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Proceedings of the Combustion Institute 31 (2007) 667–674

Proceedings  
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# Calculating the soot particle size distribution function in turbulent diffusion flames using a sectional method

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## Abstract

Soot formation in a turbulent jet diffusion flame is modeled using an unsteady flamelet approach in post-process. In the present work, we apply a detailed kinetic soot model with a sectional method, and study the evolution of the particle size distribution. Detailed information on the evolution of the soot particle size distribution function is acquired. It is found that the particle size distribution function is bimodal throughout the flame. The transition from the small to large particle size distributions is strongly influenced by surface growth and oxidation reactions. We find that large particles are most likely to be emitted from the flame. © 2006 The Combustion Institute. Published by Elsevier Inc. All rights reserved.

*Keywords:* Soot; Sectional method; Unsteady flamelets

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## 1. Introduction

The legislative regulations on particulate matter (PM) emissions from practical combustion devices have so far only considered the amount of PM emitted, despite that the toxicity of the particles depends on the particle size. Smaller particles can more easily penetrate cell membranes than large particles, they have a larger surface/volume ratio and as surface activity determines toxicity (see e.g. [1] and references therein), are more detrimental to human health than larger particles. One form of PM emissions from combustion devices is soot. As most practical combustion devices, such as diesel engines and

gas turbines, operate under turbulent diffusion flame conditions, having the possibility to accurately predict the soot particle size distribution function (PSDF) would facilitate not only legislative regulations regarding the size distribution for soot emissions, but also optimization of the combustion process towards lower emissions of hazardous particles. In this work, we present a method for predicting the soot particle size distribution under turbulent diffusion flame conditions. For this we employ the representative interactive flamelet model [2–5] to couple to turbulent flow field with a detailed kinetic soot model.

In order to accurately model soot formation, it is advantageous to consider detailed chemistry for the gas phase, soot precursor formation, and soot surface processes. In addition to the chemical processes the physical processes of particle inception, coagulation and condensation must be considered. We describe these processes using the Smoluchowski equation [6]. Finally, a mathematical model is needed to describe the development of the PSDF. From the literature two methods

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which allow formulation of conservation equations for properties of a particle size distribution are known: the method of moments [7,8] and the sectional methods [9,10]. Previous modeling work on the soot PSDF incorporate studies on laminar premixed flames using stochastic methods [11,12]. However, no conservation equations for the properties of the PSDF have yet been formulated for stochastic methods. Although the method of moments does not require any assumptions on the actual shape of the size distribution, it can only provide information on statistical quantities such as mean, variance, etc. of the distribution. Sectional methods can directly provide the actual shape of the size distribution. In Refs. [2–4] conservation equations for the moments of the soot PSDF were formulated for the RIF model. In a further development the sectional method [13] was employed in a post-process step to calculate the soot PSDF on a grid cell basis. The reaction rates needed for calculating soot precursor formation and soot surface processes were determined using spatially averaged species concentrations obtained from the RIF calculation.

In this work, we have implemented the sectional method described in [14] in an unsteady flamelet code, and investigate the evolution of the soot PSDF in a turbulent jet diffusion flame. Soot formation and oxidation processes are fully coupled and are calculated together with the gas phase processes.

## 2. Modeling

### 2.1. Soot modeling

The detailed kinetic soot model used in this work follows the approach by Mauss [15]. During the oxidation of the hydrocarbon fuel, aromatic species are formed in the gas phase of the flame and grow to poly-aromatic structures. Species with more than four rings can combine and incept soot particles [16]. They can further condense on the soot particle surface and contribute to the surface growth process. Soot particles collide and, depending on their size and the surrounding environment, combine to larger spherical particles or to agglomerates [17]. All these processes can be calculated from the Smoluchowski equation [6]. In addition heterogeneous surface growth and oxidation reactions are taken into account. The surface growth is modeled by the simple hydrogen abstraction acetylene addition mechanism (HACA) [18] with a separate ring closure [15]. Oxidation occurs via reactions of OH and O<sub>2</sub> on the soot particle surface. Details of the soot model are not part of this work, and can be found in Ref. [15]. In addition to that model, an agglomeration model similar to the model by Kazakov and Frenklach [19] was added. In the

calculations presented here the fractal dimension of the particles was kept constant  $D_f = 1.8$ .

The soot model introduces an infinite number of partial differential equations that describe the PSDF. It is not possible to solve this system of equations for one-dimensional flamelets, and it is necessary to model the particle size distribution function. The method of moments [7,8,18] and the sectional method (see for instance [9,10] and references therein) allow the formulation of convective and diffusive transport terms. In this work, we use the sectional method to describe the PSDF. According to the sectional method the soot PSDF can be divided into a discrete number of sections. All particles within a section can be represented by a single scalar, and the soot formation processes are formulated for each of these scalars rather than for each particle size. A detailed description of the model can be found in [14]. In the following a general outline is given.

A geometric and fixed division of the PSDF was chosen. The section boundaries are thus given by:

$$v_{i,\max} = v_{\text{MIN}} \left( \frac{v_{\text{MAX}}}{v_{\text{MIN}}} \right)^{i/i_{\max}} \quad (1)$$

where  $i_{\max}$  is the total number of sections and  $v_{i,\max}$  is the upper boundary of section  $i$  and  $v_{i,\min} = v_{i-1,\max}$ .  $v_{\text{MIN}}$  represents the smallest particle and  $v_{\text{MAX}}$  represents the largest particle.  $v$  signifies soot particle size, typically volume. To enhance the accuracy of the model the first section is:

$$v_{1,\min} = v_{\text{MIN}} \quad (2)$$

$$v_{1,\max} = v_{\text{MIN}} + v_{C_2}$$

where  $v_{C_2}$  is the volume of two carbon atoms in soot. In the present work calculations have been performed using 100 sections, unless otherwise stated.

The chosen conserved scalar is soot volume fraction ( $f_v$ ) within each section, denoted  $Q_i$ . Number density within section can be calculated from this scalar. In the present model the intrasectional distribution of  $f_v$  within each section is approximated with a first degree polynomial. This means that both overall number density distribution and overall  $f_v$  distribution assume near continuous shapes. The slopes of the polynomials are linear functions of the mean  $f_v$  values of the nearest neighboring sections.

To calculate the coagulation source terms each section is divided into subsections. The subsections are treated as homogeneous units and the sectional coagulation source terms are calculated by directly iterating over the Smoluchowski equation [6] for the total number of subsections (Sub). The number density of subsection  $j$  is denoted  $N_j$ . The volume of a particle in subsection  $j$  is approximated with

the representative volume  $V_j$ . A general formula for the coagulation source terms is given by:

$$\dot{Q}_i = \sum_{j=1}^{\text{Sub}} \sum_{k=j}^{\text{Sub}} \beta_{j,k} N_j N_k (\delta_{i,j,k}^{\text{gain}} (V_j + V_k) - \delta_{i,j}^{\text{loss}} V_j - \delta_{i,k}^{\text{loss}} V_k) \quad (3)$$

where  $\delta_{i,j,k}^{\text{gain}}$  is 1 if the particle formed by particles  $j$  and  $k$  falls into section  $i$  and 0 otherwise.  $\delta_{i,j}^{\text{loss}}$  is 1 if particle  $j$  belongs to section  $i$  and 0 otherwise. The  $\beta_{j,k}$  is the collision frequency. The  $\delta_{i,j,k}^{\text{gain}}$ ,  $\delta_{i,j}^{\text{loss}}$  and  $\beta_{j,k}$  are precalculated.

The nascent particles are formed by poly-aromatic hydrocarbons (PAH) colliding to form the smallest soot particles. PAH also condense onto the surface of soot particles. As particle inception and condensation are also both governed by the Smoluchowski equation these source terms are calculated as simplified instances of Eq. (3).

In the present sectional description of the heterogeneous surface reactions an implementation conserving the two first moments of the PSDF is applied. This means that the change of  $Q_i$  due to surface growth within section  $i$  is distributed between section  $i$  and  $i + 1$ . The distribution is determined by the sectionalization of the PSDF. Condensation is treated analogously to surface growth. Oxidation is distributed among section  $i$  and  $i - 1$ . Special handling is needed for section 1 and section  $i_{\text{max}}$ . For more details on this see [14]. In calculating the soot surface a fractal dimension of 1.8 is assumed for all particles. The gas phase chemistry was taken from [15].

The sectional method applied in this work has been validated using a method of moment implementation of the same detailed kinetic soot model, see [14] for details. The agreement of the soot model using a method of moments implementation for the soot PSDF has in turn been validated with experimental data, see for instance [15].

Soot formation in turbulent diffusion flames was recently investigated by Pitsch et al. [20]. In this publication it was concluded that it is necessary to include preferential diffusion effects for soot particles, to be able to predict the measured profiles for the soot volume fraction in a turbulent ethylene-air jet diffusion flame. We have recently found [21,22] that an accurate prediction of radial and axial profiles of the soot volume fraction in turbulent diffusion flames can be achieved, if the deviations of the maximum mixture fraction in the flamelet from unity are taken into account. For these calculations we did not find an influence of preferential diffusion of the soot particles on the predicted profiles. We note at this point, that we still find a sensitivity on preferential diffusion of gas molecules. We further found, that the agglomeration of soot particles has a strong influence on the amount of soot, predicted. Conditioning of the scalar dissipation rate on the location in

mixture fraction space where maximum scalar dissipation occurs, simplifies the formulation for flamelets with a maximum mixture fraction smaller than the stoichiometric mixture fraction.

## 2.2. Unsteady flamelet modeling

The unity Lewis number simplification is also used in this work as a consequence of the findings in [21,22]. With this simplification, the following transport equations in mixture fraction space for species mass fractions, enthalpy (expressed in temperature) are obtained (see e.g. Peters [23] for details) when transforming the transport equations to mixture fraction space:

$$\rho \frac{dY_i}{dt} = \frac{\rho\chi}{2} \frac{d^2 Y_i}{dZ^2} + \dot{\omega}_i \quad (4)$$

$$\rho c_p \frac{dT}{dt} = \frac{\rho\chi}{2} \left( \frac{d}{dZ} \left( c_p \frac{dT}{dZ} \right) + \sum_{i=1}^N c_{p,i} \frac{dY_i}{dZ} \frac{dT}{dZ} \right) - \sum_{i=1}^N h_i \omega_i - q_R \quad (5)$$

In the equations above  $\rho$  is the density,  $Y_i$  is the mass fraction of species  $i$ ,  $t$  is the time,  $\chi$  is the scalar dissipation rate,  $Z$  is the mixture fraction,  $c_p$  is the average specific heat capacity at constant pressure,  $c_{p,i}$  is the specific heat capacity at constant pressure of species  $i$ ,  $T$  is the temperature,  $\omega_i$  is the chemical source term for species  $i$ ,  $h_i$  the enthalpy of species  $i$ , and  $q_R$  the radiative heat loss term. The scalar dissipation rate is defined as

$$\chi \equiv 2D(\nabla Z)^2 \quad (6)$$

The transport equation for the soot sections is defined analogously to the transport equation for a general scalar without considering size-dependent transport parameters:

$$\rho \frac{dQ_i}{dt} = \frac{\rho\chi}{2} \frac{d^2 Q_i}{dZ^2} + \dot{\omega}_i \quad (7)$$

In the above equation  $Q_i$  represents the soot volume fraction of section  $i$  and  $\dot{\omega}_i$  is the soot volume fraction source term for section  $i$ .

The unsteady flamelet calculation is performed as a post-process step to the flow field calculation. The flow field used here is the same flow field which was used in [24]. This flow field was calculated using an in-house RANS code, and during the flow field calculation a steady flamelet library has been used.

In the post-process procedure for all heights in the flame, first the scalar dissipation rate, conditioned on the location of maximum scalar dissipation rate is computed and stored together with the maximum mixture fraction for use by the unsteady flamelet code. Assuming an inverse complementary error function profile for the scalar

dissipation rate, the maximum scalar dissipation rate is occurring at  $Z_{\max}/2$ . The maximum mixture fraction  $Z_{\max}$  is chosen as the mixture fraction on the centerline, and the conditional scalar dissipation rate at  $Z_{\max}/2$  is obtained as:

$$\langle \chi_{Z_{\max}/2} \rangle = \frac{c_\chi \tilde{\varepsilon} / \tilde{k} Z''^2}{\int_0^1 \text{EXP}(-2(\text{ERFC}^{-1}(2Z/Z_{\max}))^2) P(Z) dZ} \quad (8)$$

where angular brackets denote conditioned value,  $c_\chi$  is a modeling constant, here set to 2.0,  $\varepsilon$  is the turbulent kinetic energy dissipation rate,  $k$  is the turbulent kinetic energy,  $P(Z)$  denotes the probability density function (pdf) over mixture fraction, here a beta-pdf, and  $Z''$  denotes the variance of mixture fraction. Favre averaged quantities are denoted by a tilde. The second step in the post-process procedure is to perform an unsteady flamelet calculation. The unsteady flamelet calculation is initialized with a start profile from an extinguishing flamelet at high scalar dissipation rate, and the unsteady flamelet evolves in time, i.e. travels through the flame following the maximum scalar dissipation rate. There is no soot in the start profile. Finally, the results from the unsteady flamelet calculation are mapped back onto the flow field.

### 3. The experimental test flame

As in Refs. [21,22,24] we use the experiments by Young et al. [25] on a rim-stabilized ethylene turbulent jet diffusion flame. Ethylene is injected through a pipe with a diameter of 3.1 mm at a speed of 24.5 m/s into stagnant air at room temperature and atmospheric pressure. The soot volume fraction was measured by laser absorption along with mixture fraction by microprobe sampling technique, and temperature by fine wire thermocouples.

### 4. Results and discussion

Figure 1 shows a comparison between experimental data for the soot volume fraction by Young et al. [25], and the results obtained with the present sectional soot model. As has been shown recently for the moment method implementation of the present detailed kinetic soot model, the agreement with experiments is reasonable [21,22]. Figure 2 shows the evolution of the PSDF along the centerline expressed in  $f_v$ . The PSDF is such that if an integration is performed over all sections the total  $f_v$ , as shown in Fig. 1, at the given spatial coordinate is obtained. When studying the figures one should keep in mind that both  $f_v$  and particle size scales are logarithmic. The particle diameter is calculated assuming spherical particles. The seemingly

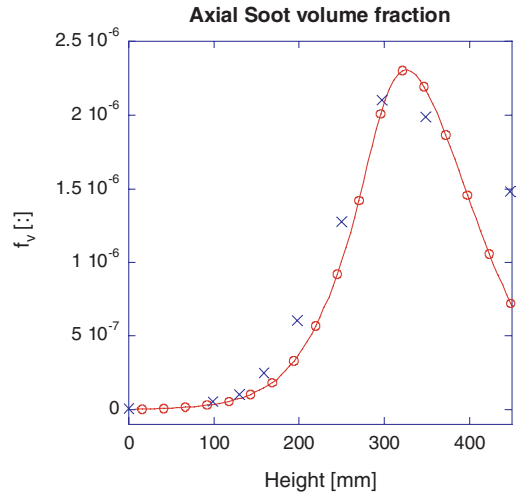


Fig. 1. The axial profile of  $f_v$  along the centerline. Crosses indicate experiments by Young et al. [25], and rings indicate numerical solution.

large  $f_v$  at an axial height of 10 mm actually corresponds to a total  $f_v$  of  $8.8 \cdot 10^{-10}$ . The lowest axial height distribution in Fig. 2 corresponds to a number averaged mean diameter of 3.5 nm. The small amount of soot found at this point in the flame diffuses from the soot formation layer to the centerline.

Moving along the centerline in the flame, the contribution of sections representing particles having a higher number of carbon atoms to total  $f_v$  increases. This is caused by coagulation and surface growth. While the PSDF shifts towards larger particles, small particles are constantly incepted creating a relatively even distribution of  $f_v$ . At 150 mm the PSDF is bimodal but the transition from the distribution of small particles to the distribution of large particles is smooth, since the valley in between is filled by the growth process of recently incepted particles. This behavior is unchanged until the total  $f_v$  peaks at a height of 300 mm. After the peak there is a large drop in medium to small particle  $f_v$  due to oxidation and reduced surface growth of the small soot particles. The reduced growth of small particles results in a strong gradient of  $f_v$  in particle radius for small particles sizes. The coagulation of particles in the medium size classes is no more balanced by a production term. At 500 mm a very distinct bimodal distribution of  $f_v$  is observed. Throughout the flame particle inception and thereby the production of small particles is found. The production rate decreases significantly at the height of 300 mm, where oxidation processes become stronger than chemical particle growth processes.

In order to assess the accuracy of the sectional method implemented here, runs were performed with 60 and 200 sections. Figure 3 shows that

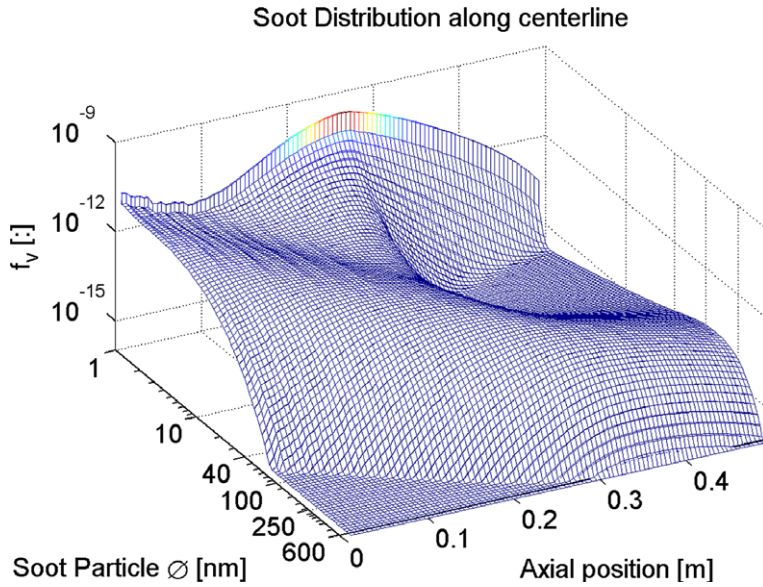


Fig. 2. Axial evolution of the PSDF along the centerline in units of  $f_v$ . Particle diameter and  $f_v$  scales are logarithmic.

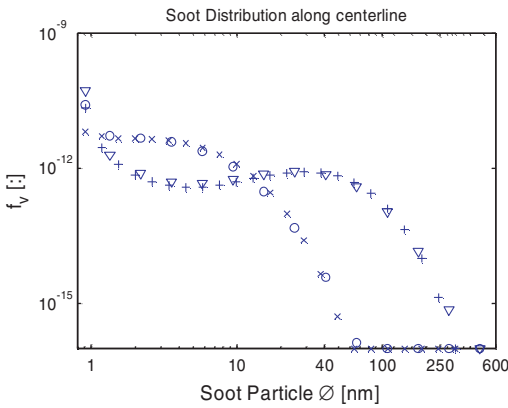


Fig. 3. A comparison between two runs using 60 and 200 sections, respectively. Sixty sections result indicated by (○) at 10 mm, (▽) at 190 mm. Two hundred sections results indicated by (×) at 10 mm, (+) at 190 mm.

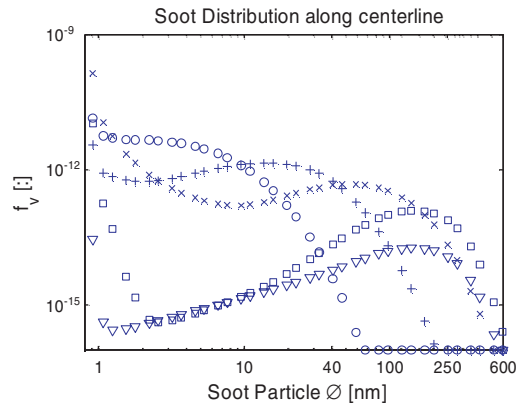


Fig. 4. PSDF in units of  $f_v$  at given heights along centerline. (○) 10 mm, (+) 130 mm, (×) 250 mm, (□) 370 mm, (▽) 490 mm.

the difference in predicted PSDF profiles at 10 and 190 mm heights is negligible. This comparison would show a highly visible difference, if numerical diffusion would have a large impact in the given flame conditions. Numerical diffusion is strongly influenced by the discretization of the PSDF.

Figure 4 extracts profiles from Fig. 2 at heights 10, 130, 250, 370, and 490 mm along the centerline clarifying the evolution of the soot PSDF. This Figure again demonstrates the strong shift of the PSDF at heights larger than 300 mm. The smooth valley between small and large particle distribution turns to a very distinct gap.

Figure 5 shows the axial evolution of the number density ( $1/\text{cm}^3$ ) instead of  $f_v$ , shown in Fig. 3. Figure 6 shows the number density at heights 10, 130, 250, 370, and 490 mm along the centerline. The history of the PSDF that was explained, when discussing Figs. 3 and 4, can also be observed in Figs. 5 and 6. However, the different regions are not as distinct.

The radial evolution of the PSDF in units of  $f_v$  at heights 100, 160, 250, and 350 mm is shown in Fig. 7. It is observed that the greatest amount of large particles is present at the centerline. In general, the PSDF does not change very much radially, if one is compensating for the radial change in total  $f_v$ . However, we note that at an axial height



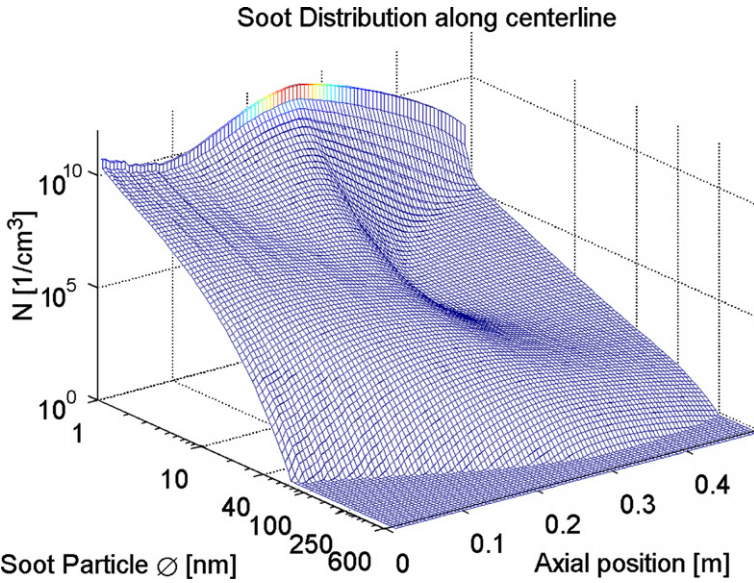


Fig. 5. Axial evolution of the PSDF along the centerline in number density ( $1/\text{cm}^3$ ).

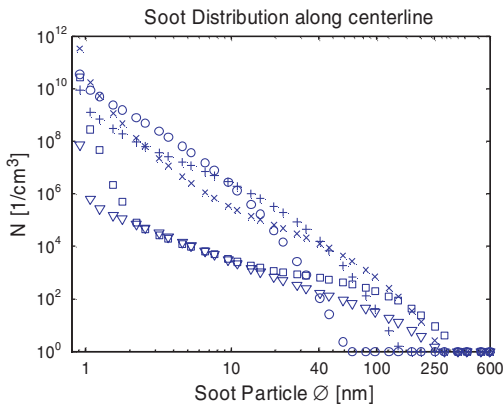


Fig. 6. PSDF (number density [ $1/\text{cm}^3$ ]) at given heights along the centerline. (○) 10 mm, (+) 130 mm, (×) 250 mm, (□) 370 mm, (▽) 490 mm.

of 350 mm, larger particles are present further out from the centerline. These particles are likely to be emitted from the turbulent diffusion flame. We also observe that the contribution to the total  $f_v$  from the mid-sized particles has decreased due to oxidation and coagulation.

## 5. Conclusion

Soot formation in a turbulent ethylene diffusion flame was calculated using the unsteady flamelet concept in post-process. A detailed chemical mechanism and a complex soot model was used together with a sectional method.

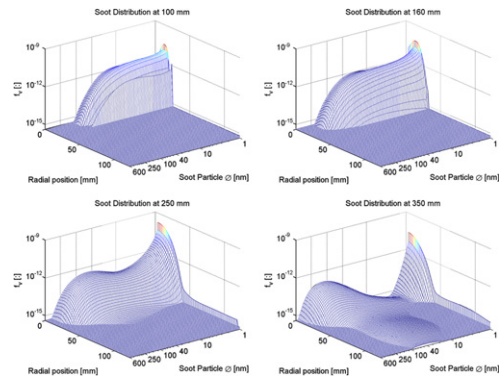


Fig. 7. Radial evolution of PSDF in units of  $f_v$  at heights 100, 160, 250, and 350 mm.

Using the sectional method, we have found that a large number of small particles resulting from particle inception is present throughout the flame. This causes that the PSDF is bimodal. In regions with strong surface growth the transition from the distribution of small particles to the distribution of large particles is smooth. In regions where oxidation processes are observed the bimodal distribution function is distinct. The soot PSDF is not changing very much radially in the flame. At a height of 350 mm we observe the break through of medium size soot particles. These particles are likely to be emitted.

A tool that provides accurate prediction of the soot PSDF in turbulent flames was presented.

Future applications will include calculations of soot PSDF in diesel engines and gas turbines using the representative interactive flamelet model. We found that the method is computationally efficient, when compared with the method of moments.

The calculation of one post-process run using the method of moments (4 moments) needed 30 min and using the sectional method (100 sections) needed 4 h on a 3.0 GHz Pentium 4 Xeon CPU. We find that the increase in CPU time is reasonable since knowledge about the PSDF provides the engineer with additional information.

## Acknowledgments

This work was supported by the European Space Agency in the project (No. 14311/00/NL/SH) “Investigations on soot concentration and primary particle sizes by advanced laser-induced incandescence”.

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## Comments

*John Hewson, Sandia National Laboratories, USA.* You have a term in the flamelet equations for soot evolution that involves the product of the scalar dissipation gradient and soot section moment, both gradients being with respect to the mixture fraction. This term is related to the evolution of the mixture fraction probability density function (PDF) [1,2]. This term advects the soot in mixture fraction space and thereby moves the location of the soot relative to, for example, the peak of the mixture fraction PDF. The presumed form of the dissipation, with respect to mixture fraction that you have chosen, would presumably affect the mean soot comparisons that you have made with the data, since they involve convolving over the mixture fraction PDF. Can you provide an estimate of how large this effect is?

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Also as a note to the authors, a consistent dissipation rate as a function of the mixture fraction can be related to the evolution of the PDF using methods in those references.

*Reply.* The term that is referred to in the question appears in the derivation of the set of equation used in this paper. This term is disappearing under the assumption that the Lewis number of a chemical species is equal to the Lewis number of the reference species that was defined in the transport equation of the mixture fraction. The Lewis number for species in turbulent flows is calculated from the sum of the laminar and turbulent diffusion coefficient. The turbulent diffusion coefficient is equal for all species. The term in question is therefore not significant if, for all species, the laminar diffusion coefficient is much smaller than the turbulent diffusion coefficient. The diffusion coefficient for soot particles is, in general, low compared

to gas phase species and can be neglected compared to the turbulent diffusion coefficient. For this answer, we define the largest section of soot particles to be the reference species. Hence, in the transport equation for the mixture fraction appears turbulent diffusion coefficient only. We find that the strongest relative motion is expected for light species as H and H<sub>2</sub>, and that the influence of the term in question decreases with decreasing laminar diffusion coefficients. We performed sensitivity calculations and found, indeed, that the convective flow of H and H<sub>2</sub> towards the soot particles influences the solution. However, we could not find any sensitivity of the diffusivity of N<sub>2</sub> on our results.

We therefore conclude that the transport of soot relative to the gas phase is not sensitive to our results. In our opinion, direct numerical simulations at different scalar dissipation rates are needed to finally answer the question.

We note that the solution of the laminar flamelet equations for soot particles is difficult. It is observed that numerical errors can cause that soot particles leave the integration domain. The inclusion of the advective term for soot particles can eliminate this numerical problem, and is then sensitive. Numerical errors must be eliminated, to estimate the physical relevance of the advective term.