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Modeling Differential Diffusion of Soot with Conditional Moment Closure

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Sandia National Laboratories

Workshop on Conditional Moment Closure
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Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

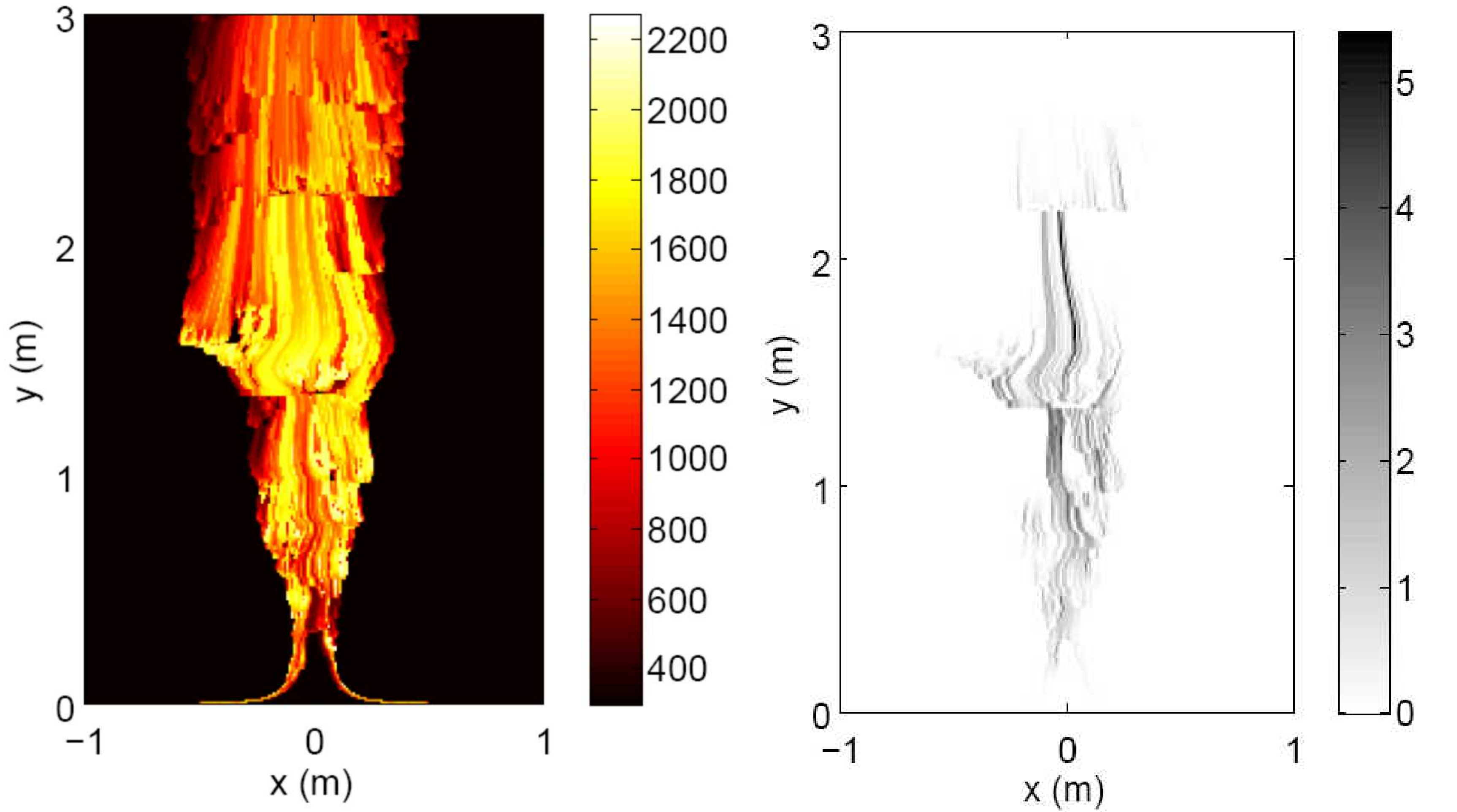
Introduction

- Conserved scalar approaches: Flamelets, CMC
  - Typically derived for unity Lewis numbers

- Differential diffusion alters relationship between reacting and conserved scalars.
  - Previous work by Kronenberg (1997), Pitsch (1998)
  - Focus here on soot.

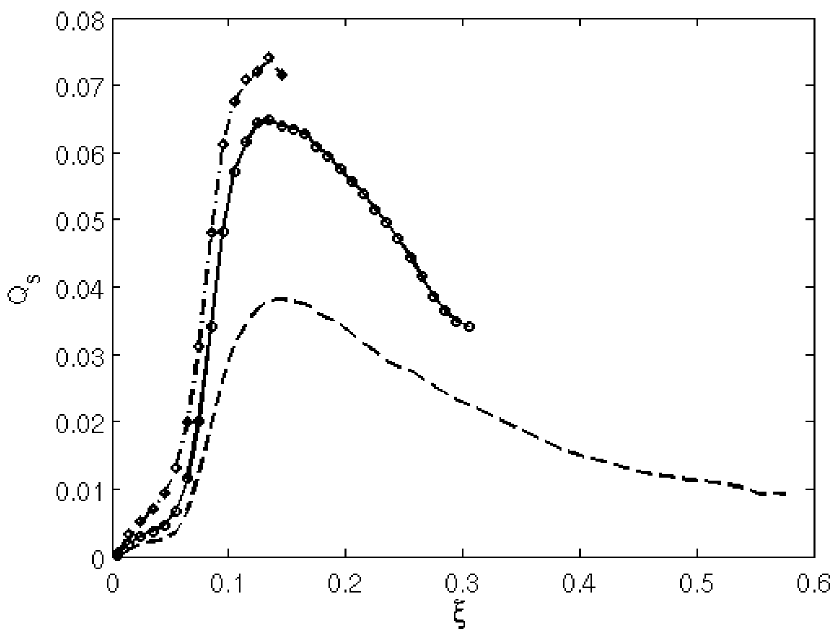
- New look at CMC formulation
  - Choices made in derivation can give slightly different formulations, require different modeling.
  - Different terms can be easier or more difficult to model depending on the scalar of interest.
- A priori analysis looks at significant terms and models for different terms.

Higher fidelity data to check closures

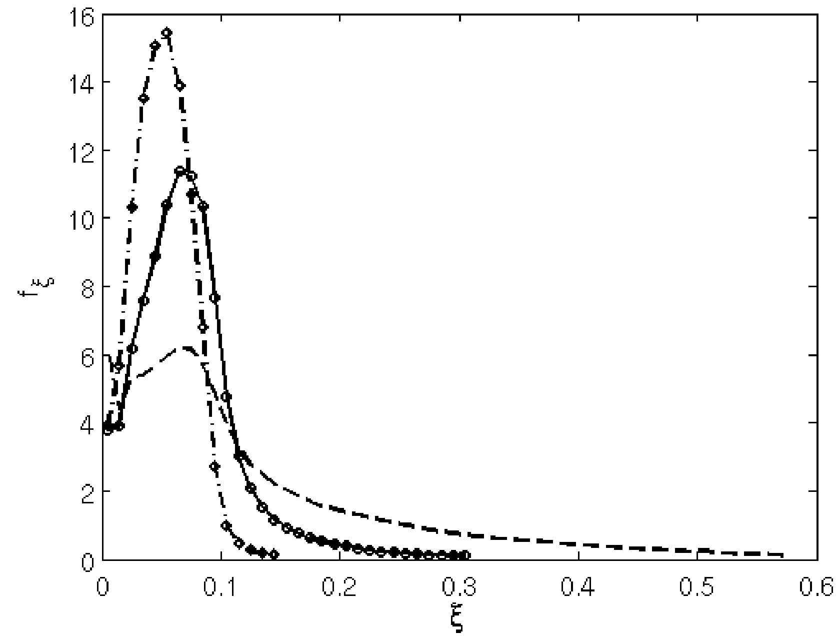


ODT results

Soot mass fractions



Mixture fraction PDF



heights are 0.9, 1.4 and 1.9 source widths

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General species conservation

$$\frac{\partial \rho Y_i}{\partial t} + \nabla \cdot \left(\rho \vec{u} Y_i \right) - \nabla \cdot \left(\rho D_i \nabla Y_i \right) + \nabla \cdot \left(\rho \vec{V}_i' \, Y_i \right) = \rho \omega_i$$

- **Allows for different diffusion models, thermophoresis, etc. for species, enthalpy, aerosols (soot).**

Joint-pdf equation

$$\frac{\partial \rho Y_i}{\partial t} + \nabla \cdot \left(\rho \vec{u} Y_i \right) - \nabla \cdot \left(\rho D_i \nabla Y_i \right) + \nabla \cdot \left(\rho \vec{V}_i' \, Y_i \right) = \rho \omega_i$$

- **From species equation, derive equation for joint-pdf evolution**

$$\frac{\partial \left[\left\langle \rho Y_k \middle| \vec{Z} \right\rangle P(\vec{Z}) \right]}{\partial t} + \nabla \cdot \left[\left\langle \rho \vec{u} Y_k \middle| \vec{Z} \right\rangle P(\vec{Z}) \right] = \sum_i \frac{\partial}{\partial Z_i} \left[\left\langle \rho \omega_i \middle| \vec{Z} \right\rangle P(\vec{Z}) \right]$$

$$- \sum_i \frac{\partial}{\partial Z_i} \left[\left\langle \nabla \cdot \left(\rho D_i \nabla Y_i \right) \middle| \vec{Z} \right\rangle P(\vec{Z}) \right]$$

$$+ \sum_i \frac{\partial}{\partial Z_i} \left[\left\langle \nabla \cdot \left(\rho \vec{V}_i' \, Y_i \right) \middle| \vec{Z} \right\rangle P(\vec{Z}) \right]$$

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Replacing diffusion terms with dissipation terms

- Mathematical identity used to replace diffusion terms in joint-pdf equation with scalar dissipation terms.

$$\nabla \cdot \left[\left\langle \rho D_k \middle| \vec{Z} \right\rangle \nabla P(\vec{Z}) \right] = -\sum_i \frac{\partial}{\partial Z_i} \left[\left\langle \nabla \cdot \left(\rho D_k \nabla Y_i \right) \middle| \vec{Z} \right\rangle P(\vec{Z}) \right] + \sum_i \sum_j \frac{\partial^2}{\partial Z_i \partial Z_j} \left[\left\langle D_k \nabla Y_i \nabla Y_j \middle| \vec{Z} \right\rangle P(\vec{Z}) \right]$$

- Can use any diffusion coefficient in this expression to replace any of the terms in the joint-pdf equation.

- Different choices result in different forms of conditional moment equations when differential diffusion is allowed.

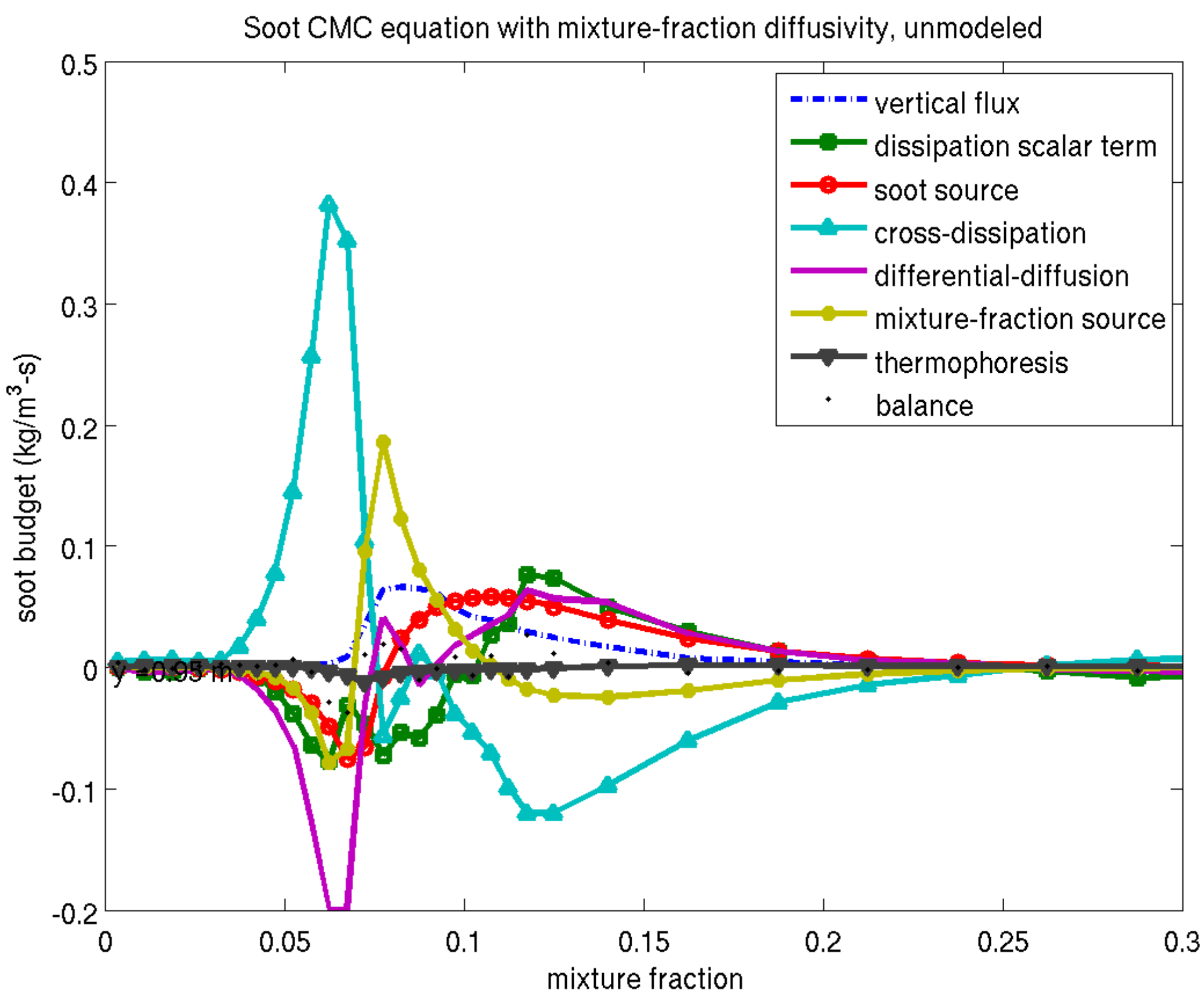
Analyzing closure requirements for traditional CMC

- **Conditional moment equation derivation**
  - **Multiply joint PDF by scalar of interest, *Y_k* ; integrate over PDF except mixture fraction, η**

$$
\begin{aligned}
\nabla\cdot\left[\left\langle \rho\vec{u}Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta})\right] = \; & \left\langle \rho\omega \middle| \vec{\eta}\right\rangle P(\vec{\eta}) \\
& + \frac{\partial}{\partial\eta}\left[\left\langle \rho D_\xi \nabla\xi\nabla Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta})\right] \\
& - \frac{\partial^2}{\partial\eta^2}\left[\left\langle \rho D_\xi \left(\nabla\xi\right)^2 Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta})\right] \\
& + \left\langle \nabla\cdot\left(\rho D_T Y_k \nabla \ln T\right) \middle| \vec{\eta}\right\rangle P(\vec{\eta}) \\
& - \left\langle \nabla\cdot\left(\rho (D_k - D_\xi)\nabla Y_k\right) \middle| \vec{\eta}\right\rangle P(\vec{\eta})
\end{aligned}
$$

  - **Equations are exact here: no modeling**

Significant terms in traditional CMC



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Cross-dissipation term in traditional CMC

- **Standard approach for closure**

$$\langle XY|\eta\rangle \approx \langle X|\eta\rangle\langle Y|\eta\rangle$$

$$\frac{\partial Y_s}{\partial x_i} = \frac{\partial Y_s}{\partial \xi}\frac{\partial \xi}{\partial x_i}$$

- **Basic term**

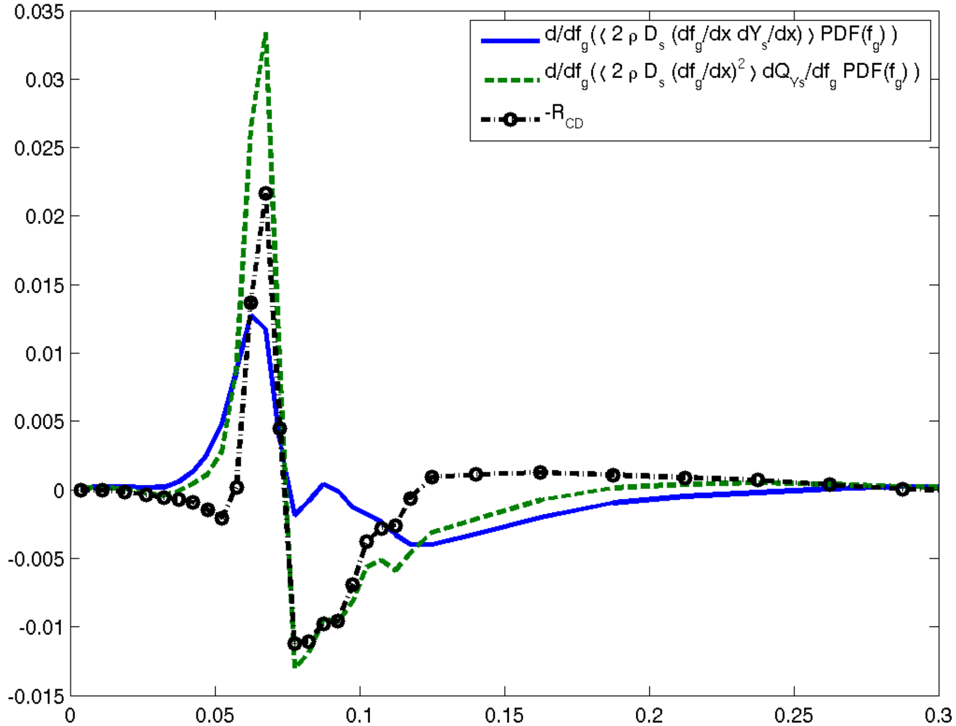
$$\frac{\partial}{\partial \eta}\left[\left\langle \rho D_\xi \nabla\xi\nabla Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta})\right]$$

- **Standard closure**

$$\frac{\partial}{\partial \eta}\left[\left\langle \rho D_\xi \left(\nabla\xi\right)^2 \middle| \vec{\eta}\right\rangle \frac{\partial Q_k}{\partial \eta} P(\vec{\eta})\right]$$

- **Residual (difference between these) is as big as standard closure**

Compare standard closure and residual term (after differentiation)



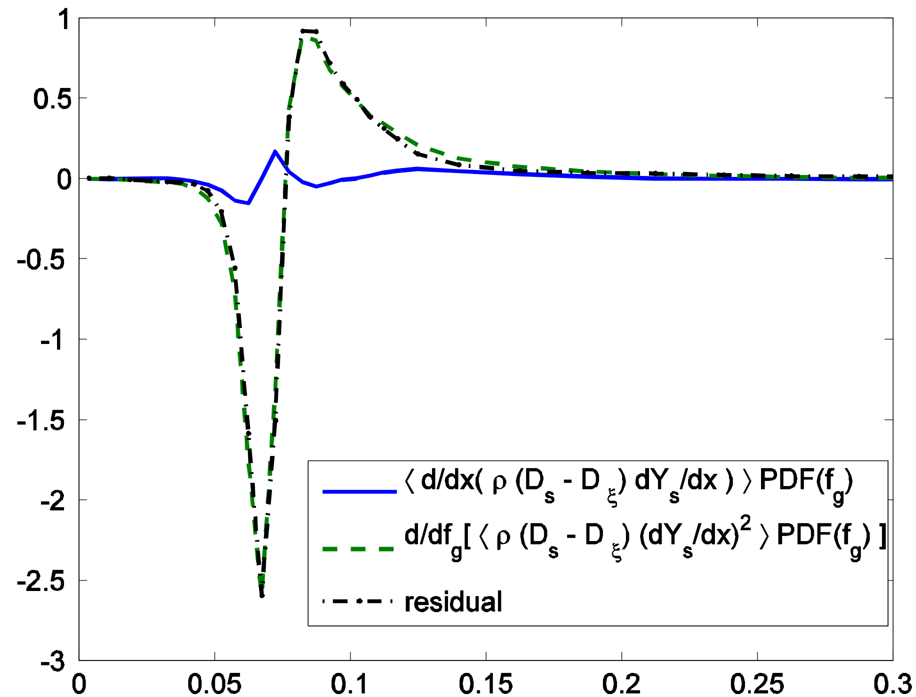
Differential-diffusion term in traditional CMC

$$\left\langle \nabla\cdot\left(\rho (D_k - D_\xi) \nabla Y_k \right) \middle| \vec{\eta} \right\rangle P(\vec{\eta})$$

- **Standard closure would use soot-dissipation rate.**

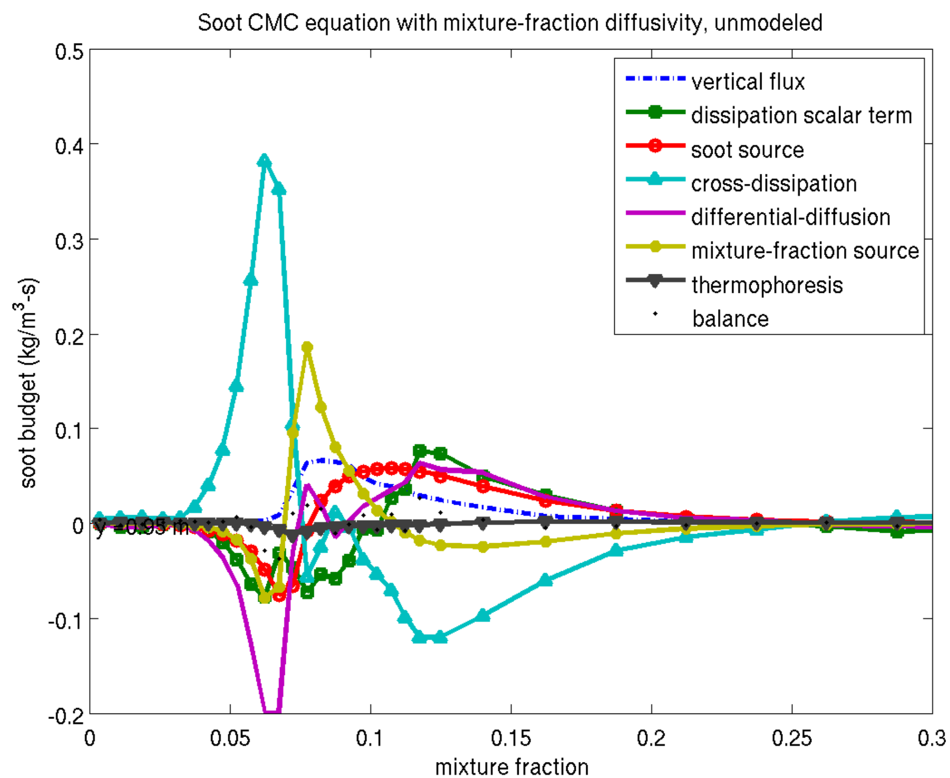
$$\left\langle \nabla\cdot\left(\rho (D_k - D_\xi) \nabla Y_k \right) \middle| \vec{\eta} \right\rangle P(\vec{\eta})$$
$$\approx \frac{\partial}{\partial \eta}\left[\left\langle \rho (D_k - D_\xi) \left(\nabla Y_k \right)^2 \middle| \vec{\eta} \right\rangle P(\vec{\eta}) \right]$$

  - **Statistics for high-Schmidt-number reacting-scalar dissipation rates are less available.**



So what about traditional CMC

- **Standard closures generally result in many large residual terms.**
  - **At least some of these residual terms will be difficult to model.**
  - **Fortunately, there is some cancellation of the residuals.**

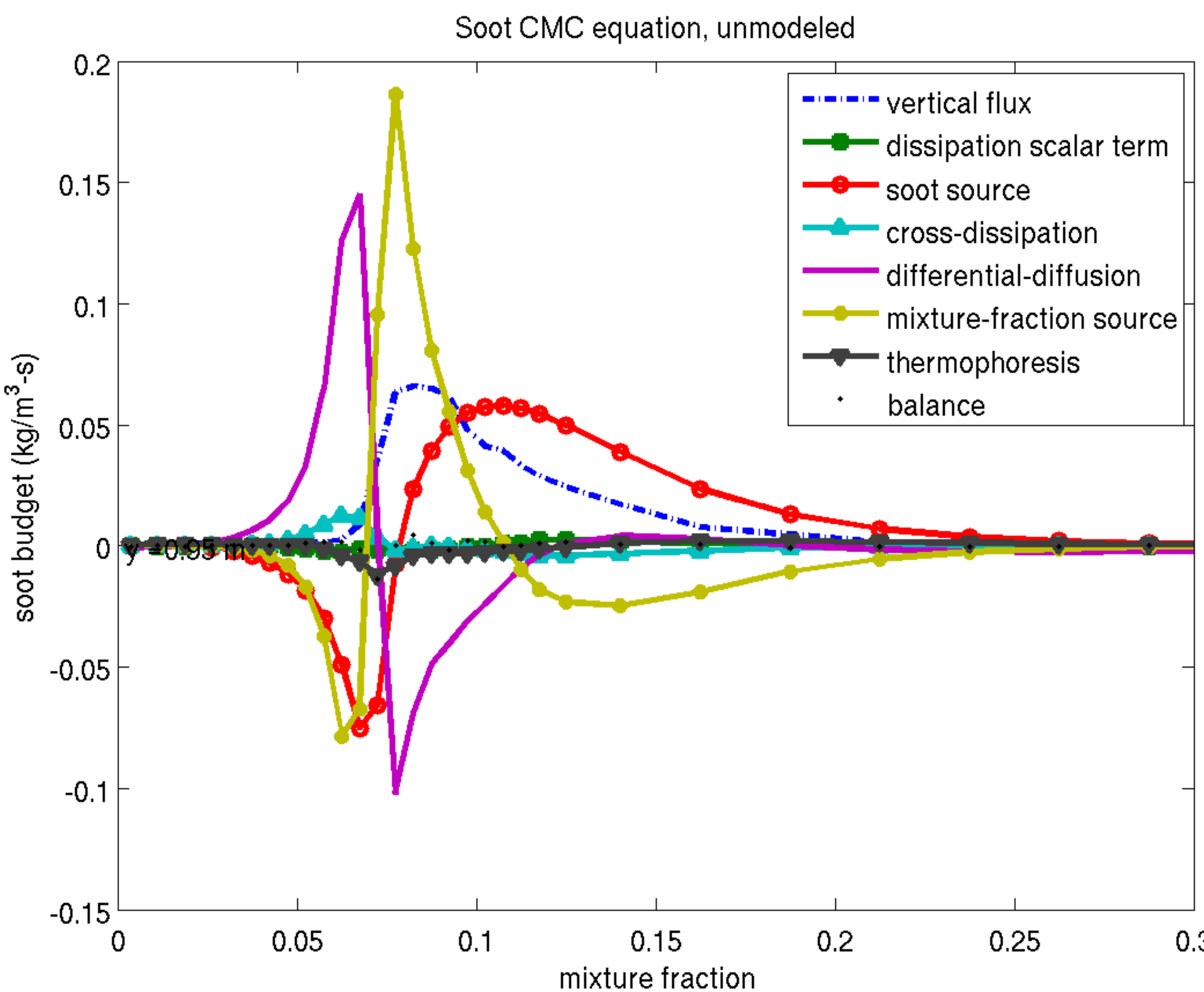


A alternate form of the CMC equations

- **It is possible to eliminate different terms in the process of deriving the conditional moment equations.**
- **Resulting CM equation has D_k, (for soot) in most places.**
  - **Many diffusive terms will be small.**

$$\begin{aligned}
\nabla\cdot\left[\left\langle \rho\vec{u}Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta})\right] = \; & \left\langle \rho\omega \middle| \vec{\eta}\right\rangle P(\vec{\eta}) \\
& +\frac{\partial}{\partial\eta}\left[\left\langle \rho D_k \nabla\xi\nabla Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta})\right] \\
& -\frac{\partial^2}{\partial\eta^2}\left[\left\langle \rho D_k \left(\nabla\xi\right)^2 Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta})\right] \\
& +\left\langle \nabla\cdot\left(\rho D_T Y_k \nabla \ln T\right) \middle| \vec{\eta}\right\rangle P(\vec{\eta}) \\
& -\frac{\partial}{\partial\eta}\left[\left\langle \nabla\cdot\left(\rho(D_\xi - D_k)\nabla\xi\right) Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta})\right]
\end{aligned}$$

Significant terms in alternate CMC



- **Soot diffusivity causes dissipation-scalar, cross-dissipation terms to be small.**
  - **Residuals remain but are trivially small.**
- **Differential diffusion is still substantial.**
  - **Standard model involves mixture fraction dissipation.**

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Differential-diffusion term in alternate CMC

$$\frac{\partial}{\partial \eta}\left[\left\langle \nabla \cdot \left(\rho (D_\xi - D_k) \nabla \xi \right) Y_k \middle| \vec{\eta} \right\rangle P(\vec{\eta})\right]$$

- **Standard closure:**

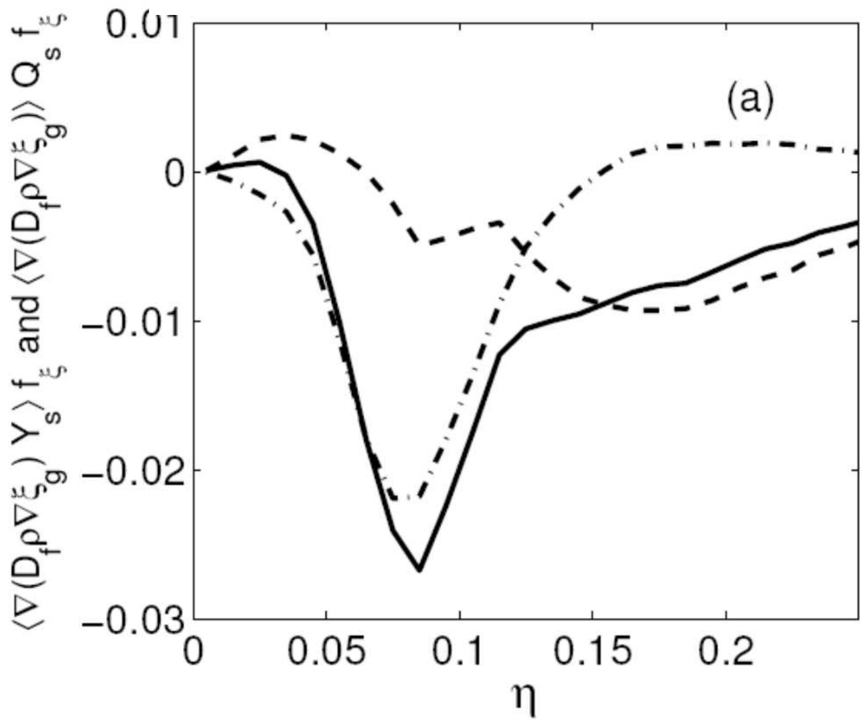
$$\left\langle \nabla \cdot \left(\rho (D_\xi - D_k) \nabla \xi \right) Y_k \middle| \vec{\eta} \right\rangle P(\vec{\eta})$$

$$\approx \left\langle \nabla \cdot \left(\rho (D_\xi - D_k) \nabla \xi \right) \middle| \vec{\eta} \right\rangle P(\vec{\eta}) Q_k$$

$$= \frac{\partial}{\partial \eta}\left(\left\langle \rho (D_\xi - D_k) \left(\nabla \xi \right)^2 \middle| \vec{\eta} \right\rangle P(\vec{\eta}) \right) Q_k$$

- **Residual is significant.**

Compare standard closure (before differentiation) and residual



So what about alternate CMC

- **Standard closures generally result in fewer large residual terms.**
  - **Need to look at differential diffusion terms. What is the physics behind these terms?**

$$\frac{\partial}{\partial\eta}\left[\left\langle \nabla\cdot\left(\rho(D_{\xi}-D_{k})\nabla\xi\right)Y_{k}\,\middle|\,\vec{\eta}\right\rangle P(\vec{\eta})\right] \approx \frac{\partial}{\partial\eta}\left[\left\langle \nabla\cdot\left(\rho(D_{\xi}-D_{k})\nabla\xi\right)\middle|\,\vec{\eta}\right\rangle P(\vec{\eta})Q_{k}\right]$$

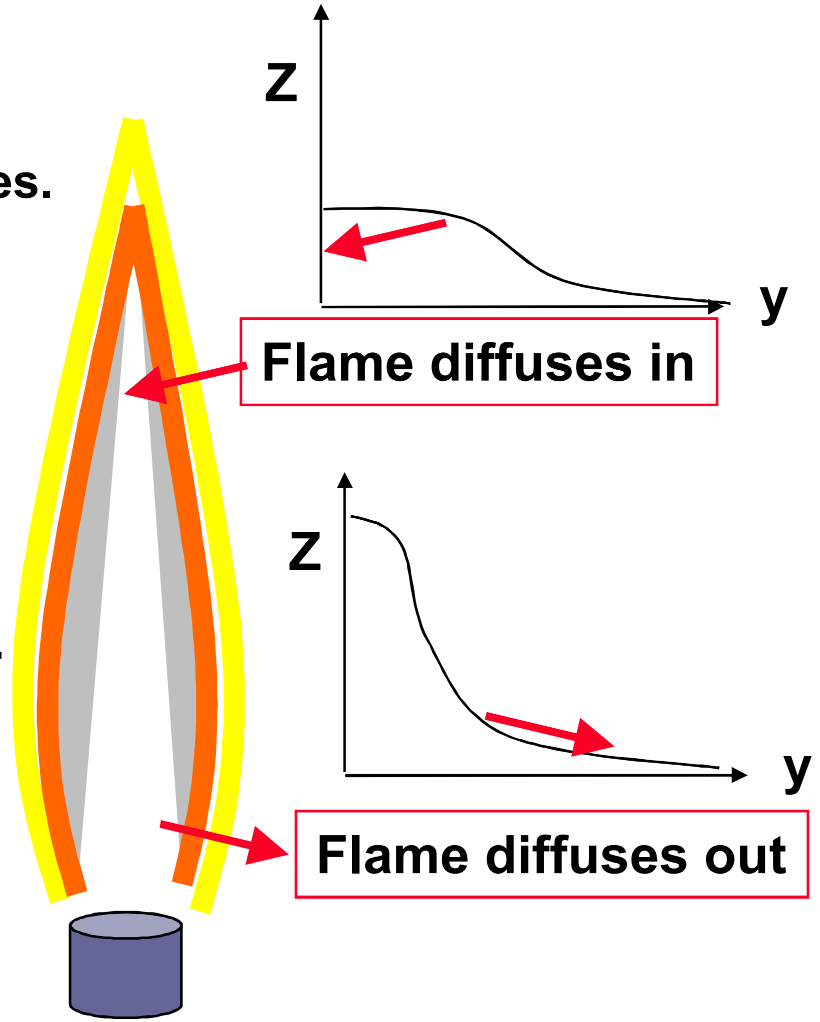
$$R_{DD} = \frac{\partial}{\partial\eta}\left[\left\langle \nabla\cdot\left(\rho(D_{\xi}-D_{k})\nabla\xi\right)Y_{k}\,\middle|\,\vec{\eta}\right\rangle P(\vec{\eta}) - \left\langle \nabla\cdot\left(\rho(D_{\xi}-D_{k})\nabla\xi\right)\middle|\,\vec{\eta}\right\rangle P(\vec{\eta})Q_{k}\right]$$

Differential diffusion of soot in flames due to mean mixture fraction evolution

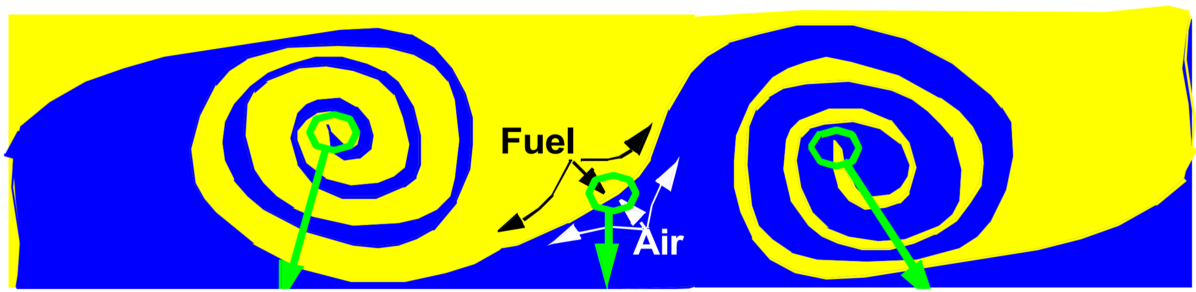
- Soot diffusion is
  - Slow relative to gas-phase species.
  - Affected by thermophoresis, etc.
- Soot is convected along with everything else.

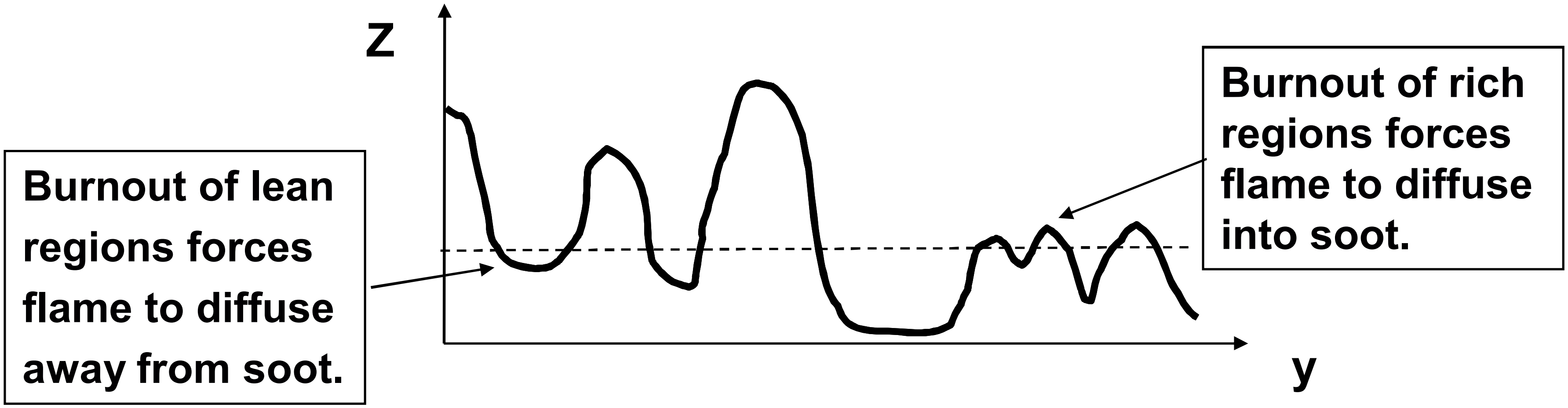
- Flames diffuse!
  - Diffuse towards location of nearest mixture fraction extrema.

  - Candle analogy suggests mean behavior: Related to evolution of mixture fraction PDF.



Differential diffusion of soot in flames due to mixture fraction fluctuations





Flame motion across fluid/soot elements is random walk about a mean

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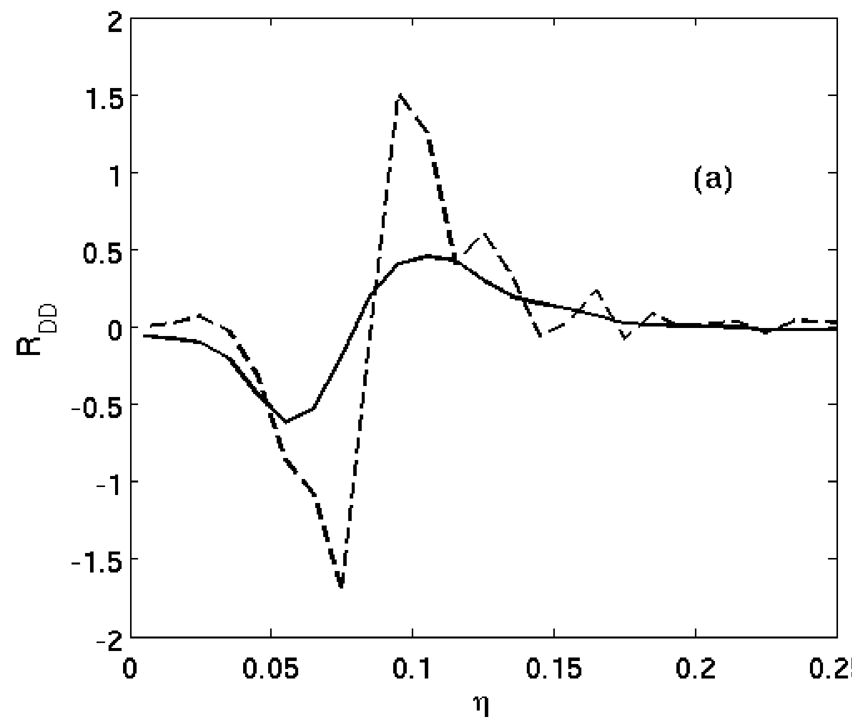
Advection due to mixture fraction fluctuations

- **Additional terms for diff-diff closure:**

$$R_{DD} = \frac{\partial}{\partial \eta}\left[\left\langle \nabla\cdot\left(\rho(D_\xi - D_k)\nabla\xi\right)Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta}) - \left\langle \nabla\cdot\left(\rho(D_\xi - D_k)\nabla\xi\right)\middle|\vec{\eta}\right\rangle P(\vec{\eta})Q_k\right]$$

- **The residual term describes the effect of fluctuating "advective" flows in mixture fraction space.**
  - **Correlation between random mixture fraction fluctuations and soot moments.**
  - **Model with diffusive process?**

$$R_{DD} \approx \frac{\rho_\eta \chi_\eta f_\xi}{2Le_{DD,t}} \frac{\partial^2 Q_s}{\partial \eta^2}$$



Advection due to mixture fraction fluctuations

- **Additional terms for diff-diff closure:**

$$R_{DD} = \frac{\partial}{\partial \eta}\left[\left\langle \nabla \cdot \left(\rho (D_{\xi} - D_{k}) \nabla \xi \right) Y_{k} \middle| \vec{\eta} \right\rangle P(\vec{\eta}) - \left\langle \nabla \cdot \left(\rho (D_{\xi} - D_{k}) \nabla \xi \right) \middle| \vec{\eta} \right\rangle P(\vec{\eta}) Q_{k} \right]$$

- **The residual term describes the effect of fluctuating "advective" flows in mixture fraction space.**
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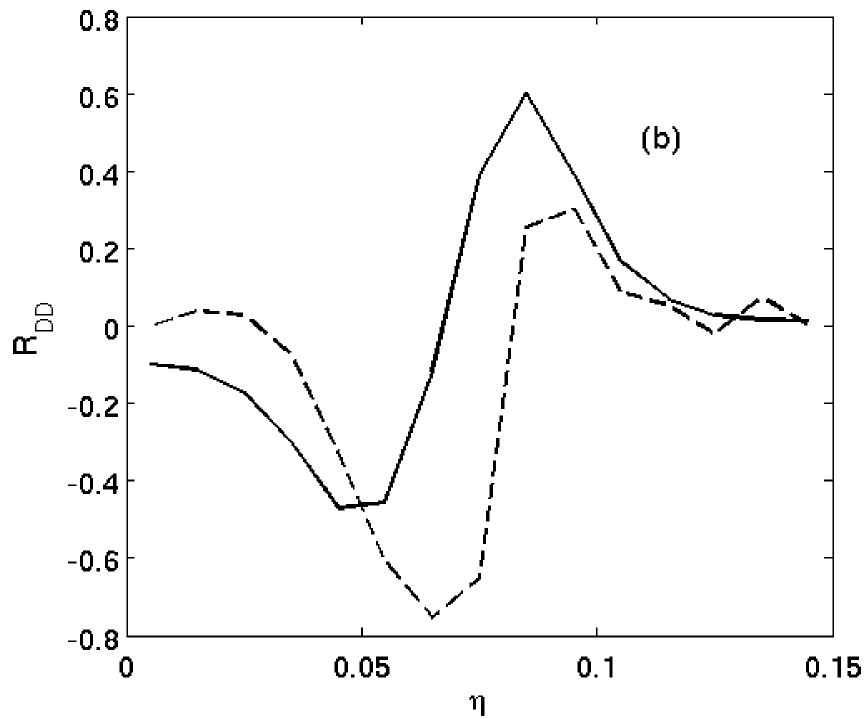

Advection due to mixture fraction fluctuations

- **Additional terms for diff-diff closure:**

$$R_{DD} = \frac{\partial}{\partial \eta}\left[\left\langle \nabla \cdot \left(\rho (D_\xi - D_k) \nabla \xi \right) Y_k \middle| \vec{\eta} \right\rangle P(\vec{\eta}) - \left\langle \nabla \cdot \left(\rho (D_\xi - D_k) \nabla \xi \right) \middle| \vec{\eta} \right\rangle P(\vec{\eta}) Q_k \right]$$

- **The residual term describes the effect of fluctuating "advective" flows in mixture fraction space.**
  - **Correlation between random mixture fraction fluctuations and soot moments.**
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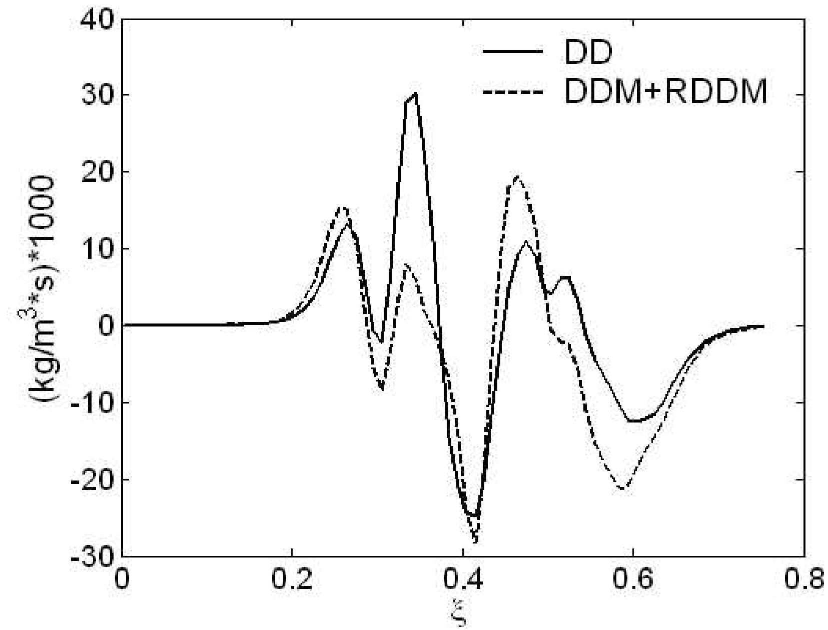

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Summary

- Conditional moment equations extended to allow general diffusion models.
- This opens options as to what terms need to be modeled.

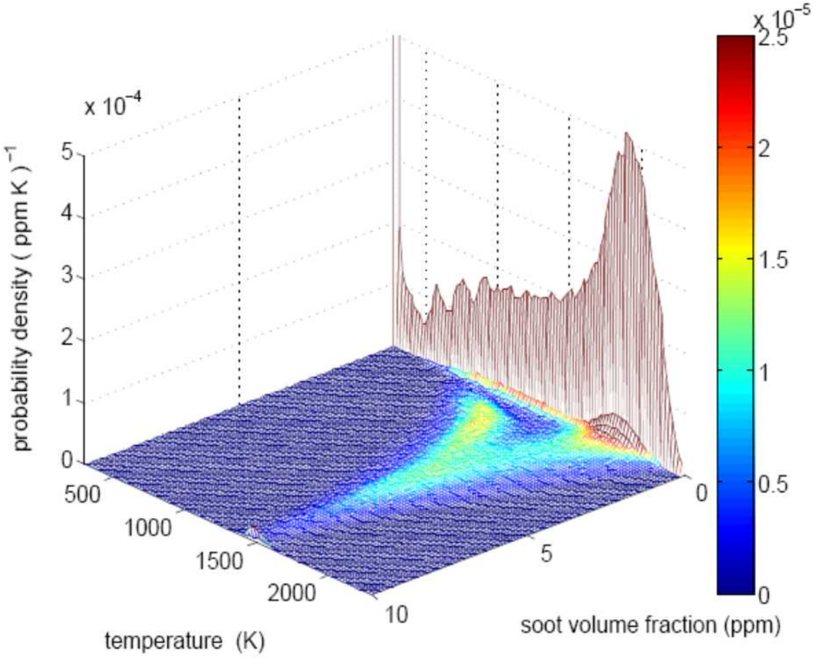
- For soot at least, the alternate approach presented here appears preferable:
  - Differential diffusion of soot relative to mixture fraction dominates.
  - Mean component of transport related to PDF(Z) evolution.
  - Fluctuating component related to dissipative processes using new model.

- Results with ODT study suggest model captures relevant physics.

Backup slides

ODT simulations provide high-fidelity data to evaluate closures

- **Buoyant 1 m wide ethene plume (line fire) spatially evolving ODT simulation.**
- **Simple soot model (Fairweather *et al.* 1992) with steady laminar flamelet source terms tabulated by enthalpy and mixture fraction.**
- **Generate statistical quantities like soot-temperature joint PDF.**



Additional key closure requirements for CMC

- For particles, differential diffusion is key physics.
- "New" CMC formulation retains a residual fluctuation term.
- Retains conservative form with PDF

Conditionally Averaged

$$\frac{\partial\left(\rho_\eta Q_i f_\xi\right)}{\partial t}+\nabla\cdot\left(\left\langle\rho\vec{u}\,|\,\eta\right\rangle Q_i f_\xi\right)=\left\langle\rho\omega_i\,|\,\eta\right\rangle f_\xi+\frac{\rho_\eta\chi_\eta f_\xi}{2Le_i}\frac{\partial^2 Q_i}{\partial\eta^2}-\frac{\partial^2}{\partial\eta^2}\left(\frac{\rho_\eta\chi_\eta f_\xi}{2}\right)Q_i$$

$$-\left(\frac{D_\xi-D_i}{D_\xi}\right)\frac{\partial}{\partial\eta}\left(\frac{\rho_\eta\chi_\eta f_\xi}{2}\right)\frac{\partial Q_i}{\partial\eta}-R_{DD}$$

- These terms describe the effect of the mean and fluctuating "convective" flows in mixture fraction space.
  - Diffusion of mixture fraction relative to soot

Additional key closure requirements for CMC

- For particles, differential diffusion is key physics.
- "New" CMC formulation retains a residual fluctuation term.

Conditionally Averaged

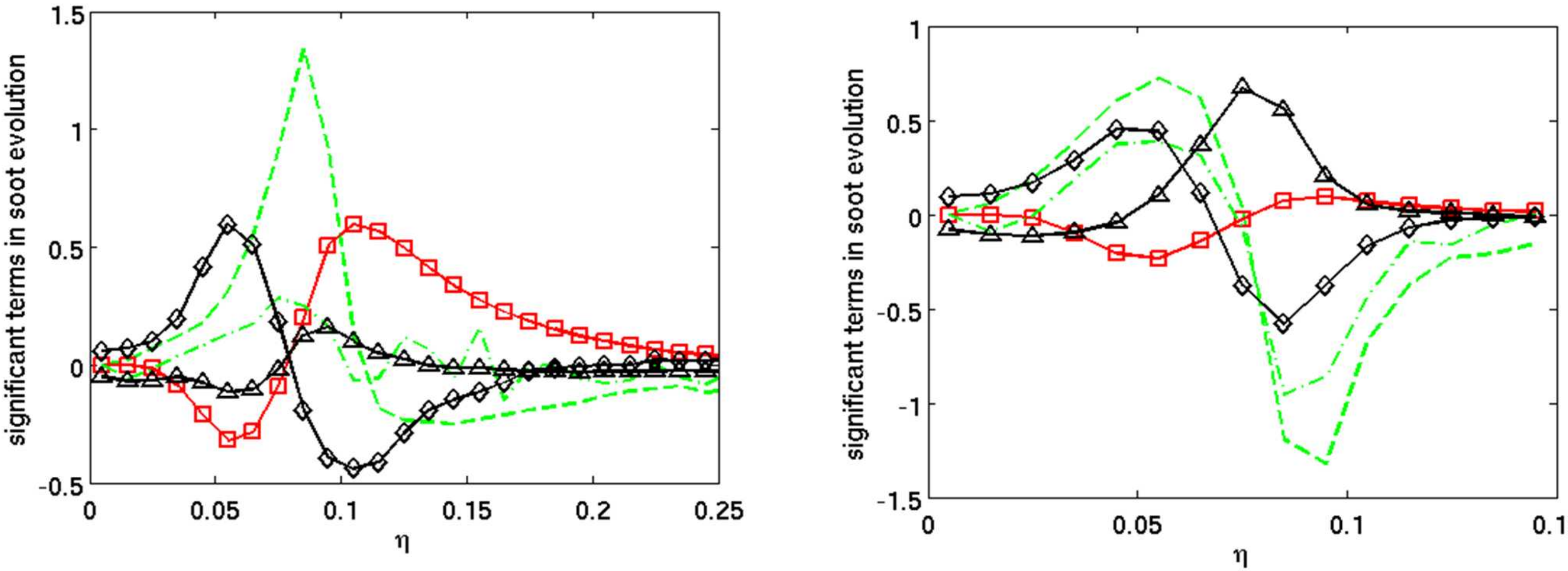
$$\frac{\partial\left(\rho_\eta Q_i f_\xi\right)}{\partial t}+\nabla\cdot\left(\left\langle\rho\vec{u}\,|\,\eta\right\rangle Q_i f_\xi\right)=\left\langle\rho\omega_i\,|\,\eta\right\rangle f_\xi+\frac{\rho_\eta\chi_\eta f_\xi}{2Le_i}\frac{\partial^2 Q_i}{\partial\eta^2}-\frac{\partial^2}{\partial\eta^2}\left(\frac{\rho_\eta\chi_\eta f_\xi}{2}\right)Q_i$$

$$-\left(\frac{D_\xi-D_i}{D_\xi}\right)\frac{\partial}{\partial\eta}\left(\frac{\rho_\eta\chi_\eta f_\xi}{2}\right)\frac{\partial Q_i}{\partial\eta}-R_{DD}$$

- Boxed term represents the mean evolution of the mixture composition

ODT results

•Terms plotted below for heights in ODT simulations where mixture fraction pdf is centered on production (left) and on oxidation (right).



Advection (dash), pdf flux (dash-dot) -- long-term evolution of soot.
Soot source (squares).
Diff-diff by evolution of pdf (triangles) -- long-time advection in mixture fraction.
Diff-diff fluctuations R_{DD} (diamonds) -- short-time diffusion in mixture fraction.

Dissipation-scalar term in Klimenko-Bilger CMC

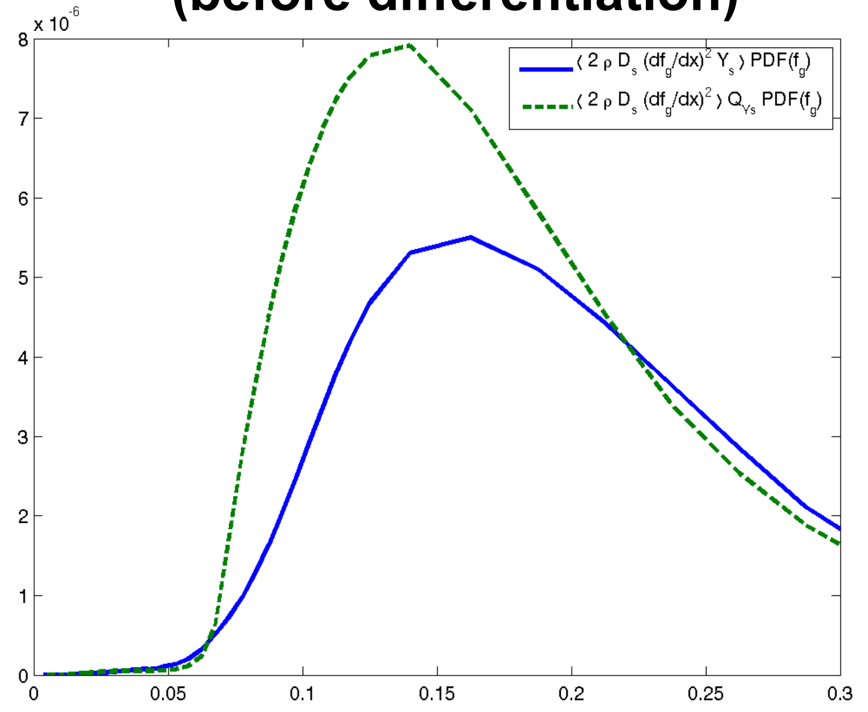
$$\frac{\partial^2}{\partial \eta^2}\left[\left\langle \rho D_\xi \left(\nabla \xi\right)^2 Y_k \middle| \vec{\eta} \right\rangle P(\vec{\eta})\right]$$

- **Standard closure**

$$\left\langle \rho D_\xi \left(\nabla \xi\right)^2 Y_k \middle| \vec{\eta} \right\rangle P(\vec{\eta})$$

$$= \left\langle \rho D_\xi \left(\nabla \xi\right)^2 \middle| \vec{\eta} \right\rangle Q_k P(\vec{\eta})$$

Compare standard closure (before differentiation)



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Dissipation-scalar term in Klimenko-Bilger CMC

- **Basic term**

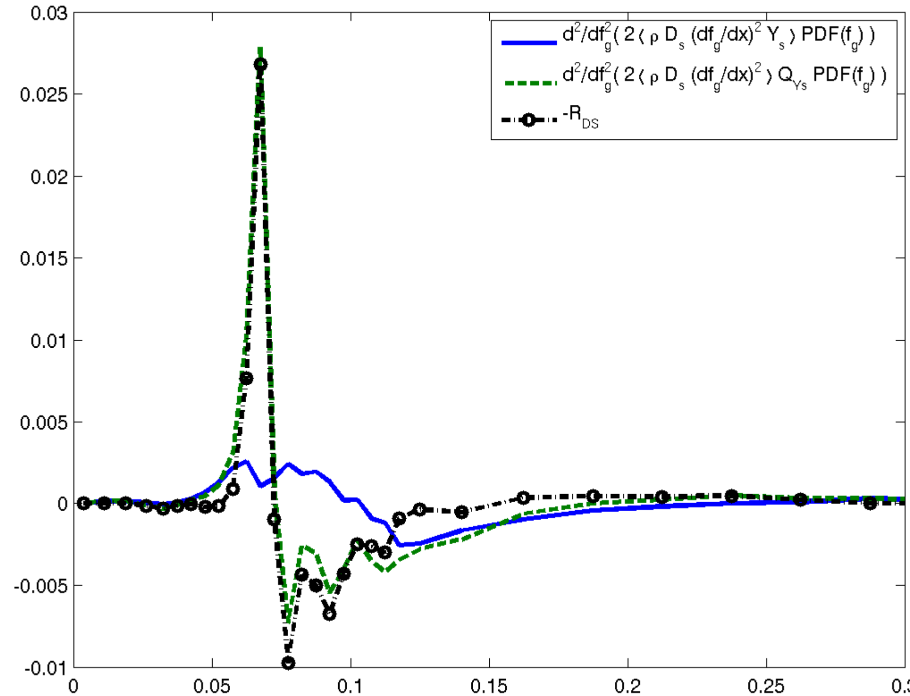
$$\frac{\partial^2}{\partial \eta^2}\left[\left\langle \rho D_\xi (\nabla \xi)^2 Y_k \middle| \vec{\eta} \right\rangle P(\vec{\eta})\right]$$

- **Standard closure**

$$\frac{\partial^2}{\partial \eta^2}\left[\left\langle \rho D_\xi (\nabla \xi)^2 \middle| \vec{\eta} \right\rangle Q_k P(\vec{\eta})\right]$$

- **Residual (difference between these) is as big as standard closure**

Compare standard closure and residual term (after differentiation)



Cross-dissipation term in Klimenko-Bilger CMC

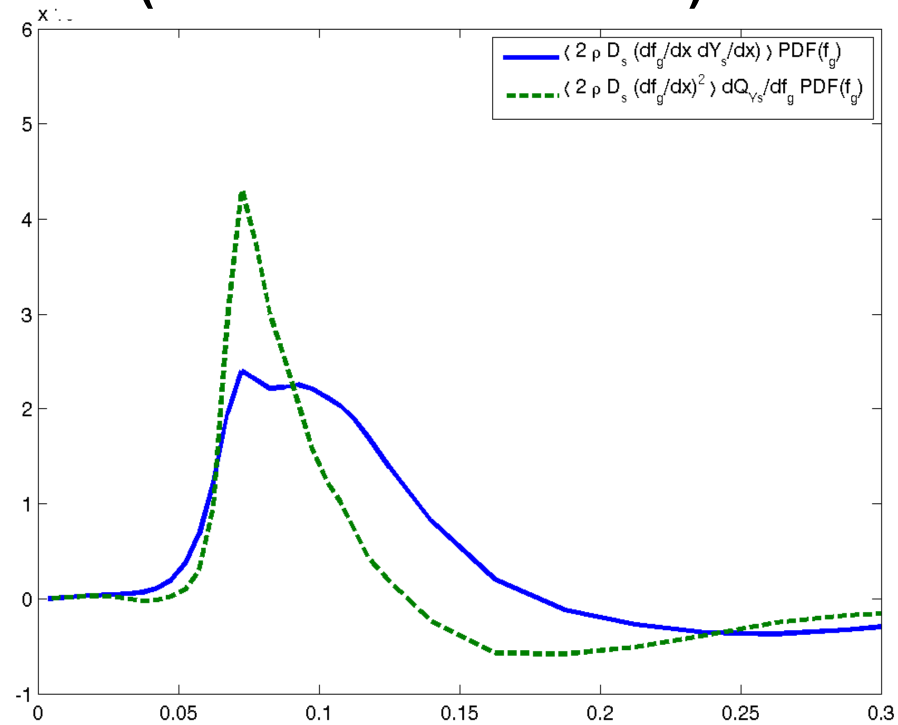
$$\frac{\partial}{\partial \eta}\left[\left\langle \rho D_{\xi} \nabla \xi \nabla Y_{k} \middle| \vec{\eta} \right\rangle P(\vec{\eta})\right]$$

- **Standard closure**

$$\left\langle \rho D_{\xi} \nabla \xi \nabla Y_{k} \middle| \vec{\eta} \right\rangle P(\vec{\eta})$$

$$\approx \left\langle \rho D_{\xi} \left(\nabla \xi\right)^{2} \middle| \vec{\eta} \right\rangle \frac{\partial Q_{k}}{\partial \eta} P(\vec{\eta})$$

Compare standard closure (before differentiation)



Cross-dissipation term in Klimenko-Bilger CMC

- **Basic term**

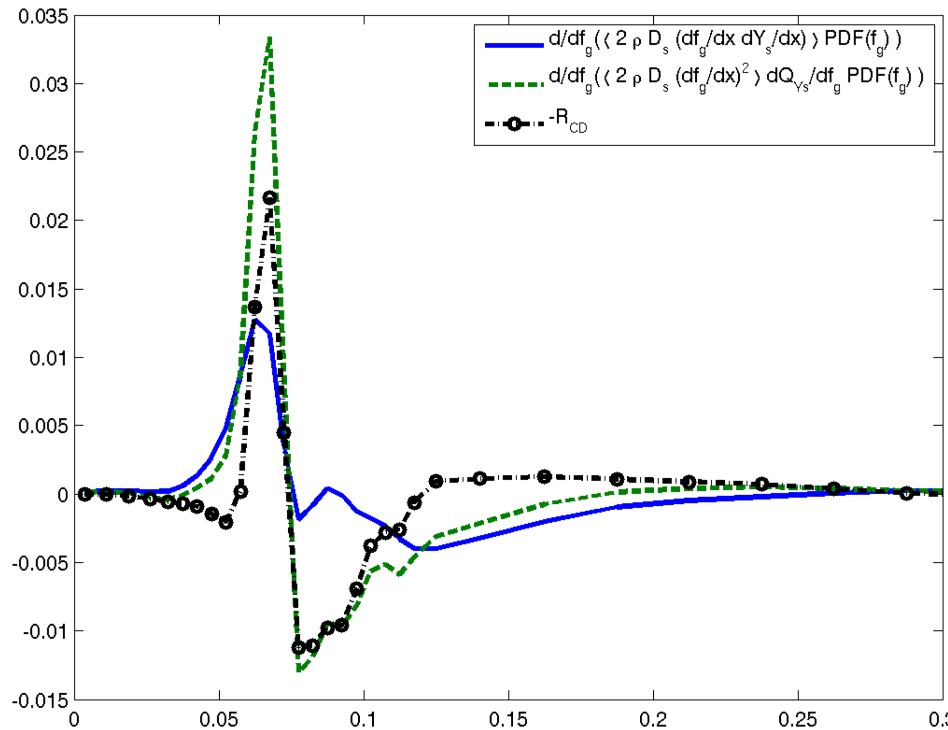
$$\frac{\partial}{\partial \eta}\left[\left\langle \rho D_{\xi} \nabla \xi \nabla Y_{k} \,\middle|\, \vec{\eta} \right\rangle P(\vec{\eta})\right]$$

- **Standard closure**

$$\frac{\partial}{\partial \eta}\left[\left\langle \rho D_{\xi} \left(\nabla \xi\right)^{2} \,\middle|\, \vec{\eta} \right\rangle \frac{\partial Q_{k}}{\partial \eta} P(\vec{\eta})\right]$$

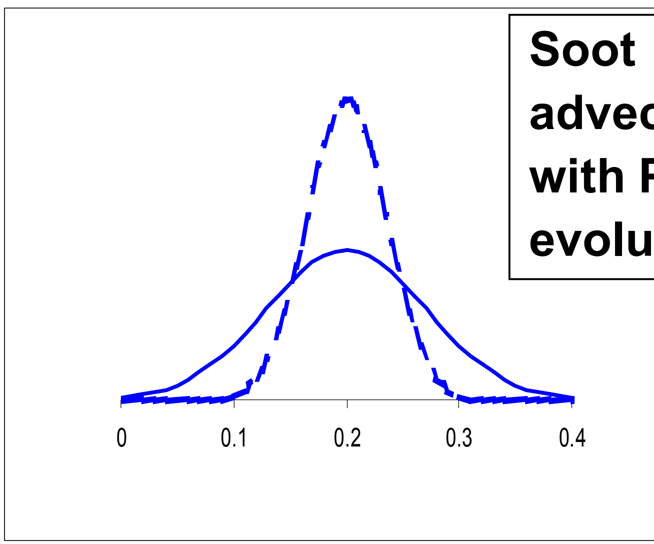
- **Residual (difference between these) is as big as standard closure**

Compare standard closure and residual term (after differentiation)



Soot advection in mixture fraction space

- **How does soot advection work in turbulent flows?**
  - **In CMC-based approach there is advection in the mixture fraction coordinate.**
  - **Candle analogy suggests mean behavior: Related to evolution of mixture fraction PDF.**



Soot advected with PDF evolution

$$\frac{\partial}{\partial\eta}\left\langle \nabla\cdot\left(\rho(D_\xi - D_k)\nabla\xi\right)Y_k \middle| \vec{\eta}\right\rangle P(\vec{\eta})$$

$$\approx \frac{\partial}{\partial\eta}\left[\left\langle \nabla\cdot\left(\rho(D_\xi - D_k)\nabla\xi\right)\middle|\vec{\eta}\right\rangle P(\vec{\eta})Q_k\right]$$