Soot Property Reconstruction from Flame Emission Spectrometry

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MOTIVATION

Characterization of Soot in Flames

- Strong sensitivity of soot mechanisms to temperature and flow gradients implies the necessity for in-situ, nonintrusive measurement techniques.
- Electromagnetic radiation serves as a suitable agent for nonintrusive characterization due to convenient probing in hostile environments and high resolution
- Radiative properties of soot particles depend on a wide range of soot characteristics: spherule size, aggregate morphology (fractal dimension), aggregate size, refractive index, temperature, volume fraction

MOTIVATION

Flame Emission Spectrometry for Soot Diagnostics

Within the spectral windows where combustion gases are transparent, soot emission prevails as a continuum spectra

For optically thin flames, line-of- sight flame emission at near IR range is determined by the following soot properties

- Temperature,
- Volume fraction,
- Refractive index function $E(m) = \mathrm{Im}\left(\frac{m^2-1}{m^2+2}\right)$

Emission spectrometry measurements can be used to infer these properties

MOTIVATION

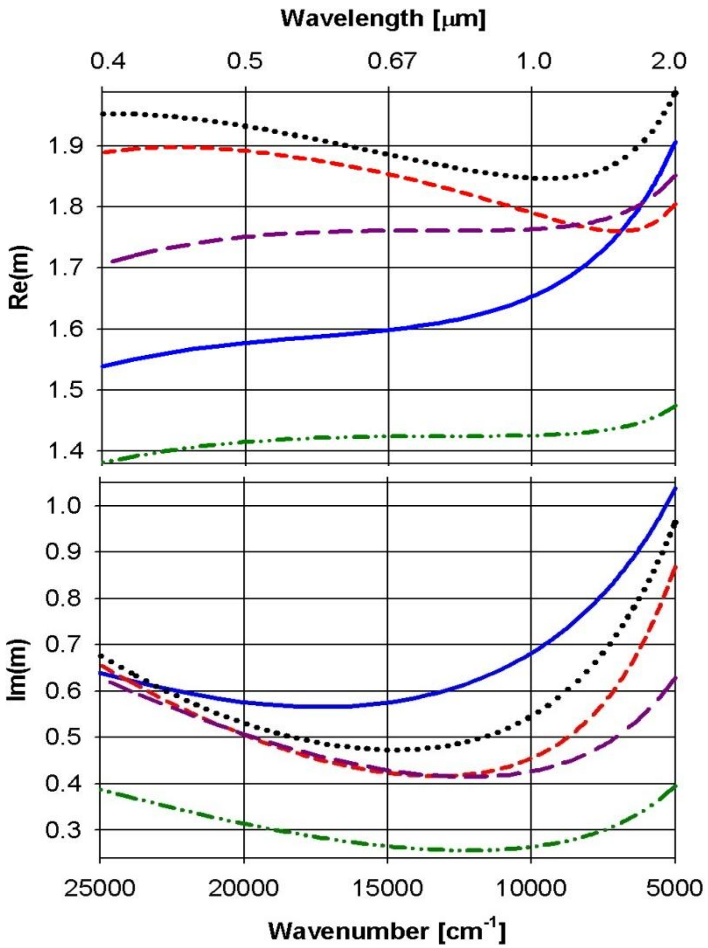
- In previous studies, reconstruction of T, f_v or T, κ from tomographic analysis of flame emission spectrometry was carried out by selecting a refractive index from literature
- Spectral variation of E(m) is commonly neglected

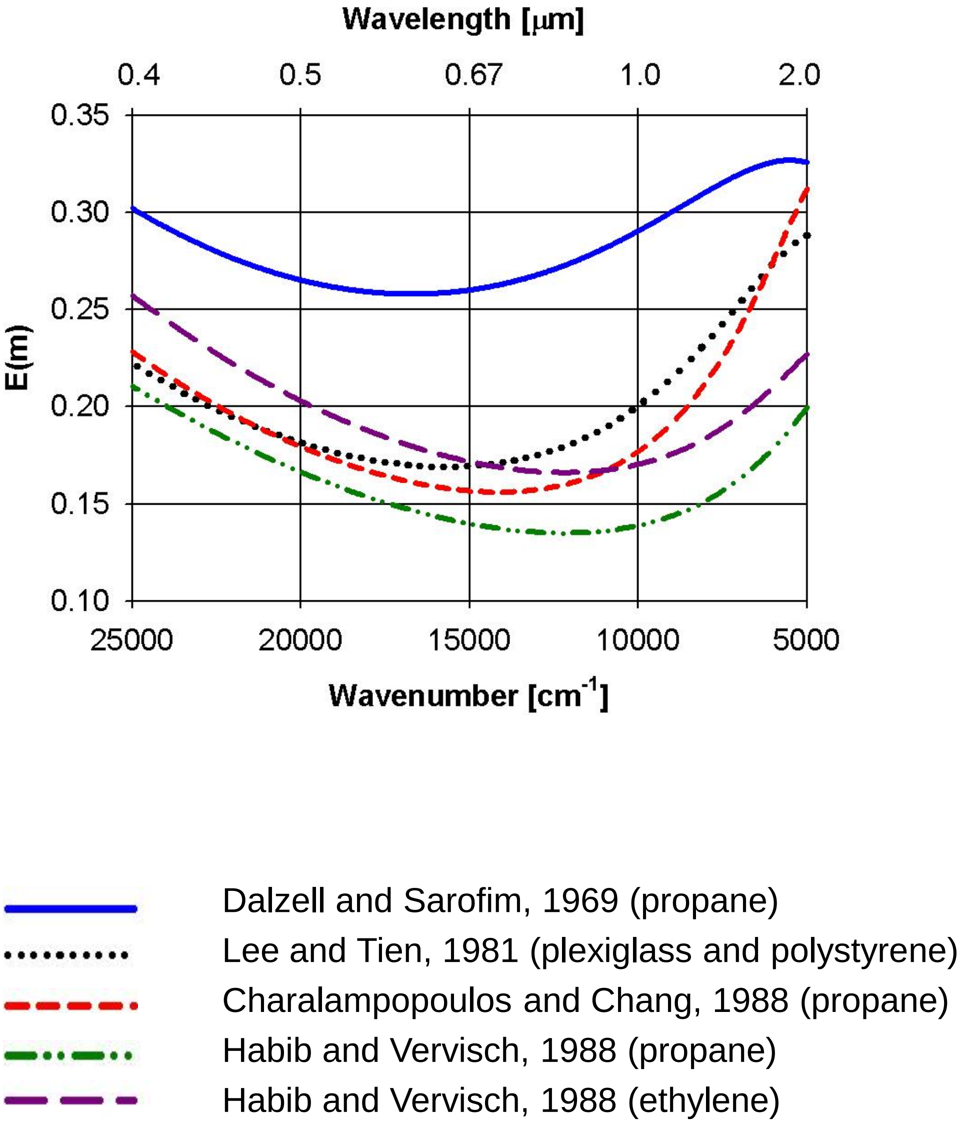
Towards near IR

- Sensitivity of E(m) to wavenumber increases
- Spectral variation of emission intensities depend on T and E(m)

Characteristic information on refractive index can be extracted from spectral variation of emission

OPTICAL CONSTANTS





- Drude-Lorenz dispersion model
- Dispersion parameters suggested in literature yield significant variation

OPTICAL CONSTANTS

Soot refractive index

- One of the most important limitations of optical soot characterization techniques is uncertainty in complex refractive index of soot.
- Considerable variation in reported values point out the sensitivity of this parameter to flame conditions.
- Local variations of refractive index within a flame, its dependence on temperature, fuel type and H/C ratio are among reported findings.

OBJECTIVE

To develop, validate and apply a nonintrusive soot diagnostics methodology for in-situ determination of temperature, volume fraction and refractive index of soot aggregates formed inside small-scale flames by using near-infrared emission spectrometry.

METHODOLOGY

Principle of radiative transfer based nonintrusive particle characterization

Particle properties (complex index of refraction, temperature, concentration, size, shape, morphology)

- Direct Model -
Physical model that governs propagation of radiation in particulate medium

Radiative parameters (**emission**, transmission, scattering)

- Measurement -
Radiative parameters

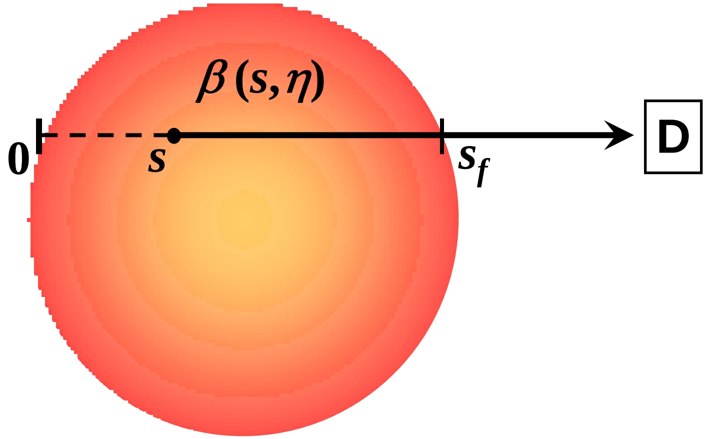
- Inverse model -
Inverse analysis of direct model

DESIRED PARAMETERS
Particle properties

OUTLINE

- INVERSE MODEL – Soot property reconstruction
    - Description of methodology
    - Data conditioning in the presence of noise
- VALIDATION – Simulated experiments by a direct model
    - Effects of physical assumptions
    - Effects of experimental limitations
    - Performance of data conditioning
- APPLICATION – Ethylene diffusion flame

L-o-S TRANSFER OF RADIATIVE EMISSION



Cross section of a vertical, axisymmetric, lab-scale flame

Line of sight spectral emission intensity

$$I_\eta\Big|_x = \int_0^{s_f} \underbrace{\left[\kappa(s,\eta)\cdot I_{b\eta}(s)\right]}_{\text{Emission}} \cdot \underbrace{\exp\left[-\int_s^{s_f}\beta(s',\eta)ds'\right]}_{\text{Self-extinction}} ds$$

Assumption: **Optically thin flame ➡ negligible self-absorption**

DETERMINATION OF REFRACTIVE INDEX

Extraction of a refractive index parameter Ψ**-function** from l-o-s emission intensities

$$I_\eta\Big|_x = \int_0^{s_f} \left[\kappa(s,\eta)\cdot I_{b,\eta}(s)\right] ds$$

- Wien's approximation to Planck function $$I_{b\eta}(s) = 2hc_0{}^2\eta^3 \exp\left(-\frac{hc_0\eta}{kT(s)}\right)$$
- soot absorption coefficient follows Rayleigh regime $$\kappa(s,\eta) = 6\pi\eta\cdot f_v(s)\cdot E_m$$
- Spatial dependence of *E*$_m$ function negligible along the path $$E_m = \mathrm{Im}\left(\frac{m^2-1}{m^2+2}\right)$$

$$I_\eta = B_0\eta^4 \boldsymbol{E_m(\eta)} \int_0^{s_f} \boldsymbol{f_v(s)}\cdot \exp\left[-\frac{B_1\eta}{\boldsymbol{T(s)}}\right] ds$$

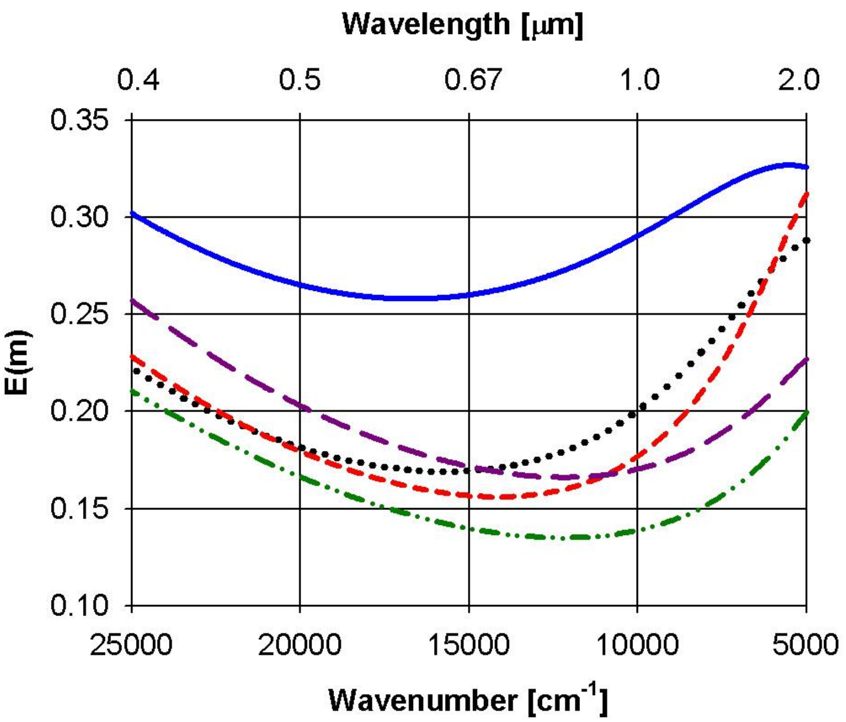
it can be shown that

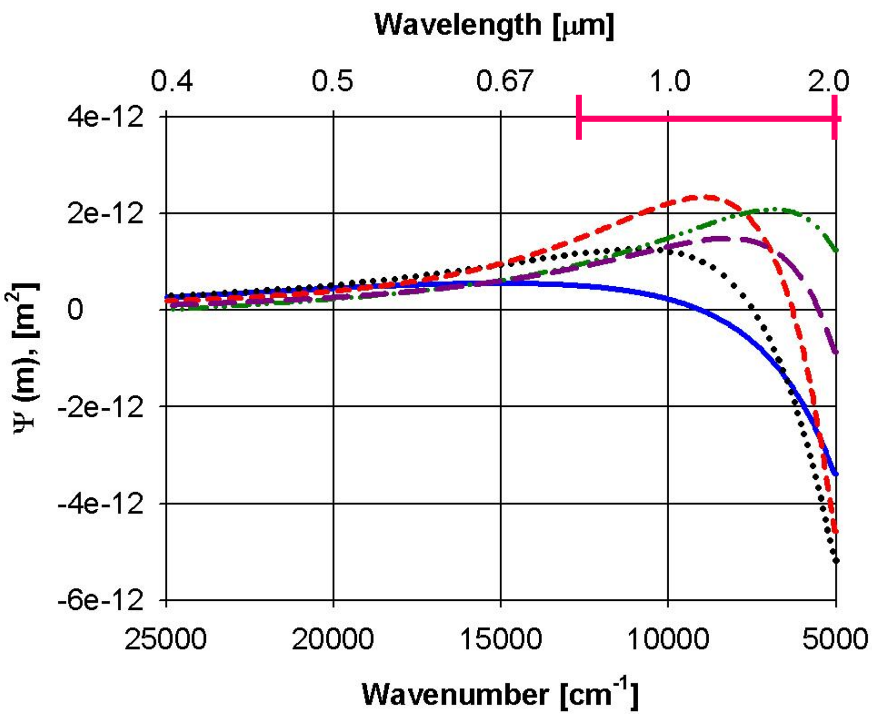
$$\boxed{\frac{\partial}{\partial\eta}\left(\frac{1}{I_\eta/\eta^4}\cdot\frac{\partial\ I_\eta/\eta^4}{\partial\eta}\right) = \frac{\partial}{\partial\eta}\left(\frac{1}{E_m}\cdot\frac{\partial\ E_m}{\partial\eta}\right) = \Psi}$$

depends only on refractive index

➡ It is possible to isolate refractive index information from measured radiative intensities

REFERENCE REFRACTIVE INDEX FUNCTION

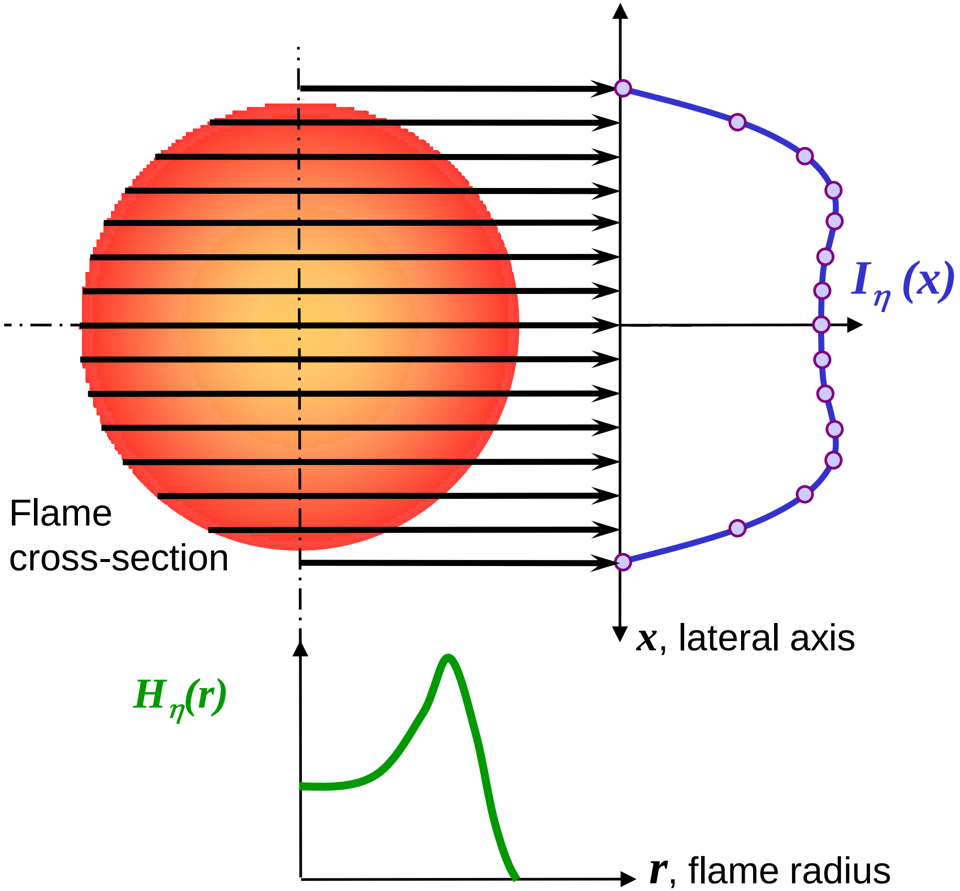




✓ Ψ-function can be used as a characteristic function

➡ The set which provides best fit to experimental Ψ-function is used to determine spectral refractive index of soot

TOMOGRAPHIC RECONSTRUCTION



$$I_\eta\Big|_x \approx \int_0^{s_f}\left[\kappa(s,\eta)\cdot I_{b,\eta}(s)\right]ds = \int_0^{s_f} H_\eta(s)ds$$

Line-of-sight spectral emission intensity

$I_\eta\,(x)$

MEASURED

⬇

1-D Tomography

Assumptions:
- Axisymmetry
- Infinitely small beam width

⬇

Radial emission source term field

$H_\eta(r)$

INFERRED

TEMPERATURE AND VOLUME FRACTION

Once refractive index dispersion model is selected E(m) can be evaluated

$$\Phi_\eta(r) = \ln\underset{\checkmark}{\underline{\left(\frac{H_\eta(r)}{\eta^4}\right)}} - \ln\ \underset{\checkmark}{\underline{E_m(r,\eta)}}\ = -\frac{B_1}{\underset{?}{\underline{\boldsymbol{T(r)}}}}\eta + \ln(B_0 \cdot \underset{?}{\underline{\boldsymbol{f_v(r)}}})$$

Linear regression to Φ_η vs η yields

$$T(r_i) = -B_1 / slope$$

$$f_v(r_i) = \exp(intercept) / B_0$$

DATA CONDITIONING FOR NOISE REDUCTION

- Spectral smoothing
  - moving average filtering,
  - model function fitting

$$I_\eta\Big|_x = B_0\eta^4 \boldsymbol{E_m(\eta)} \int_0^{s_f} \boldsymbol{f_v(r)} \cdot \exp\left[-\frac{B_1\eta}{\boldsymbol{T(r)}}\right] ds \quad ⇨ \quad I_{fit}(\eta) = \eta^4 \cdot \exp(a_0 + a_1\eta + a_2\eta^2 + a_3\eta^3)$$

$$\Psi = \frac{\partial}{\partial\eta}\left(\frac{1}{I_\eta/\eta^4} \cdot \frac{\partial\ I_\eta/\eta^4}{\partial\eta}\right) = 2a_2 + 6a_3\eta$$

- Flame center determination

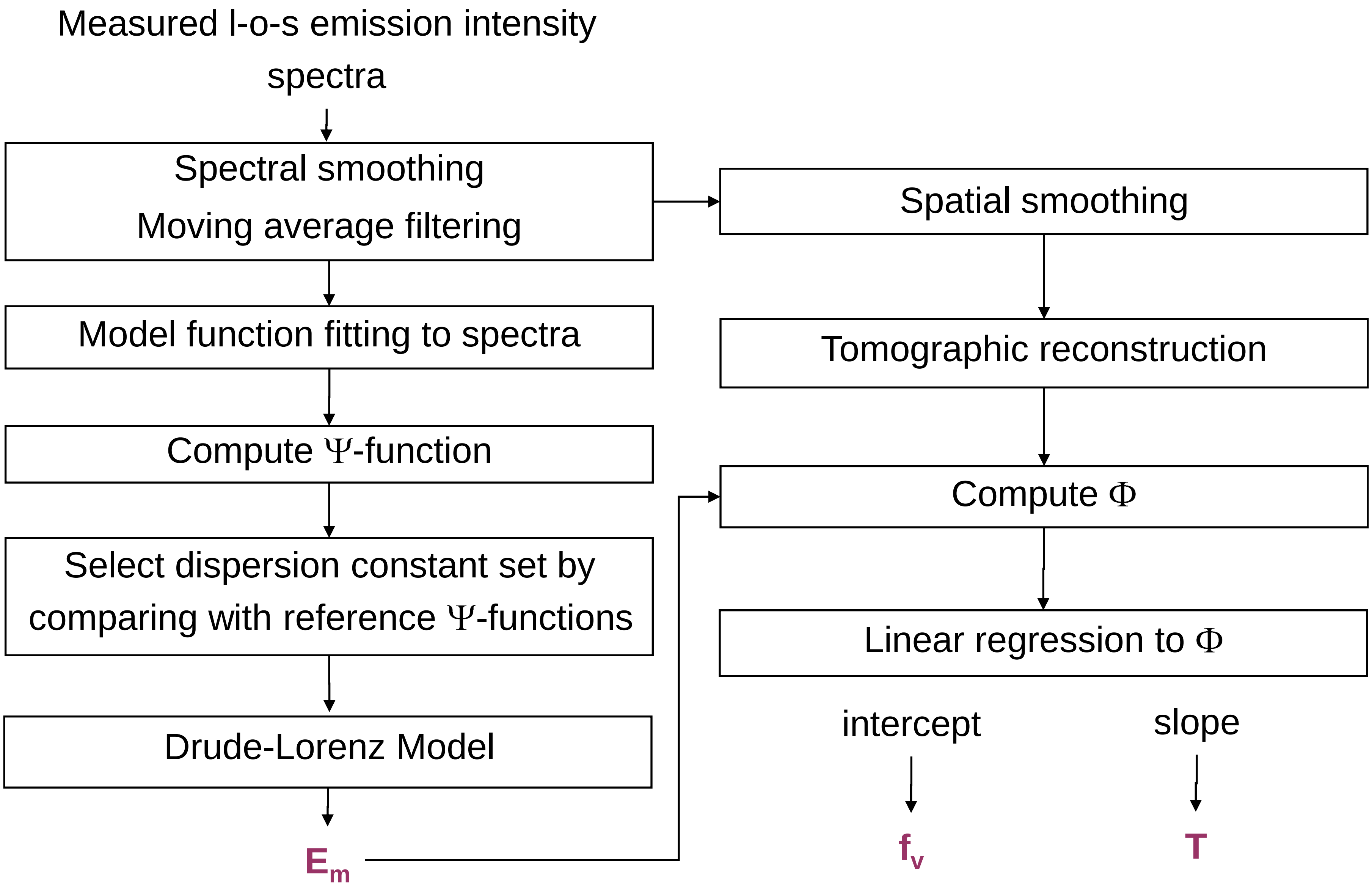
  6th order polynomial fitted to lateral profiles

  derivative=0 point accepted as centre

- Spatial smoothing

  B-Splines: piecewise 4th order polynomials fitted with matching derivatives at intersection knots

INVERSION ALGORITHM



Measured l-o-s emission intensity spectra

Spectral smoothing

Moving average filtering

Model function fitting to spectra

Compute Ψ-function

Select dispersion constant set by comparing with reference Ψ-functions

Drude-Lorenz Model

E_m

Spatial smoothing

Tomographic reconstruction

Compute Φ

Linear regression to Φ

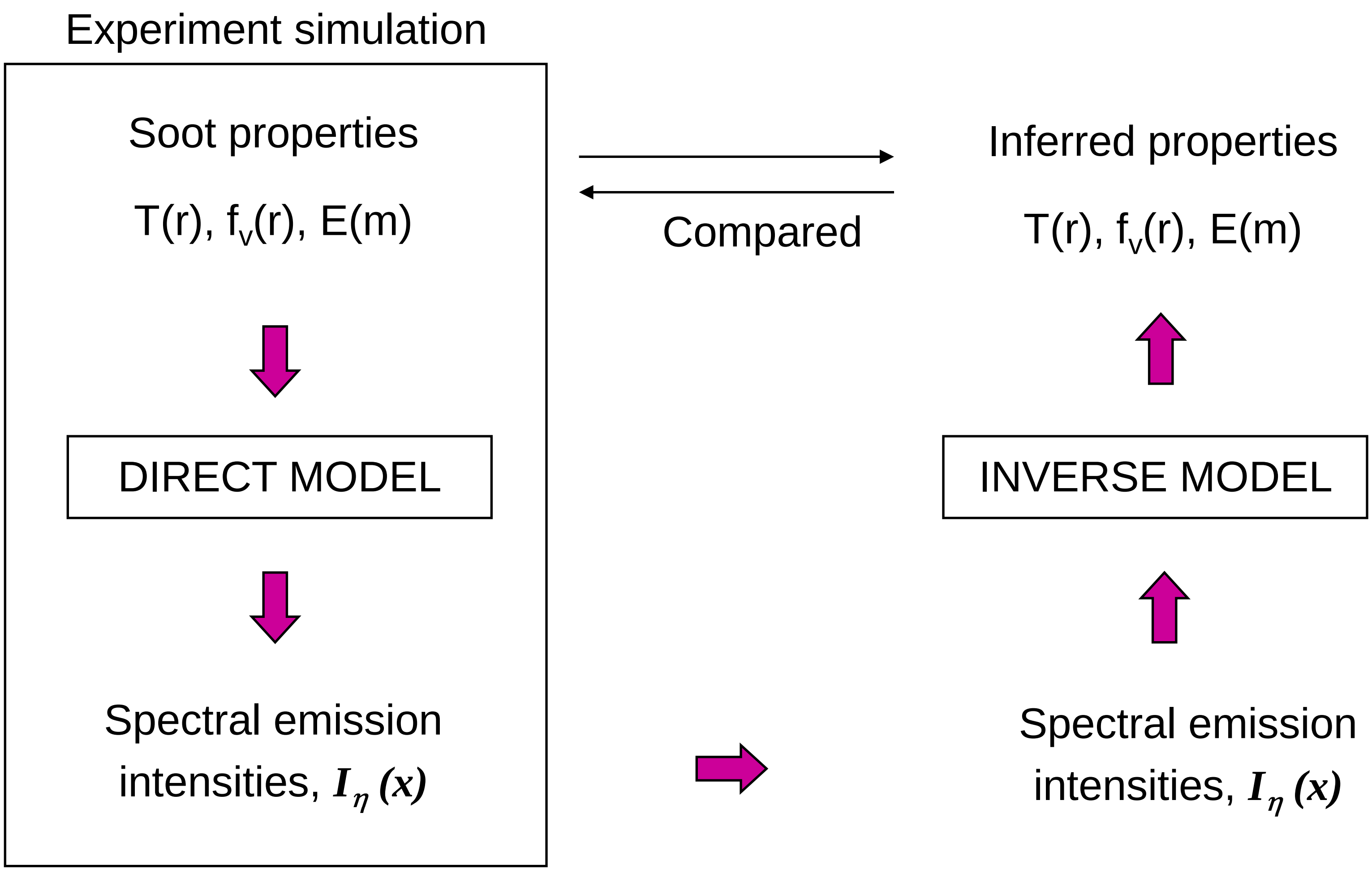
intercept

f_v

slope

T

METHOD OF VALIDATION



Experiment simulation

Soot properties

T(r), f_v(r), E(m)

DIRECT MODEL

Spectral emission intensities, $I_\eta (x)$

Compared

Inferred properties

T(r), f_v(r), E(m)

INVERSE MODEL

Spectral emission intensities, $I_\eta (x)$

DIRECT MODEL

Simulation of Radiative Transfer in Soot-Laden Media

Generation and propagation of radiative energy emitted by soot particles from the high-temperature, non-homogeneous, sooty combustion environment to the measuring equipment is governed mainly by three physical phenomena:

- line-of-sight radiative transfer through the participating medium
- radiative properties of soot agglomerates
- optical constants of soot bulk material

Work Done

Modelling 3 governing physical phenomena and isolated validation of models

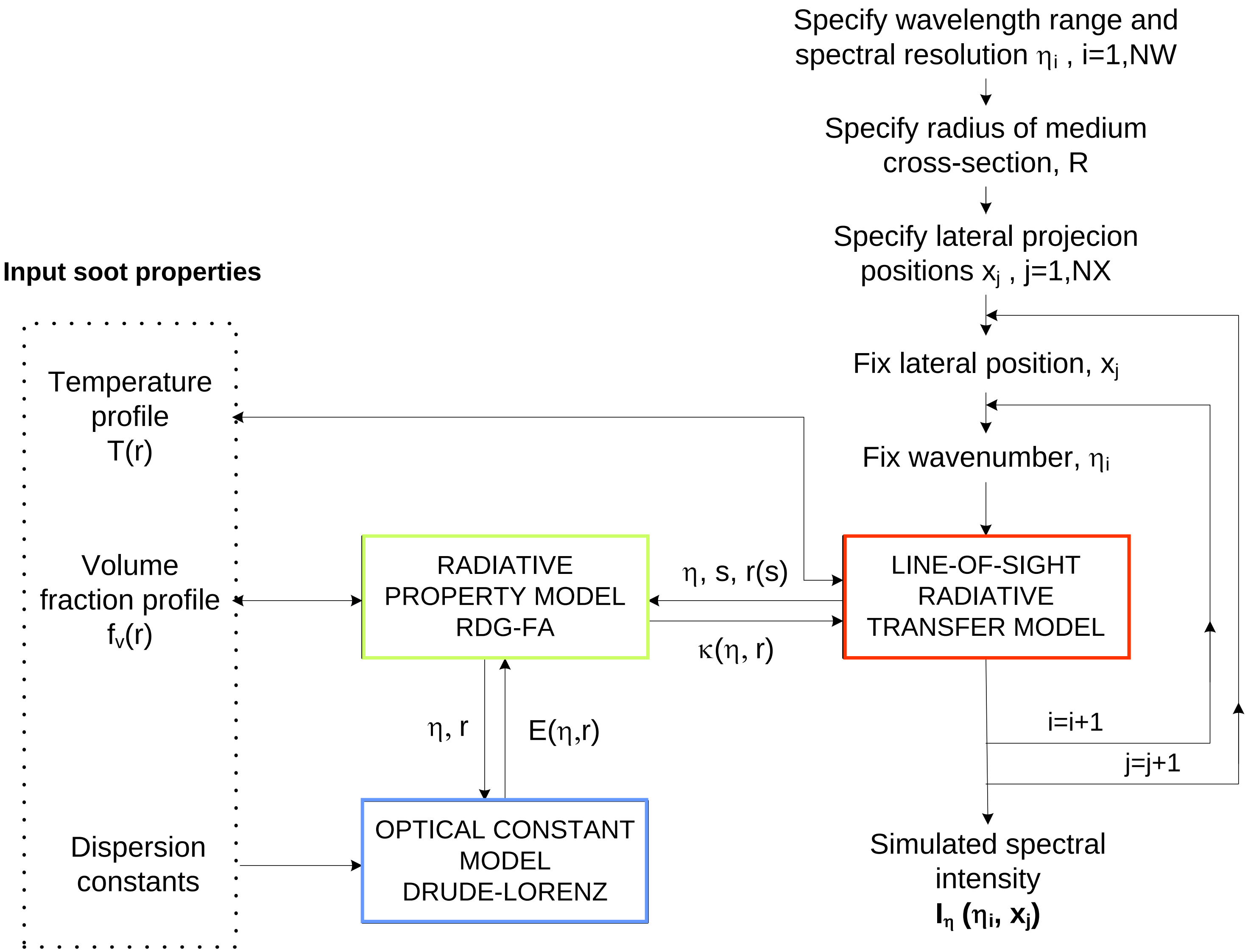
↓

Coupling the sub-models

↓

Applying coupled direct model to a well characterized flame from literature to simulate flame emission spectra

DIRECT MODEL - COUPLING PROCEDURE



Specify wavelength range and spectral resolution η_i , i=1,NW

Specify radius of medium cross-section, R

Specify lateral projecion positions x_j , j=1,NX

Fix lateral position, x_j

Fix wavenumber, η_i

Input soot properties

Temperature profile T(r)

Volume fraction profile $f_v(r)$

Dispersion constants

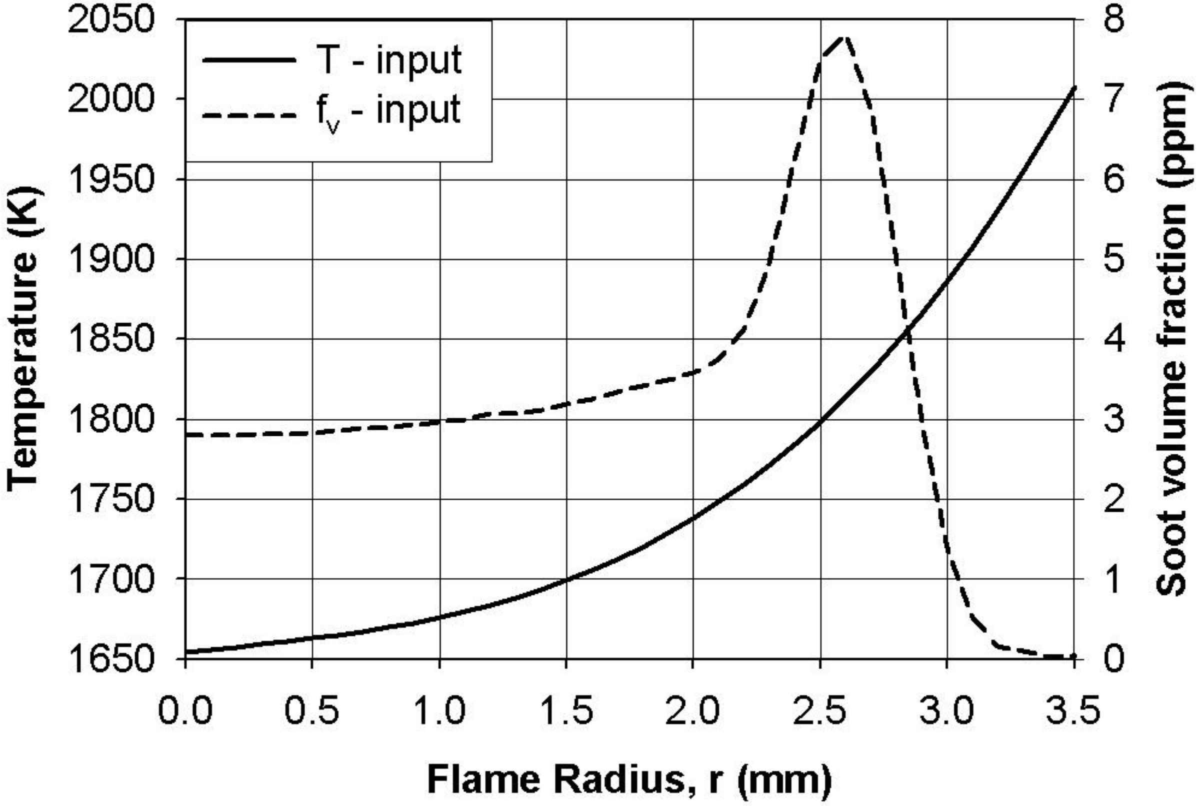
RADIATIVE PROPERTY MODEL RDG-FA

LINE-OF-SIGHT RADIATIVE TRANSFER MODEL

OPTICAL CONSTANT MODEL DRUDE-LORENZ

Simulated spectral intensity **I_η (η_i, x_j)**

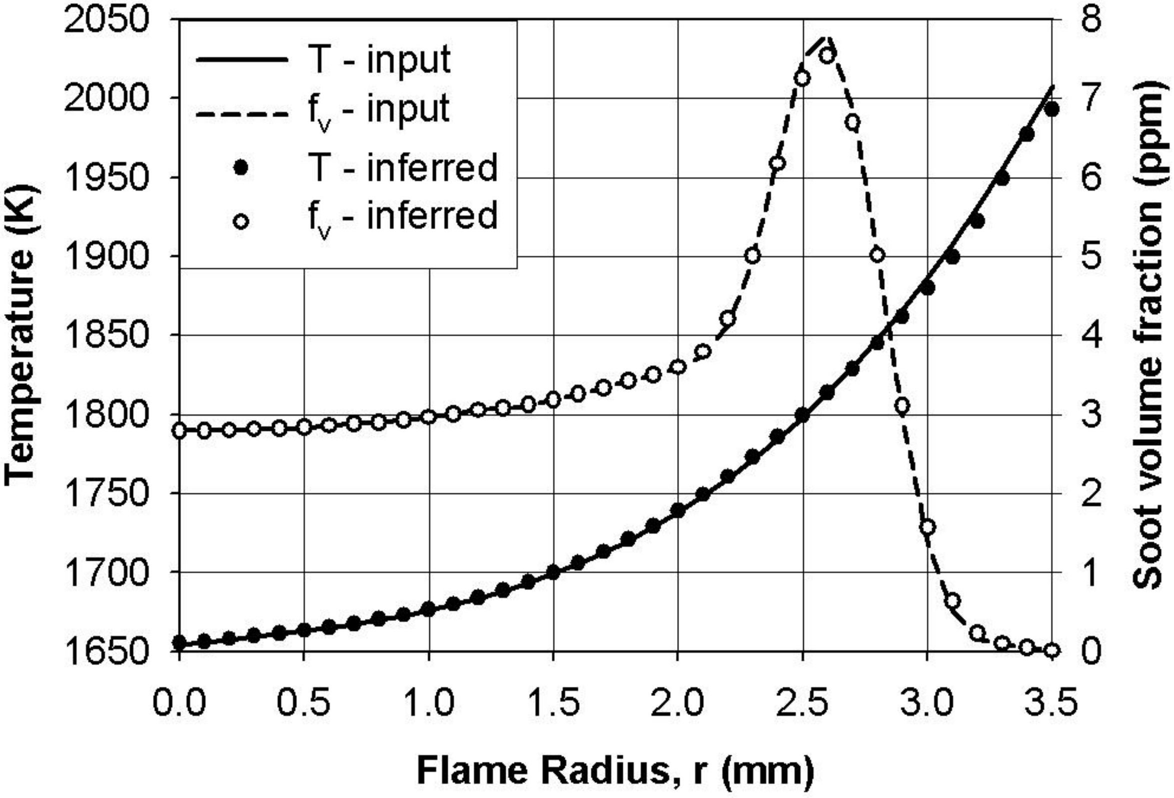
TEST PROBLEM



Laminar ethylene diffusion flame (Snelling et al., 2002)
ID=10.9mm, h=30mm

Representative of an axisymmetric sooting flame in laboratory conditions

VALIDATION OF INVERSION ALGORITHM



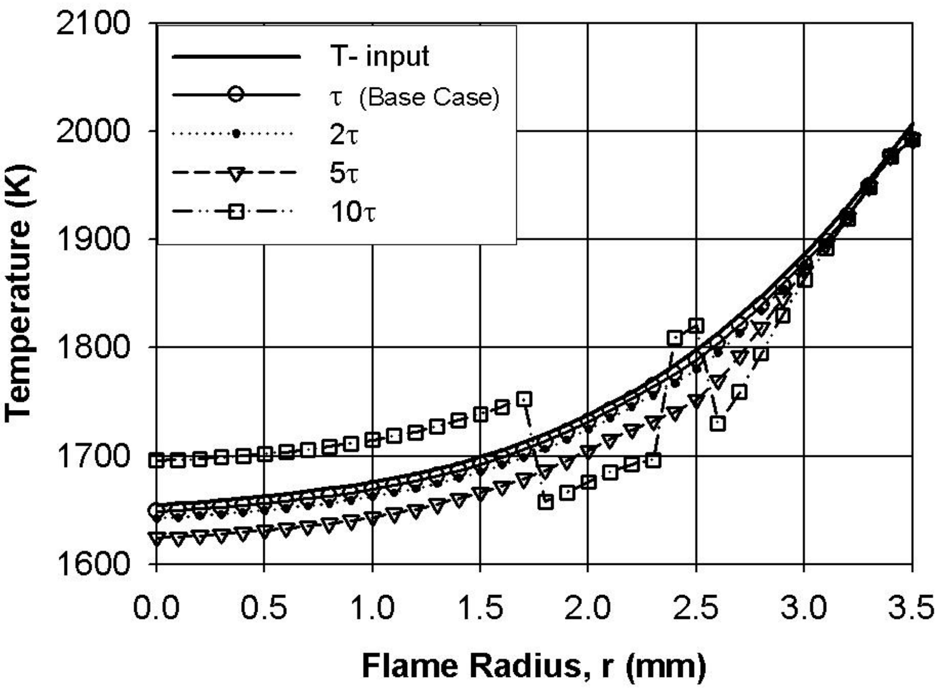
Same result for each input set of dispersion constants

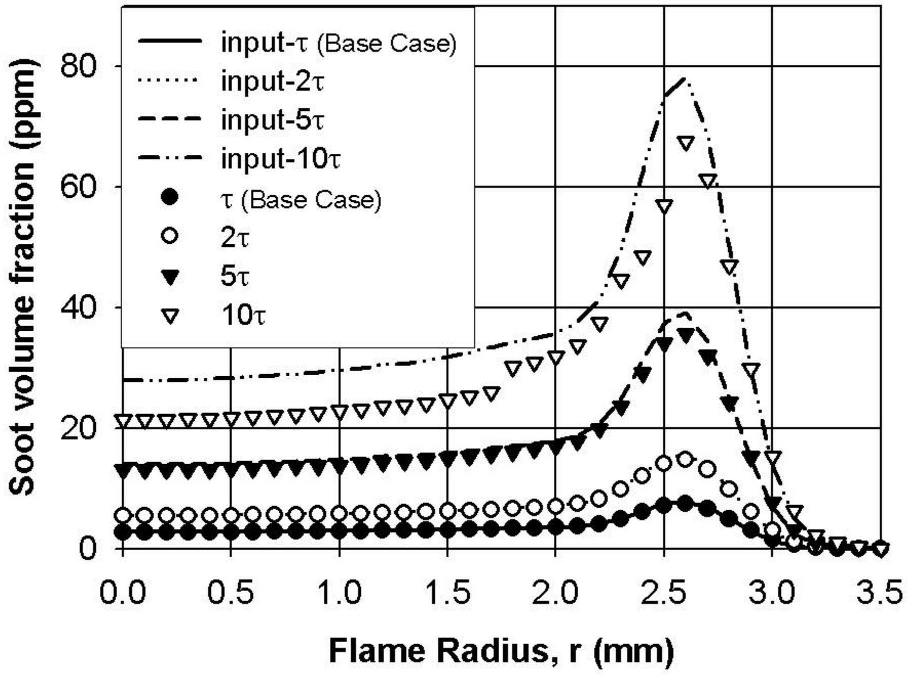
Algorithm performs successfully for optically thin flames with ideal line-of-sight measurements of high spatial resolution

EFFECT OF PHYSICAL ASSUMPTIONS

Effect of optical thickness

Negligible self-absorption assumption checked

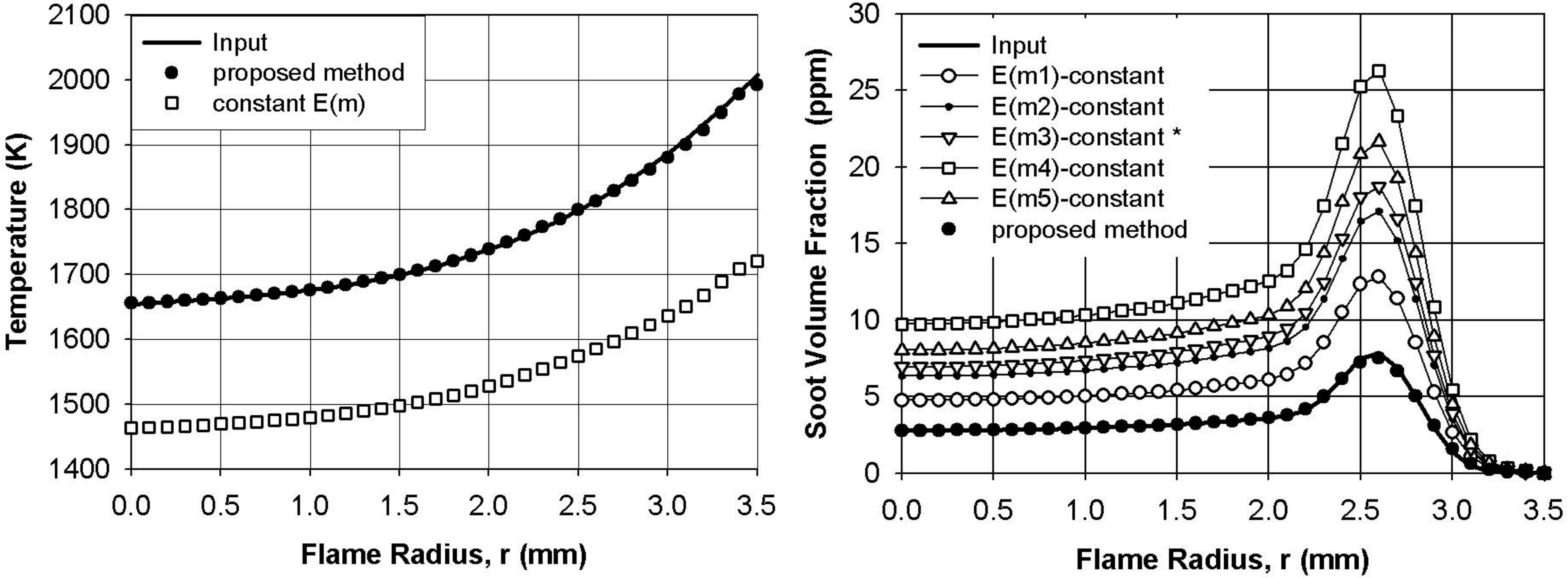




It is found that the proposed method which is based on negligible self-attenuation assumption can be confidently applied to flames with optical thickness less than 1.5.

EFFECT OF PHYSICAL ASSUMPTIONS

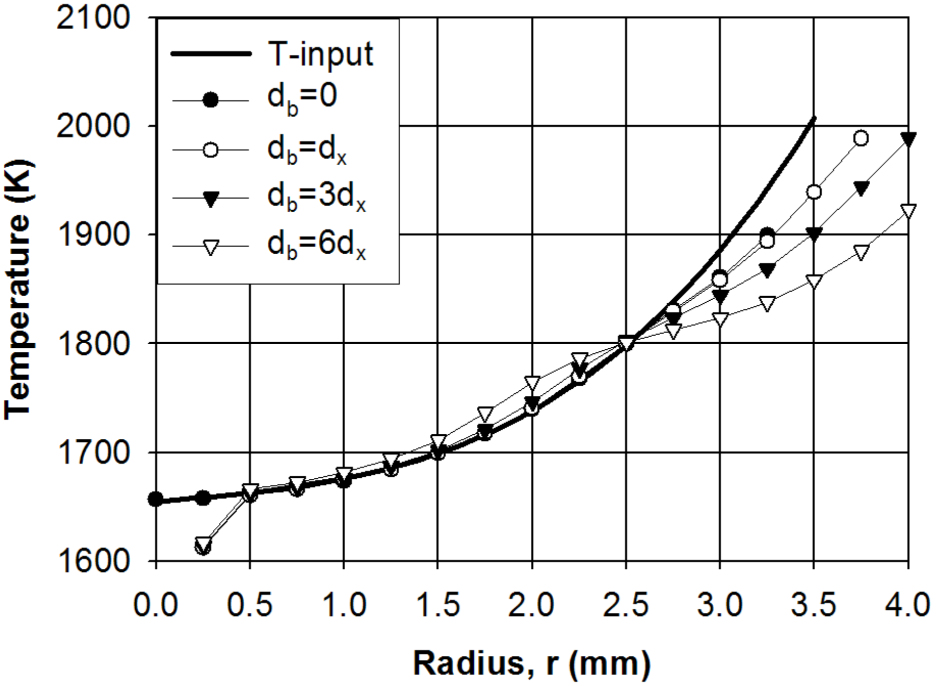
Effect of spectral variation of E_m

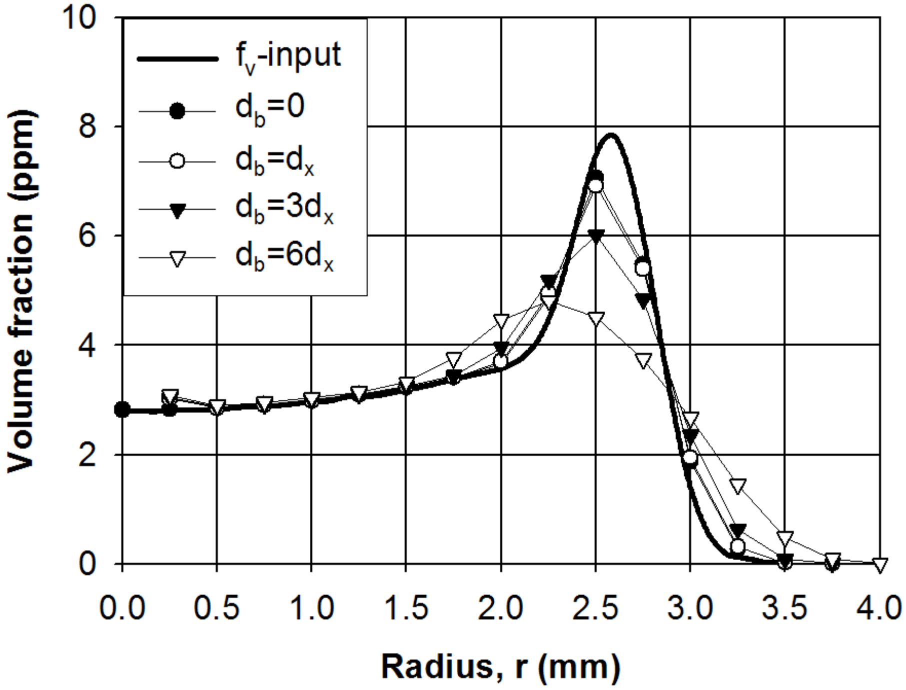


7500-8500 cm^{-1} (1180-1333 nm)

EFFECTS OF EXPERIMENTAL LIMITATIONS

Effect of beam diameter for coarse scanning resolution

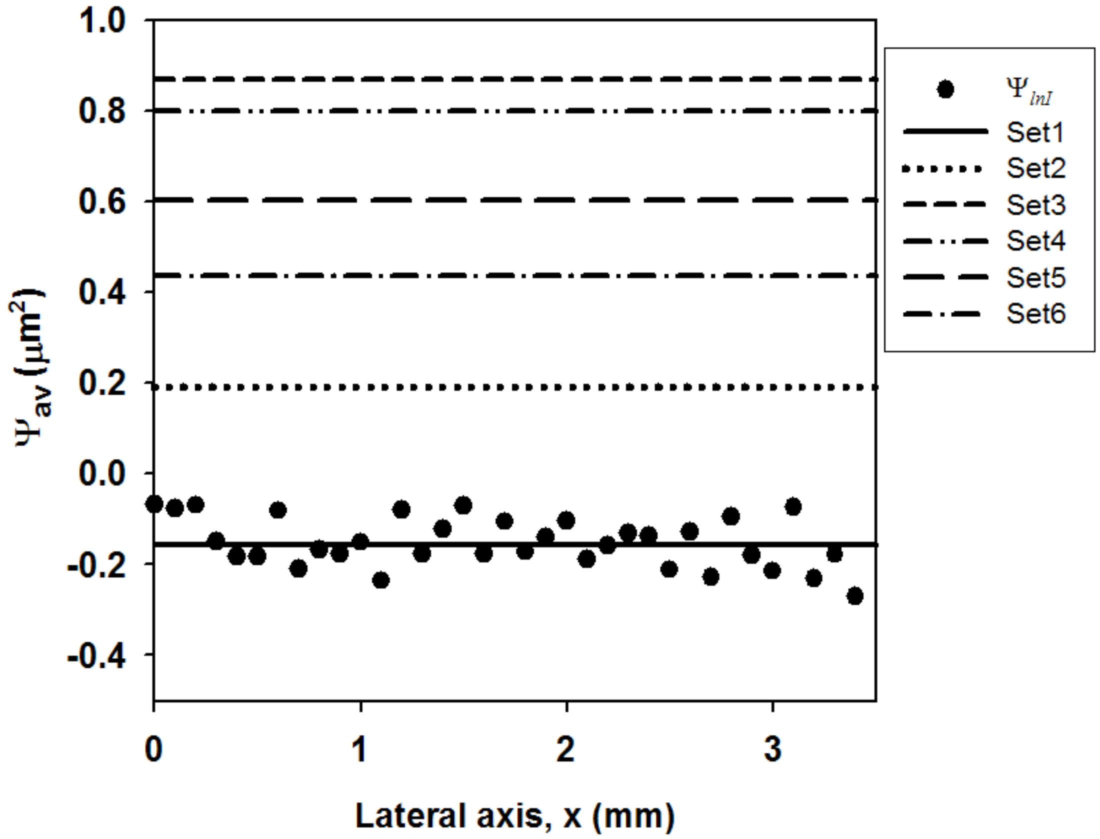




PERFORMANCE OF DATA CONDITIONING

Ψ-function retrieval and refractive index selection

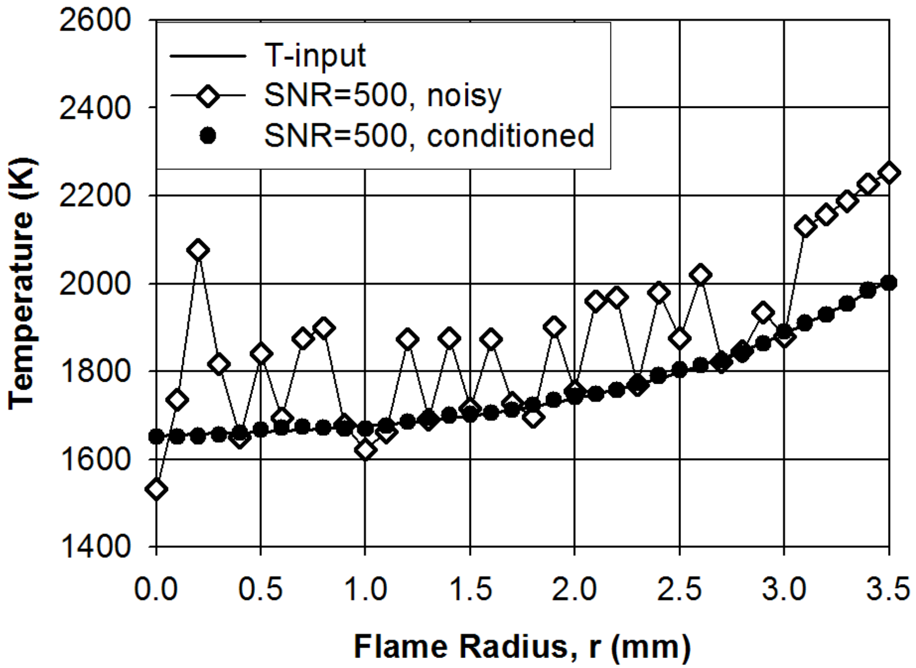
Noisy intensities simulated with Set 1 optical constants were supplied to inversion algorithm

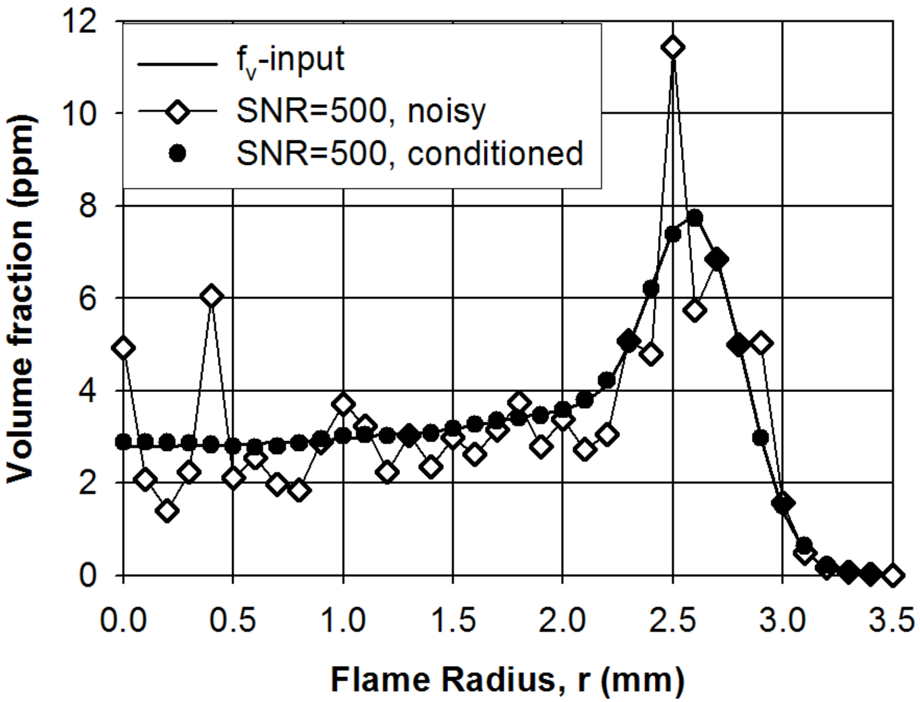


✓ Inversion algorthim with data conditioning retrieves correct Ψ-function and refractive index

PERFORMANCE OF DATA CONDITIONING

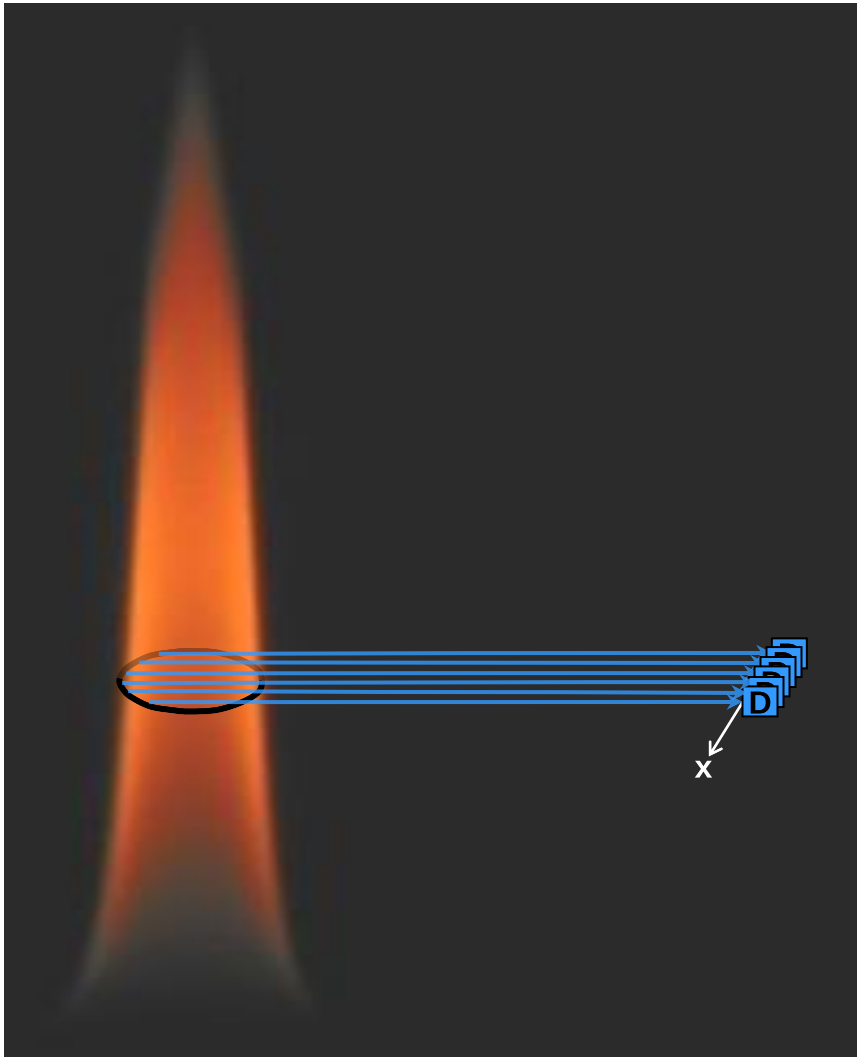
Improvement of inferred soot property profiles





EXPERIMENTAL METHODOLOGY

Flame Emission Spectroscopy Measurements



Ethylene-Air Diffusion Flame

Burner diameter	2 cm
Luminous flame height	91 mm
Fuel flow rate	18.3 0.4 cm^3/s
Exit velocity, u_0	5.8 cm/s
Re ($u_0.D_0/\nu$)	155

Flame Scanning Parameters

Beam diameter	3 0.2 mm
Horizontal spatial resolution	0.5 0.2 mm
Vertical spatial increment	10 0.5 mm

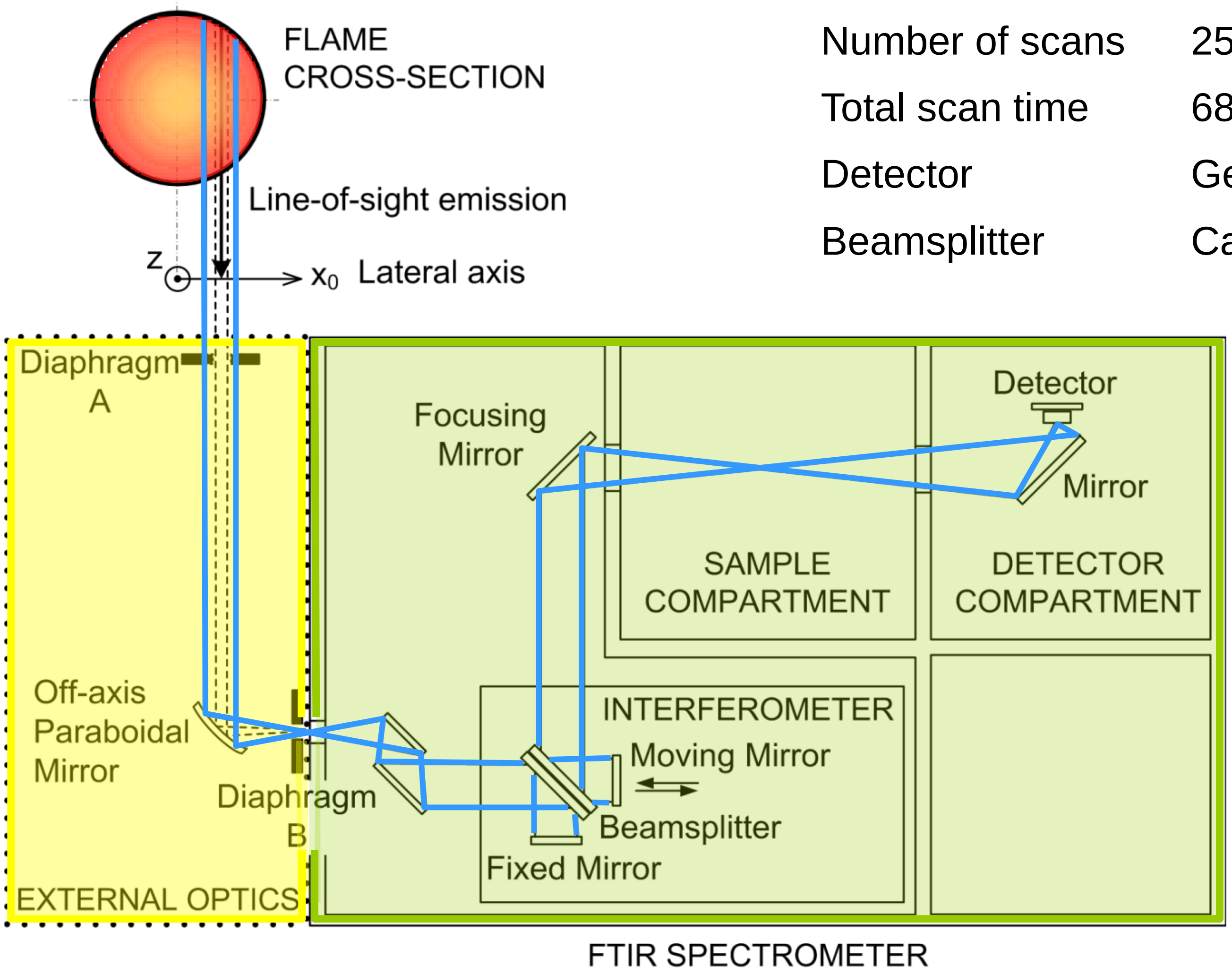
Measured parameter:

Line-of-sight Emission spectra

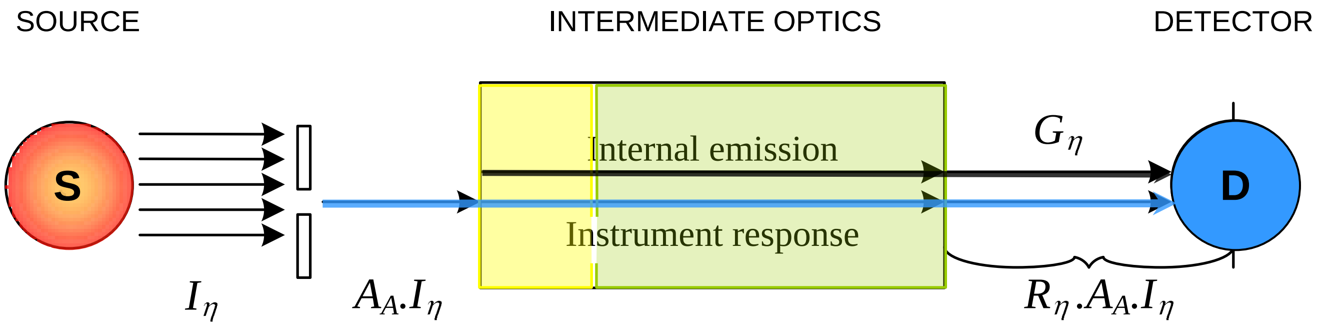
EXPERIMENTAL METHODOLOGY

NIR Emission Spectroscopy

Spectral range	9000-6000 cm^{-1} (1.1-1.7 μm)
Spectral resolution	$\Delta\eta$ = 25 cm^{-1}
Number of scans	256
Total scan time	68 s
Detector	Germanium Photodiode
Beamsplitter	CaF_2



CALIBRATION WITH BLACKBODY



Components of detected energy

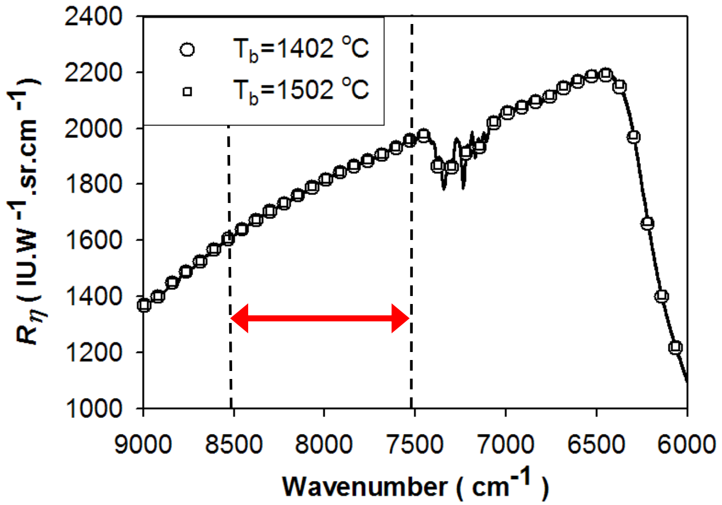
$$S_\eta = G_\eta + R_\eta \cdot A_A \cdot I_\eta$$

recorded in instrument units (IU)

Blackbody experiments: flame replaced by blackbody furnace @ $T=T_b$

Instrument function of the spectrometer evaluated from blackbody emission spectra

$$R_\eta = \frac{S_{\eta,\mathrm{b}} - G_{\eta,\mathrm{b}}}{A_{\mathrm{A,b}} \cdot I_{\eta,\mathrm{b}}(T_b)}$$



Calibration of flame emission spectra

$$I_\eta = \frac{S_\eta - G_\eta}{A_A \cdot R_\eta}, \left[\frac{\mathrm{W}}{\mathrm{m}^2 \cdot \mathrm{sr} \cdot \mathrm{m}^{-1}}\right]$$

- ✓ **Instrument effects quantified**
- ✓ **IU → physical units**

UNCERTAINTY ASSESSMENT

Calibration Eqn. $I_\eta = 2hc_0^2\eta^3 \cdot \boxed{A_{A,b}}/\boxed{A_A} \cdot S_\eta/S_{\eta,b} \cdot \left[\exp\ hc_0\eta/T_b k\ -1\right]$

Emission intensity is a function of 3 independently measured quantities ➡ $\left(\frac{\Delta I_\eta}{I_\eta}\right)^2 = \sum_{j=1}^{3}\left(\frac{\partial I_\eta}{\partial Q_j}\right)^2$

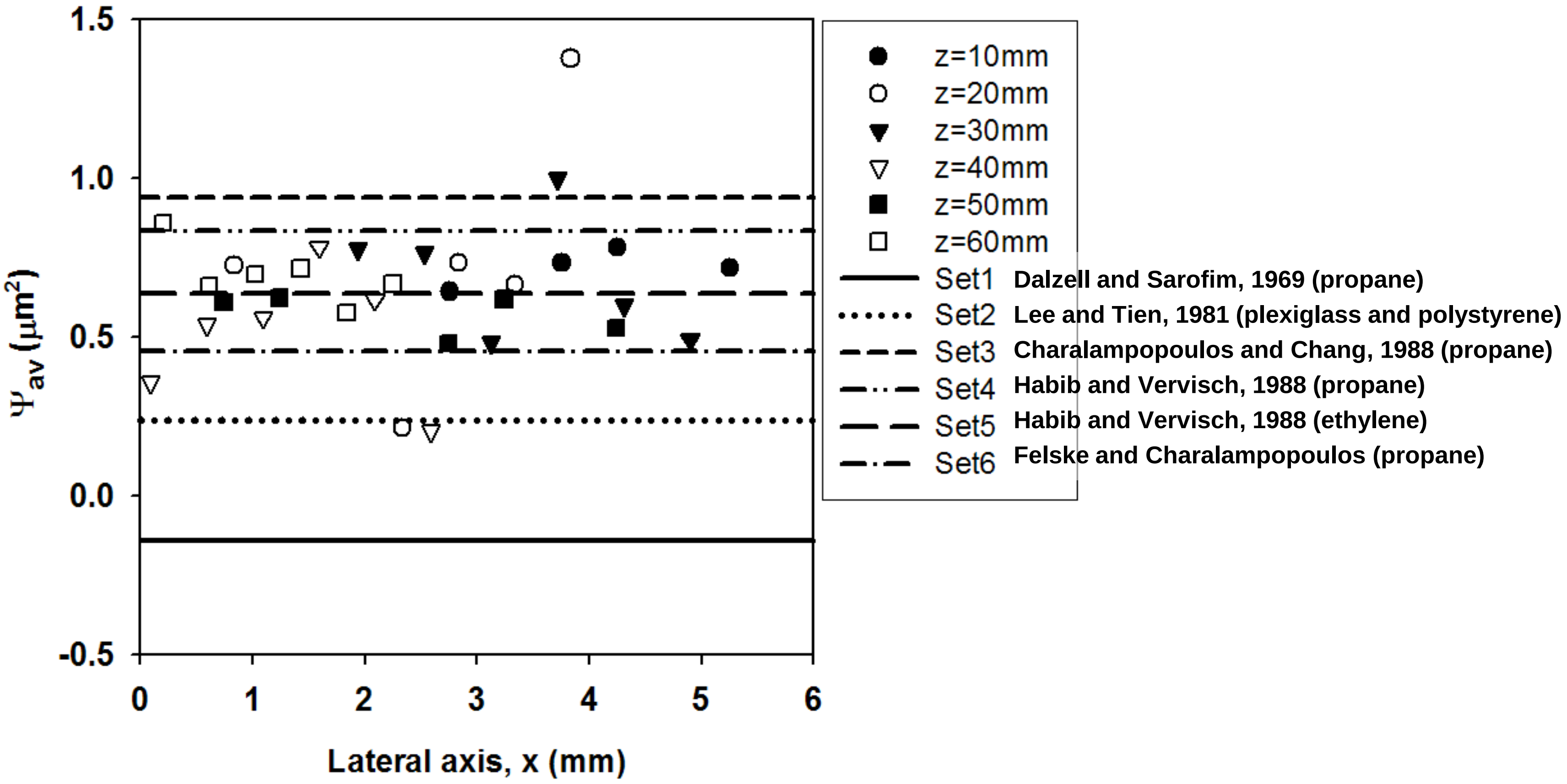
Combined Error $\Delta I_\eta = I_\eta \cdot \left[{\Delta S_\eta/S_\eta}^2 + {\Delta S_{\eta,b}/S_{\eta,b}}^2 + {hc_0\eta\Delta T_b/kT_b^2}^2 \right]^{1/2}$

Measured Quantity, Q	***Uncertainty limit, ΔQ*** [a]	$\partial I_\eta/\partial Q$	***Rel. uncertainty level***
Blackbody temperature, T_b	± 2°C	$\eta hc_0\Delta T_b/kT_b^2$	**< 0.4 %**
Blackbody emission spectrum [c] $S_{\eta b}(\eta)$	$\pm 97.5\times10^{-5}$ IU	$\Delta S_{\eta b}/S_{\eta b}$	**< 1.1 %**
Flame emission spectrum [d] $S_\eta(\eta, x_0, z)$	$\pm 13.7\times10^{-5}$ IU for $x_0 < 6$mm $\pm 4.18\times10^{-5}$ IU for $x_0 > 6$mm	$\Delta S_\eta/S_\eta$	**≤ 10 %** **(20-30 % at weak signal zones)**
Flame emission intensity $I_\eta(\eta)$	Avg. limits [b]: ± 0.014 W/(m^2.sr.cm^{-1}), $x_0 < 6$mm ± 0.004 W/(m^2.sr.cm^{-1}), $x_0 > 6$mm	$\Delta I_\eta/I_\eta$	**≤ 10 %** **(20-30 % at weak signal zones)**

[a] 99% confidence level; [b] spatially variable; [c] 0.085 IU < $S_{\eta b}$ < 0.16 IU; [d] S_η < 0.0083 IU

APPLICATION TO MEASURED FLAME

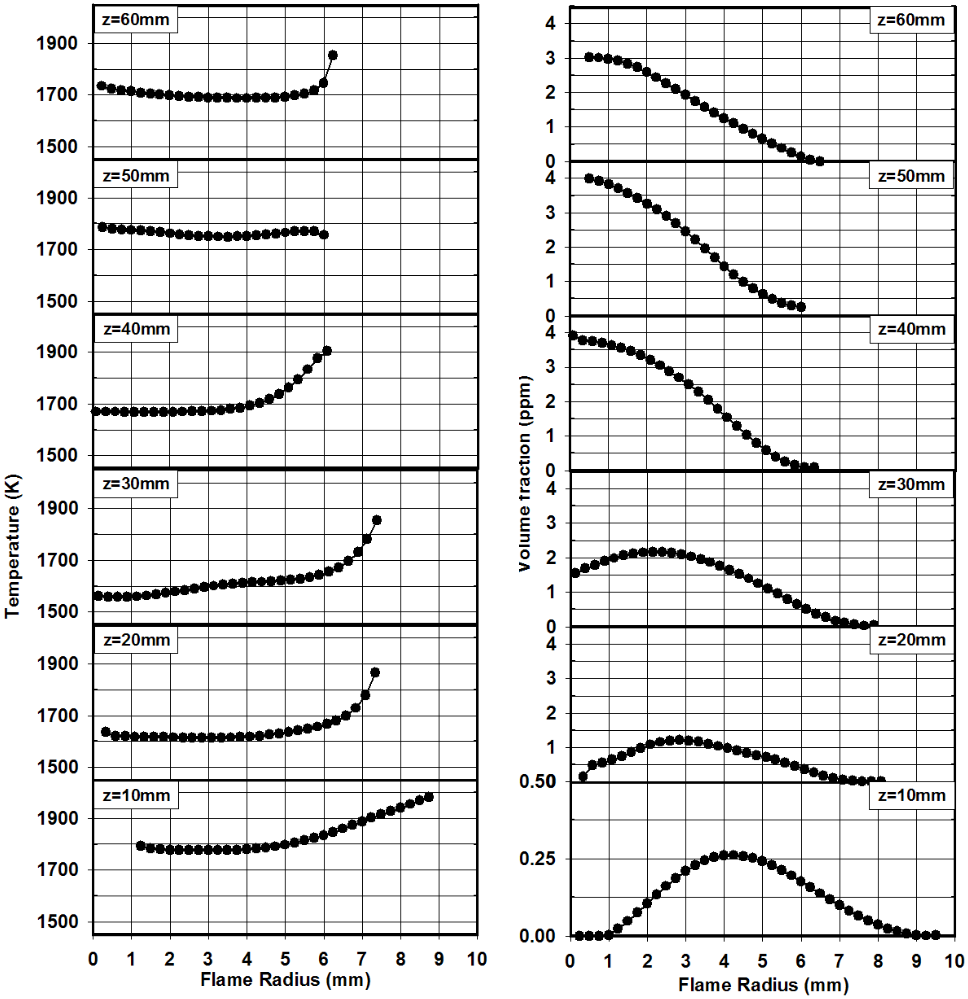
Ψ-function retrieval and refractive index selection



Inferred Ψ-function leads to selection of Set 5 by Habib and Vervisch

APPLICATION TO MEASURED FLAME

Inferred Soot Property Profiles



CONCLUSIONS

- A nonintrusive soot diagnostics methodology was developed, validated and applied for in-situ determination of temperature, volume fraction and refractive index of soot aggregates formed inside flames by using near-infrared emission spectrometry.
- Reconstructive capabilities of the method was validated on a realistic test case representing flame conditions by using a direct model as an experiment simulator and comparing inferred properties with simulator inputs.
- The effects of physical approximations on the method were analyzed. It was found that the proposed method which is based on negligible self-attenuation assumption can be confidently applied to flames with optical thickness less than 1.5. Assuming constant refractive index assumption within near-infrared range spectrum leads to considerable errors both in temperature and volume fraction profiles.
- Lateral scanning resolution needs to be adequately fine to resolve sharp soot volume fraction gradients. It was found that the beam diameter which is limited by experimental possibilities introduce considerable dispersing effects especially when the scanning resolution is coarse.

CONCLUSIONS

- A set of data conditioning steps were developed to accommodate noisy data commonly encountered in practical soot diagnostics. Use was made of simulated noisy intensities to demonstrate effectiveness of the data conditioning procedure.
- Application of the proposed soot diagnostics methodology on the experimentally investigated ethylene/air diffusion flame was realized by inferring soot properties from spectral intensities measured by Fourier Transform Infrared Spectrometry.
- Inferred properties are found to display expected effects of experimental limitations.
- Validation with simulated data and favorable application to measurements indicate that proposed methodology is a promising option for nonintrusive soot diagnostics in flames.

RELATED PUBLICATIONS

Ayrancı I., Vaillon R., Selçuk N., André F. and Escudié D., Determination of soot temperature, volume fraction and refractive index from flame emission spectrometry, Journal of Quantitative Spectroscopy and Radiative Transfer, Vol. 104 (2), p. 266-276, 2007.

Ayrancı I., Vaillon R., Selçuk N., Near-infrared emission spectrometry measurements for nonintrusive soot diagnostics in flames, Journal of Quantitative Spectroscopy and Radiative Transfer, Vol. 109 (2), p. 349-361, 2008.

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