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In-Situ Soft X-ray Absorption Spectroscopy of Flames

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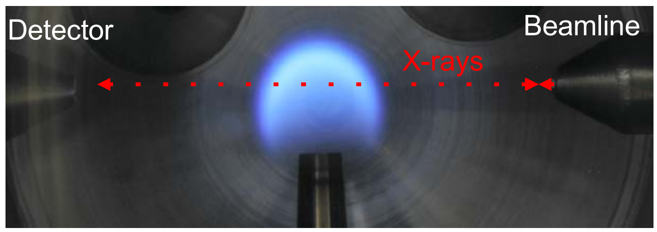
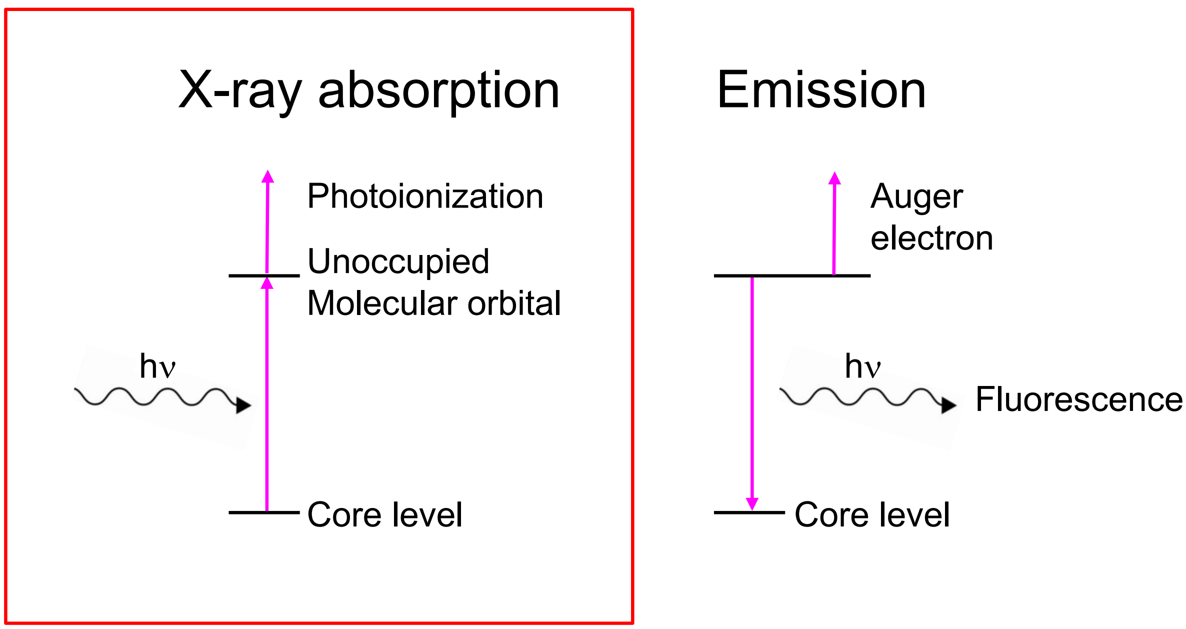
Motivation for Soft X-ray Diagnostic Techniques in Flames



Core-level spectroscopy may provide in-situ flame measurements that are complementary to conventional valence spectroscopy techniques.

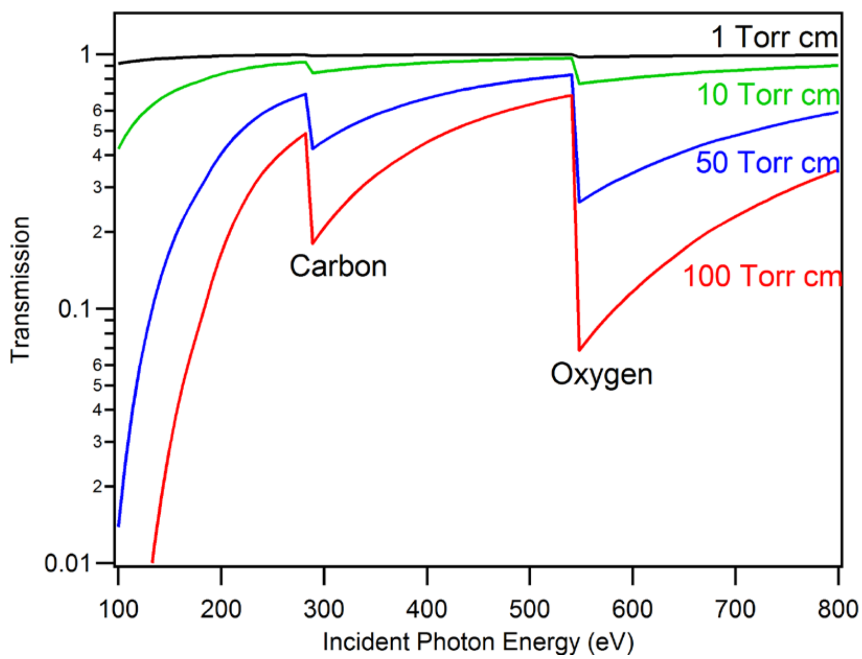
- Tomographic imaging of all carbonaceous species
- Carbon-based mixture fraction imaging
- Species specificity using near-edge spectral region
- Probe combustion of flame conditions that are not amenable to laser diagnostic techniques
- Overcome limitations of valence electron spectroscopy in flames

Soft X-ray Absorption Probes Core Level Electrons



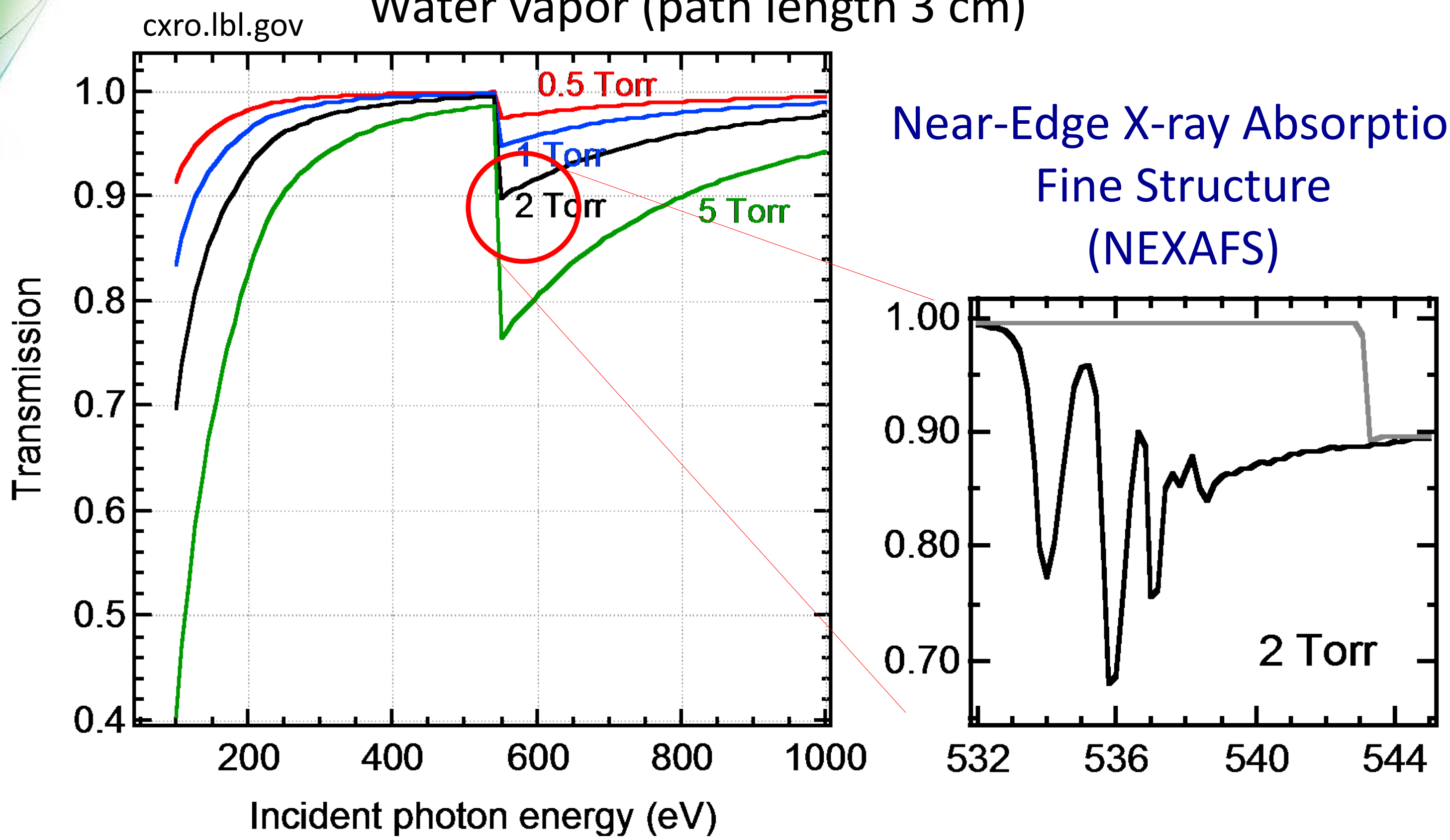
Absorption of X-rays at the Carbon and Oxygen K-edges

$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$



Near-Edge vs Far-Edge Information

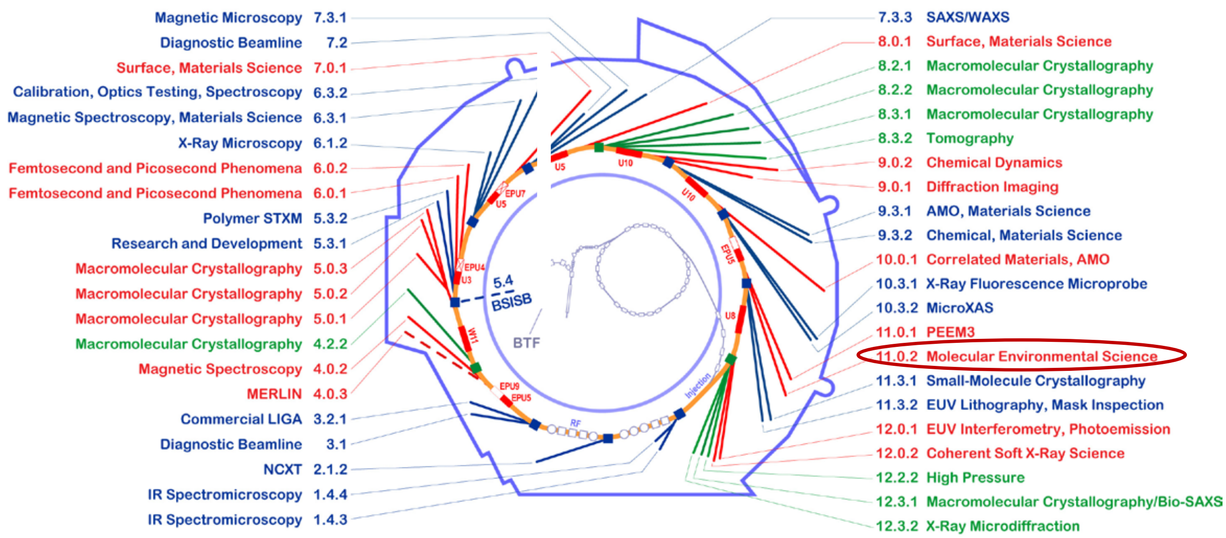
Water vapor (path length 3 cm)



Near-Edge X-ray Absorption Fine Structure (NEXAFS)

Far-edge: Elemental composition; Near-edge: Chemical speciation

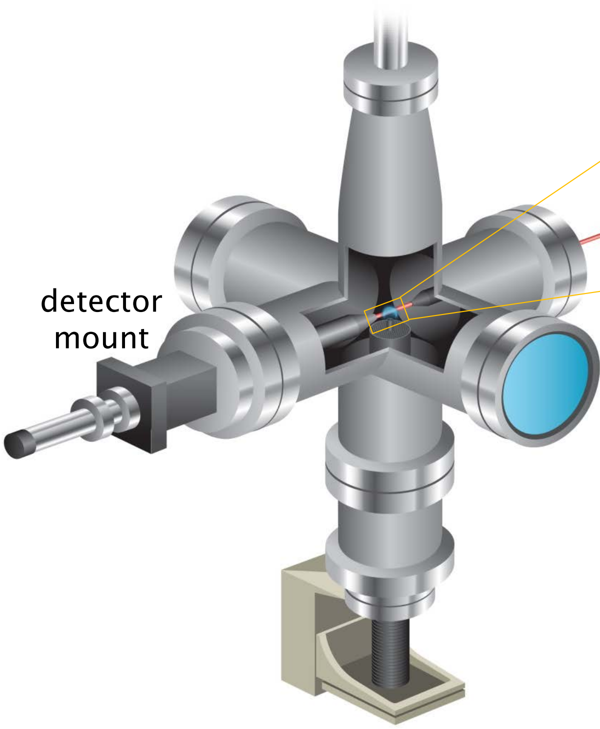
ALS Beamlines

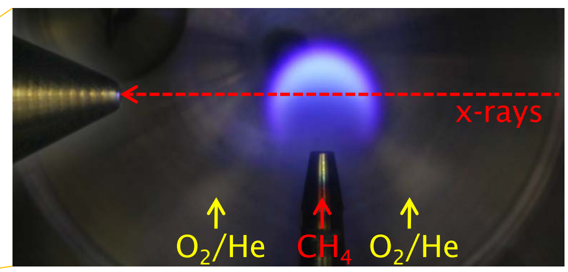


Beamline Specifications

- Energy range: 75-2000 eV
- Undulator beamline
- Photon flux: 10^{11}-10^{12} ph/s
- Resolving power: $E/\Delta E$ = 2300-7500
- Flame experiment requires dedicated roll-up endstation

Flame Vacuum Chamber Endstation





- Silicon nitride window separates beamline from flame chamber.
- Detector: water-cooled AXUV photodiode with 100 nm aluminum window
- Beam diameter: 200µm
- Beam path purged with helium

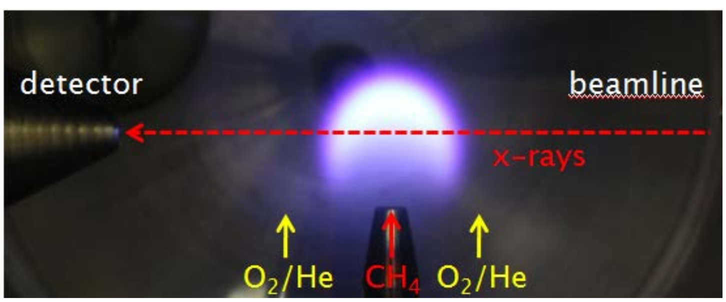
Line-of-sight Measurements
Superposition of Species Absorption Spectra

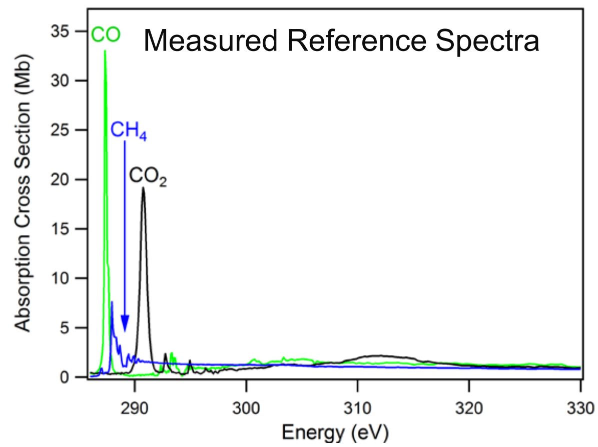
Beer-Lambert relation

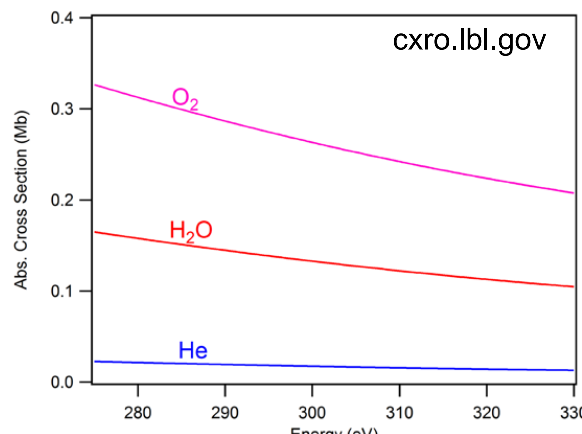
$$T_{hv} = \left(\frac{I_T}{I_0}\right)_{hv} = e^{-\sum_i \sigma_{i,hv} N_i l}$$

$\sigma_{i,hv} N_i l =$ Absorbance of species i over distance l

absorption cross section ($\sigma_{i,hv}$)   number density (N_i)

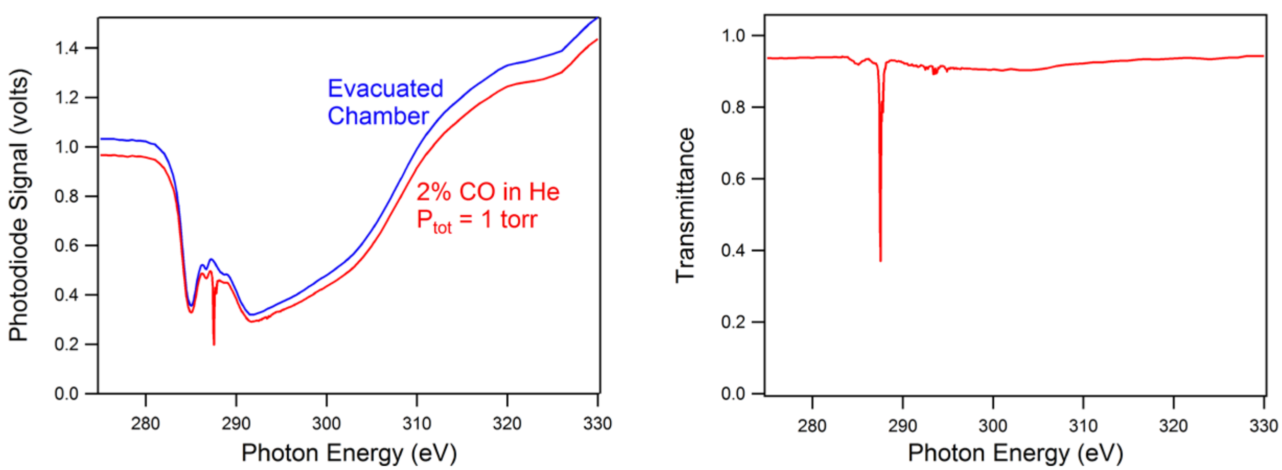




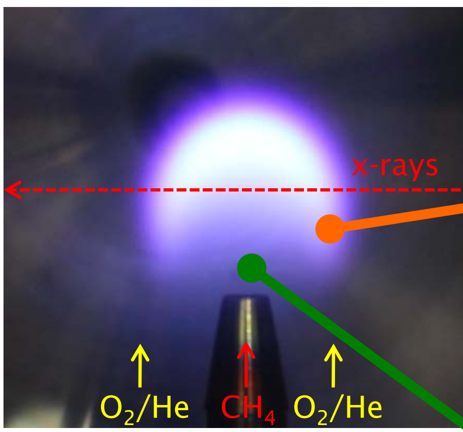


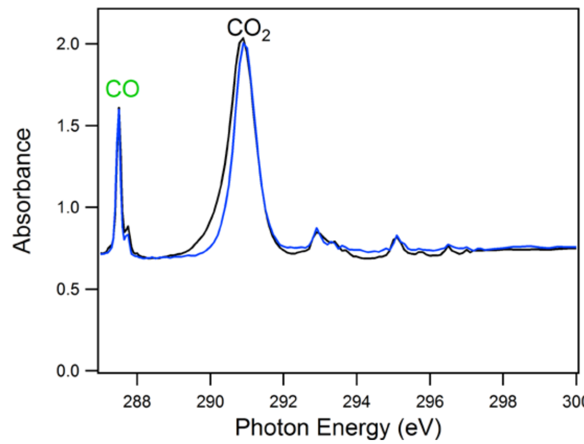
Correct for Background Absorption

Use evacuated chamber or helium-filled chamber at known pressure

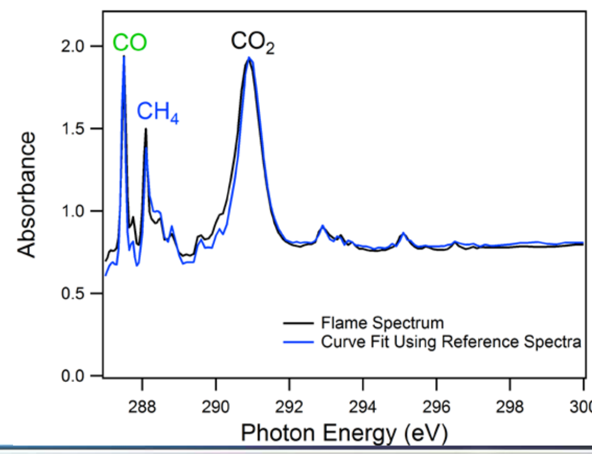


Molecular Speciation in Flames by Carbon K-edge X-ray Absorption

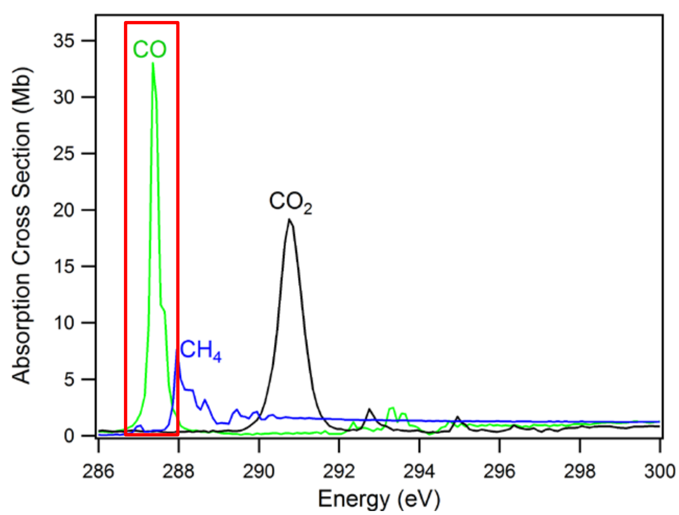


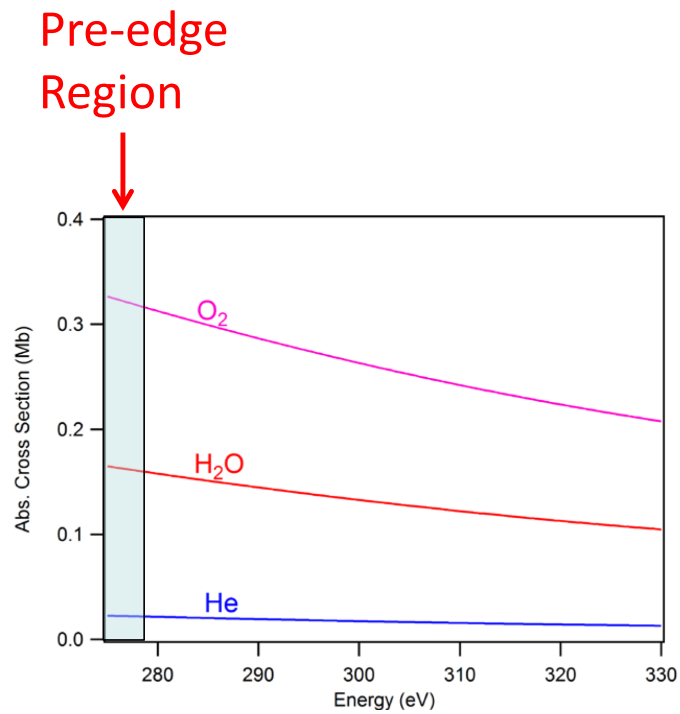


Jet: CH_4
Coflow: 60% O_2, 40% He
P_{total} = 20 Torr
d_{nozzle} = 3.1 mm
d_{coflow} = 44 mm



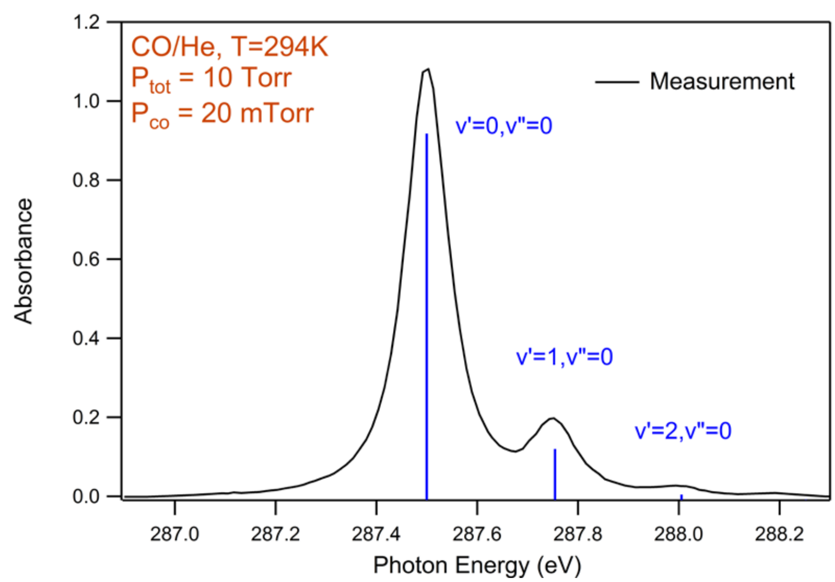
Pre-edge Spectral Region Correction for Oxygen and Helium Absorption





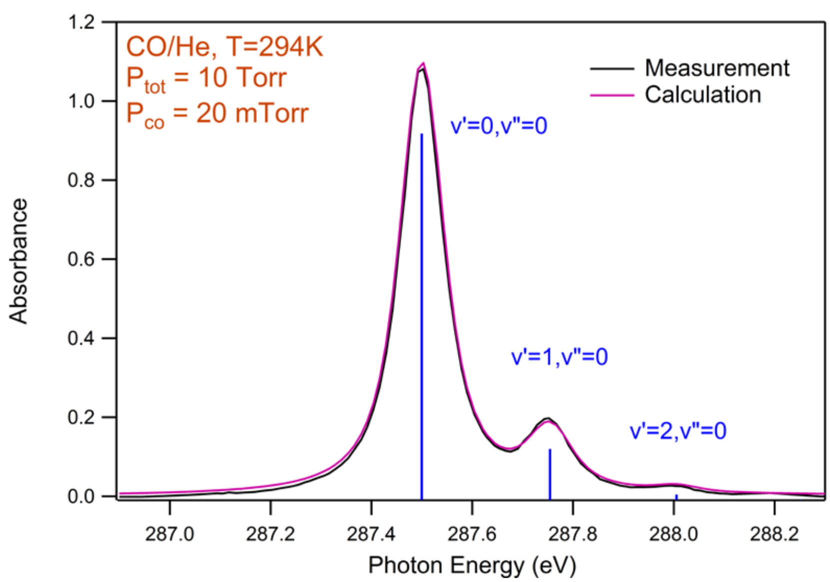
X-ray beam linewidth: 0.12 eV

Vibrational Structure of CO NEXAFS Spectra



X-ray beam linewidth: 0.02 eV

Vibrational Structure of CO NEXAFS Spectra



X-ray beam linewidth: 0.02 eV

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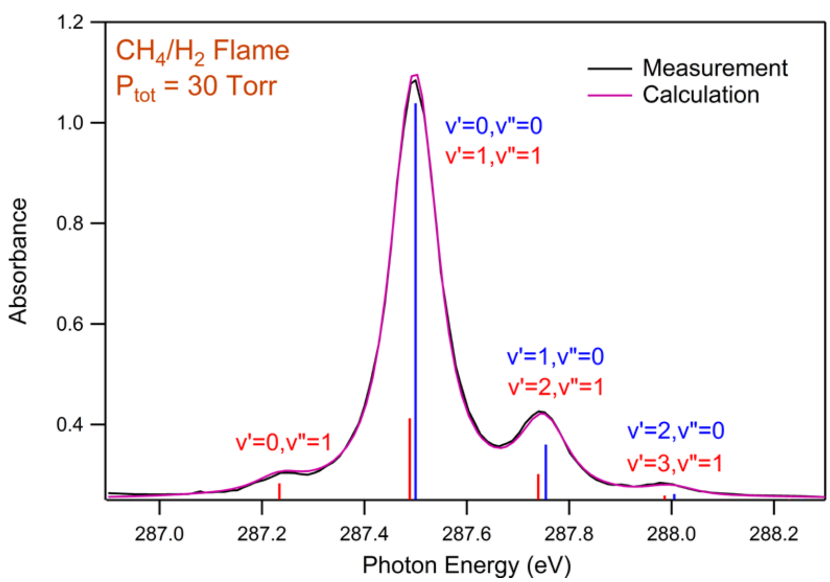
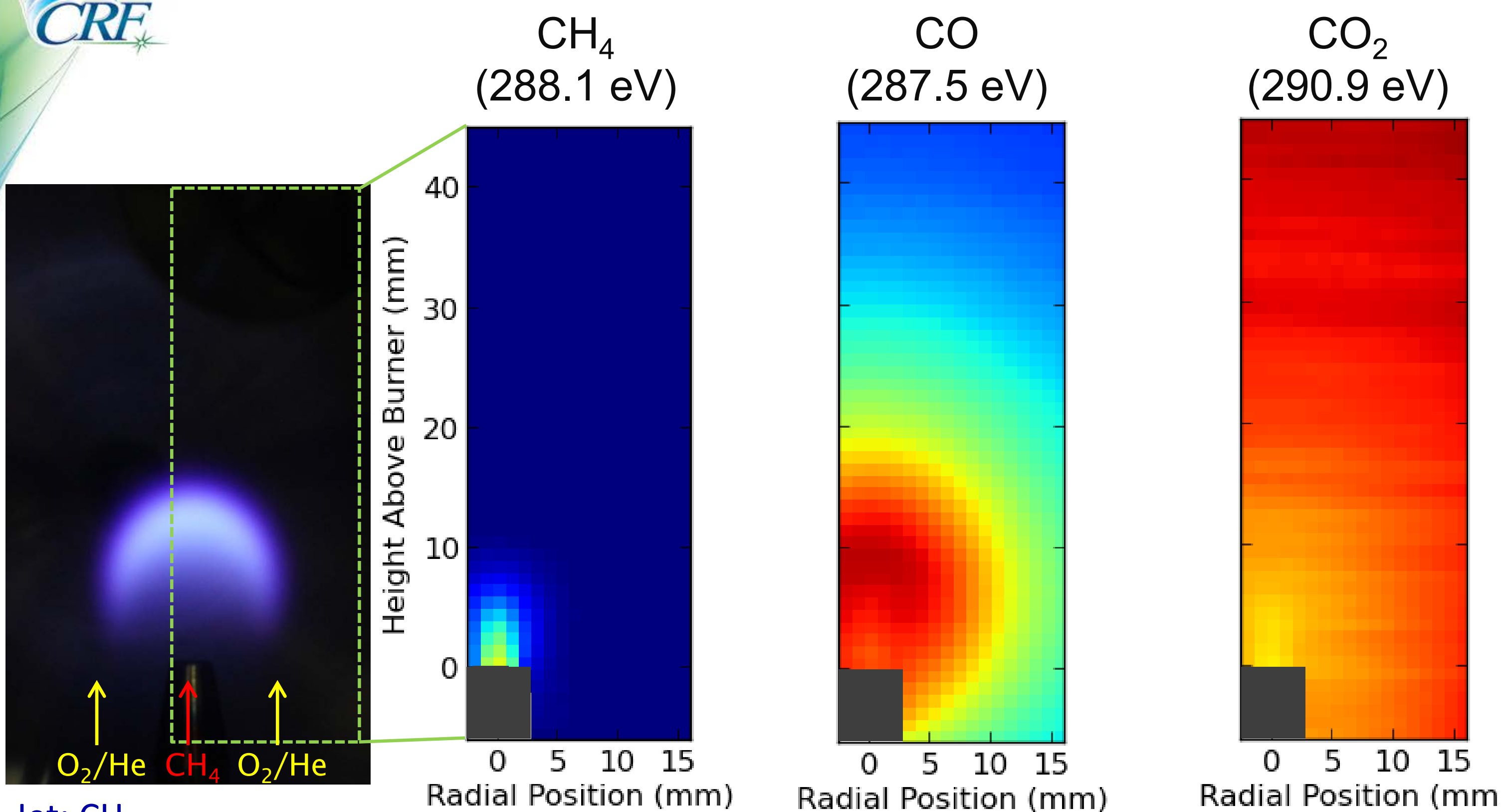


X-ray beam linewidth: 0.02 eV

Molecular Speciation in Flames



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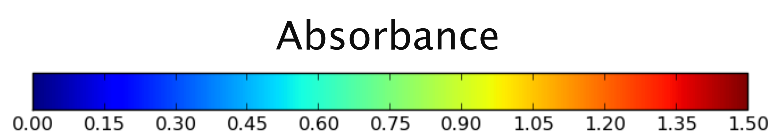
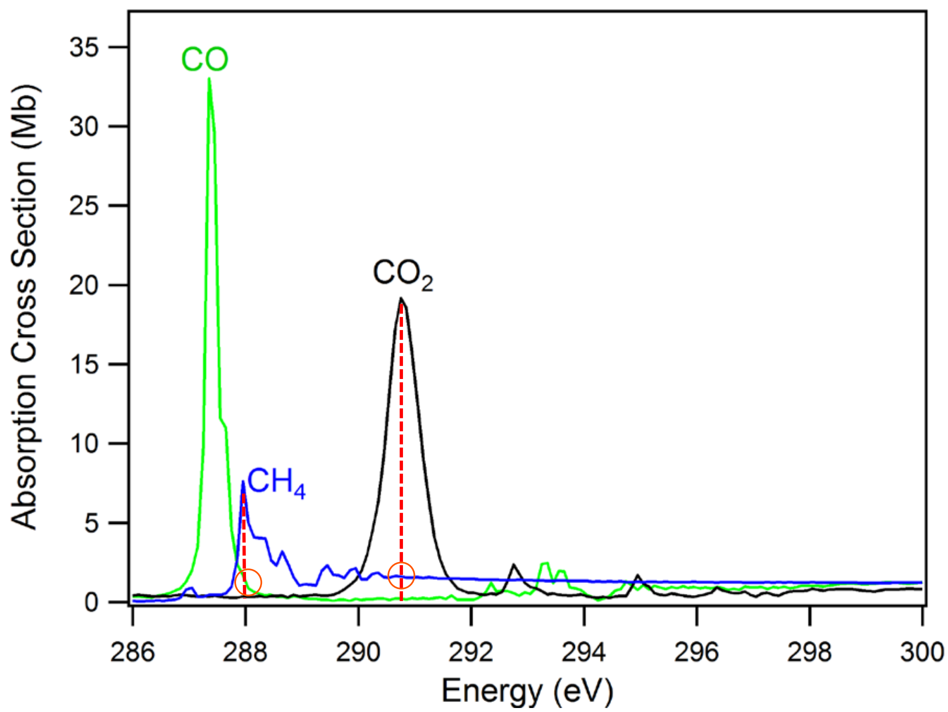
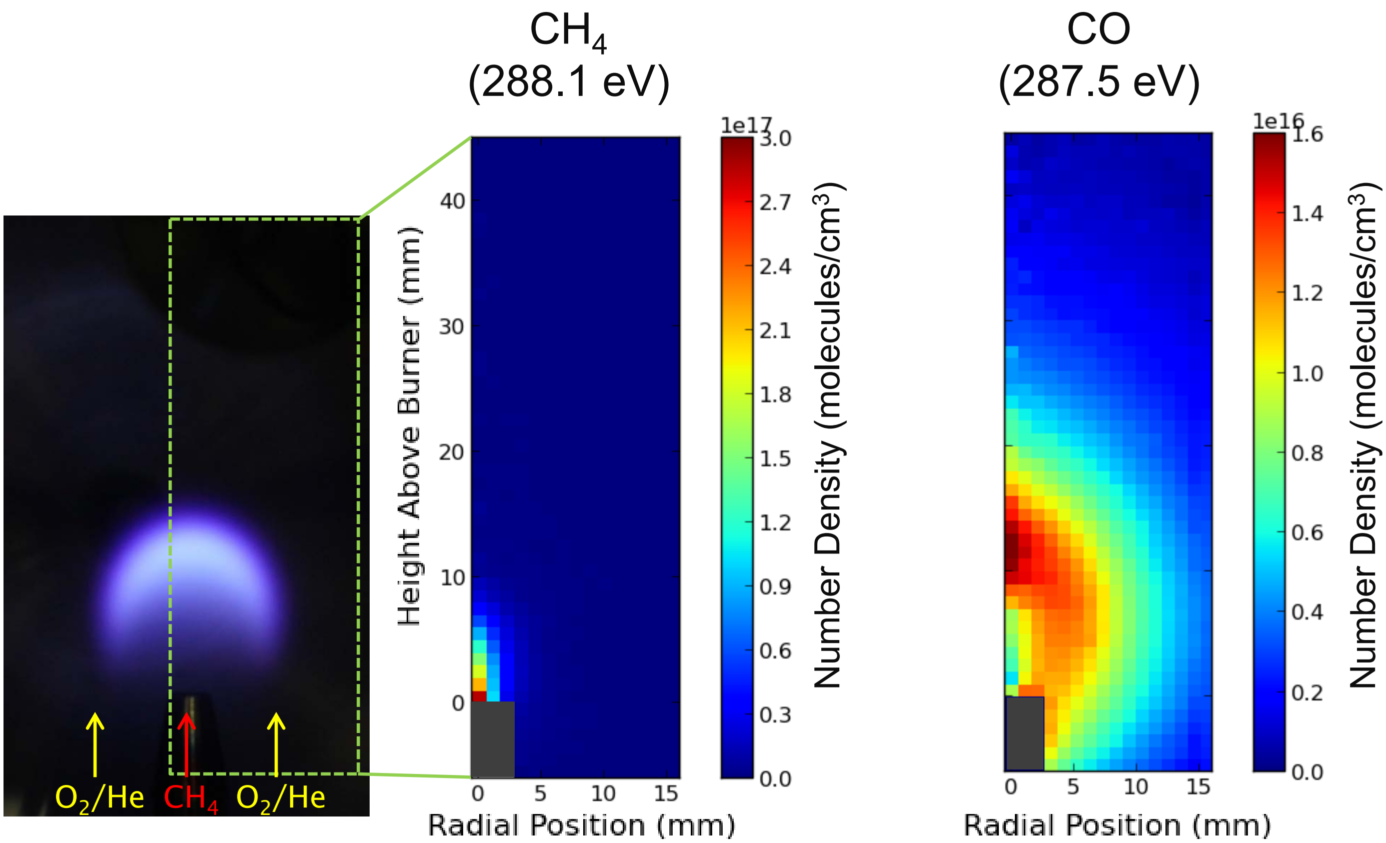


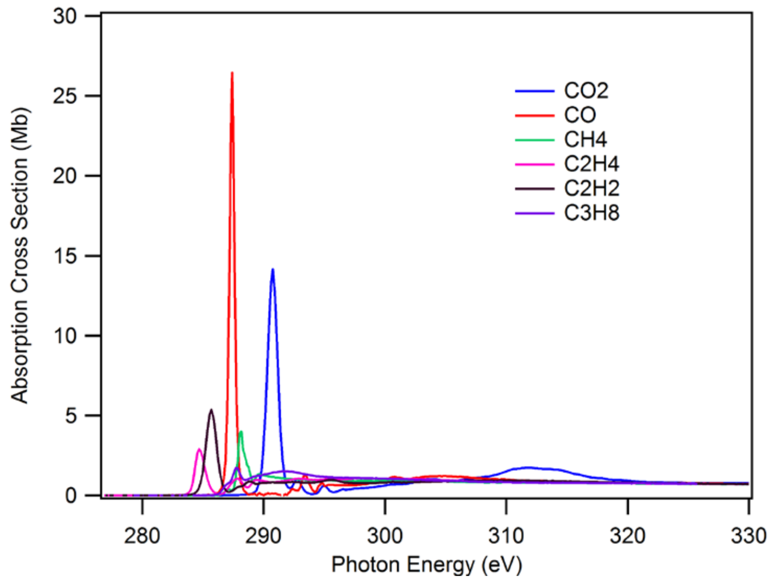
Corrections of 2-D Absorption Measurements for Spectral Cross-Talk



In-Plane Number Density Measurement
Abel Inversion

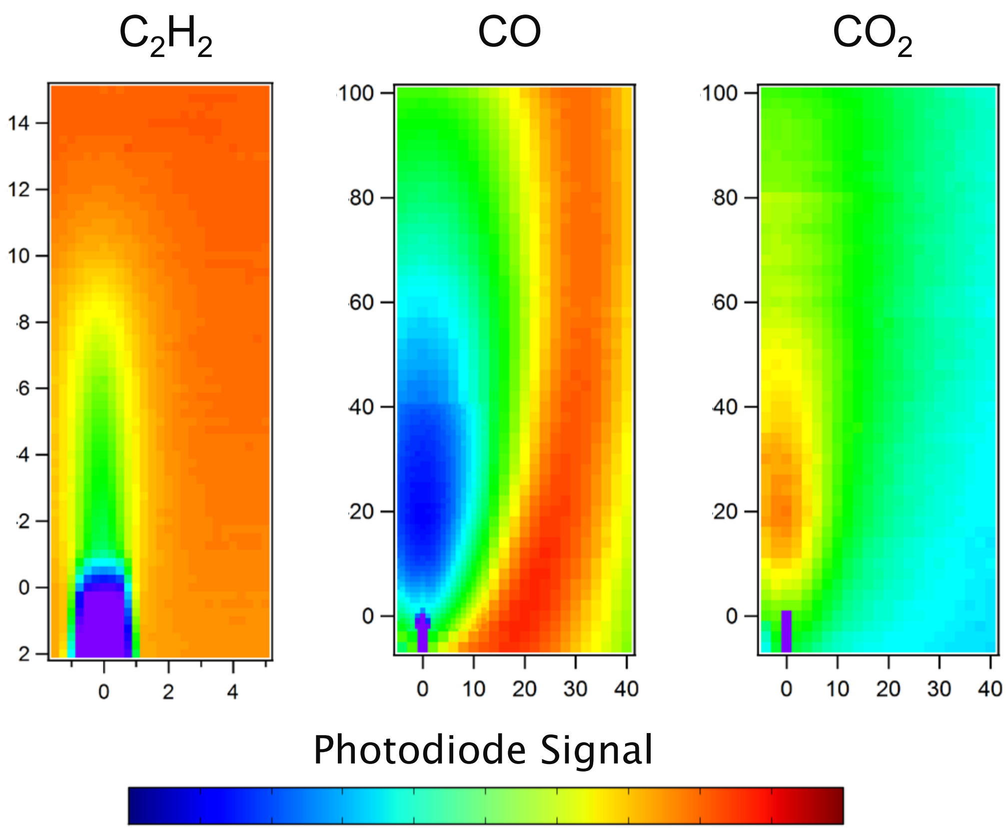


Extending In-situ NEXAFS of Flames to Larger Hydrocarbon Fuels



Acetylene Flame

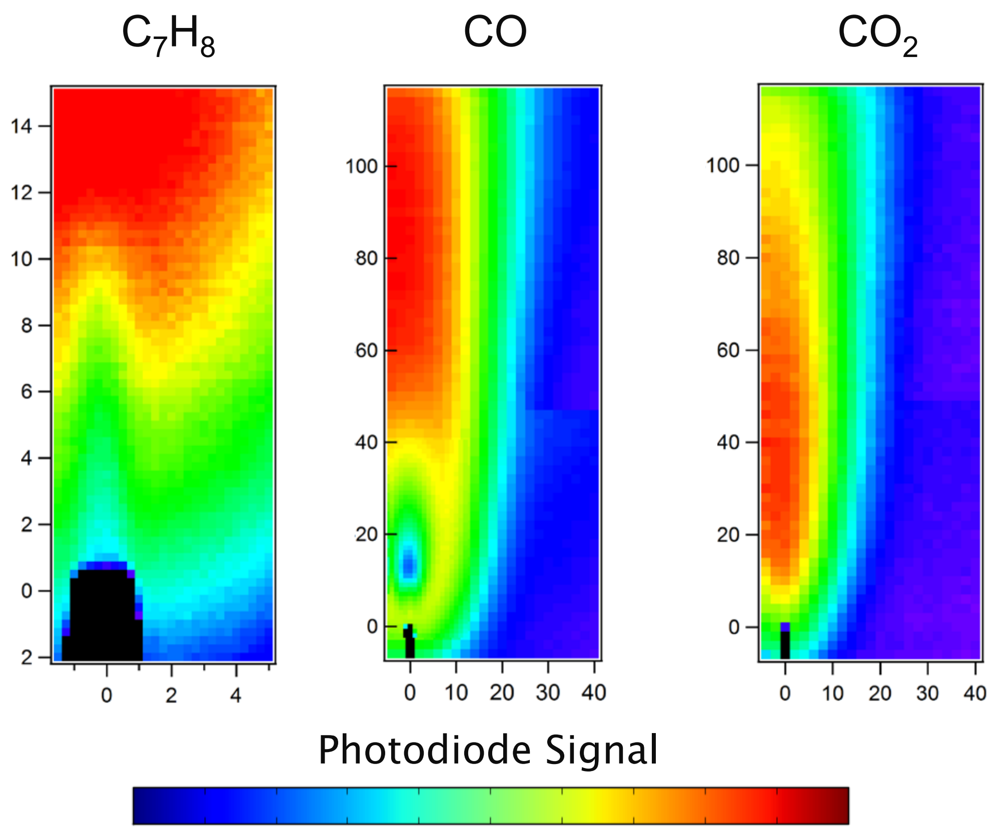
P = 11 Torr



Preliminary Data – Not corrected for background absorption

Toluene/Hydrogen Flame

P = 30 Torr



Preliminary Data – Not corrected for background absorption

Summary

- Detection of major carbonaceous species
- Advantages
  - Facilitates spectral fitting
  - Spectral interferences are unstructured
  - No beam steering
- Imaging of axisymmetric flames
  - Tomographic imaging
- Diagnostic development in steady low-pressure laminar flames
  - Progression from simple to complex fuels
  - Flames with soot precursors
- New opportunities using other light sources?

Acknowledgements

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