Van der Geld and Vermeer [26] proposed a model to predict the diameter distribution of satellite droplets produced during the breakup of a cylindrical liquid column, the main droplets being distributed according to a Gaussian distribution. This model is based on three constraints, namely, the normalization, the surface energy conservation and the mass conservation constraints and reports quite realistic results with the prediction of bi-modal diameter distributions as often reported in the literature. However, in this approach the MEF was used to account for a part of the atomization process only—the satellite formation—and requires the prior knowledge of the main-drop distribution.

Bi-modal drop-diameter distributions were also obtained by Chin *et al*. [27] who generalized Chin *et al*. [24] formulation by accounting for the three components of drop velocity, i.e., $d\Psi = dVdU_x dU_y dU_z$. Following Sellens' approach [21], three momentum conservation constraints were written, one for each component (see Equations 34 and 35), leading to a solution similar to Equation 41 but introducing seven parameters. Comparisons between this new solution and PDT measurements performed over the whole spray and reorganized as a global distribution following Li *et al*.'s procedure [22] reported an acceptable fit on the number-based drop-diameter distribution of a spray produced by a swirl atomizer. To apply their model, Chin *et al*. [27] used as much experimental information as necessary to estimate the source terms. They demonstrated that increasing the number of velocity components in the model considerably improved the agreement (especially along the right distribution tail) and that the use of the generalized three velocity-component model predicted a bi-modal distribution, the enhancement of bimodality being controlled by the azimuthal momentum source term.

Chin *et al*. [28] proposed another extension of Chin *et al*.'s approach [24] to model the production of main and satellite drops produced by a cylindrical liquid jet subject to a Rayleigh instability. The formulation was conducted in the volume-velocity space ($d\Psi = dVdU$) taking one component of velocity into account but introducing two supplementary constraints, one related to the surface area to volume ratio of small drops and the other related to the non-sphericity of main droplets. These two constraints were respectively written as:

$$\begin{cases} \iint_{\Psi} \frac{f}{\delta} d\delta du = C_{-1} \\ \iint_{\Psi} f\delta d\delta du = C_{+1} \end{cases} \tag{45}$$

and led to the following 7-parameter diameter-velocity distribution:

$$f = 3\delta^2 exp\left(-\lambda_0 - \lambda_1\delta^3 - \lambda_2\delta^3 u - \lambda_3\delta^3 u^2 - \lambda_4 B\delta^2 - \lambda_5\delta^{-1} - \lambda_6\delta\right) \tag{46}$$

It is interesting to note here that one of the two constraints (first equation in Equation 45) is identical to Sellens' partition constraint (Equation 38) and is therefore appropriate to limit the small drop production. The second additional constraint is actually identical to the definition of the D_{10} mean-diameter. This new formulation allowed bi-modal distributions to be obtained. Considering this approach and the one due to Chin *et al*. [27], it seems that a minimum of seven parameters is required if one has to represent bi-modal drop-size distribution. Chin *et al*. [28] performed an experiment where droplets produced by a liquid column were visualized by a high-speed camera (1,500 images/s). The analysis of the images allowed the measurement of an equivalent diameter (based on the volume conservation) and of the velocity of each drop. They investigated a jet at a flow rate appropriate to enhance the satellite drop production. The bi-modal drop-diameter distribution they measured was satisfactorily represented by the maximum entropy distribution given by Equation 46 (both peak diameters and the width of each distribution mode were well estimated). Note that in this application, as in the one presented by Chin *et al*. [27], the information required to determine the seven parameters were deduced from the experiments. Thus, these applications did demonstrate the ability of the mathematical solution to fit experimental drop-diameter distribution but did not fully validate the model.

As mentioned above, the specification of the information required by the constraints, i.e., the moments $\langle g_r \rangle$ in Equations 9 and 15, constitutes a critical step to the application of the MEF model to predict spray drop-diameter and velocity distributions. This point is crucial since, as demonstrated by van der Geld and Vermeer [26], inappropriate values of these moments can lead to unrealistic distributions. The determination of the moments introduced by the constraints must be based on a physical description of the atomization process. In this respect, one should mention Mitra and Li's attempt [29] in developing a deterministic sub-model to derive the Weber number as well as the mass, momentum and energy source terms required by Li and Tankin's solution (Equation 30) to model the atomization of a flat liquid sheet. To be applicable, Mitra and Li's model requires the detailed knowledge of the breaking sheet including its thickness, its length, the wavelength of the perturbation controlling the disintegration, the initial amplitude of this perturbation, as much as information it is usually difficult to know with a sufficient accuracy. Thus, the problem of the determination of the moments is shifted but not solved.

Dobre and Bolle [30] considered also this point in the application of their model. They formulated a MEF model to predict drop-diameter distributions of sprays produced by ultrasonic atomizers. These atomizers produce a spray by making vibrate the free surface of a liquid film at an ultrasonic frequency. Their model was based on the normalization, mass conservation and energy (surface and kinetic) conservation constraints. The resulting number-based drop-size distribution suffers from the same drawback as Sellens and Brzustowski's solution and presents an unrealistic behavior at the small drop-diameter distribution tail. This aspect was not too penalizing in their work since Dobre and Bolle [30] performed measurements with a laser diffraction technique (LDT). Contrary to the PDT, the LDT is a line-of-sight measurement technique: the measuring volume is a laser beam and the measurement is space integrated. As explained by Dodge *et al*. [20], this technique is a spatial sampling instrument, such as photographic, and registers signals proportional (in one dimension) to spatial frequency (counts/m). Thus, the nature of the information provided by LDT is different than the one provided by PDT. Furthermore, LDT reports a volume-based drop-size distribution and Dobre and Bolle conducted their comparison work on this type of distribution using Equation 6 to calculate the volume-based drop-size distribution from the maximum entropy number-based solution. As shown by Equation 6, the unrealistic behavior at small drop-diameter doesn't affect the volume-based distribution too much since small droplets have a small contribution to the total liquid volume. For the model application, Dobre and Bolle [30] deduced an expression of the moment required by the energy constraint. This expression contains information such as the mass mean-diameter and the droplet velocity that are not known a priori. They finally determined the missing information from the solution that ensured the best fit with their measurements. Despite the fact that the left distribution tail and the peak diameter were well estimated, their comparison revealed a low ability of their solution to correctly represent the big drop-diameter distribution tail (right part of the distribution).

The MEF formulations due to Sellens' and to Li and Tankin's groups present another difficulty of application for reasons of mathematical and numerical nature. Indeed, the specific mathematical form of the distributions provided by these formulations (Equations 23, 30, 39, 41 or 46) requires a numerical resolution for the determination of the Lagrangian multipliers λ_i. As demonstrated by van der Geld and Vermeer [26], such numerical resolutions are not easy to perform and become more and more difficult as the number of constraints, i.e., the number of parameters to be determined increases. These observations motivated the development of other MEF formulations.

The model due to Cousin *et al*. [31] made use of Ahmadi and Sellens' conclusions, i.e., 1 – the determination of the drop-diameter distribution doesn't need to go through the calculation of the joint size-velocity distribution, 2 – the drop-diameter distribution can be determined from the writing of constraints expressing mean drop-diameters. Cousin *et al*. [31] proposed a formulation to determine the drop-diameter distribution only. Furthermore, instead of basing the set of constraints on conservation laws as in the previous investigations, they assumed the existence of a single mean drop-diameter D_{q0}, called the constraint diameter, that would carry enough information to predict the drop-size distribution. Thus, Cousin *et al*.'s formulation is based on the following set of constraints:

$$\begin{cases} \int_0^\infty f_n(D)dD = 1 \\ \int_0^\infty f_n(D)D^q dD = {D_{q0}}^q \end{cases} \tag{47}$$

The distribution that satisfies this set of constraints and that maximizes Equation 16 has an analytical expression. Indeed, making use of Equations 12, 13 and 17, it can be shown that the number-based drop-diameter distribution is given by:

$$f_n(D) = q\frac{\left(\frac{1}{q}\right)^{1/q}}{\Gamma\left(\frac{1}{q}\right)}\frac{1}{D_{q0}}exp\left(-\frac{1}{q}\left(\frac{D}{D_{q0}}\right)^q\right) \tag{48}$$

where Γ is the Gamma function. This solution depends on two parameters, namely, the constraint diameter D_{q0} and its order q. Cousin *et al*. [31] proposed also a second formulation to determine the volume-based drop-diameter distribution. In order to ensure a coherent formulation, i.e., the number-based and volume-based drop-diameter distributions satisfy Equation 6, they demonstrated that the entropy to be maximized by the volume-based drop-diameter distribution $f_v(D)$ should be:

$$S_v = -k\int_0^\infty f_v(D)ln\left(\frac{f_v(D)}{U_v(D)}\right)dD \tag{49}$$

where U_v is a volume-based drop-diameter distribution whose corresponding number-based drop-diameter distribution is uniform. Therefore, if no information is available, i.e., no constraint is specified, the maximization of Equation 16 reports a constant distribution $f_n(D)$ and the maximization of Equation 49 reports $f_v(D) = U_v(D)$. The entropy S_v given by Equation 49, referred in the literature as the Bayesian entropy, is a more general expression for the statistical entropy given by Equation 16. It is actually identical to the Kullback-Leibler number (see [13]), which is the measure of the nearness of two distributions. The necessity of adapting the statistical entropy to the type of distribution sought is another manifestation of the fact that, as mentioned in section 2, from a mathematical point of view, only the number-based drop-diameter distribution is a probability density function. Thus, the volume-based drop-size distribution solution of Cousin *et al.*'s formulation [31] is given by:

$$f_v(D) = q\frac{\left(\frac{1}{q}\right)^{4/q}}{\Gamma\left(\frac{4}{q}\right)}\frac{D^3}{{D_{q0}}^4}exp\left(-\frac{1}{q}\left(\frac{D}{D_{q0}}\right)^q\right) \tag{50}$$

It can be checked that Equations 48 and 50 satisfy Equation 6. The analytical nature of the solution provided by this formulation is an advantage compared to the previous formulations since it allows calculations to be performed. For instance, it can be demonstrated that the mean drop-diameter series introduced by Equation 5 is given by:

$$D_{kl} = q^{\frac{1}{q}}D_{q0}\left(\frac{\Gamma\left(\frac{1+k}{q}\right)}{\Gamma\left(\frac{1+l}{q}\right)}\right)^{\frac{1}{k-l}} \tag{51}$$

The volume-based drop-diameter distribution given by Equation 50 succeeded in representing measured distributions in many different situations. For instance, Cousin *et al.* [31] applied it for water sprays produced by swirl atomizers used at low injection pressures. The measurements were performed with a laser-diffraction technique (LDT). Consequently, the comparisons were conducted on the volume-based drop-size distributions. Cousin *et al.* [31] coupled their MEF model with a linear theory that applied to the liquid sheets produced by the swirl atomizers allowed the Sauter mean-diameter D_{32} to be calculated. Using this mean diameter, they determined the constraint diameter D_{q0} with Equation 51 testing several values of the constraint order q. For four different atomizers tested at several injection pressures, they obtained a reasonable agreement between the mathematical and the measured distributions (good prediction of the peak diameter as well as of the distribution width) for a constant value of q, namely, $q = 1$. Therefore, for their specific domain of investigation, the MEF model coupling with the deterministic linear theory provided a fully deterministic model to predict volume-based drop-diameter distributions.

Sindayihebura *et al.* [32] followed an identical approach to derive a model to predict the volume-based drop-diameter distribution of sprays produced by ultrasonic atomizers similar to those used by Dobre and Bolle [30]. Several atomizers were used as well as several liquids and the measurements were performed with a LDT. A stability analysis of the vibrating liquid free surface was used to determine the Sauter mean-diameter. The agreement between the measured and the mathematical distributions was acceptable with a constant constraint order, namely, $q = 3$. And a similar behavior was reported by Dumouchel and Boyaval [33] who investigated gasoline sprays produced by high-pressure swirl atomizer. For the four injection systems used, the best fit was obtained with a constant value of q of the order of 0.54. In their application, Dumouchel and Boyaval used the experimental mean-diameter D_{43} to calculate the constraint diameter. As noted in Section 2 (Table 1), this diameter corresponds to the first moment of the volume-based drop-diameter distribution.

These results induced the enthusiastic idea that each atomization process could be characterized by a specific value of q, giving credit to the information status of this parameter. Dumouchel and Boyaval [33] demonstrated that the relative span factor of the volume-based drop-diameter distribution given by Equation 50 depends on the parameter q only. The relative span factor measures the relative width of the distribution (see [1,2]). Thus, if liquid atomization processes are to be related to a constant parameter q, they should be as well characterized by a constant relative distribution width. This point has never been clearly established so far.

Dumouchel and Boyaval [33] reported another result that is worth mentioning here. They proposed another MEF formulation to determine volume-based drop-size distribution but based on four constraints expressing the four first moments of the distribution, namely, the four mean-diameters D_{43}, D_{53}, D_{63} and D_{73}. The resulting four-diameter distribution could be therefore written under the following form:

$$f_v(D) = U_v(D) exp\left(-\lambda_0 - \lambda_1 D - \lambda_2 D^2 - \lambda_3 D^3 - \lambda_4 D^4\right) \quad (52)$$

For one water spray produced by a swirl atomizer and measured by a LDT they compared the ability of Equations 50 and 52 to represent the experimental distribution. In Equation 50, the parameter q and D_{q0} were determined on the basis of the relative distribution width and the measured D_{43}, and, the application of Equation 52 was achieved by using the four experimental mean-diameters D_{43}, D_{53}, D_{63} and D_{73}. It was observed that the two mathematical distributions offer a different fit. The four-parameter distribution (Equation 52) showed a better fit for the small drop population whereas the two-parameter distribution (Equation 50) provided a better fit from the distribution peak to the large drop population. As mentioned by Kapur [13] this result shows that in the MEF application the quality of information is more important than the number of information introduced in the procedure. In other words, the information provided by the relative distribution width and its arithmetic mean is sufficient to predict a volume-based drop-size distribution. This result was used by Boyaval and Dumouchel [34] in a subsequent work to predict volume-based drop-diameter distributions in situations where the relative distribution width and the mean-diameter D_{43} could be measured only. This work demonstrated to which extend the MEF model can be used to provide a completed spray characterization from partial experimental information.

However, the solution of Cousin *et al*. [31] (Equations 48 and 50) leaves us with a problem: similar to the maximum entropy distribution provided by Sellens and Brzustowski [12,14] (Equation 24), the probability associated with the smaller drop diameters is much larger than is generally observed, i.e., the number-based drop-size distribution doesn't decrease towards zero when D approaches zero. This limitation is not too penalizing when using the volume-based drop-diameter distribution since the small drops represent a rather negligible proportion of the liquid volume. This is mathematically illustrated in Equation 50 by a distribution proportional to D^3. However, this unrealistic behavior of the number-based distribution prevails applying the volume-based drop-diameter distribution to characterize spray with a minimum diameter different than zero. This problem was overcome by Dumouchel and Malot [35] who wrote the previous solution (Equation 50) in a new coordinate system where the abscissa axis had been shifted of the quantity D_{min} corresponding to the minimum drop diameter in the spray. The new volume-based drop-size distribution, expressed in terms of D_{43} instead of D_{q0}, is given by:

$$f_v(D) = q\frac{\Gamma^4\left(\frac{5}{q}\right)}{\Gamma^5\left(\frac{4}{q}\right)}\frac{\left(D - D_{min}\right)^3}{\left(D_{43} - D_{min}\right)^4} exp\left(-\frac{\Gamma\left(\frac{5}{q}\right)}{\Gamma\left(\frac{4}{q}\right)}\left(\frac{D - D_{min}}{D_{43} - D_{min}}\right)^q\right) \quad (53)$$

which is a three-parameter distribution, i.e., q, D_{43} and D_{min}. This new mathematical distribution provided a very good fit of sprays produced by low-velocity cylindrical liquid jet, such sprays being characterized by a non-zero minimum diameter [36]: the minimum diameter, the peak diameter and the distribution width were well estimated. It allows also the representation of ultrasonic sprays to be improved [37]. Furthermore, the most enthusiastic aspect of this new volume-based drop-diameter distribution is that the corresponding number-based drop-diameter distribution calculated from Equation 6 decreases towards zero when the diameter approaches D_{min} and the non-physical behavior at small drop diameters is no longer observed. Thus, in agreement with Ahmadi and Sellens [25], the authors concluded that the determination of spray drop-size distribution required at least three parameters, one of which being related to the small-drop population.

However, as far as the MEF is concerned, the solution (Equation 53) obtained by shifting of the initial volume-based drop-size distribution is not satisfactory. The combination of Equations 6 and 53 reports a number-based distribution proportional to $(D - D_{min})^3/D^3$ which forces $f_n(D)$ to decrease towards zero when D approaches D_{min} (with $D > D_{min}$). However, in the absence of information, i.e., if no constraint is specified, the number-based drop-size distribution $f_n(D)$ is not constant anymore, which contradicts the formalism. This indicates that this distribution is not the one that maximizes the statistical entropy but is a biased solution. Another way of demonstrating this consists in calculating the number-based distribution from the shift of the surface-based distribution instead of the volume-based distribution. The resulting number-based distribution would be proportional to $(D - D_{min})^2/D^2$ instead of $(D - D_{min})^3/D^3$. This is due to the fact that the number, surface and volume of droplet are not similarly distributed in the diameter space. In such conditions, the MEF doesn't tolerate variable change as the invariance of the statistical entropy is not ensured.

As illustrated by the different approaches presented in this review, the behavior of the number-based drop-size distribution's tail at small drop diameter has always been a major point in MEF formulation. Sellens [21] explained that the unrealistic behavior of the number-based drop-size distribution he obtained with Brzustowski (Equation 23) at small drop diameter was due to the fact that the momentum, kinetic energy and surface energy constraints used in their formalism did not carry any information on the small drop diameter. To overcome this, problem, Sellens [21] introduced a supplementary constraint, the partition constraint (Equation 38) whose effect is to limit the surface area/volume ratio of the whole spray.

Li and Tankin [15,16] circumvented this problem: they formulate their approach in the volume-velocity solution space and performed a change of variable to end up in the diameter-velocity solution space. This change of variable, described by Equation 26, imposes the distribution to be proportional to $3D^2$, which enforces the distribution to report the expected behavior at small drop-diameter. However, such a procedure is inconsistent with the MEF. Indeed, the exact statistical entropy of a continuous probability density function *f(x)* is (see [38]):

$$S_{cont} = -k\int f(x)ln\left(\frac{f(x)}{m(x)}\right)dx \quad (54)$$

where *m(x)* is a measure of the solution space (Borel-Lebesgue measure). If the reachable states are equally distributed in the solution space, i.e., if the density of states is constant over the solution space, this measure can be taken equal to 1. Equation 54 becomes then identical to Equation 16. The measure *m(x)* assures the invariance of the entropy under transformations such as variable changes. Therefore, as mentioned by van der Geld and Vermeer [26], this measure should have been taken into account by Li and Tankin in order to satisfy Jaynes consistency principle that reads "Two problems with the same relevant physical information show the same pdf's". The formulation due to Li and Tankin [16] and applied in many other investigations, does not satisfies this principle. To demonstrate this, let us imagine writing Li and Tankin's formulation in the drop surface area-velocity solution space, instead of the drop volume-velocity solution space, on the basis of the same conservation laws (mass, momentum and energy). The final variable change required to go back in the drop diameter-velocity solution space would report a distribution proportional to $2D$ instead of $3D^2$. We see that, despite using the same information, Li and Tankin's formulation can lead to several solutions according to the initial solution space in which the approach is formulated. In consequence, the solution provided by Li and Tankin [16] is not the one with the maximum statistical entropy but is a biased solution. This problem is equivalent to the one discussed above and concerning the solution provided by Dumouchel and Malot [35] (Equation 53) and comes from the fact that the number, surface and volume of a drop don't have identical density in the solution space.

It is interesting to mention here the formulation due to Kim *et al*. [39] that proposed a different approach to deal with the problem of the small diameter drops. They used the same system of constraints as the one formulated by Li and Tankin [16] and determined the distribution that satisfied this set of constraint and maximized the Bayesian entropy given by:

$$S_B = -k\iint_{\Psi} f(\delta,u)ln\left(\frac{f(\delta,u)}{f_0(\delta)}\right)d\delta du \quad (55)$$

where $f_0(\delta)$ is a prior number-based drop-diameter distribution that had to be specified. The solution of this formulation is given by:

$$f(\delta,u) = f_0(\delta)exp\left(-\lambda_0 - \lambda_1\delta^3 - \lambda_2\delta^3 u - \lambda_3\left(\delta^3u^2 + B\delta^2\right)\right) \quad (56)$$

Considering the atomization of a flat liquid sheet, Kim *et al*. [39] related the prior-distribution to the linear growth rate of the unstable waves. This approach imposed a minimum drop diameter much greater than the one expected. Kim *et al*. argued that this problem came from the fact that linear theory ignored the satellite drop formation. To overcome this problem, they imposed the prior-distribution $f_0(\delta)$ to evolve as $m\delta^2$ in the small drop diameter region without giving any physical justification for that. This characteristic of the prior-distribution forces the distribution to decrease towards zero when δ tends towards 0, and comparisons between measured and calculated distributions revealed acceptable agreement as far as the peak diameter and the distribution width are concerned. We can note that, as for

Li and Tankin's solution (Equation 30), the distribution given by Equation 56 is proportional to δ^2. Therefore, the solutions given by Equation 30 and Equation 56 are likely to be same if $m = 3$.

Dumouchel [40] proposed an extension of Cousin *et al.*'s formulation [31] addressing the question of the small drop diameter distribution. This extension was inspired by Griffith [41] and Lienhard and Meyer [42]. The most objective distribution that satisfies the constraint introduced by Cousin *et al.* (Equation 47) is the one that maximizes the statistical entropy (Equation 16). This distribution, given by Equation 48, is based upon the implication that a given particle had just as good a chance of being produced as any other, regardless of its size. However, as explained by Griffith [41] in the theory on the size distribution of particles in a comminuted system, there are a priori reasons to believe that particles of certain sizes are more likely to be produced than others. Generally speaking, this means that diameter classes should not be, a priori, equivalently reachable. Dumouchel [40] exploited this idea in the calculation of liquid spray number-based drop-size distribution. An a priori probability, $g(D)$, was introduced to represent the likelihood that a particle class of drop would be reached. This probability was expressed by:

$$g(D) = AD^{\alpha-1} \tag{57}$$

where A is assumed to be a constant and α, a parameter to be determined. Using statistical mechanics, Dumouchel [40] demonstrated that the most objective distribution that satisfies the set of constraints Equation 47, when the probability of reaching a diameter D is given by $g(D)$ (Equation 57) is the one that maximizes the statistical entropy given by:

$$S = -k\int_0^\infty f_n(D)\, ln\left(\frac{f_n(D)}{g(D)}\right)dD \tag{58}$$

Note that this entropy is similar to the one maximized by Kim *et al.* [39]. The solution of this formulation is given:

$$f_n(D) = \frac{q}{\Gamma\left(\frac{\alpha}{q}\right)}\left(\frac{\alpha}{q}\right)^{\frac{\alpha}{q}}\frac{D^{\alpha-1}}{D_{q0}{}^{\alpha}}\, exp\left(-\frac{\alpha}{q}\left(\frac{D}{D_{q0}}\right)^q\right) \tag{59}$$

This solution introduces three parameters, i.e., two coefficients q and α that control the shape of the distribution, and the constraint diameter D_{q0} that positions the distribution in the diameter space. Note that for $\alpha = 1$, i.e., $g(D) = \text{Cte}$, Equation 59 is identical to Cousin *et al.*'s solution (Equation 48). It is referred in the literature as the three-parameter Generalized Gamma function [42]. Dumouchel [40] demonstrated that it is identical to the empirical Nukiyama-Tanasawa distribution, which is an empirical distribution often used in the literature to represent liquid spray drop-diameter distribution. Paloposki [17] identified the permissible range of the parameters q and α of the Nukiyama-Tanasawa distribution in order to allow any drop-diameter distribution type (number-based, length-based, surface based and volume-based) to be physically representative. This permissible range defines two regions in the (q, α) space, namely:

$$\begin{cases} q > 0 \quad \text{and} \quad \alpha > 1 \\ \text{or} \\ q < 0 \quad \text{and} \quad \alpha < -3 \end{cases} \tag{60}$$

In particular, when the parameters q and α satisfy Equation 60, the number-based drop-diameter distribution always reports the expected behavior at small diameter drops. Dumouchel [40] pointed out the role of each equation introduced in the formalism. When the parameters q and α are positive (first case in Equation 60), the constraint expressed by the second equation of Equation 47 ensures the existence of a maximum diameter, and Equation 57 ensures the existence of a minimum diameter. In the second case (q and α negative), a maximum drop-diameter is imposed by Equation 57 whereas the D_{q0} constraint ensures the existence of a minimum diameter. As for Cousin *et al.*'s solution, the analytical nature of the solution is interesting and allows calculations to be performed. Dumouchel [40] reported the expressions of the other drop-diameter distribution types as well as their modal-diameters and the mean-diameter series. Furthermore, the fact that this solution is identical to the Nukiyama-Tanasawa distribution ensures a good capability of representing liquid spray drop-diameter distribution. Indeed, exploring the potentiality of several empirical distributions to represent experimental drop-size distributions, Paloposki [9] found that the Nukiyama-Tanasawa distribution is one of the best. However, he pointed out a problem of parameter stability that manifests by drastic variations of their values for reasonable changes in the initial conditions. This behavior, reported for parameters q and α, questions the validity of these parameters as far as their physical meaning is concerned.

This very point was addressed by Lecompte and Dumouchel [43] who identified a characteristic feature of the three-parameter Generalized Gamma function. This distribution is almost insensitive to a modification of the parameters q and α (the third parameter D_{q0} being kept constant whatever the value of q) if these parameters correlate as:

$$|\alpha||q|^n = \text{constant} \tag{61}$$

Thus, if several spray drop-diameter distributions have parameters q and a satisfying Equation 61, they can be satisfactorily represented by a unique couple (q; α). Lecompte and Dumouchel [43] applied the Generalized Gamma function to represent measured drop-diameter distributions produced by ultrasonic atomizer, compound atomizer and a twin-fluid atomizer. In each situation, the set of parameters (q, α, D_{q0}) ensuring the best fit was determined. The working conditions are summarized in Table 2. This table indicates the measurement technique used for each situation. When LDT is used, the comparison procedure was based on the volume-based drop-size distribution. The Scanning Mobility Particle Sizer is a particle counter like the PDT. Thus, when these measurement techniques are used, comparisons were conducted on the number-based drop-diameter distribution. For all situations, the agreement between the measured and the mathematical distributions were of very good quality: the minimum, peak and maximum diameters as well as the width and height of the distributions were very well estimated. Furthermore, an analysis of the results that took into account the insensitivity of the Generalized Gamma distribution if Equation 61 is satisfied reported that in each situation, one or two parameters were constant whereas the remaining parameters reported a clear dependency with the working conditions. These results are summarized in Table 2. Note that, as obtained by the application of Cousin *et al*.'s model, the parameter q is found to be representative of the atomizer, i.e., of the liquid atomization process. All these results led the authors to conclude that the set of constraints given by Equations 47 and 57 contain enough information to correctly predict drop-diameter distribution and that the parameters introduced by these constraints are physically representative.

Table 2. Summary of the different situations analyzed by Lecompte and Dumouchel [43] (MT: measuring technique, LDT: laser diffraction technique, PDT: Phase Doppler Technique, SMPS: Scanning Mobility Particle Sizer, f: ultrasonic atomizer frequency, σ: surface tension, ρ_L: liquid density, Q_v: liquid volume flow rate, ΔP_i: injection pressure, P_a: air pressure).

Atomizer and Working Conditions	MT	Parameters		
		q	α	D_{q0}
Ultrasonic atomizer ▪ $f \in$ [41 kHz; 135 kHz] ▪ $\sigma \in$ [31 mN/m; 73 mN/m] ▪ $\rho_L \in$ [900 kg/m^3; 1000 kg/m^3] ▪ $Q_v \in$ [0.4 l/h; 1 l/h]	LDT	–1	–23.6	$= \left(0.86 \dfrac{\sigma}{\rho_L f^2}\right)^{1/3}$
Compound injector ▪ Fluid: Ethanol ▪ $\Delta P_i \in$ [0.2 MPa; 0.5 MPa]	LDT	0.2	$\propto \Delta P_i^{-0.7}$	$\propto \Delta P_i^{-1.7}$
Twin-fluid atomizer (internal mixing) ▪ Fluid: Normafluid ▪ $P_a \in$ [1 MPa; 6 MPa] ▪ $Q_v \in$ [0.3 ml/mn; 6 ml/mn]	SMPS PDT	–0.3	–6.3	$\propto P_a^{(0.03Qv-0.2)}$

In agreement with Ahmadi and Sellens' result (Equation 44), the solution provided by Dumouchel's formulation (Equation 59) indicates that three parameters are sufficient to represent liquid spray drop-size distributions. However these solutions have a limitation: they are mono-modal distributions. In other word, they are not adapted to represent bi-modal distributions. Bi-modal liquid spray drop-diameter distributions have been reported in the literature (see [26,28] for instance). Yongyingsakthavorn *et al*. [44] also reported bi-modal volume-based drop-diameter distributions for sprays produced by high injection pressure gasoline injector. Their measurements, performed with a LDT for a wide range of injection pressures, were satisfactorily fitted by a combination of two three-parameter Generalized Gamma functions, i.e.:

$$f_v(D) = (1-\beta) f_{v1}(D) + \beta f_{v2}(D) \tag{62}$$

This distribution depends on seven parameters: (q_1, α_1, $(D_{q0})_1$) for f_{v1}, (q_2, α_2, $(D_{q0})_2$) for f_{v2} and the blending parameter β that is comprised between 0 and 1. Furthermore, as for the applications reported by Lecompte and Dumouchel [43], the parameters were found to organize very well with the working conditions. In particular, for all tested injection pressures, it was found $q_1 = q_2 = -0.13$, $\alpha_1 = \alpha_2 = -21.5$ and the three remaining parameters were found dependent of the injection pressure.

To complete this review, a different approach proposed by Li *et al*. [45] should be mentioned. According to those authors, the MEF, widely used to predict spray drop-size distributions, is, strictly speaking, applicable for isolated systems in thermodynamics equilibrium. Therefore, it is not physically consistent with real conditions of atomization processes. This motivated the authors to formulate a new model for the prediction of the droplet-size distribution based on the thermodynamically consistent concept—the maximization of entropy generation (MEG) during the spray formation, which is a non-isolated and irreversible process. The approach consists in calculating the entropy generated during the atomization, i.e., between the nozzle exit down to a location where the droplets are formed. The problem is written in the volume solution space ($d\Psi = dV$) and any length is non-dimensionalized by the mass mean-diameter D_{30}. Making the assumption that an atomization process is isothermal and therefore, that the liquid internal energy is unchanged during the process, Li *et al*. [45] obtained the following expression for the generated entropy per unit of mass:

$$s_{gen} = \alpha \sum_i P_i \ln P_i - \sum_i P_i \left(\beta\delta + \gamma\delta^2\right) - \frac{\sigma}{T} a_1 \tag{63}$$

where a_1 is the surface area per unit mass at the initial stage (nozzle exit), T is the temperature, σ is the surface tension and α, β and γ involve physical characteristics of the problem such as the surface tension, the temperature, the specific volume, the isothermal compressibility. The most appropriate drop-size distribution at the final stage is the one that satisfies the normalization and the mass conservation constraints and that maximizes the generated entropy (Equation 63)]. Performing the same variable change as Li and Tankin [15] (Equation 26), Li *et al*. ended up with the following number-based drop-size distribution:

$$f_n(\delta) = 3\delta^2 \exp\left(-\lambda_0 - \lambda_1\delta - \lambda_2\delta^2 - \lambda_3\delta^3\right) \tag{64}$$

The authors realized that this solution is similar to the one that would be obtained from the MEF formulation with the normalization and the three constraints expressing the mean diameters D_{10}, D_{20} and D_{30}. Li *et al*. [45] tested the ability of this new distribution to represent drop-size distribution of sprays produced by an air-blast atomizer and measured with a PDT. Several air and liquid velocities were tested. For each case, the experimental mean drop diameters D_{10}, D_{20}, D_{30} were used to numerically calculate the distribution given by Equation 64. The agreement between the measurement and the model was average. In some situations, the estimation could not even predict the peak diameter. One must note here that, having claimed that the MEF is not appropriate to develop liquid spray production model, the authors should not have used their MEG model as a MEF model which is what they did when using the experimental mean drop diameters D_{10}, D_{20}, D_{30}. The appropriate information required by their model should not be the one carried by these mean-diameters. This might be a reason why the comparisons they provided demonstrate the weak ability of Equation 64 to represent drop-size distribution.

5. Summary and Conclusions

The MEF as described in Section 2 of this paper allows a probability density function to be determined provided that some information related to the distribution sought is known. Thus, the first point to be mentioned is that its application to liquid spray characteristics must be restricted to the number-based distribution (either mono or bi-dimensional) since, from a mathematical point of view, this is the only type of distribution that is a probability density function. As demonstrated by Cousin *et al*. [31], the prediction of any other spray distribution types from the MEF requires the statistical Shannon entropy to be replaced by the more general Bayes entropy whose expression must be adapted to the distribution sought.

The success and relevance of a MEF model lies in the writing of the appropriate set of constraints that expresses the available information. The initial formulations for spray drop-size and velocity distribution prediction due to Sellens and Brzutowski [12,14] and to Li and Tankin [16] based the set of constraints on conservation laws that any physical process is expected to satisfy, i.e., the conservation of mass, momentum and surface and kinetic energies. Such an approach was questioned by Sirignano and Mehring [46] who reminded the importance of viscous dissipation in any atomization process. The viscous dissipation, that is associated to change of liquid internal energy or enthalpy, characterizes the irreversibility of the formation process. Thus, surface energy and kinetic energy in the final spray cannot be directly related to the initial energy values without considering the intermediate thermal processes. According to Sirignano and Mehring [46], maximum entropy formulations should be corrected by conserving the sum of liquid kinetic energy, surface energy and liquid internal energy or enthalpy. Finally they emphasized that entropy as defined in droplet distribution literature is based on information theory and not on thermodynamics. Sellens and Brzustowski [12] explained that their approach is not an application of thermodynamic principles to atomization in the classical sense but an application of statistical inference in which some of the constraints are expressed in terms of thermodynamic variables. In other words, they wonder whether some physical conservation laws could carry the appropriate amount of information to predict spray drop-size and velocity distributions.

The drop diameter-one velocity joint distributions obtained by Sellens and Bzustowski [12] and by Li and Tankin [16] reveal a similarity: both are dependent on four parameters (four Lagrangian multipliers for Sellens and Brzustowki's solution and three Lagrangian multipliers and a liquid Weber number based on the mass mean-diameter for Li and Tankin's solution). However, these two formulations show three differences that it is instructive to list.

First, Sellens and Brzustowski's formulation applies right in the breakup region whereas Li and Tankin's approach applies on a control volume that extends from the atomizer nozzle exit down to a plane where droplets are formed. Thus, Sellens and Brzustowski's model must incorporate the liquid system deformation prior to the breakup. For instance, applied to an atomizing liquid sheet, this requires at least two velocity components to be taken into account [21]. This aspect is not required by Li and Tankin's model. This difference also indicates that the source terms introduced in the constraints to account for energy losses during the process are not equivalent in each approach. For instance, whereas the liquid-gas interface surface area is globally increased during an atomization process, it is reduced by surface tension forces when the bulk liquid flow experiencing its maximum deformation breaks up into drops.

Second, the energy constraints are differently written. Sellens and Brzutowski [12] wrote two separate constraints for the directed kinetic energy and the surface energy whereas Li and Tankin combined these two energies in a single equation. This point has been largely debated in the different papers. Sellens and Brzustowski [12] and Ahmadi and Sellens [25] emphasized that the irreversibility of the process where kinetic energy is readily transformed into surface energy but the reverse transformation is not possible imposes conserving these two energies separately. On the other hand, Mitra and Li [29] explained that liquid kinetic energy is largely responsible for liquid atomization in practical sprays and atomization increases the surface area and hence the surface energy of the liquid. Therefore, separate conservation of liquid kinetic and surface energy violates the physics involved. (In fact the literature dedicated to liquid atomization processes (see [5]) demonstrates that the energy source largely responsible for atomization is not the directed kinetic energy but the turbulence and circulatory kinetic energies.) A decisive argument is due to Chin *et al*. [24] who noted that the shortcoming of considering a combined energy constraint is that no information is provided to how the total energy source is distributed between the kinetic energy and the surface energy. Thus, any combination of constant total energy source would result in the same probability density function. This argument pleads in favor of Sellens and Brzustowski's formulation.

Third, Sellens and Brzustowski [12] identified the droplet by their diameter and velocity and Li and Tankin [16] formulated their model in the drop volume-velocity space and performed a variable change to express their solution in the diameter-velocity space. The enthusiastic aspect of this latter approach is that, contrary to Sellens and Brzustowski's solution, Li and Tankin's number-based drop-diameter distribution decreases towards zero when the drop diameter approaches zero. As discussed by several authors [26,40] and as explained in this paper, such a mathematical manipulation must be prohibited without taking the appropriate precaution to ensure entropy invariance and to respect Jaynes consistency principle. In consequence, the solution provided by Li and Tankin is not a maximum entropy distribution but a biased distribution. This comment applies for all formulations that made use of this mathematical treatment [18,22-24,27,28,45]. As explained in this paper, it also concerns the proposition due to Dumouchel and Malot [35]. To be complete on this point, on should note that Kim *et al*.'s model [39] is not concerned by this criticism since they incorporate in their formulation a prior-distribution that can be seen as a supplementary constraint. The similarity of their solution with the one due to Li and Tankin comes from the fact that they imposed the prior-distribution in consequence. A more appropriate approach would have been to consider this prior-distribution as a supplementary parameter. This does not mean that the mathematical distribution derived by Li and Tankin [16] as well as those build on an extension of their approach [24,27,28,39] are inappropriate to represent drop diameter-velocity distributions. Most of these works reported an acceptable ability of these distributions to reproduce measured drop-diameter and velocity distributions. This demonstrates that these solutions are mathematically flexible enough to do so, i.e., they contain enough parameters to fit a wide variety of distributions.

The unrealistic behavior of Sellens and Brzustowski's solution for the left distribution tail (small drop-diameter range) indicates that the conservation of mass, momentum, surface energy and kinetic energy constitutes an insufficient or inappropriate amount of information to predict spray drop-diameter and velocity distributions. As explained by Kapur [13], if the MEF distribution is not correct either more information is needed or information is needed in some other form. Sellens [21] noticed that the constraints he used didn't carry any information on the small drop diameter and concluded to the necessity of introducing a supplementary constraint whose role is to limit the surface area to volume ratio of the entire spray. This new constraint, called the partition constraint and identical to the definition of the mean diameter D_{-10}, introduced a supplementary parameter in the solution and avoided the unrealistic behavior at the small drop-diameter distribution tail.

The investigation due to Ahmadi and Sellens [25] allowed a major turning to be passed in the prediction of spray characteristic from MEF. They demonstrated that drop-size and velocity can be considered independently and derived a simplified MEF model to predict number-based drop-diameter distributions. Their work shows that the conservation of mass and of surface energy and the partition constraint contain enough information for that. In other words, number-based drop-diameter distributions can be predicted from the three mean-diameters D_{-10}, D_{30} and D_{32}. These results gave birth to the formulation due to Dumouchel's group.

First, this formulation is developed for the determination of the drop-diameter distribution only. Second, it is not based on the set of constraints expressing physical conservation laws but addresses the question of the minimum information required for the determination of a spray drop-size distribution. The first approach suggested using a single constraint based on the definition of a mean diameter D_{q0}, the constraint diameter, keeping both D_{q0} and q as parameters. The resulting maximum entropy distribution was acceptable provided that it was used to represent volume-based drop-diameter distributions of sprays whose minimum diameter was sufficiently near zero. But the solution was not good to represent number-based distributions as the same drawback as for the solution provided by Sellens and Brzustowski was observed. This problem was analyzed as a lack of information introduced in the problem. Dumouchel [40] proposed an extension of this approach by including a prior-distribution that expresses the fact that each drop has not the same chance of being produced. This prior-distribution introduces a supplementary parameter. The solution of this extension was identified as a three-parameter Generalized Gamma function. This distribution is analytical. This constitutes an advantage compared to the previous maximum entropy distributions since it is easier to manipulate and allows calculations to be performed. Furthermore, this distribution is identical to the empirical Nukiyama-Tanasawa distribution and therefore its ability to represent spray drop-diameter distributions has not to be proved since this has been reported by many investigations over the past fifty years.

Finally, a different model based on entropy maximization has been suggested by Li *et al*. [45]. Excluding the traditional use of the MEF, these authors used the thermodynamically consistent concept of maximization of entropy generation. This approach was developed for the prediction of number-based drop-diameter distributions. Despite the fact that the solution reported by this formulation fails in representing some measured drop-size distributions, which, as discussed above might be a consequence of an inappropriate application, this approach assumes that an atomization process is an isothermal process, allowing the change of liquid internal energy to be neglected. As demonstrated by Sirignano and Mehring [46] (see comment above) this assumption is simply not acceptable for liquid atomization processes.

The MEF models to determine liquid spray drop-size distributions have been developed and applied for sprays produced by several atomizer designs and concepts including low-pressure swirl atomizers [15,16,21-23,25,27,31,39], ultrasonic atomizers [30,32,37,43], low-pressure single cylindrical orifice nozzle [24,28,36], low pressure gasoline injector [43] and high pressure gasoline injector of different designs [34,44], and twin fluid atomizers with internal [43] or external [45] interaction between the liquid and the gas flows. Being based on a limited number of constraints, most of these MEF models return a mono-modal distribution. However, models allowing the description of more complex distribution shapes (bi-modal) have been developed according to two approaches; 1 – increase the number of constraints [27,28], 2 – combine two mono-modal MEF solutions [44]. These models agree on the fact that seven parameters are required to describe bi-modal spray drop-size distributions.

The validation of the maximum entropy distributions reported by the different models presented in this paper is usually discussed by comparing them with measured spray distributions. However, the application of the models requires the prior determination of the moments introduced in the constraints and that constitute the available information. Therefore these moments must be specified. Here, one should note that the determination of the source terms in the constraints based on physical conservation laws is a difficulty that has not been solved so far. Furthermore, for other approaches, the parameters introduced in the constraints are not even clearly related to physical bases which rends their prior-determination more complex. Therefore, several procedures to apply the model have been proposed. They can be classified in three groups: 1 – use as much experimental information (such as moments of the distribution) as necessary to allow the parameters of the mathematical distribution to be calculated; 2 – determine the set of parameters that ensures the best fit with the measured distribution; 3 – a mixing of the two first procedures. For non-analytical maximum entropy distributions (those obtained from physical conservation laws), the final solution had to be determined by numerical procedures which appeared to be a tricky task. Indeed, inappropriate values of the moments or/and of the integration limits could result in unrealistic solutions. Such a problem was avoided with the analytical three-parameter Generalized Gamma function. Despite these numerical difficulties, one should underline that, in almost all the situations, comparisons between measured and mathematical distributions reported good agreements. For instance, adopting the first procedure, Ahmadi and Sellens [25] used the three mean-diameters D_{-10}, D_{30} and D_{32} provided by the measurements and obtained good representations of the measured number-based drop-size distributions. Thus, if a model to predict these three mean-diameters is derived from the analysis of the measurements, the maximum entropy model becomes autonomous and predictive. On the other hand, Lecompte and Dumouchel [43] and Yongyingsakthavorn *et al*. [44] adopted the second procedure and determined, for each investigated situation, the set of parameters that ensured the best fit with the measured distributions. Here again, the agreements were very good. Furthermore, these parameters were correlated to the working conditions. Besides the fact that these correlations demonstrate the physical relevance of the parameters of the maximum entropy distribution, they allow the whole model to be predictive.

The good agreements between measured and predicted distributions found in the literature demonstrate at least that most of the maximum entropy distributions have a good ability to reproduce liquid spray drop-size distribution. Therefore, from a theoretical point of view, the MEF is appropriate to derive predictive models for liquid spray drop-size distributions. The problem in the application of these models is the prior knowledge of the information—or the constraints—required by the formalism.

One must emphasize at this stage that this aspect doesn't affect the potentiality of the MEF: indeed, the MEF is not supposed to determine constraints for a given situation. Therefore, the MEF shifts the problem of drop-size distribution model development to the one of appropriate information determination. The advantage is that, thanks to the MEF, we know how much information is required and we have interesting clues concerning the form to be given to this information. For instance, the MEF models agree that three pieces of information are required to determine mono-modal drop-size distributions. For the determination of number-based drop-size distributions, this information can be provided by three mean diameters (D_{-10}, D_{30}, D_{32}) or by a constraint diameter together with a prior probability distribution expressing the diameter-class accessibility. Efforts should be done now to model these pieces of information. Beside this, one should add that the MEF can provide a practical help. For instance, it can be used to extend the partial information reported by an experimental diagnostic and provide the whole drop-size distribution. This was successfully demonstrated and applied for high density gasoline sprays [34]. Furthermore, the set of parameters that ensures the best representation can appear to be physically relevant characteristics to categorize and investigate the atomization process and the spray. This could constitute an interesting starting point for the constraint investigation.

The application of the MEF to determine spray drop-size distributions rests on information theory and not on thermodynamics. As mentioned above, the validity of MEF models requires comparisons with measurements to be performed and this comparison procedure often introduces partial measurement results in the models. As far as this point is concerned, it is important to bear in mind that the diagnostics available so far to measure drop-size distribution are sensitive to different spray characteristics and report information that depends on their working principle. Therefore, the validity of any model that incorporates an experimental support will be limited by the experimental procedure. In other words, the formulation of a universal model based on the MEF or on any other formalism to predict liquid spray drop-size distribution would require a universal definition of this characteristic as well as an appropriate diagnostic to measure it.

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