An Experimental and Modeling Study of NO_x- CO Formation in High Hydrogen Content (HHC) Fuels Combustion in Gas Turbine Applications DE-FE0012005

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Project Participants

University of South Carolina and Princeton University

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- Sheikh Farhan Ahmed – Graduate Student
- Fahd Ebna Alam – Graduate Student

Co-PI: **Dr. Bihter Padak**, Assistant Professor
University of South Carolina– Chemical Engineering

- Nazli Asgari – Graduate Student

Co-PI: **Dr. Frederick Dryer**, Professor
Princeton University – Mechanical and Aerospace Engineering

- Mac Haas – Graduate Student

Objectives

- Detailed and validated coupled HHC + NO_x kinetic model

.• New experimental data for speciation for the oxidation kinetics of HHC fuel compositions in presence of impurities.

- Understanding of CO, NO and NO_2 formation and concentration in shear layer flow regimes (hot and cold flow interactions). NO → NO_2 conversion in hot-cold shear layer interaction and EGR.

- Detailed and reduced kinetic model for HHC fuels including detailed fuel compositions and NO_x.

Tasks

- Study of reactivity and speciation data of NO_x under various conditions

- Studies of CO, NO, NO_2 formation and conversion for NO → NO_2 in shear/mixing layers

- Studies of high pressure HHC fuel kinetics using High Pressure Laminar Flow Reactor (HPLFR)

- Kinetic assessment, validation and development of a comprehensive HHC fuel + NO_x kinetic mechanism

Presentation Outline

- Research Team Members
- Project Objectives
- Research Tasks
- Year 1 progress
  - ❑ Kinetic Modeling of NO_x formation in HHC Fuels
    - – Tanvir Farouk
  - ❑ Experimental Setup for Speciation Measurements
    - – Bihter Padak
  - ❑ Measurement of Small Species Data
    - – Frederick Dryer
- Summary

Kinetic Modeling

Mechanisms

Saudi Aramco C_1-C_4 Mechanism (2012)
- 253 species
- 1536 reactions

Princeton C_0 Mechanism (2011)
- 13 species
- 27 reactions

USC C_1-C_4 Mechanism (2007)
- 111 species
- 784 reactions

SanDiego C_1-C_3 Mechanism (2002)
- 21 species
- 93 reactions

GRI Mechanism (1999)
- 53 species
- 325 reactions

C_0 – C_4 Models

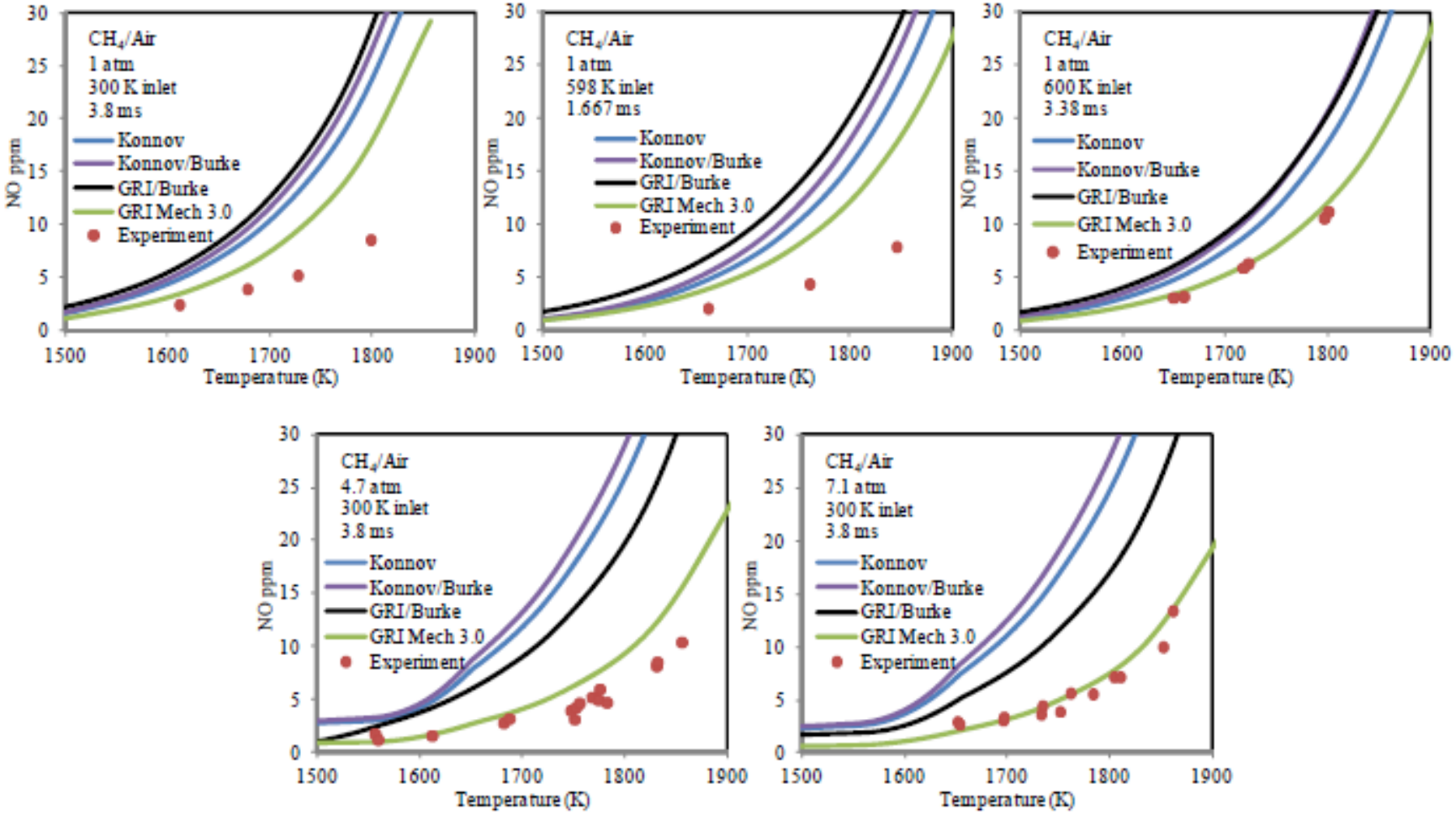
Konnov NO_x Mechanism (2009)
- Fenimore
- N_2O

Rasmussen NO_x Mechanism (2008)
- N_2O absent
- N_xH_y absent

Dagaut NO_x Mechanism (2006)
- N_2O
- Limted C_xN_y

NO_x Models

Model Performance: Jet Stirred Reactor

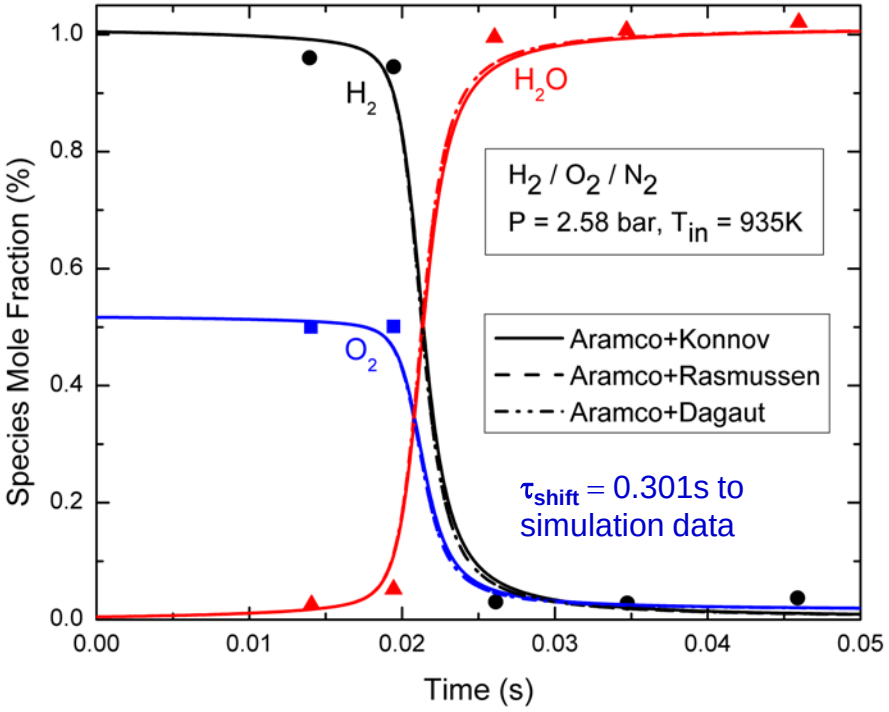


- Jet stirred characteristic reaction time determined based upon predicted laminar flame speed. Burke et al hydrogen model was substituted f for the comparable submodel in the named mechanisms.
- Model predictions suggest that experimental NO_x results are principally dependent on thermal NO_x generation.
- The GRI-Mech 3.0 reaction rate constant for N2+O-NO+N is substantially less than in Konnov (which uses a critical review result)

Konnov model: Combust Flame 156 (11) (2009) 2093-2105
GRI-Mech 3.0: GRI-Mech 3.0. http://www.me.berkeley.edu/gri_mech/
Burke et al. (2012) ; Int. J. Chem. Kinet 44 (7) (2012) 444-474

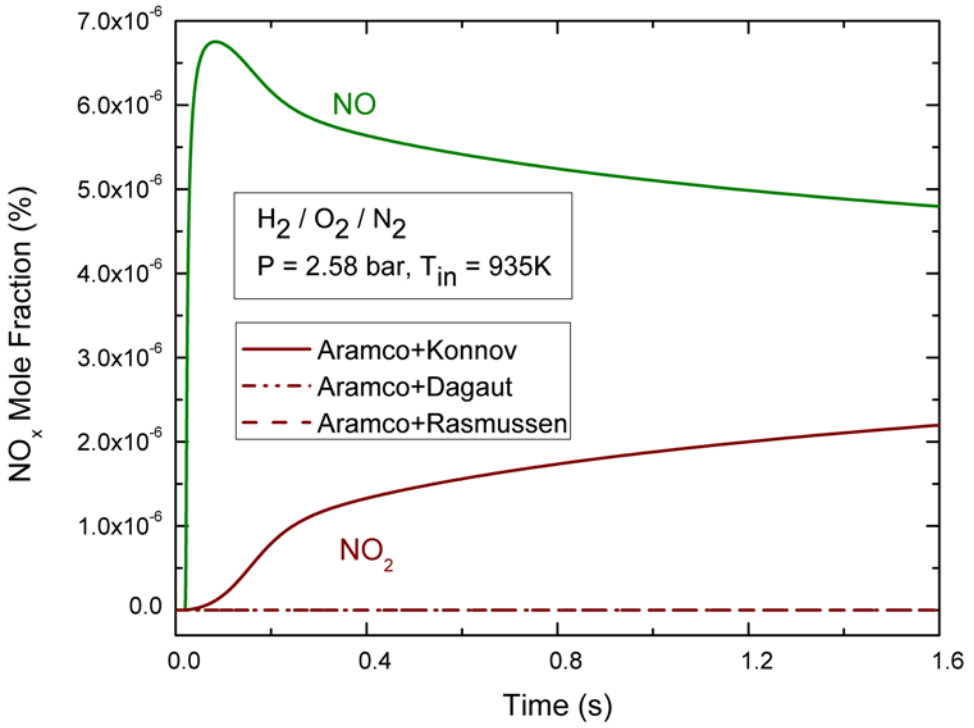
Data from Steele, Ph.D, University of Washington, 1995.
Predictions from prior Industrial sponsored research at Princeton (SIEMENS)

Performance of H_2- NO_x Models



Temporal evolution of species evolution for H_2 oxidation at elevated pressure. Experimental data of Mueller et al. IJCK, (1999) 113-125.

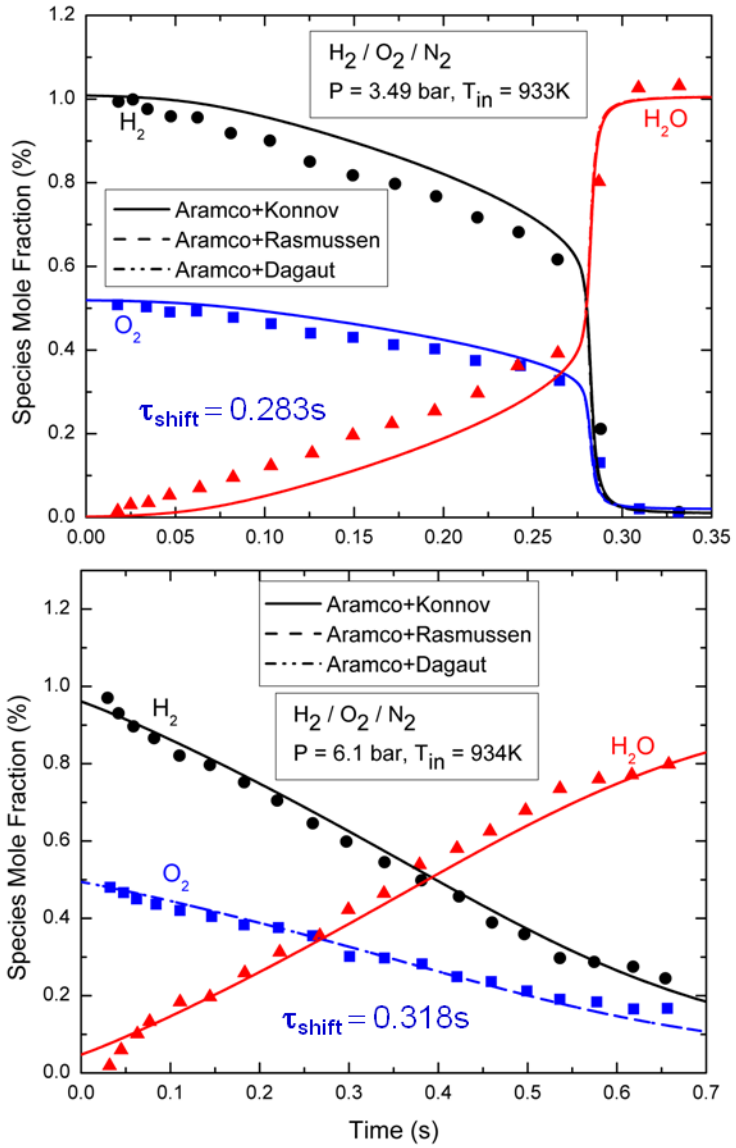
- Fuel oxidation kinetics – very good agreement with experimental measurements.
- Different NO_x pathways – inconsistency in NO_x concentration predictions.
- $No_x$for pure H_2 is not consistent.



Temporal evolution of NO, NO_2 evolution for H_2 oxidation at elevated pressure.

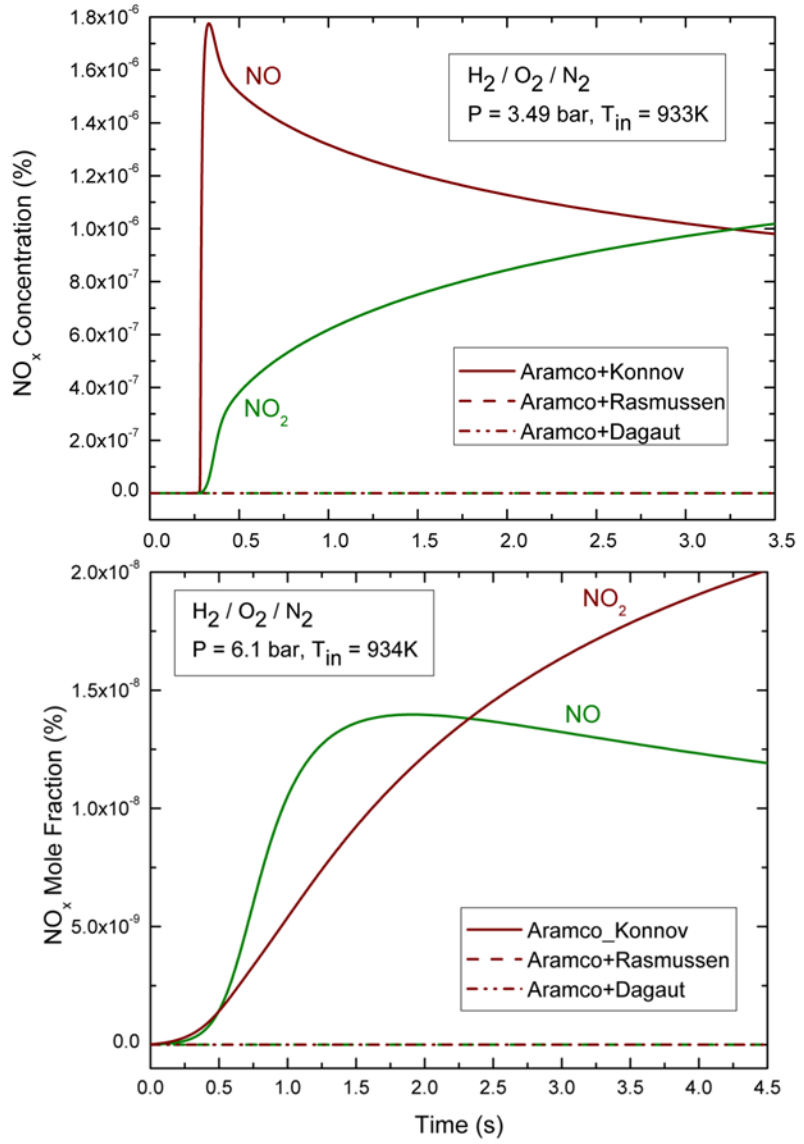
More target data for NO_x !

Performance of H_2- NO_x Models



Temporal evolution of species evolution for H_2 oxidation at elevated pressure. Experimental data of Mueller et al. IJCK, (1999) 113-125.

Konnov – predicts higher NO_x . Rasmussen and Dagaut shows negligible concentration

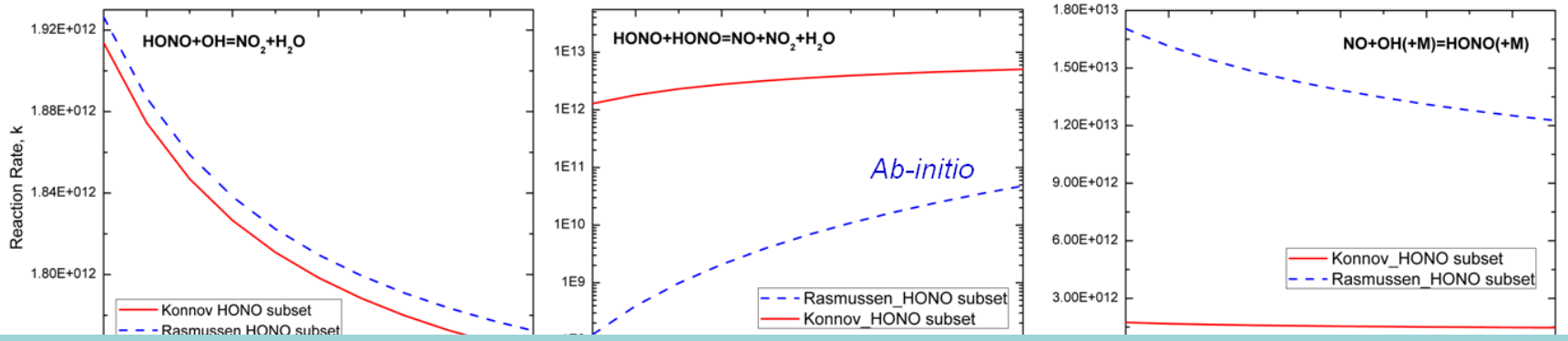


H_2- NO_x Models

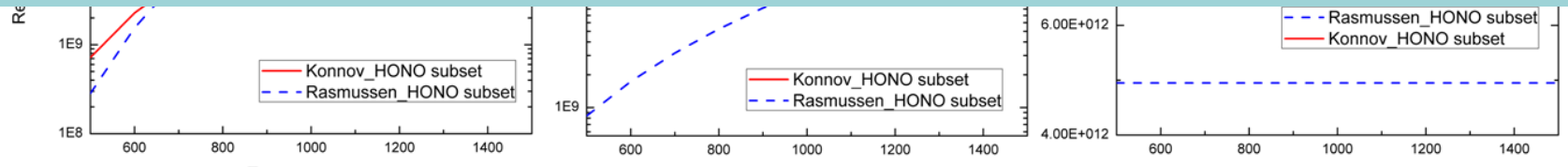
Reactions	Rasmussen	Konnov	Dagaut
NO+OH(+M)=HONO(+M)	√	√	√
NO_2+H_2=HONO+H	√	√	√
$NO_2+HO_2=HONO+O_2$	√	√	√
NO_2+HCO=HONO+CO	√	√	√
NO_2+CH_2O=HONO+HCO	√		√
HNO+NO_2=HONO+NO	√	√	√
HONO+O=NO_2+OH	√		√
HONO+OH=NO_2+H_2O	√	√	√
HONO+$NO_2=HONO_2$+NO	√		
HONO+HONO=NO+NO_2+H_2O	√	√	√
$NO_2+CH_2O=HNO_2$+HCO	√	√	
HNO_2+O=NO_2+OH	√		√
HNO_2+OH=NO_2+H_2O	√		√
NO_2+OH(+M)=$HONO_2$(+M)	√		√
$HONO_2+H=H_2+NO_3$	√		
$HONO_2$+H=H2O+NO_2	√		
$HONO_2+OH=H_2O+NO_3$	√		
HNO_2(+M)=HONO(+M)	√		
$HONO_2$+H=OH+HONO	√		
$NO_2+H_2=HNO_2$+H	√		√
$NO_2+HO_2=HNO_2+O_2$	√		

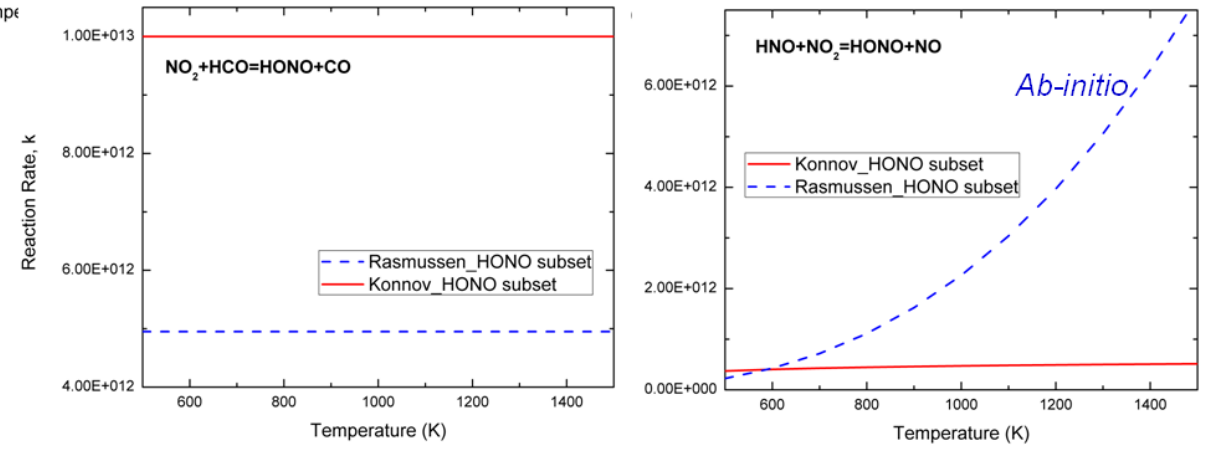
- HONO pathway is a source of difference.
- Most models have different HONO pathways – HNO_2, $HONO_2$ are unaccounted for.
- Updates to HONO pathways and species.

H_2- NO_x Models – Variation in Reaction Rates



Orders of magnitude differences in the rates





H_2- NO_x Models – Proposed Updates

Reactions
$NO_2+CH_2O=HONO+HCO$
$HONO+O=NO_2+OH$
$HONO+NO_2=HONO_2+NO$
$NO_2+CH_2O=HNO_2+HCO$
$HNO_2+O=NO_2+OH$
$HNO_2+OH=NO_2+H_2O$
$NO_2+OH(+M)=HONO_2(+M)$
$HONO_2+H=H_2+NO_3$
$HONO_2+H=H2O+NO_2$
$HONO_2+OH=H_2O+NO_3$
$HNO_2(+M)=HONO(+M)$
$HONO_2+H=OH+HONO$
$NO_2+H_2=HNO_2+H$
$NO_2+HO_2=HNO_2+O_2$

- HONO pathway is a source of difference.
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H_2- NO_x Models – Proposed Updates

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$NO_2+CH_2O=HONO+HCO$
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$NO_2+CH_2O=HNO_2+HCO$
$HNO_2+O=NO_2+OH$
$HNO_2+OH=NO_2+H_2O$
$NO_2+OH(+M)=HONO_2(+M)$
$HONO_2+H=H_2+NO_3$
$HONO_2+H=H2O+NO_2$
$HONO_2+OH=H_2O+NO_3$
$HNO_2(+M)=HONO(+M)$
$HONO_2+H=OH+HONO$
$NO_2+H_2=HNO_2+H$
$NO_2+HO_2=HNO_2+O_2$

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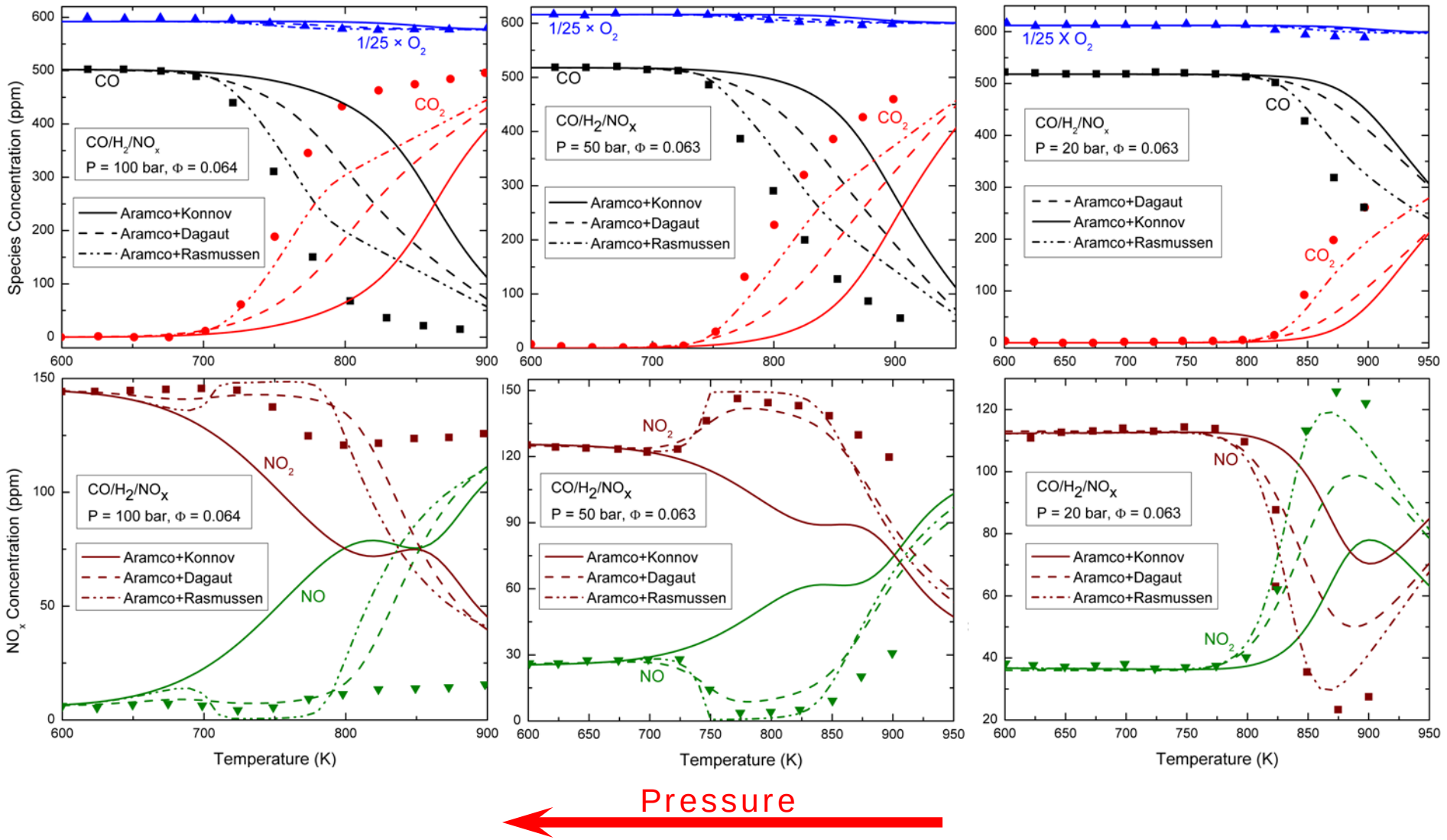
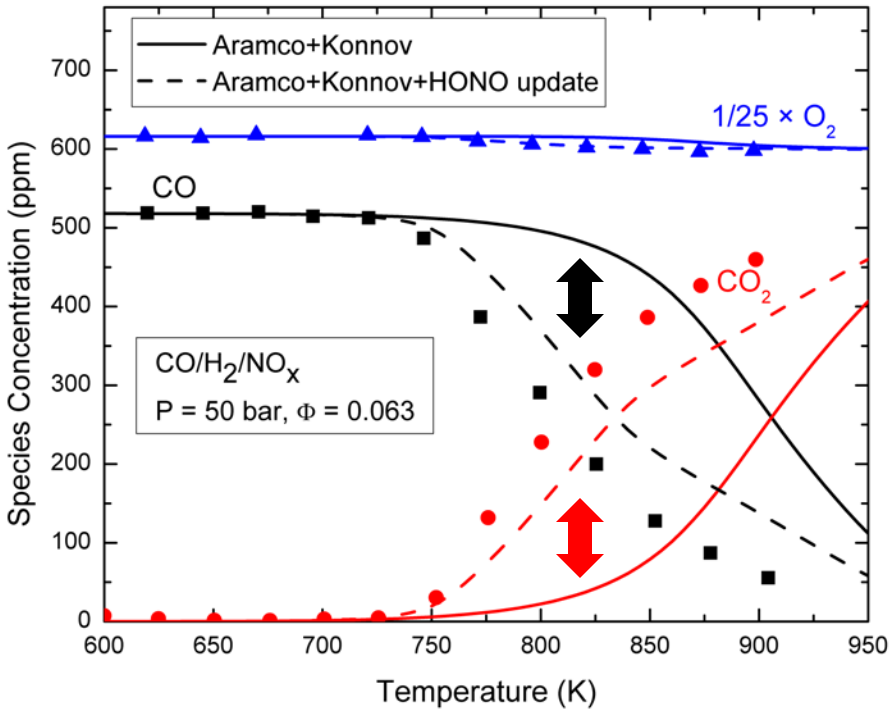
Flow Reactor Reactivity – Base Cases

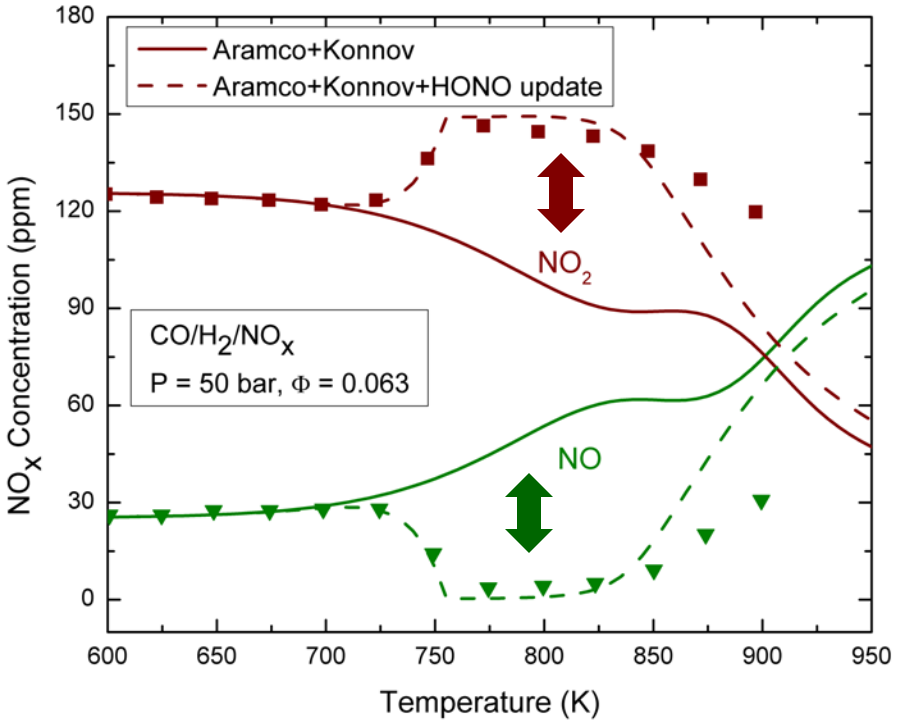


Discrepancy between predictions and measurements – NO_x but also H_2/CO oxidation !!!

Rasmussen C. L., Hansen J., Marshall P, Glarborg P., IJCK, (2008), 454-480

Flow Reactor Reactivity – HONO updates



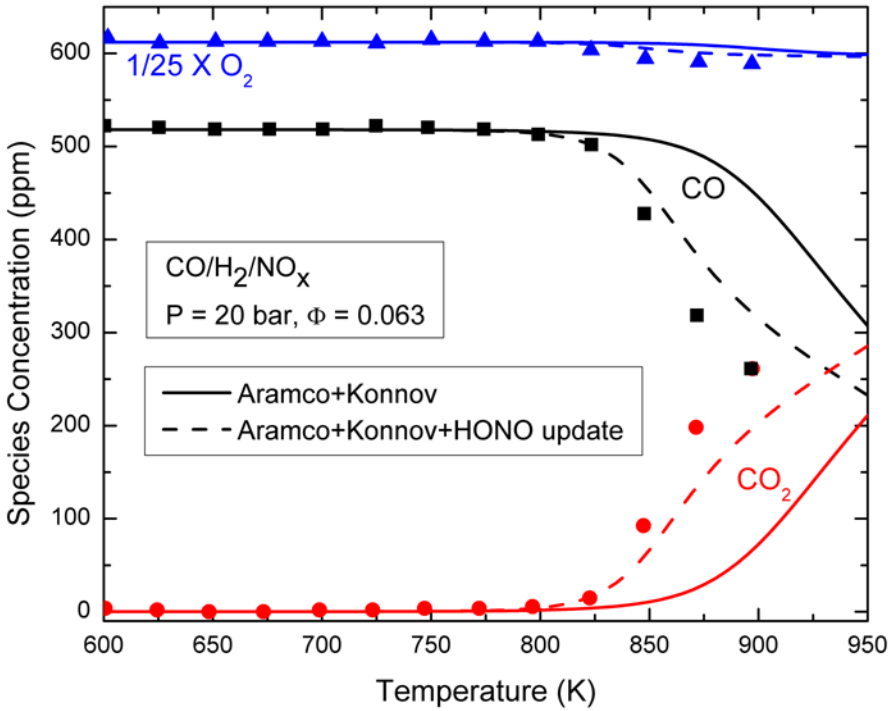


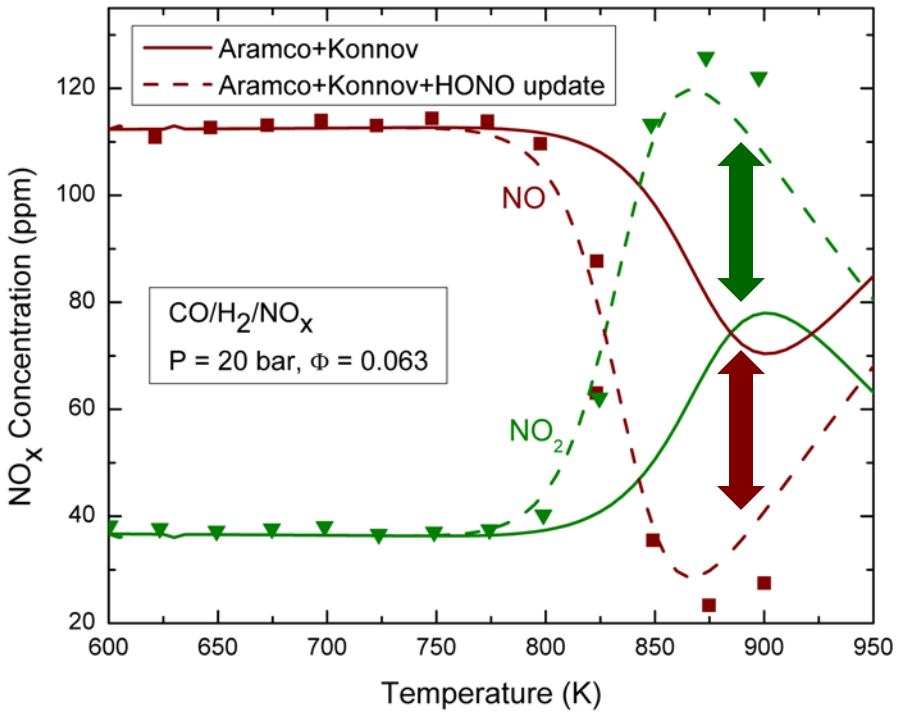
Flow reactor reactivity data for $CO/H_2/NO_x$ oxidation at 50 bar and 0.063 equivalence ratio. Experimental data for that of Rasmussen et al. IJCK, (2008) 454 – 480. Konnov NO_x model.

- Inclusion of additional HONO pathways significantly improves the NO_x predictions.
- An increase in the reactivity of CO is also apparent

– Oxidation of fuel is influenced!

Flow Reactor Reactivity – HONO updates

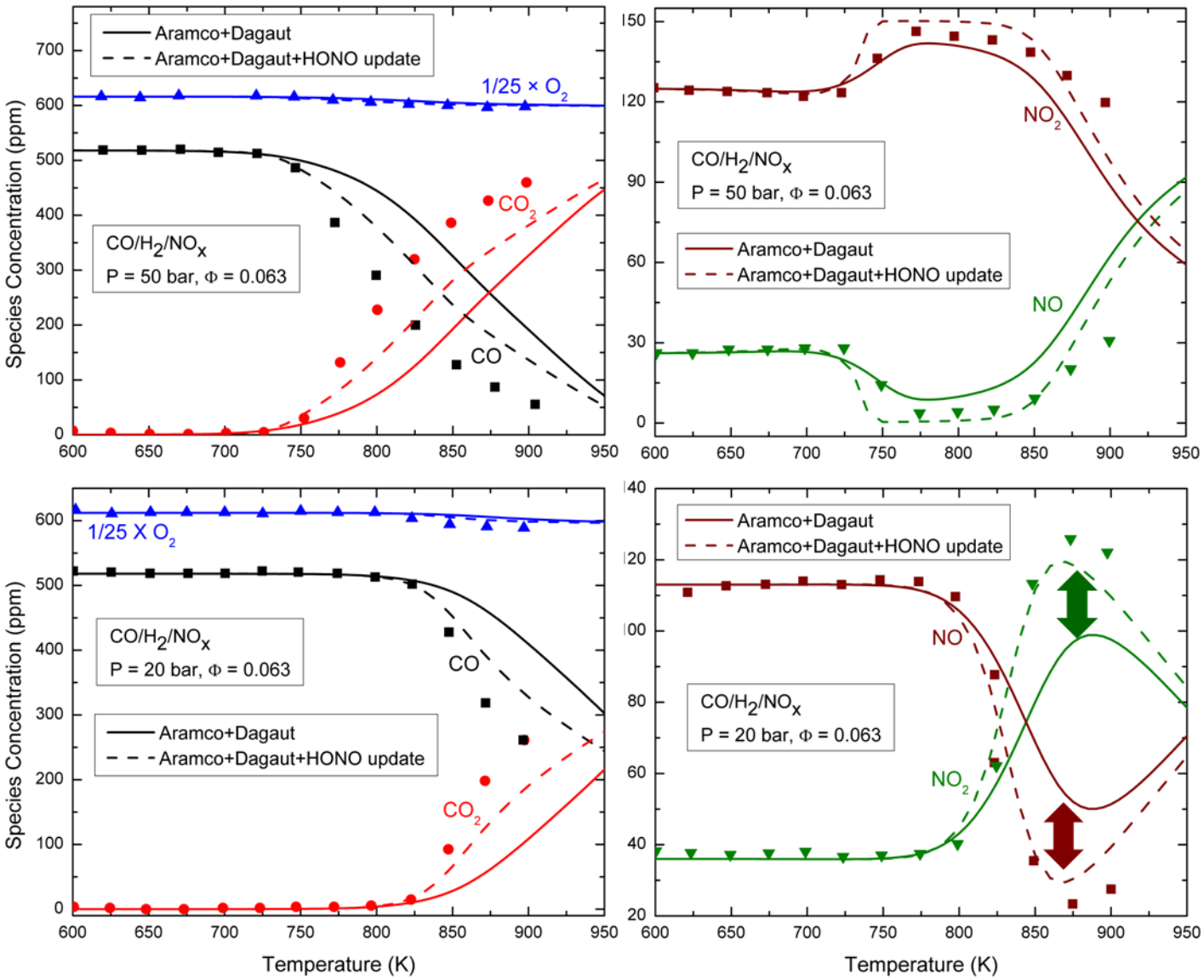




Flow reactor reactivity data for $CO/H_2/NO_x$ oxidation at 20 bar and 0.063 equivalence ratio. Experimental data for that of Rasmussen et al. IJCK, (2008) 454 – 480. Konnov NO_x model.

- Similar trends are observed for 20 bar conditions.

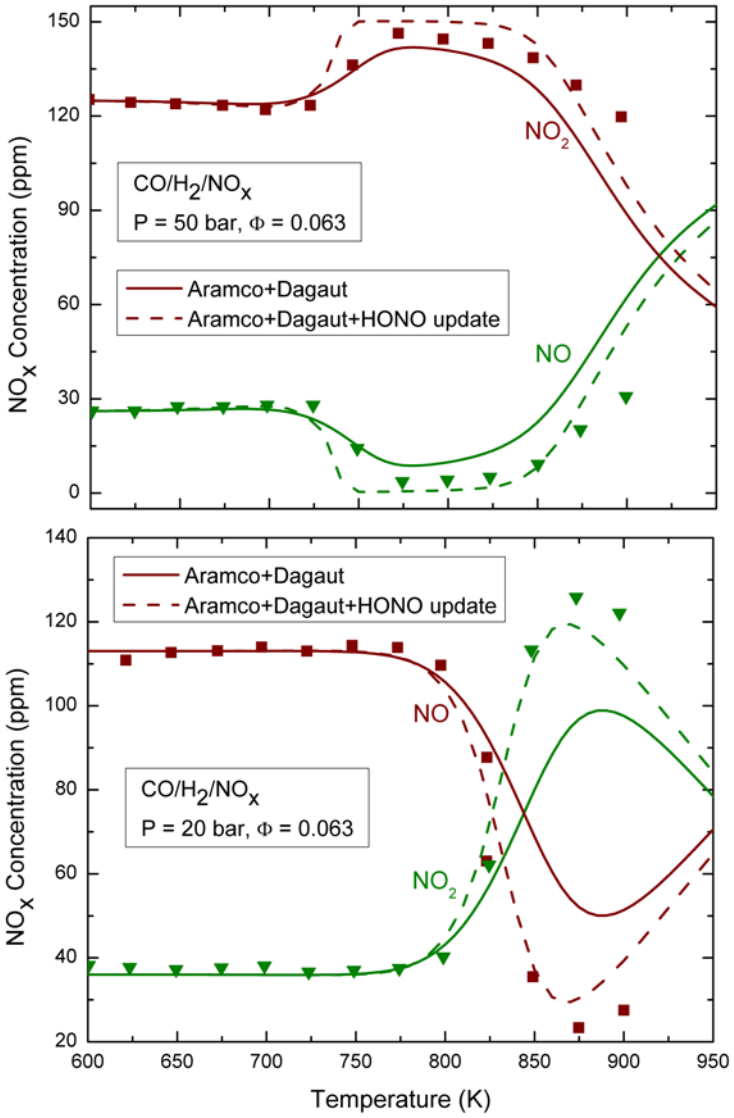
Flow Reactor Reactivity – HONO updates



- Similar performance improvement for the Dagaut model.
- Dagaut NO_x contains a limited subset of HONO pathway.

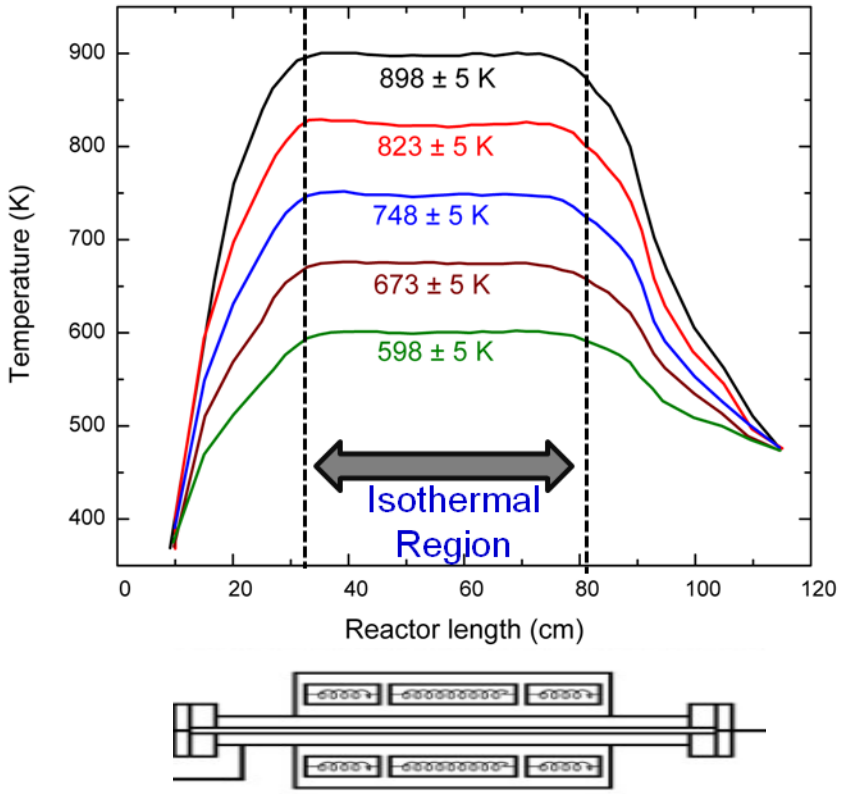
Flow reactor reactivity data for $CO/H_2/NO_x$ oxidation at 50 bar and 20 bar for 0.063 equivalence ratio. Experimental data for that of Rasmussen et al. IJCK, (2008) 454 – 480. Dagaut NO_x model.

Possible Sources of Discrepancy



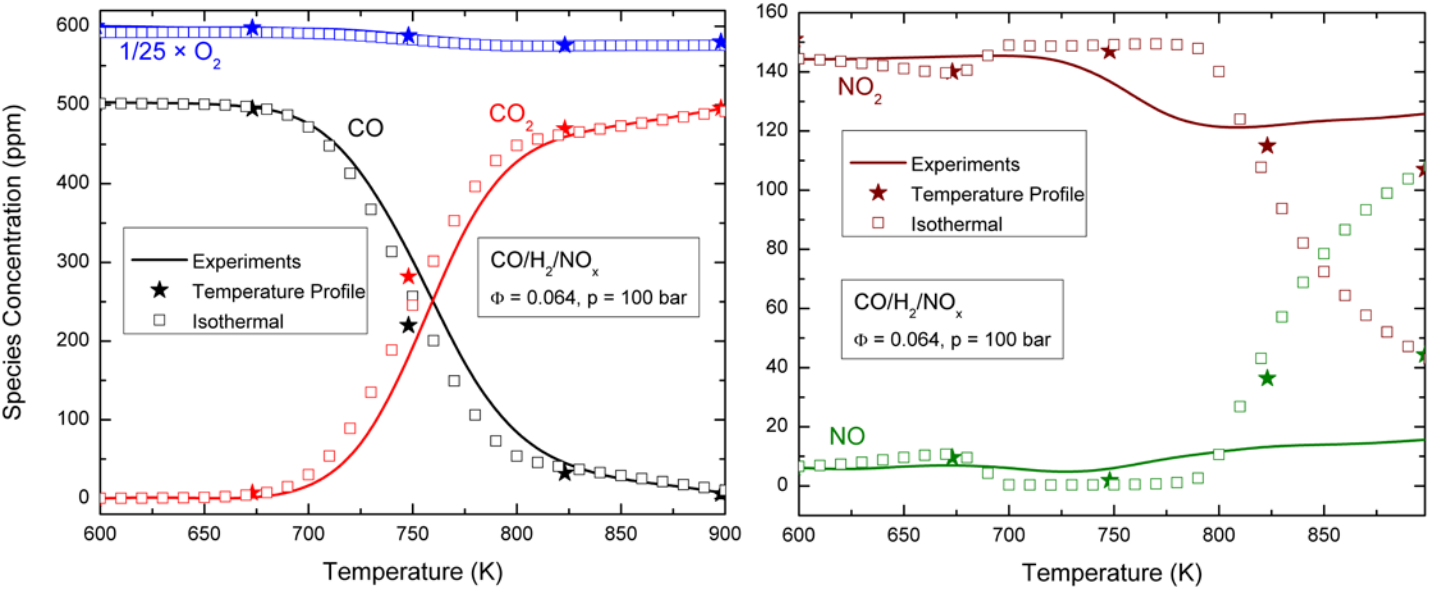
- Simulations are conducted for an isothermal configuration!!

Ramp up and cool down regions in experiments

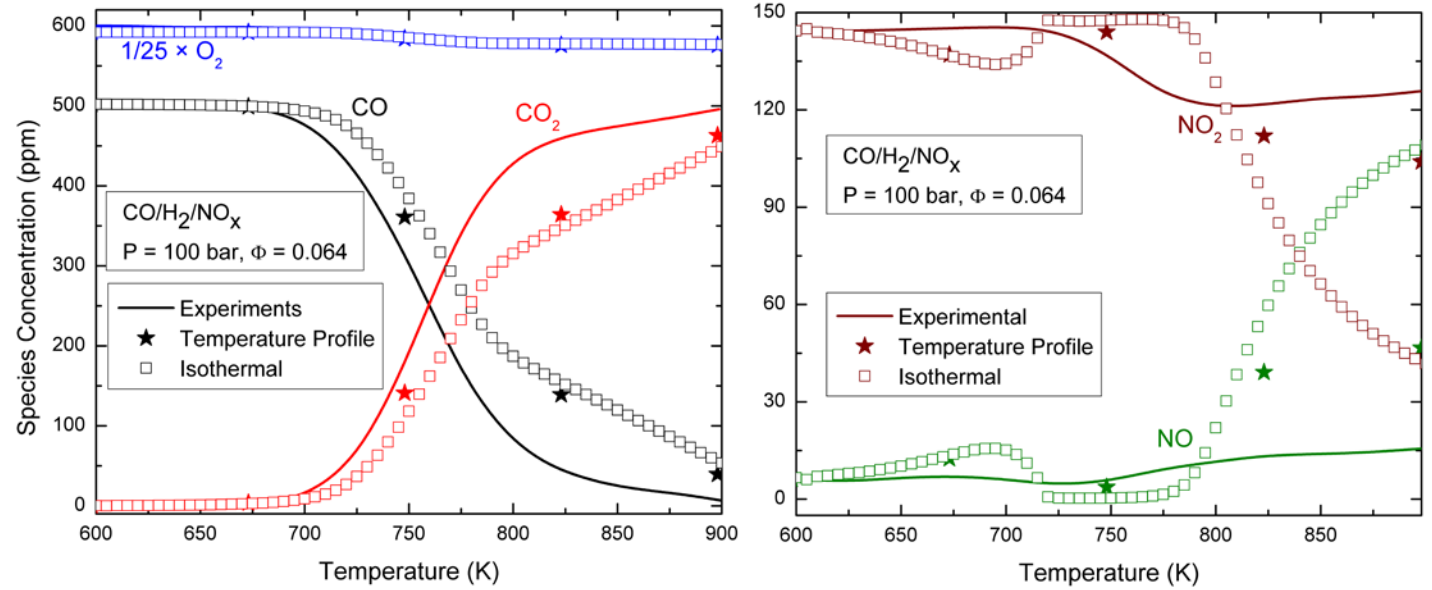


Prescribed temperature profile in the Rasmussen et al. flow reactor experiments

Influence of Prescribed Temperature Profile



Rasmussen model (HONO updates) with temperature profile

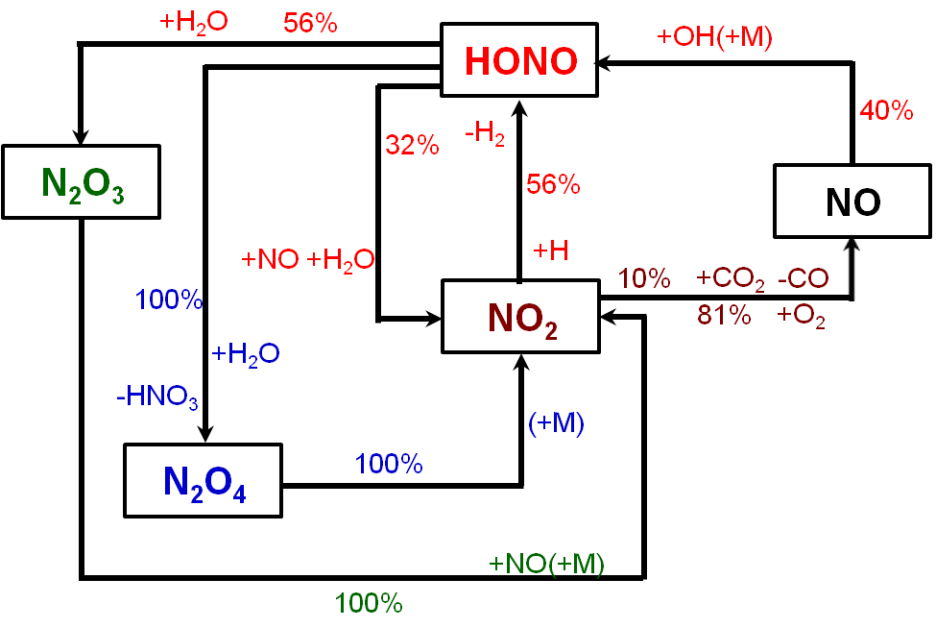


Konnov model (HONO updates) with temperature profile

- Limited temperature profile simulations were conducted – due to the limited set of available data.
- Temperature profile does affect the predictions – ramp up and cool down influences the kinetics.

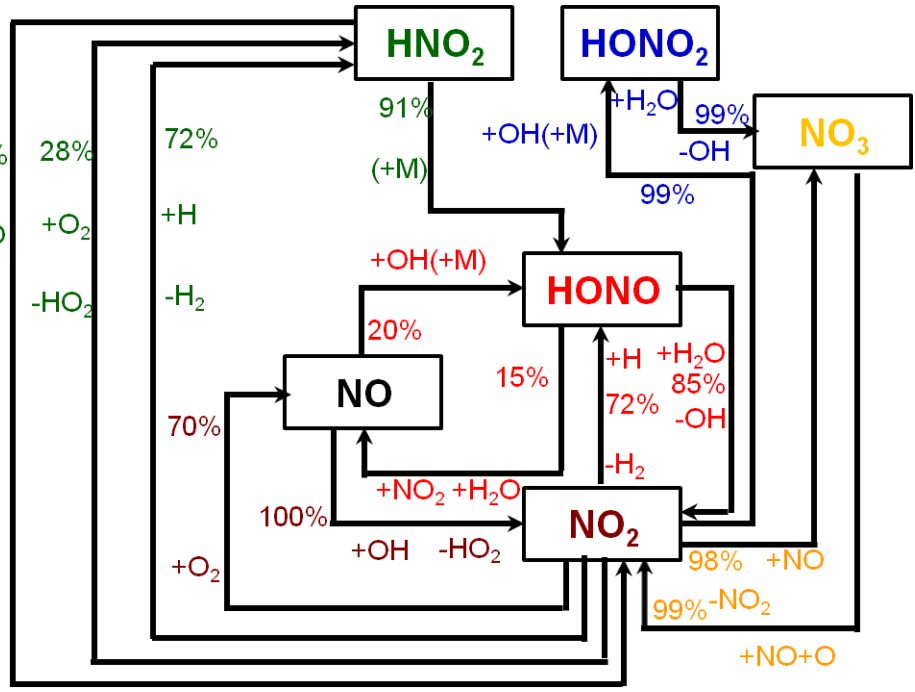
Differences in HONO

Konnov HONO



• HONO is solely produced from NO and NO_2 and is consumed only 32% to NO_2 and the rest to N_2O_3 and N_2O_4, which are eventually contributing in the formation of NO_2.

HONO updates

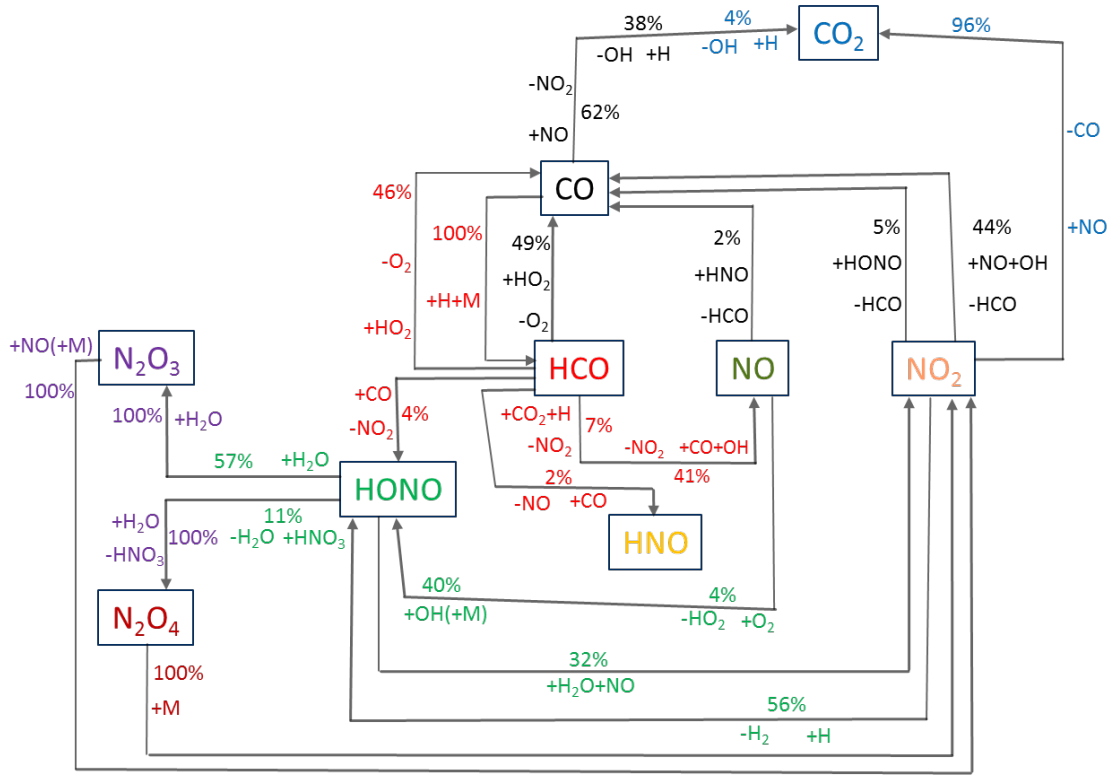


• Significantly different from the Konnov-HONO subset.

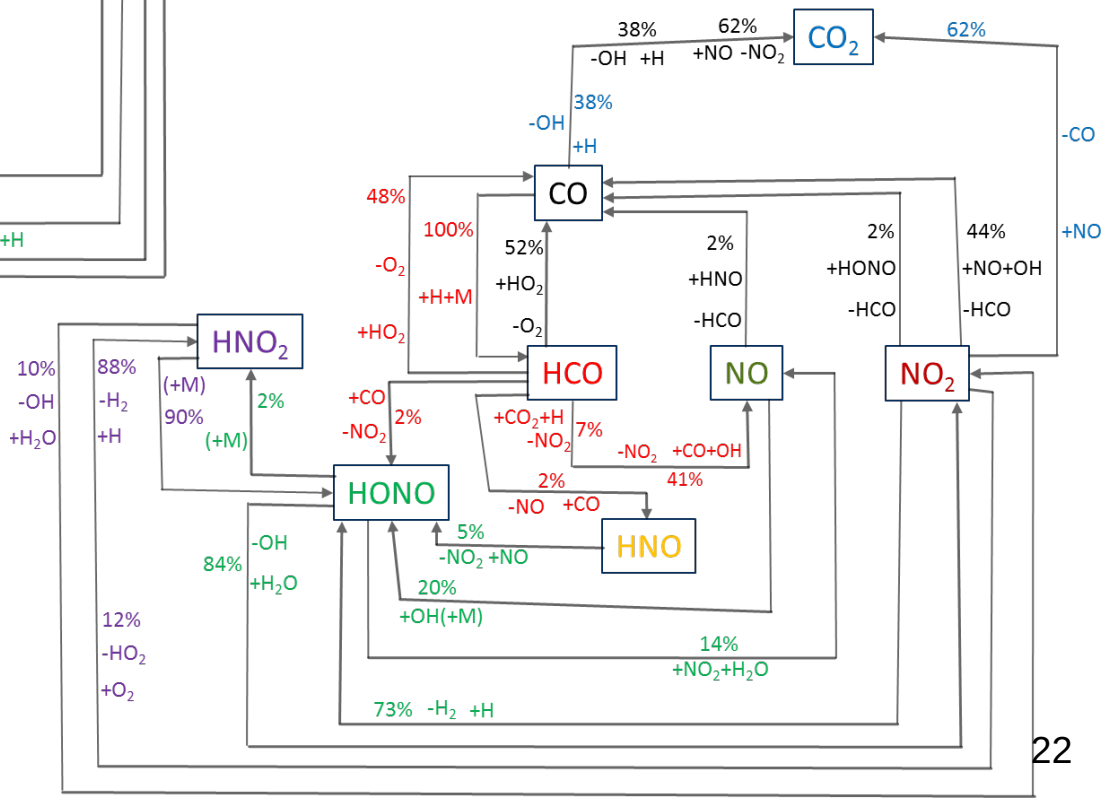
• HONO is produced from NO_2, NO and HNO_2 and is consumed mainly to form NO_2 and NO.

• The species HNO_2 and $HONO_2$ have contribution in the HONO paths through NO_2 and NO_3 respectively.

HONO Pathway - CO Oxidation



HONO updates

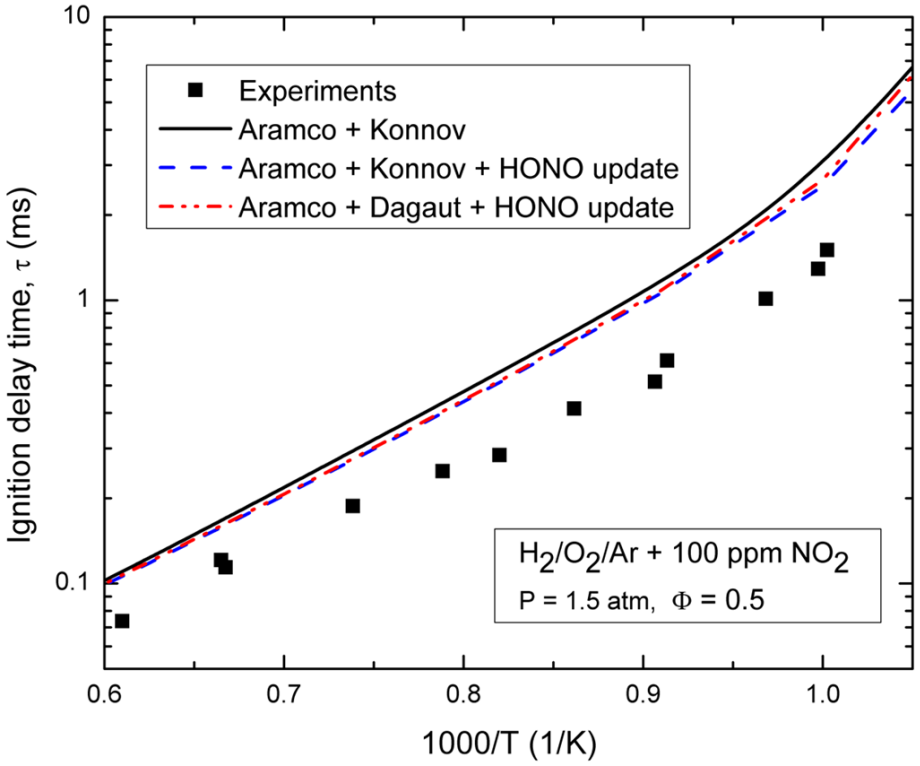


- HONO forms NO_2

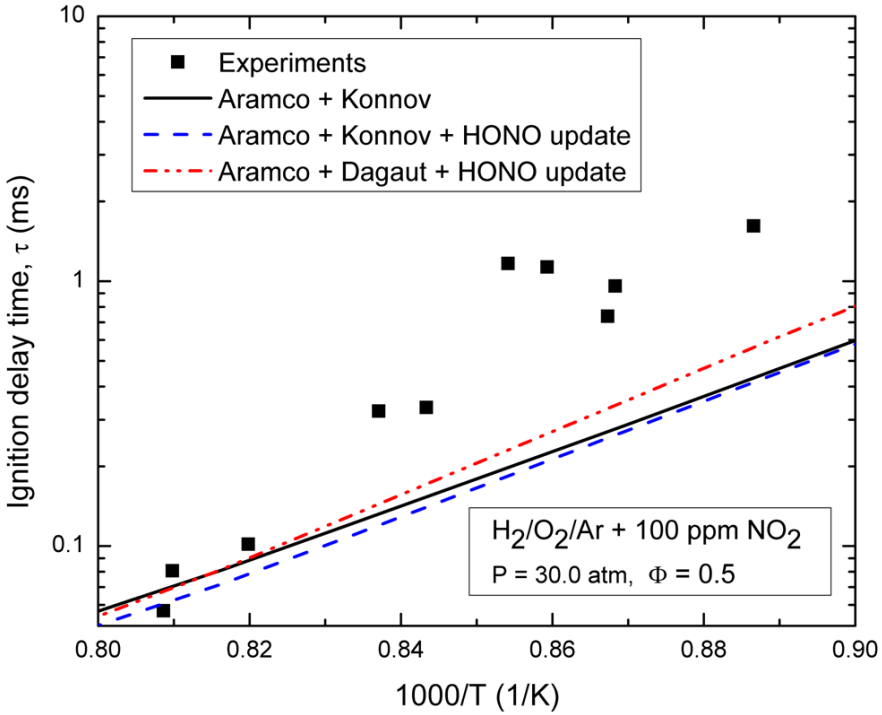
HONO + OH = H_2O + NO_2 in the updated model

- 38% of CO_2 formed by OH + CO

HONO Updates on Ignition Delay Predictions



Ignition delay time does not show significant variation with HONO-updates for H_2/O_2 system.



A. Keromnes, et al., "*An Experimental and Detailed Chemical Kinetic Modeling Study of Hydrogen and Syngas mixture oxidation at elevated pressures*", Combustion and Flame, 160 (2013), 995 – 1011.

Summary

- Potential critical pathways in H_2-NO_x has been identified

.

- HONO, HNO_2, $HONO_2$ updates are proposed

- The updates are found to have significant effect on the over all predictions of NO_x concentration in flow reactor reactivity over a broad range of pressures.

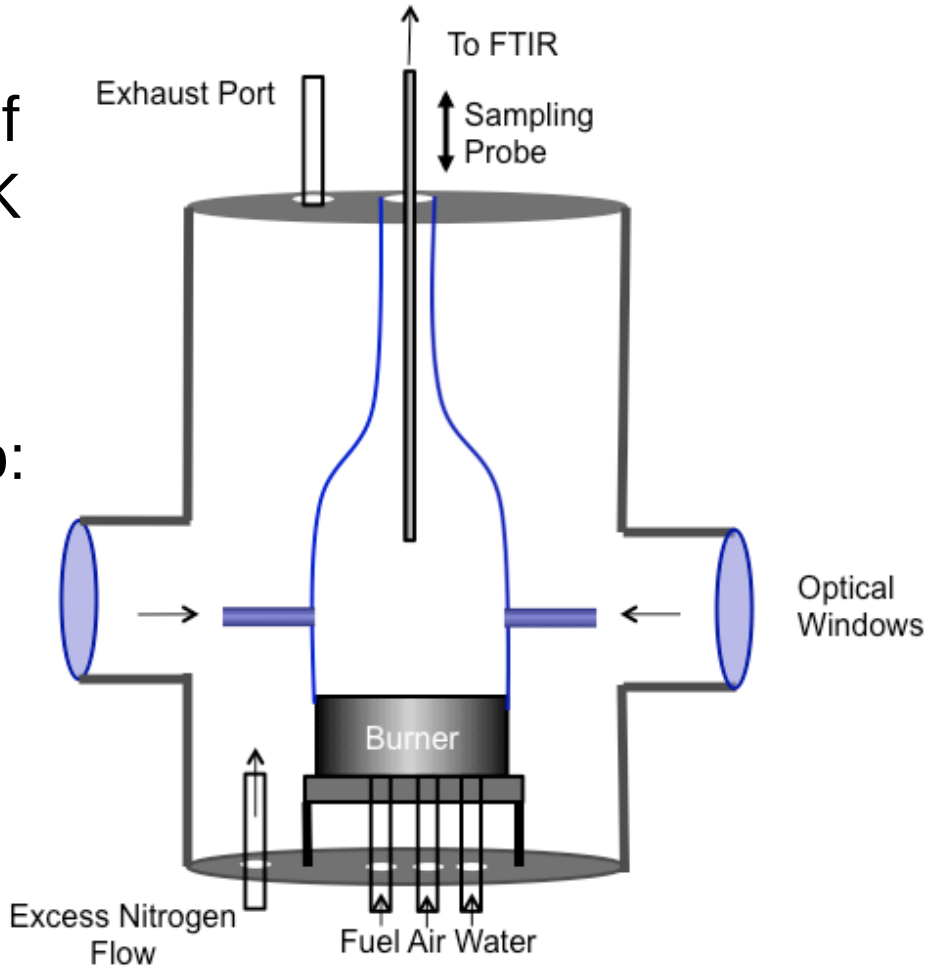
- The updates are also found to have a major influence on the CO oxidation which is critical to syngas combustion

Experimental Setup for Speciation Measurements

High Pressure Combustion Experiments

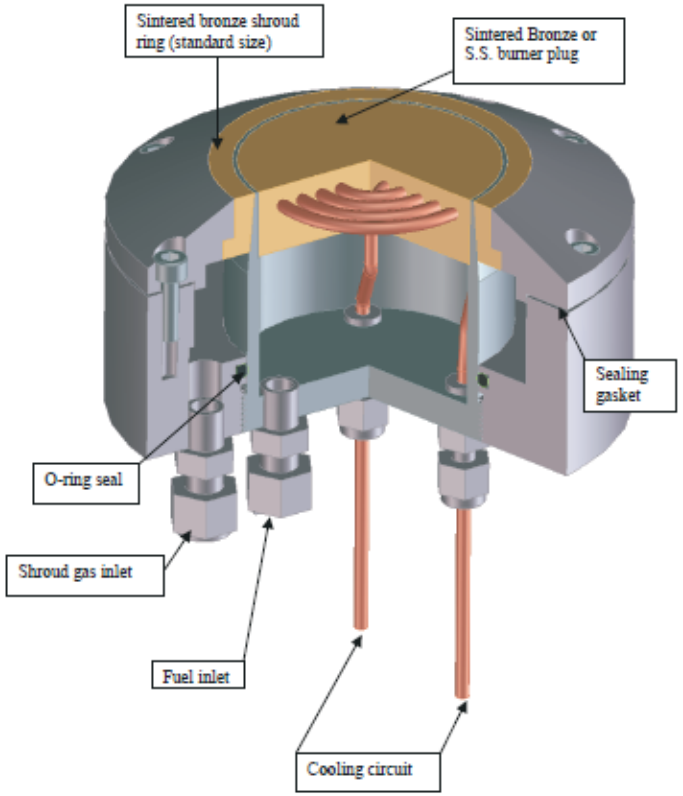
- Reactivity and speciation data of NO_x at 1 – 15 atm, 600 – 1400 K

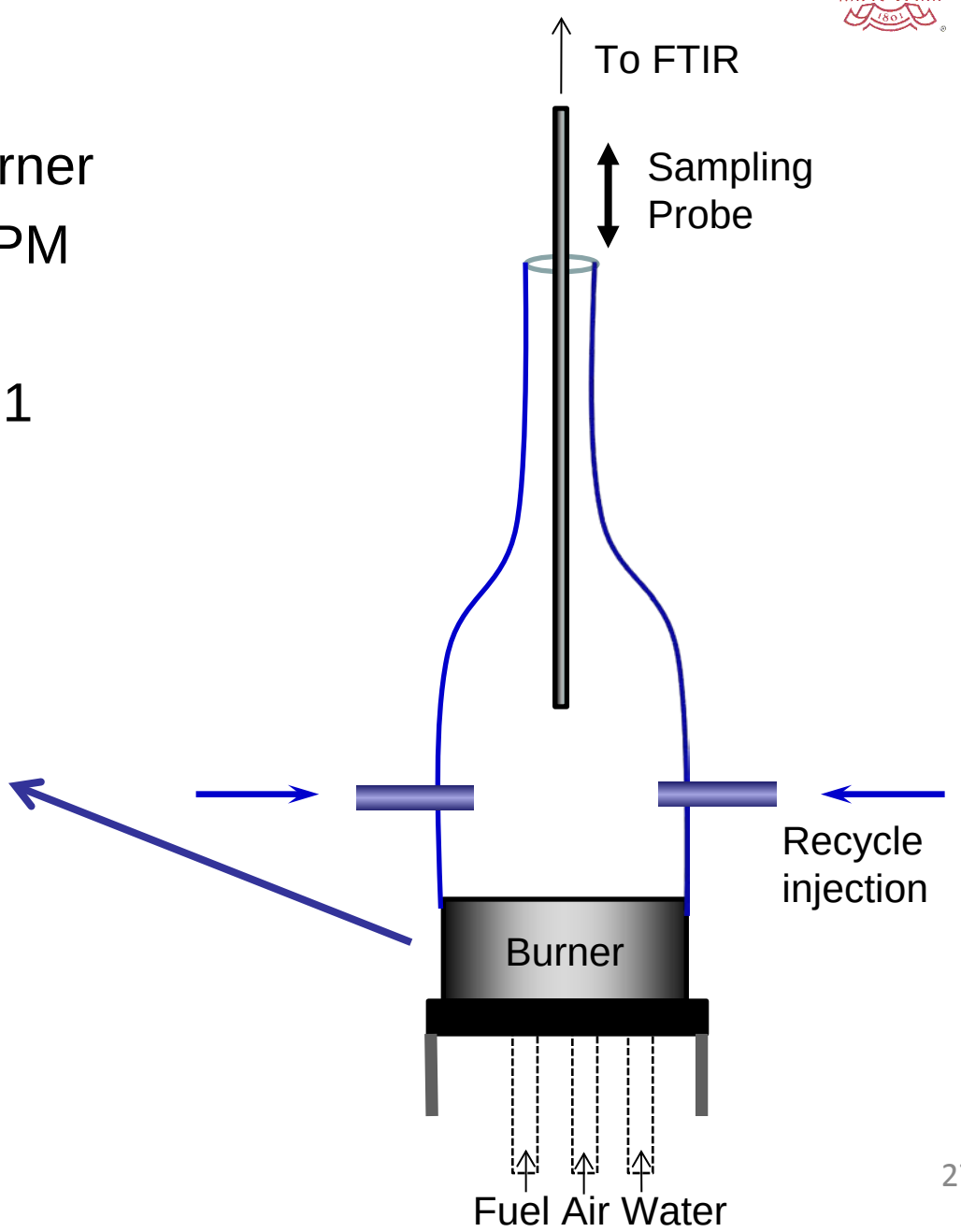
- Post-combustion NO_X formation will be measured with respect to:
  - Composition
  - Radial shearing from recycle



Combustion Experiments

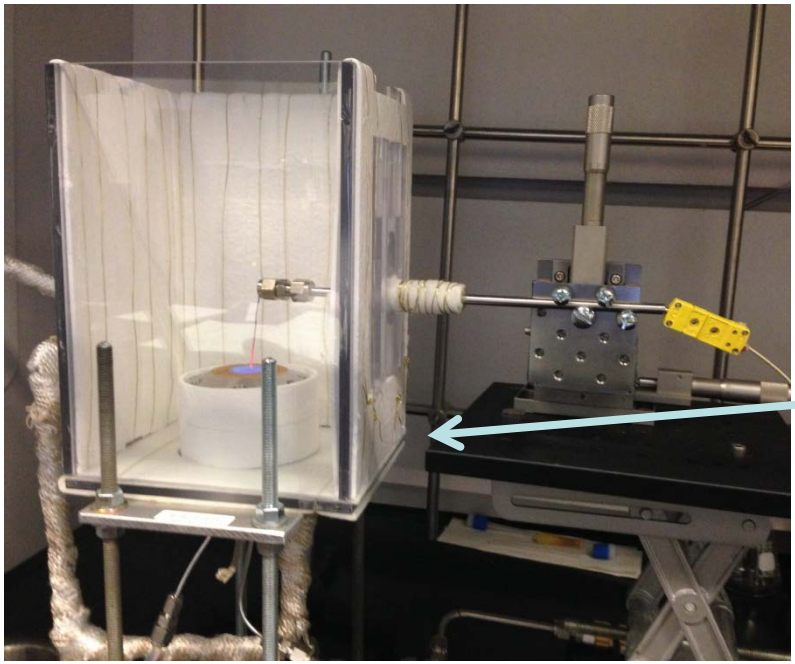
- Low-flow McKenna burner
- Total flow rate: 3-4 SLPM
- H_2/CO: 0.4-1
- Equivalence ratio: 0.4-1





Experimental Setup

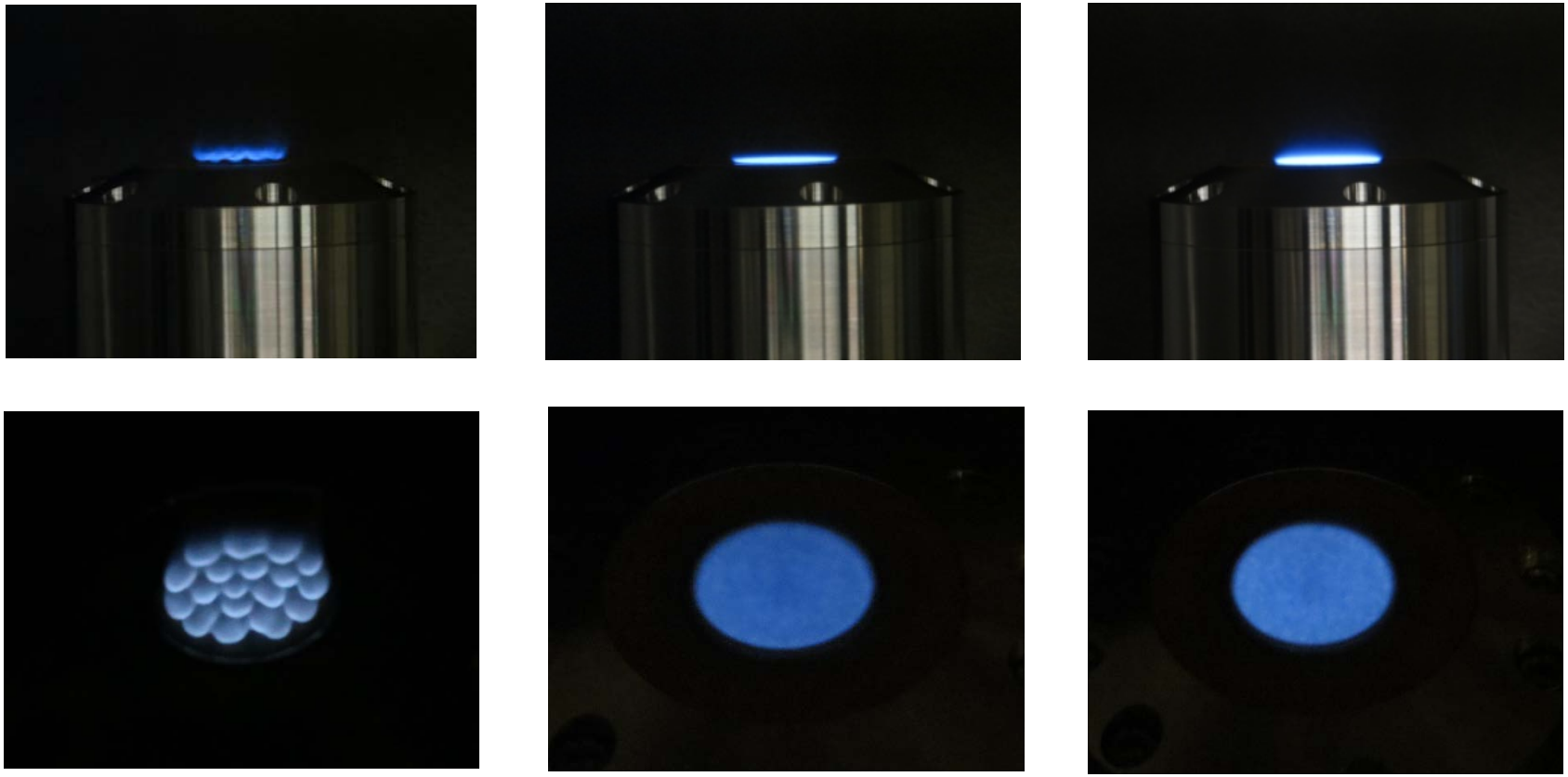
Experimental setup for atmospheric pressure experiments



Burner Tests

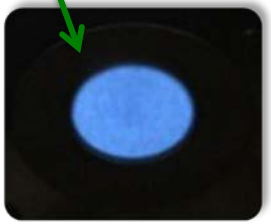
Changing ϕ (0.4-1) at constant flow rate of 4LPM, H_2/CO:1

ϕ = 0.4 **ϕ = 0.7** **ϕ = 1**



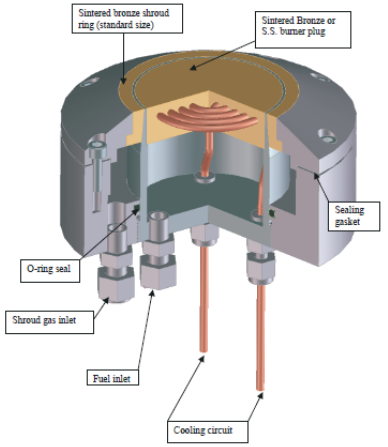
- Fuel-lean mixture (ϕ=0.4) results in cellular flame

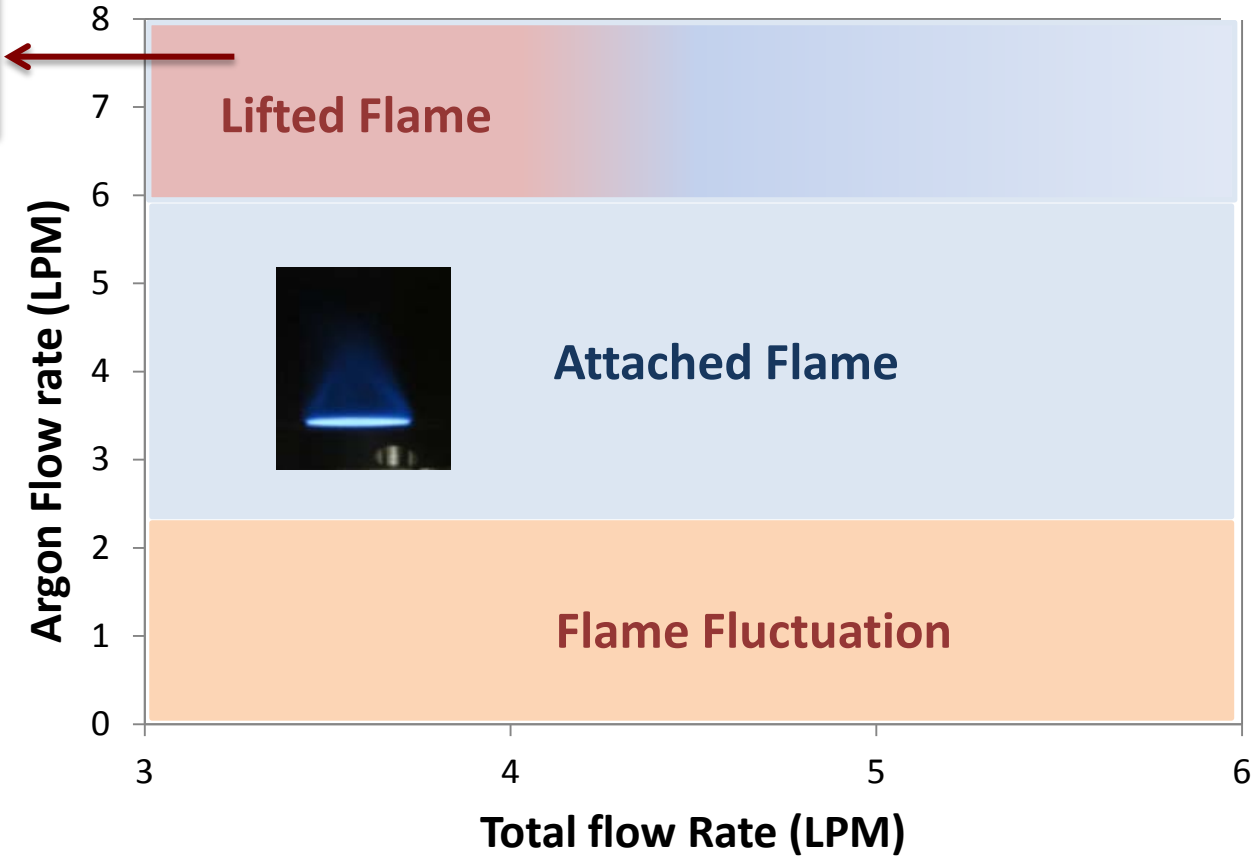
Flame Stability



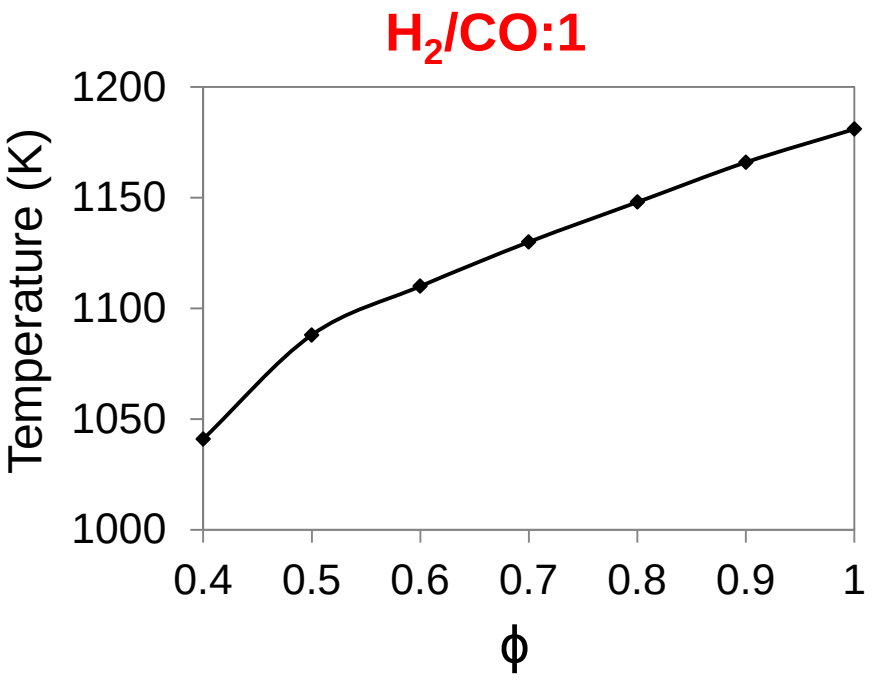
Flame Stability With Argon

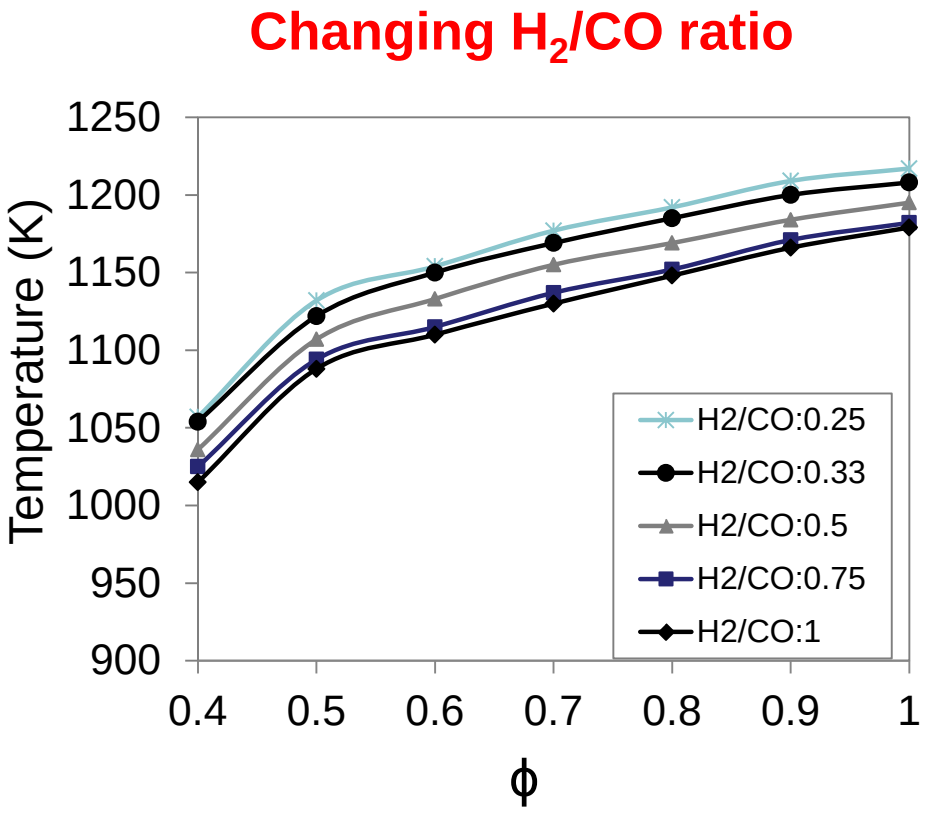
- Flame perturbations due to air entrainment
- Use argon as shroud gas





Flame Temperature vs Equivalence Ratio

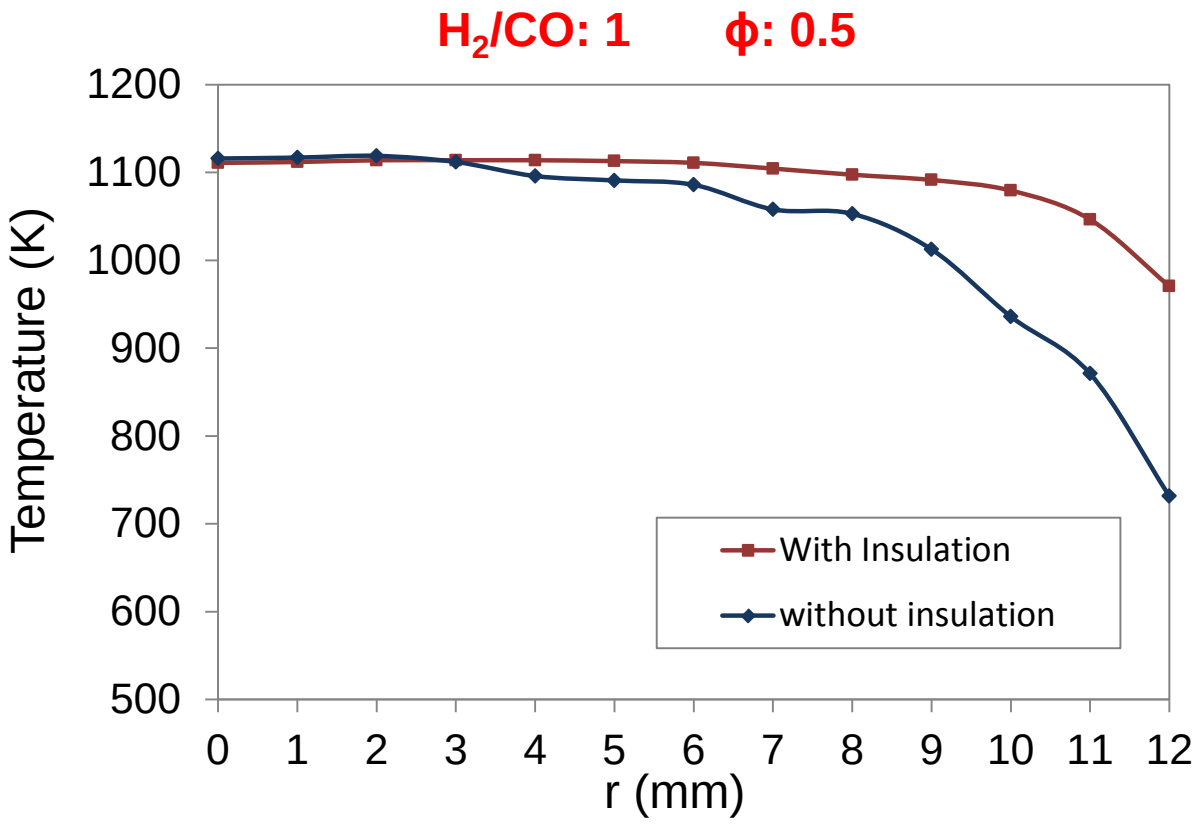


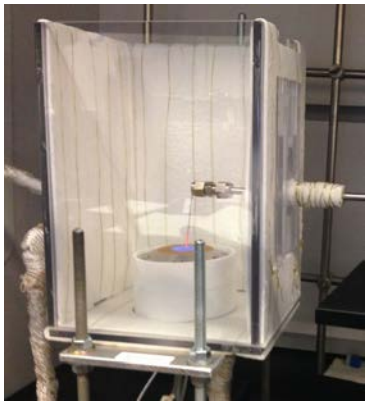


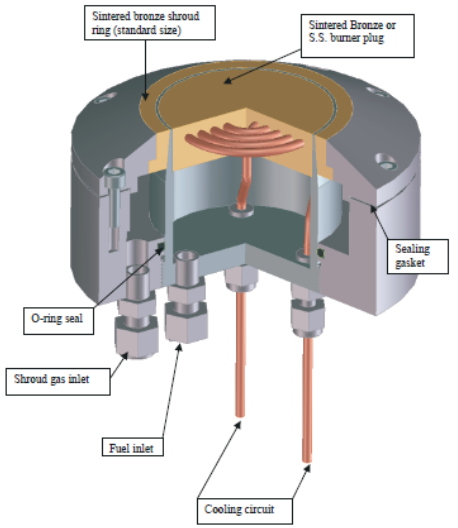
Decreasing H_2/CO ratio results in an increase in flame temperature

Radial Temperature Profile

Radial temperature profile 2 mm above the burner surface



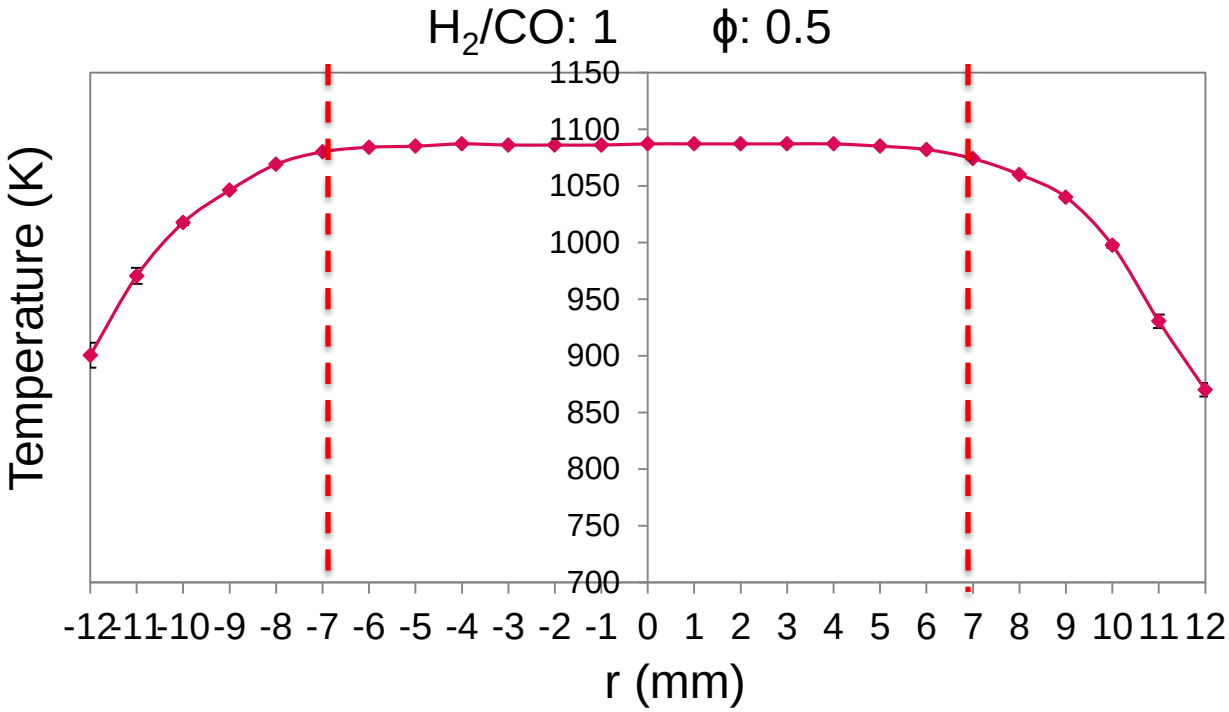


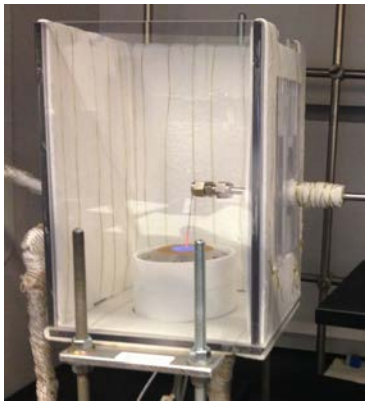


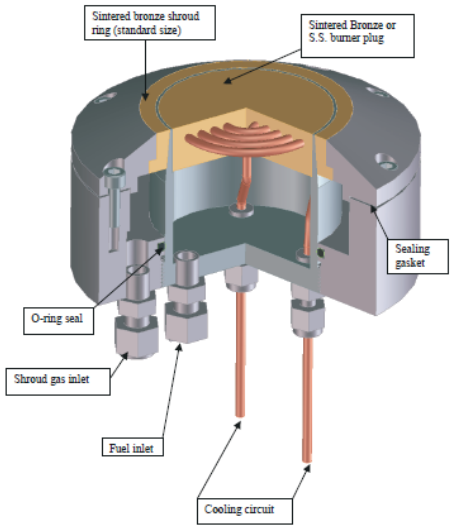
Radial profile is more uniform with insulation

Radial Temperature Profile

Radial temperature profile 2 mm above the burner surface





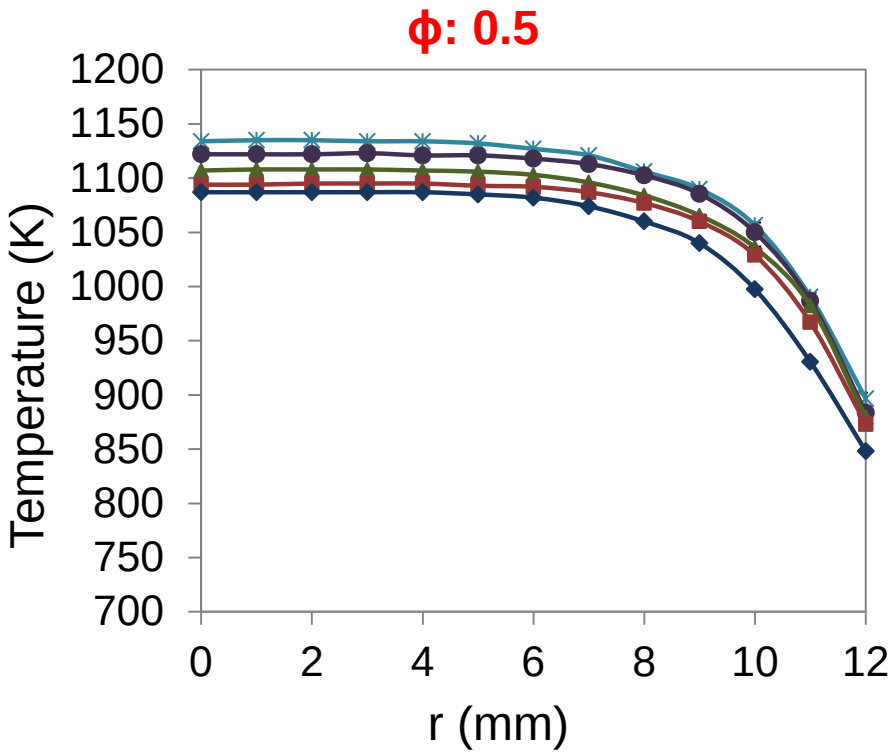


Temperature/concentration gradients towards the outer parts of the flame in McKenna burners *

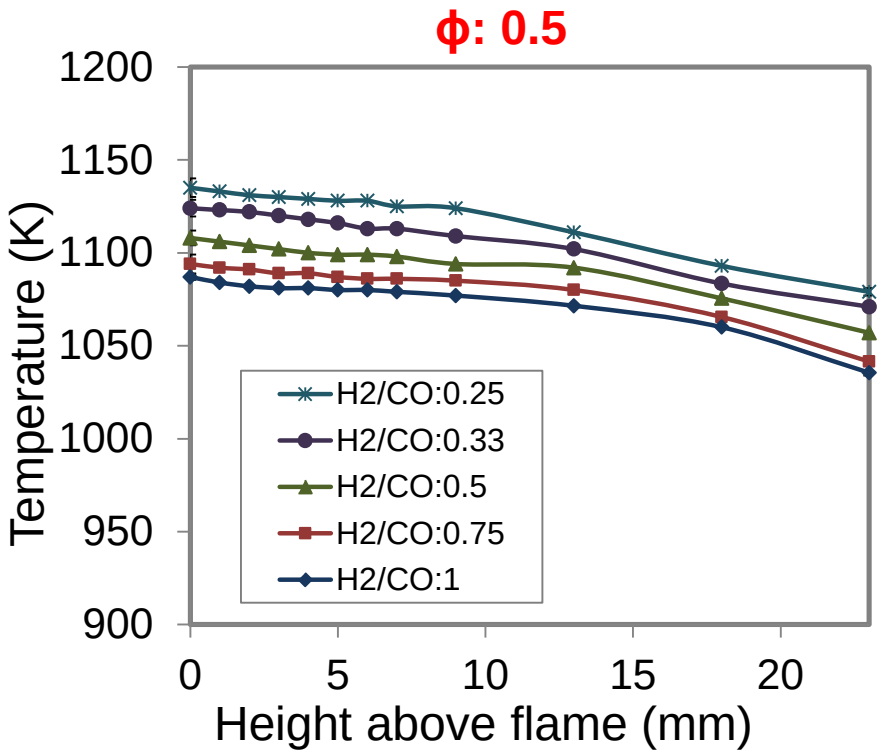
*Kastelis et al. WWAI Conference 2008.

Radial and Axial Temperature Profiles

Radial temperature profile

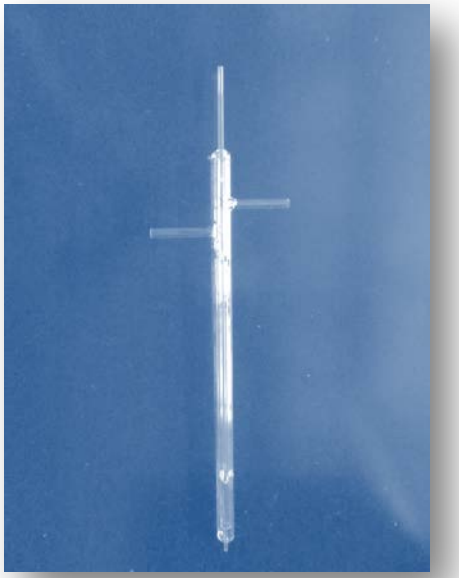


Axial temperature profile

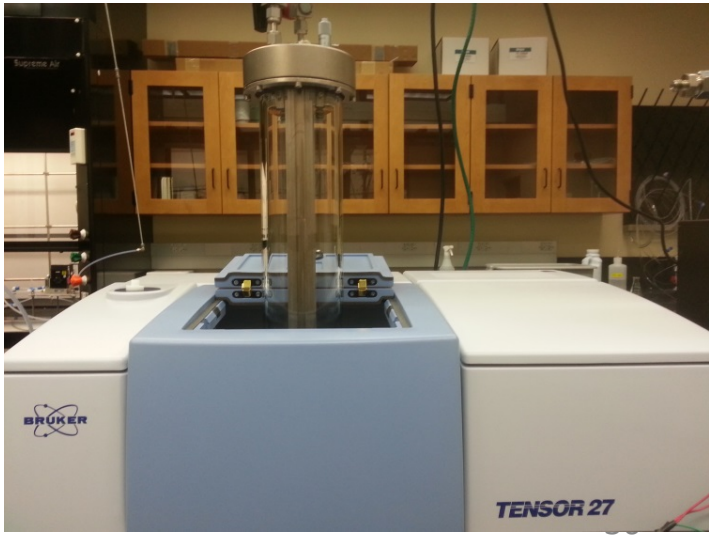


Sampling

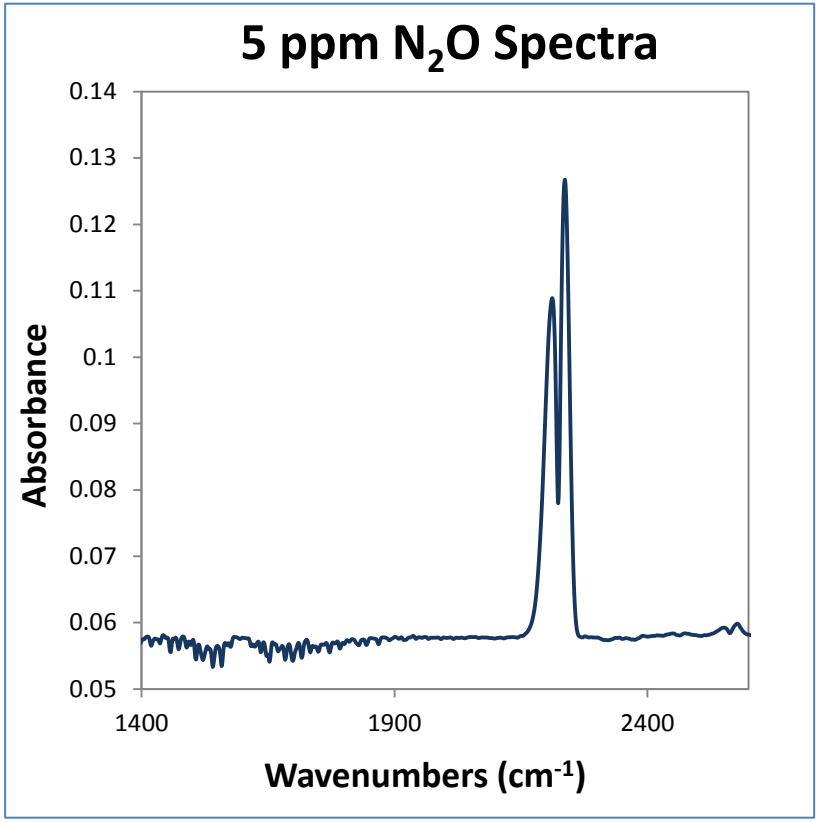
- Species profiles vs. single point measurements
  - Movable probe with a translation stage
  - Water cooled probe made of quartz

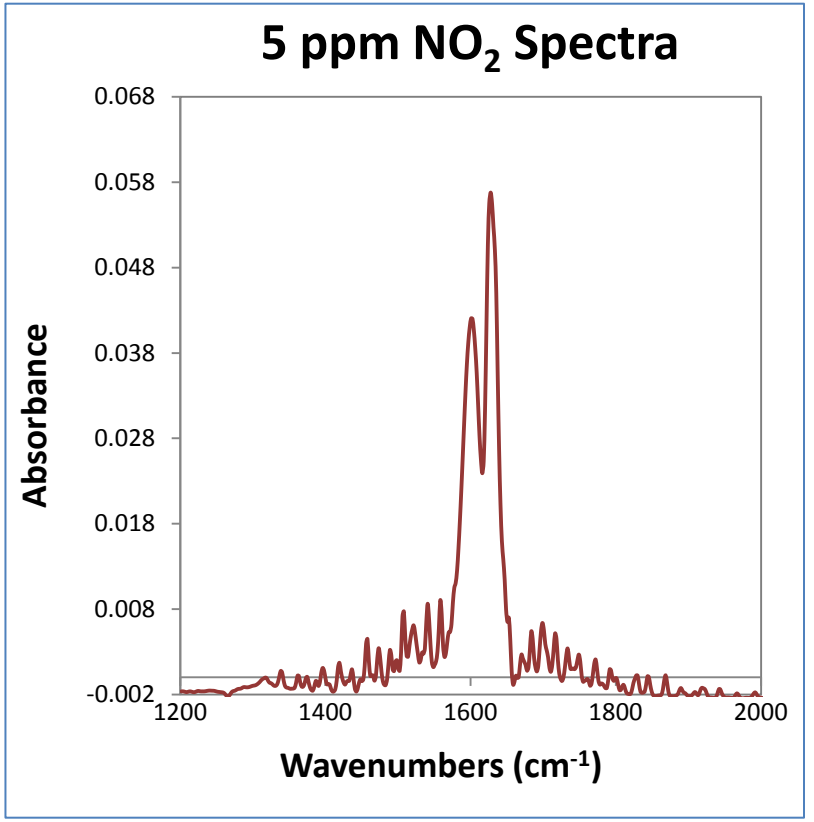


- Speciation of nitrogen oxides (NO, N_2O, NO_2) vs. total NO_x data
  - Employ FTIR for measurements
- Bruker Tensor 27 IR bench
  - coupled with MCT detector for increased sensitivity
- Variable path length cell with maximum optical length of 8m



NO_x Measurements





Summary

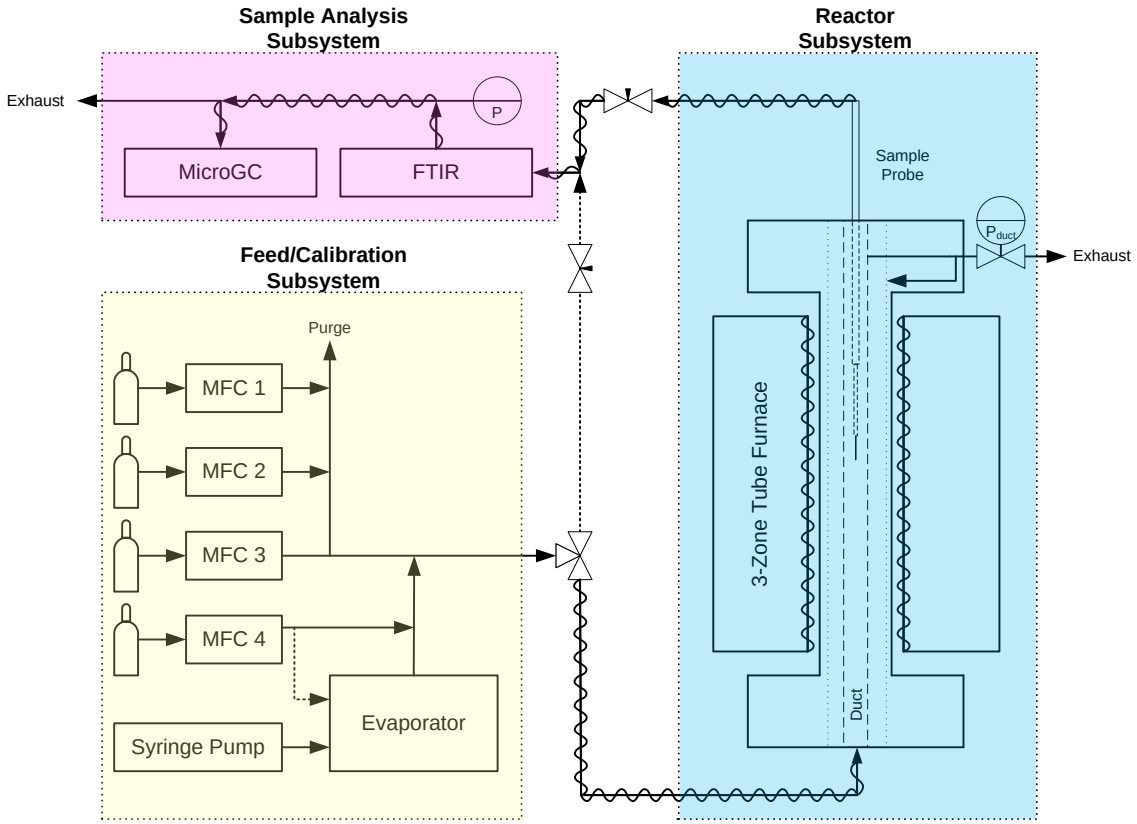
- Detailed NOx speciation data is needed to validate/develop kinetic models
- Experiments
  - Burner has been characterized
  - Temperature profiles were obtained for the model
  - Calibration of FTIR for NO_x species was done

- Future work
  - NO_x measurements from the burner
  - Construction of high-P combustion chamber
  - NO_x measurements at high P
  - Effects of diluents
  - NO_x, CO measurements in shear/mixing layer

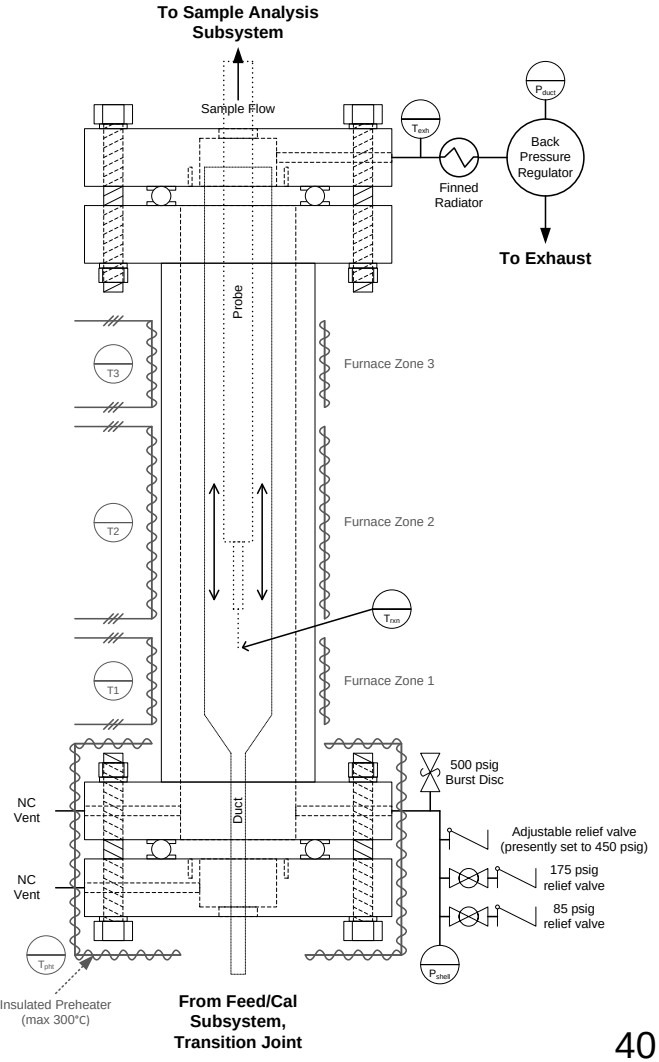
Measurement of Small Species Data

HPLFR Schematics

Facility Overview



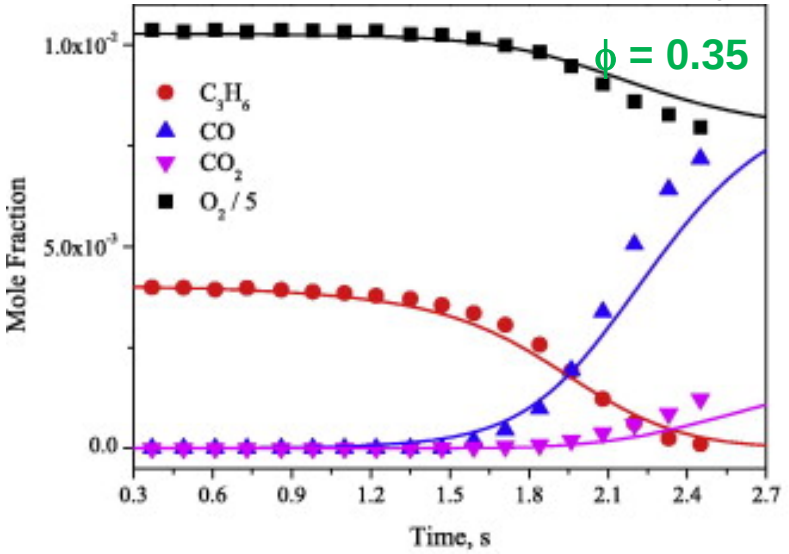
Reactor Subsystem Detail



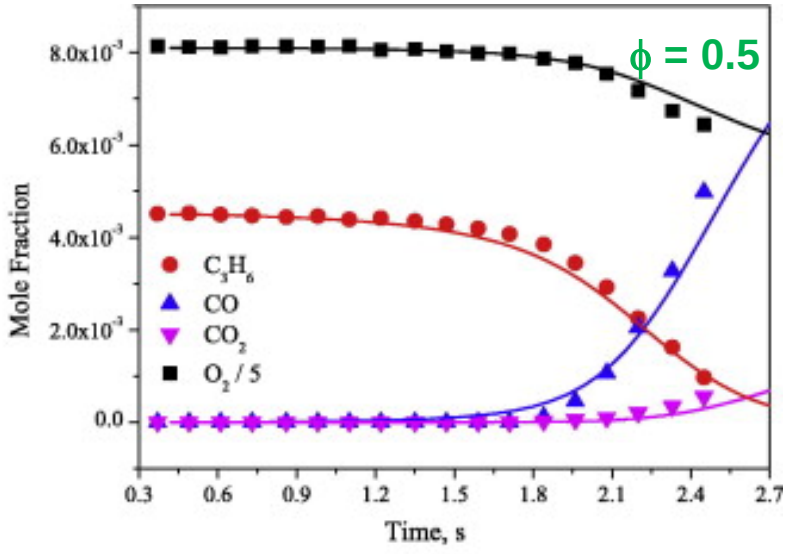
Propene (C_3H_6) Oxidation in HPLFR

P = 15 atm → Highest pressure for species-resolved validation data appearing in the literature

T = 800 K → Enhances sensitivity to reactions responsible for fuel destruction



(a) 0.40% C_3H_6, 5.14% O_2 in N_2, φ = 0.35



(b) 0.45% C_3H_6, 4.05% O_2 in N_2, φ = 0.5

Symbols – HPLFR measurements; Lines – 0-D Kinetic model predictions of recent comprehensive NUI Galway Model[1]

- Resonantly-stabilized allyl radical (aC_3H_5) suppresses reactivity relative to radicals formed from other small olefins and saturated hydrocarbons
  - This suppression is a particular feature of propene otherwise not represented by most natural gas/LPG fuel components
- Many older models did not include treatment of chemistry subsequent to allyl self reaction (aC_3H_5+ aC_3H_5→ products)
  - This led to especially poor predictions of ignition
- Depending on composition, the sensitivity to aC_3H_5 chemistry may be masked by higher reactivity of blend components (e.g., H_2 or C_3H_8)

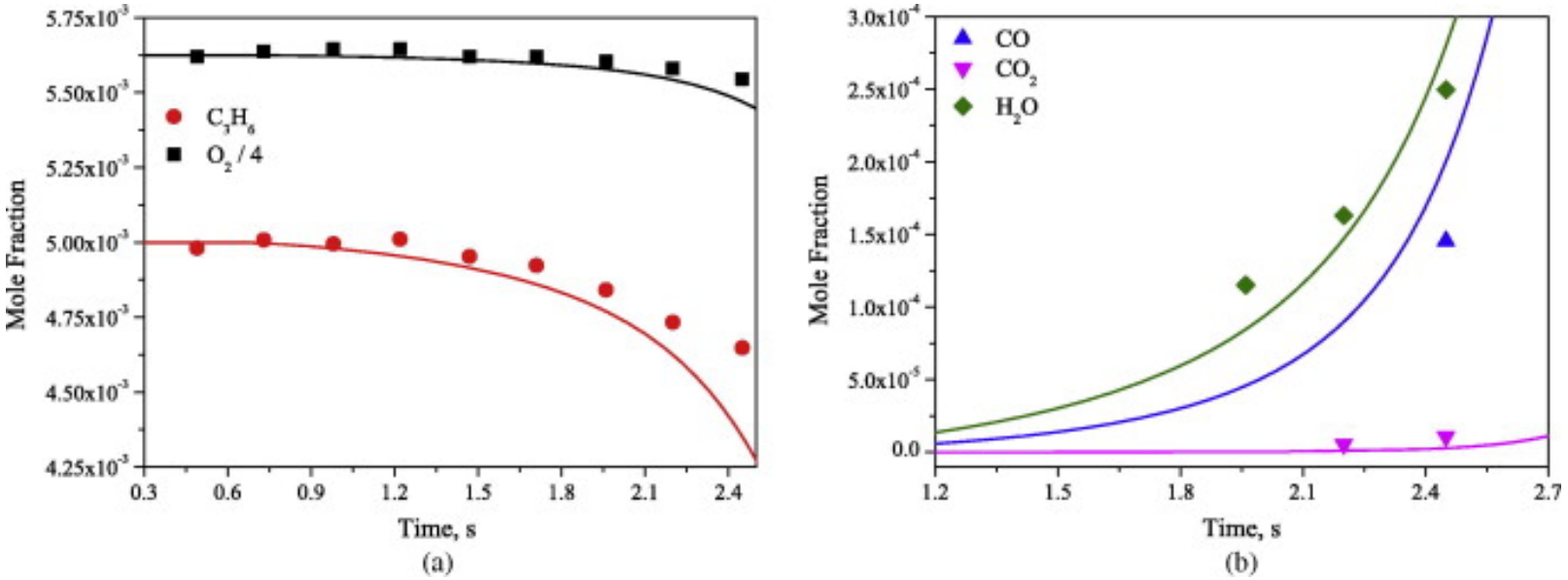
1. Burke et al. *Combust Flame.* 161:2765 (2014)

Propene (C_3H_6) Oxidation in HPLFR[1]

P = 15 atm → Highest pressure for species-resolved validation data appearing in the literature

T = 800 K → Enhances sensitivity to reactions responsible for fuel destruction

ϕ = 1.0



Symbols – HPLFR measurements; Lines – 0-D Kinetic model predictions of recent NUI Galway Model[1]

- Good agreement between measurements and model predictions

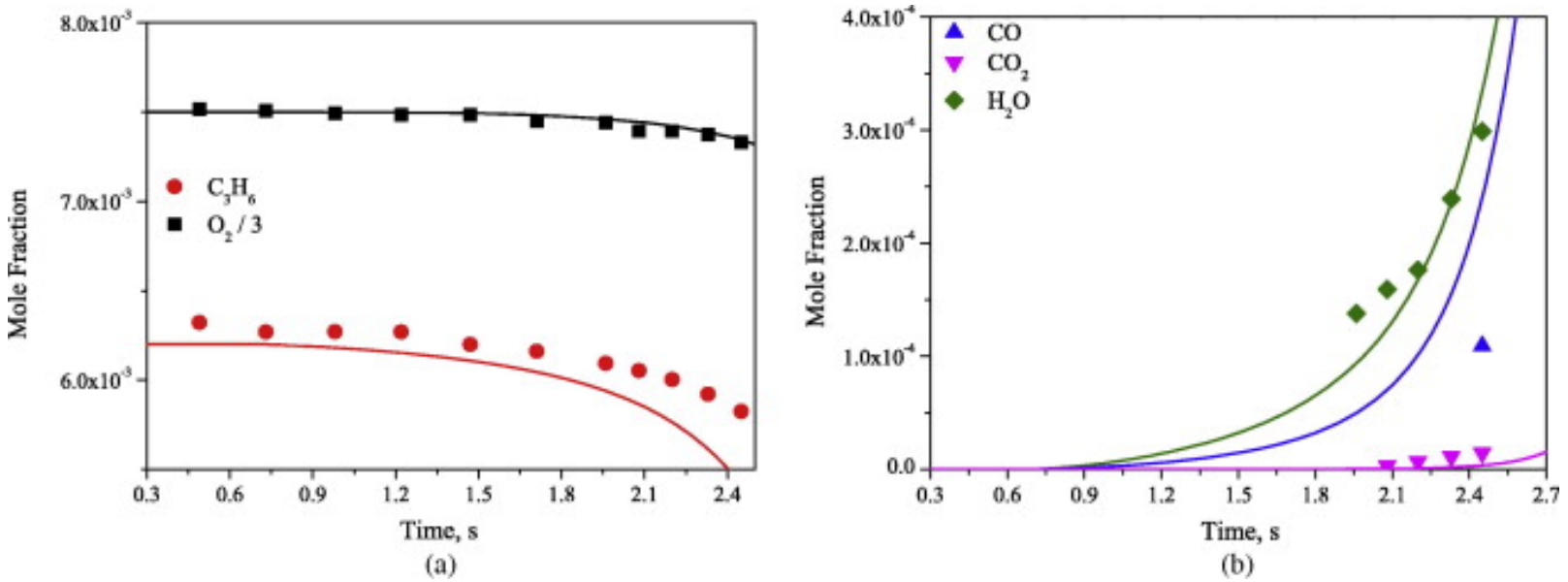
1. Burke et al. *Combust Flame.* 161:2765 (2014)

Propene (C_3H_6) Oxidation in HPLFR[1]

P = 15 atm → Highest pressure for species-resolved validation data appearing in the literature

T = 800 K → Enhances sensitivity to reactions responsible for fuel destruction

ϕ = 1.25



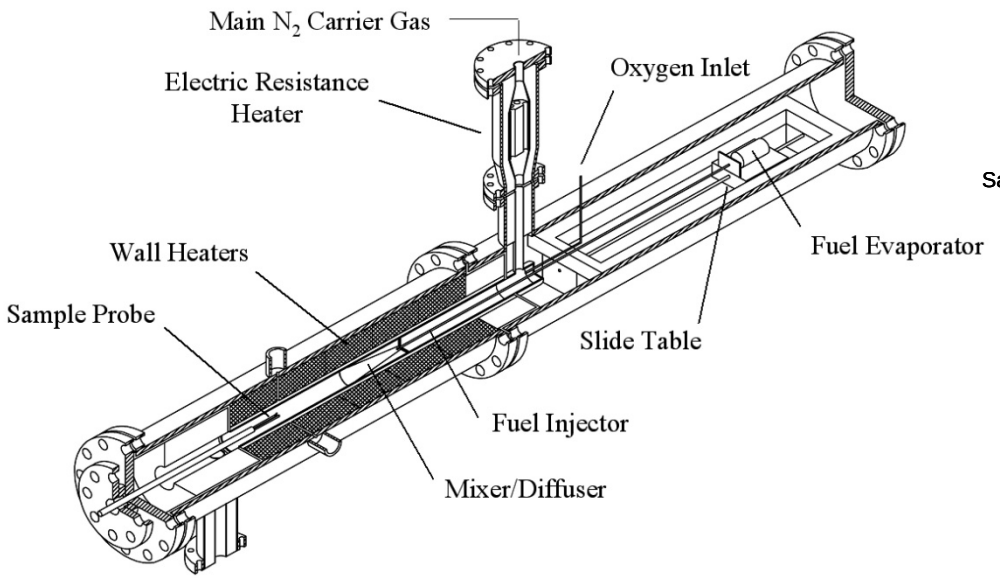
Symbols – HPLFR measurements; Lines – 0-D Kinetic model predictions of recent NUI Galway Model[1]

- Good agreement between measurements and model predictions

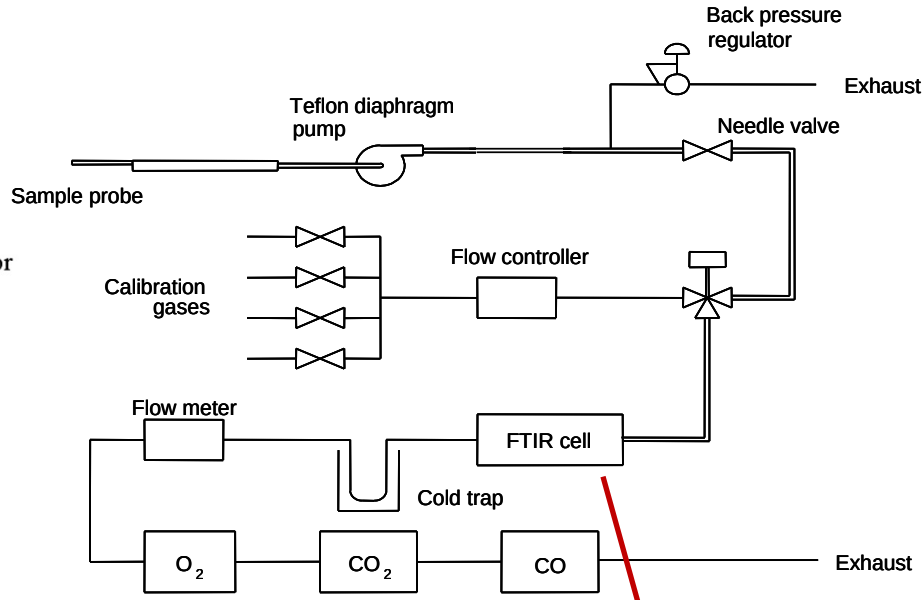
1. Burke et al. *Combust Flame.* 161:2765 (2014)

VPFR Schematics

Reactor Detail



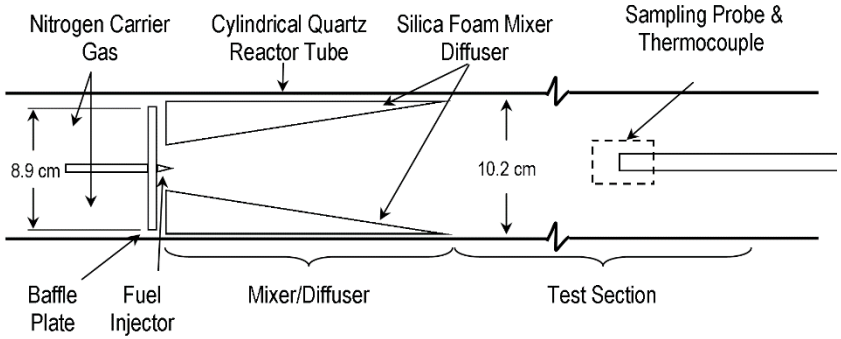
Sampling Subsystem



Reactor Duct Material: Fused Silica
Pressure: 0.2 - 20 atm
Temperature: 300 - 1200 K

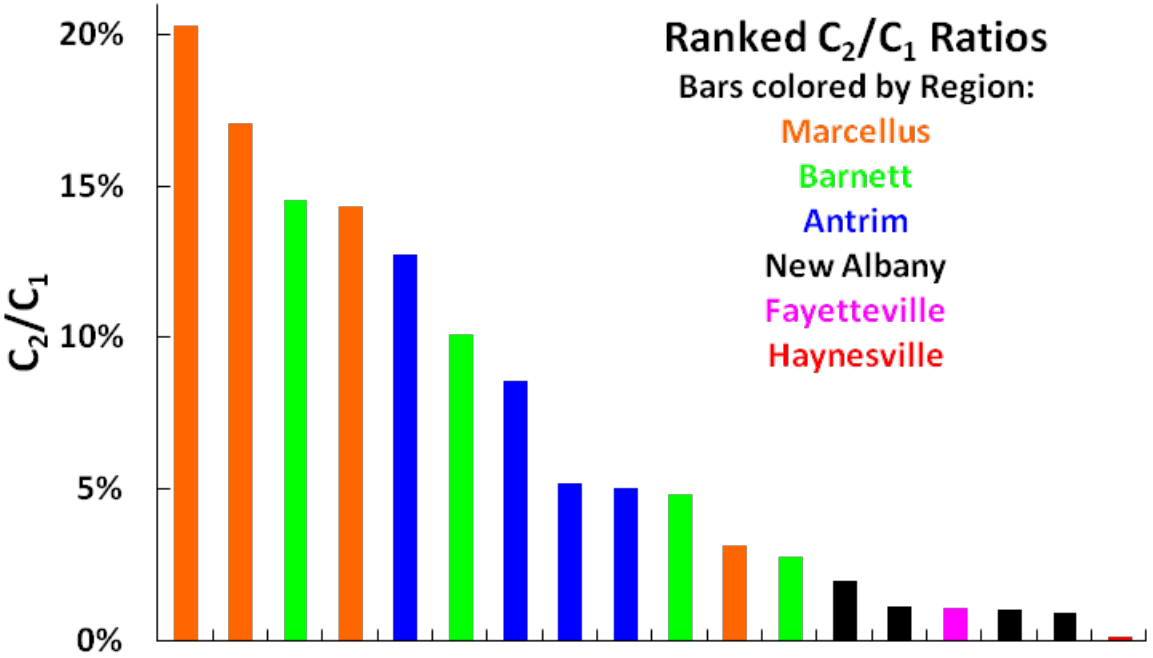
Reactor Section Dia.: 10 cm
Mass Flow rate: 10 - 30 g/s
Residence Time: 0.0015 - 5 s

Mixer/Diffuser Detail



CH_4, C_2H_6, C_2H_4, CH_2O, H_2O

CH_4 / C_2H_6 Blend Oxidation in VPFR



- Wetter natural gas to be expected from shale gas "revolution"
- Present figure is illustrative – does not consider gas separation/conditioning
  - e.g., associated with Mariner West/East pipelines from Marcellus Shale
- As will be shown, C_2H_4 is a key intermediate of C_2H_6 oxidation, so C_2H_6 fairly represents the C_2 fraction

1. Figure adapted from Bullin & Krouskop. *Oil & Gas J.* 107:50 (2009)

CH_4 / C_2H_6 Blend Oxidation in VPFR

Run Number	P (atm)	$T_{initial}$ (K)	CH_4 Mole Fraction (ppm)	C_2H_6 Mole Fraction (ppm)	O_2 Mole Fraction (ppm)	φ (approx.)
1	10	1052	4972	0	10105	1.0
2	10	1050	5170	52	10118	1.0
3	10	1048	4938	101	10507	1.0
4	10	1050	5089	212	9940	1.0
5	10	1029	5800	0	23600	0.5
6	10	1029	5550	180	11750	1.0
7	10	1026	5500	180	23800	0.5
8	18	970	5900	0	23200	0.5

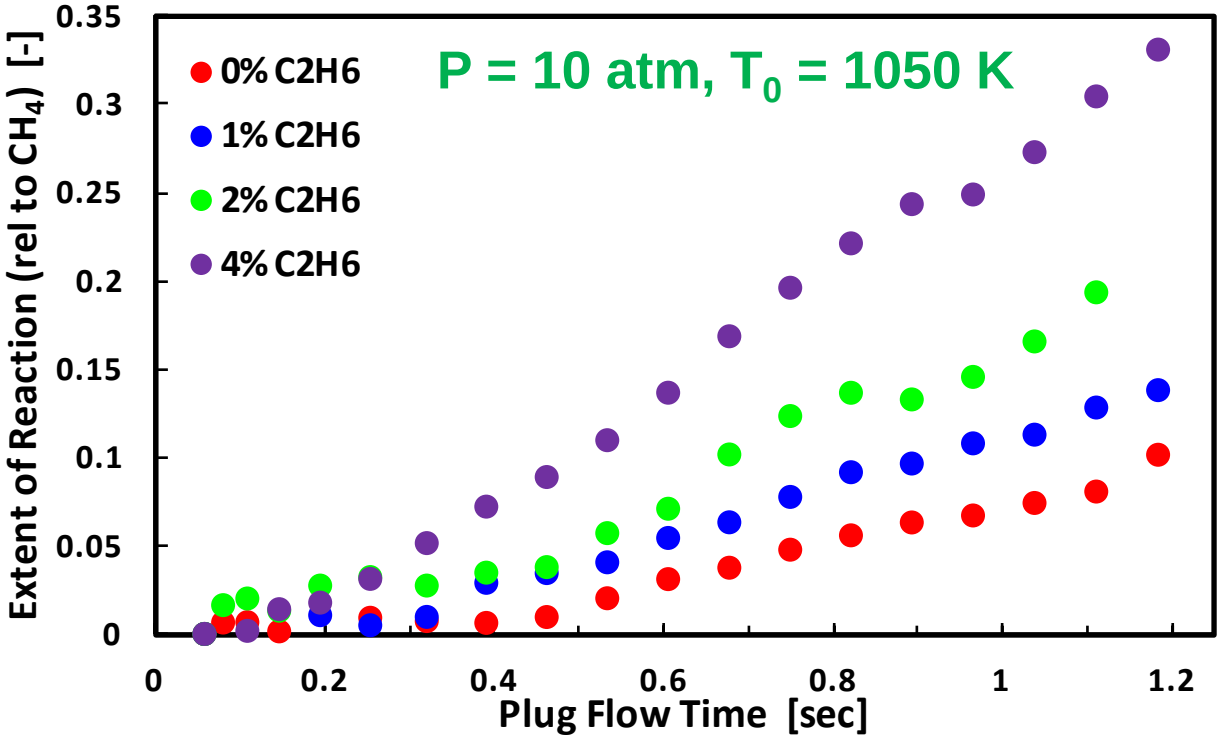
Present Focus

Design of Experiments

- Runs 1-4: compare methane reactivity as ethane fuel fraction increases from 0→4%
- Runs 5&7: compare lean reactivity for C_2H_6 fuel fractions of 0% and 3%
- Runs 6&7: compare reactivity of 3% C_2H_6 blends at phi of 0.5 and 1.0
- Run 8: higher pressure increases overall reactivity for CH_4, requires lower temperature for same reaction timescale

CH_4 / C_2H_6 Blend Oxidation in VPFR

Runs 1 – 4: Effect of increased C_2H_6 doping in CH_4

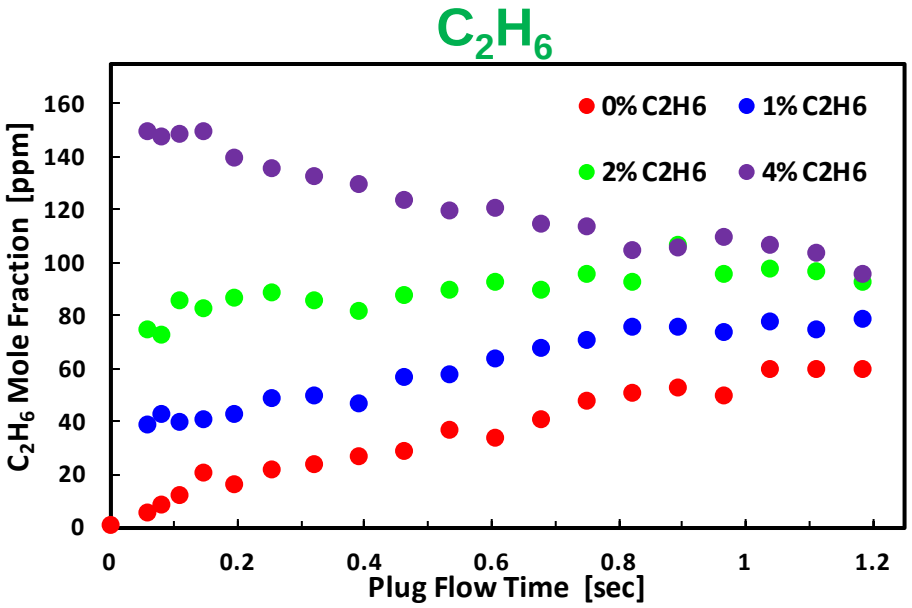


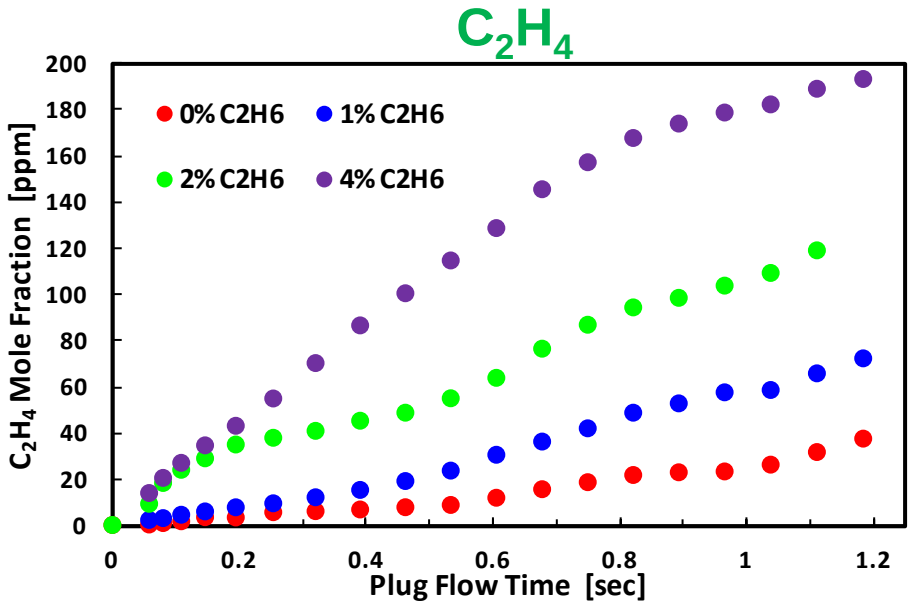
- Overall initial reactivity increases *fourfold* for just 4% C_2H_6 in CH_4
- Enhanced reactivity due to ethane doping should also have significant effect on ignition behavior
  - Significant "extra" reactivity will diminish at flame conditions

CH_4 / C_2H_6 Blend Oxidation in VPFR

Runs 1 – 4: Effect of increased C_2H_6 doping in CH_4

P = 10 atm, T_0 = 1050 K



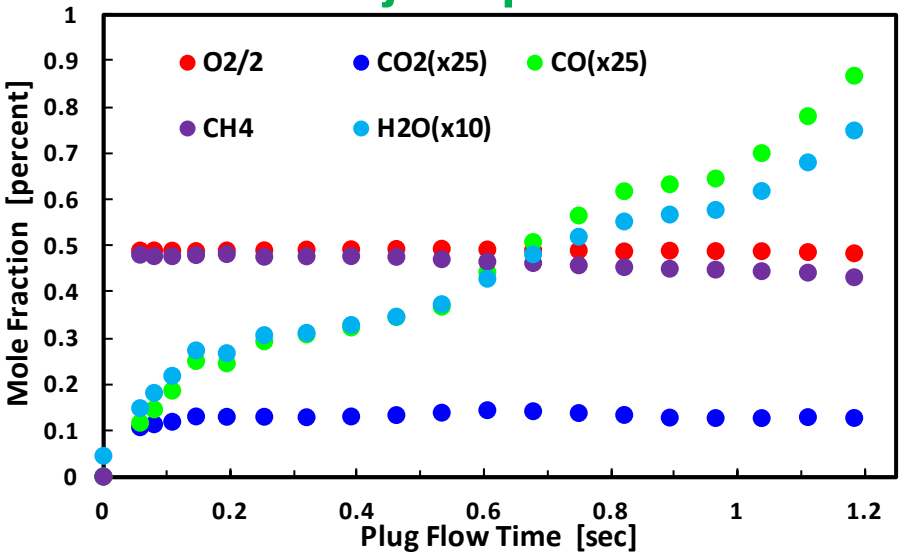


- Methane generates own pool of C_2H_6 via $CH_3+CH_3(+M)\rightarrow C_2H_6(+M)$
- Significant flux of $C_2H_6\rightarrow C_2H_4$ via $C_2H_5(+M)\rightarrow C_2H_4+\mathbf{H}(+M)$
- Enhanced initial reactivity due to ethane doping partly due to chemistry subsequent to production of **H** atom from C_2H_5

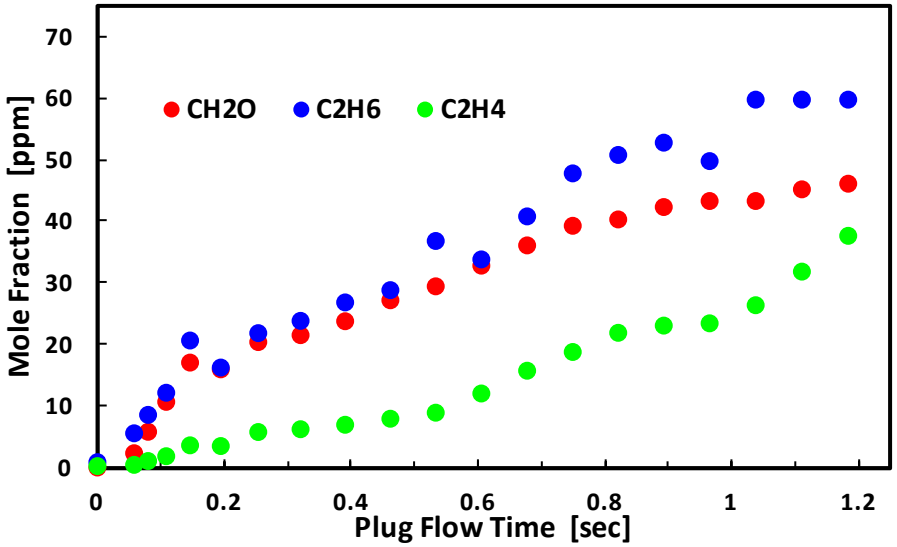
CH_4 / C_2H_6 Blend Oxidation in VPFR

Run 1: CH_4 Fuel, P = 10 atm, T_0 = 1050 K, φ ~ 1.0

Major Species



Minor Species



- The CO_2 generated is a facility effect (not explained by homogeneous gas phase kinetics) – for modeling purposes, can be treated by **reinitialization**
- Mixer/Diffuser-affected region extends to ~0.2 seconds – can also be treated by **re-initialization**
- Alternatively, a **time shift** will accurately predict major species profiles

Dryer et al. , Prog. En. Combust. Sci. (2014) 44:19-39

Overall Summary

- Potential critical pathways in H_2-NO_x has been identified.

.

- HONO, HNO_2, $HONO_2$ updates are proposed.

- The updates are found to have significant effect on the over all predictions of NO_x concentration in flow reactor reactivity over a broad range of pressures.

- The updates are also found to have a major influence on the CO oxidation which is critical to syngas combustion.

- Experiments - Burner has been characterized
  -Temperature profiles were obtained for the model
  - Calibration of FTIR for NO_x species was done

- HPLFR has been characterized and experiments are being conducted to obtain data on small hydrocarbon oxidation.

.

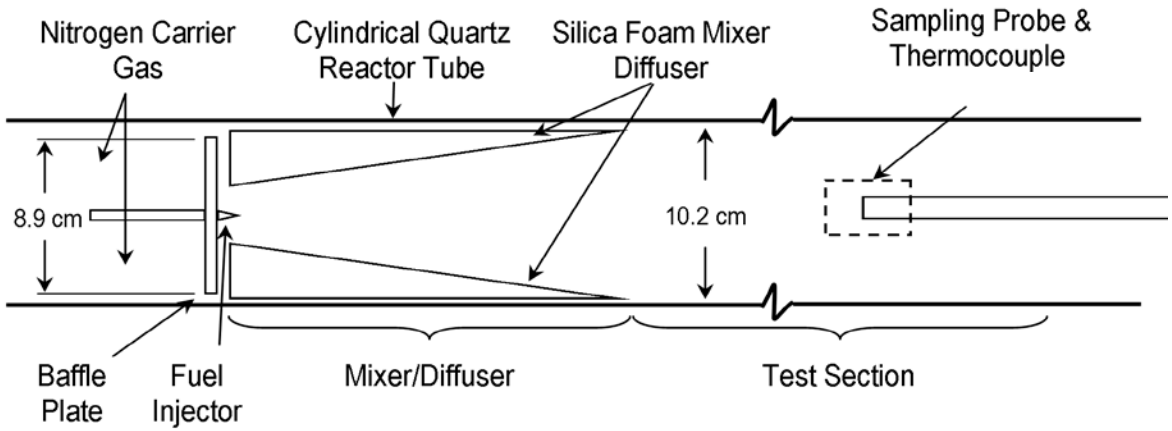
- Oxidation of methane, ethane blends at various ratios are conducted to identify the influence of trace hydrocarbon species on oxidation kinetics.

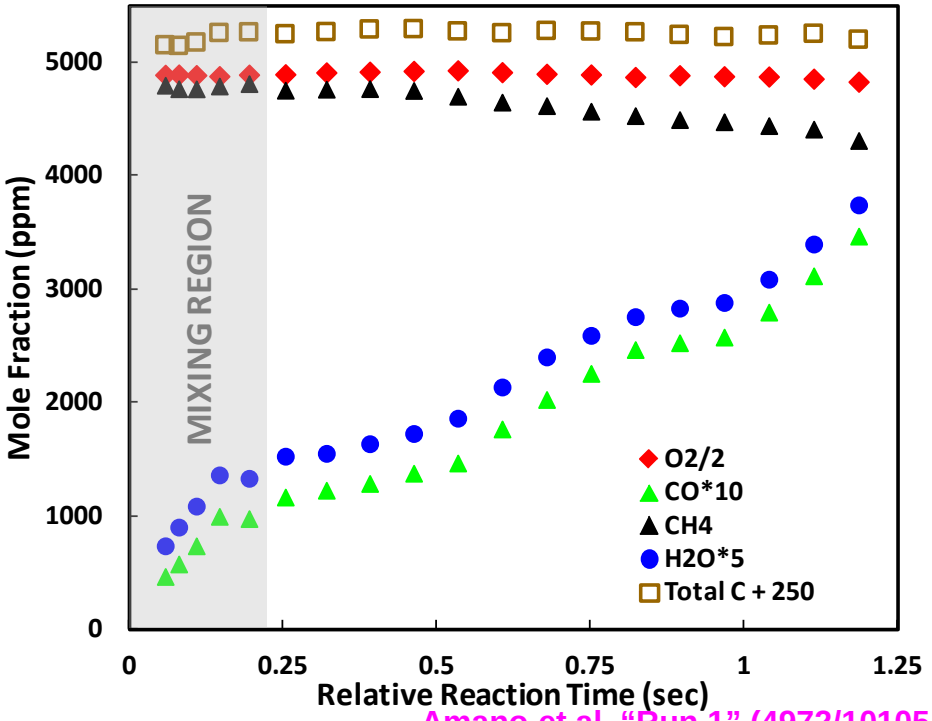
- Computational re-initialization for homogenous calculations to accurately simulate small species kinetics has been developed.

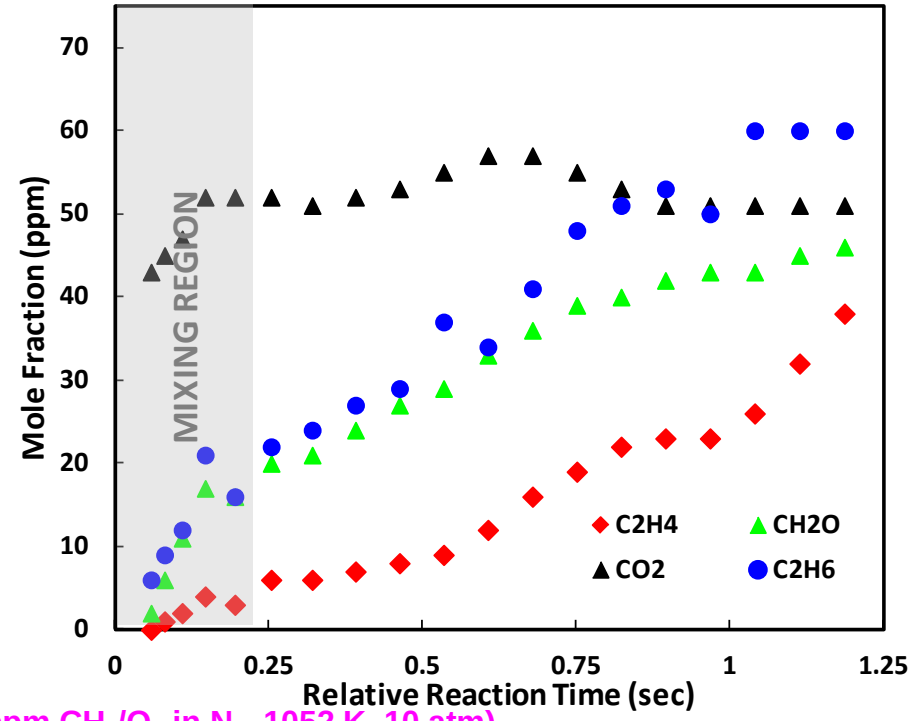
Thank You

Diluent CH_4 Oxidation at High P, Intermediate T

VPFR Schematic



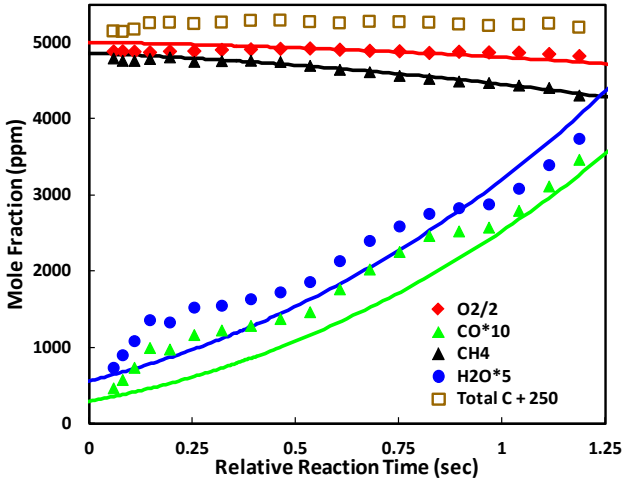


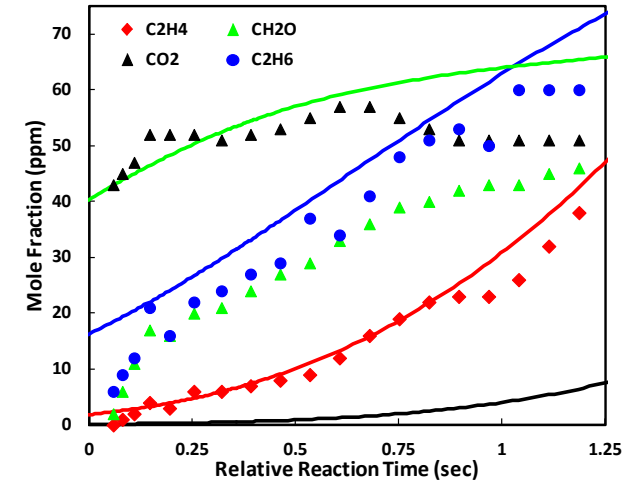


Amano et al. "Run 1" (4972/10105 ppm CH_4/O_2 in N_2, 1052 K, 10 atm)

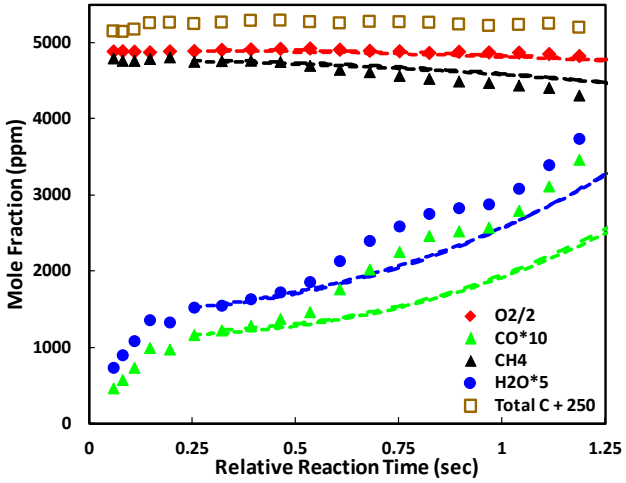
Model Comparisons to Flow Reactor Data – Initialization Approaches

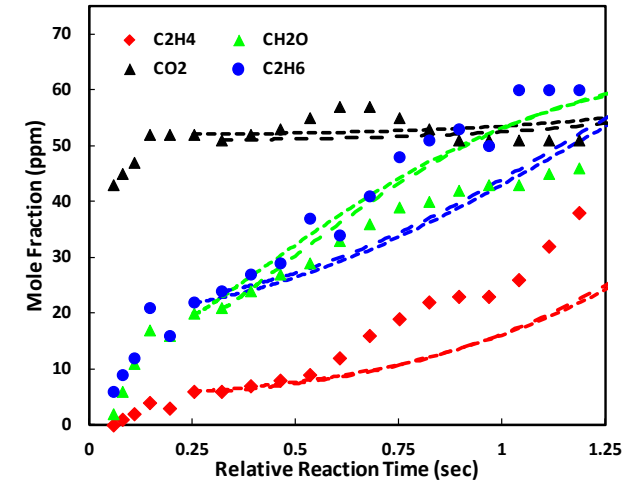
Coordinate-shifting
t_{shift} = 0.9 sec



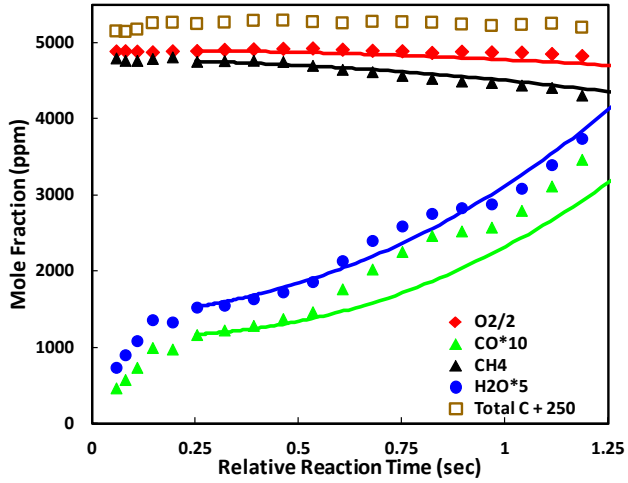


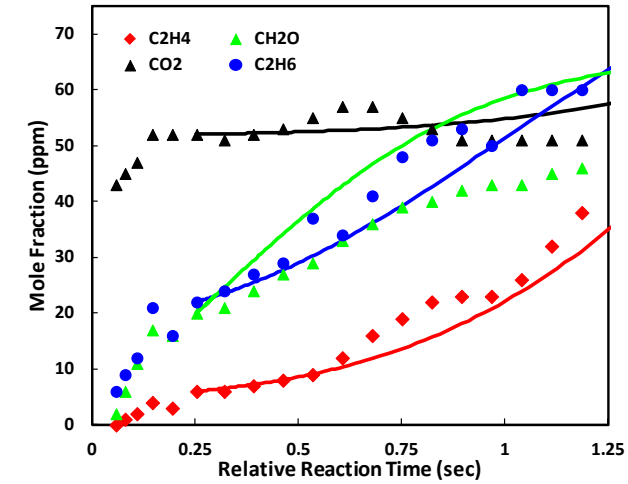
Simple Reinitialization
x_o = 35 & 40 cm





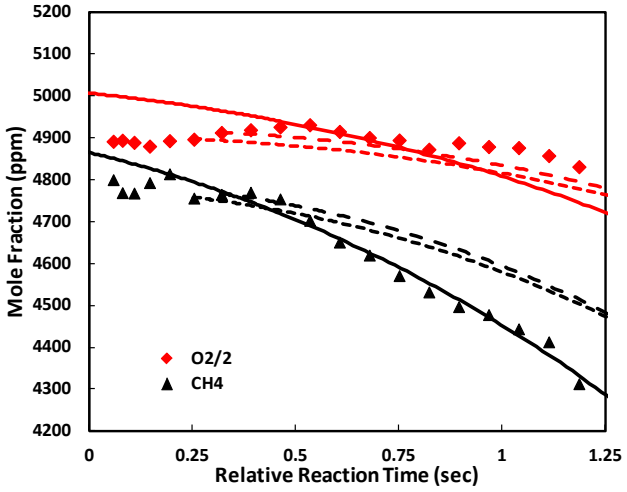
Computational Reinitialization
x_o = 35 cm

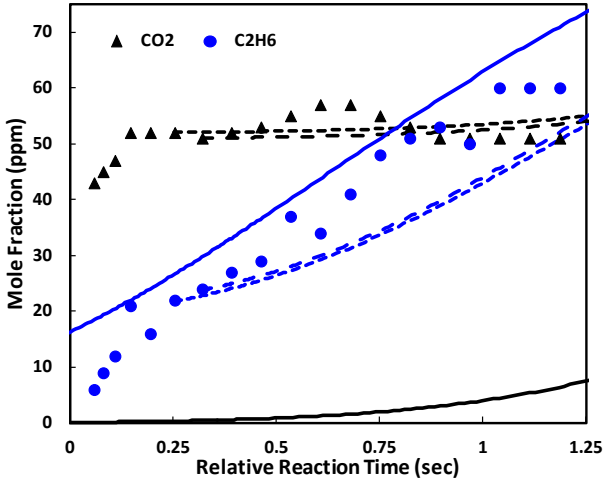


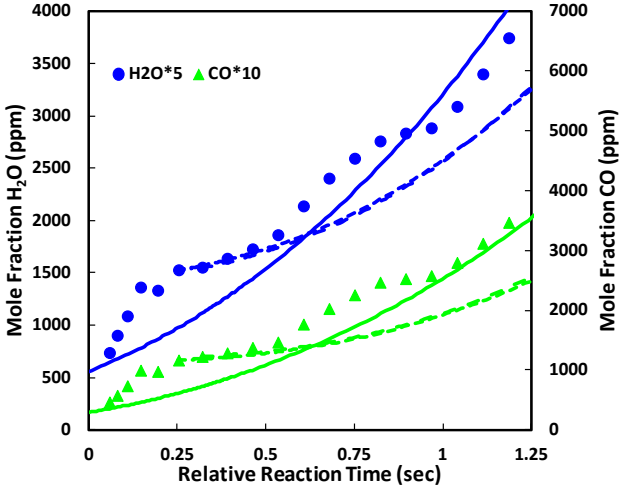


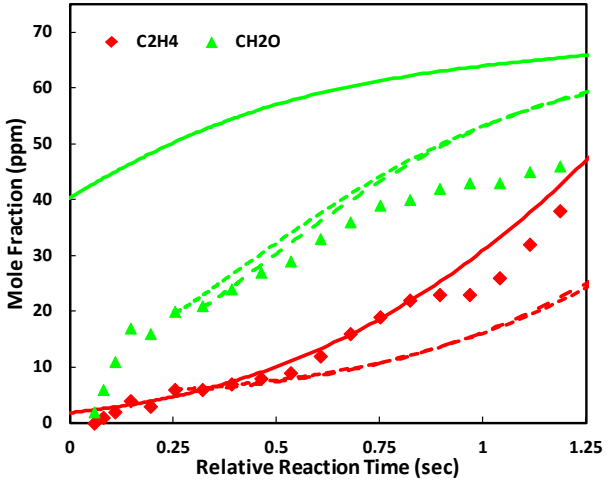
Kinetic Model – Aramco Mech 1.3; Data – Amano et al. "Run 1" (4972/10105 ppm CH_4/O_2 in N_2, 1052 K, 10 atm)

Comparison of Initialization Approaches









- **Coordinate (Time)-Shift** matches overall **reactivity gradient**; however, **cannot predict initial mole fraction deviations** – see CH_2O, CO_2 profiles

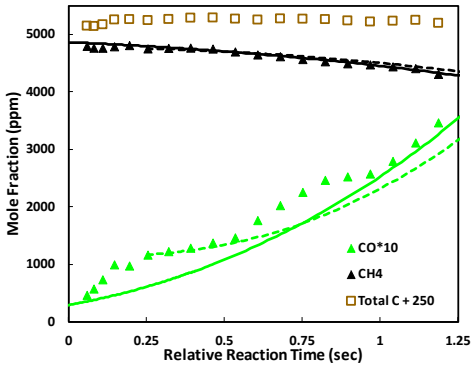
- **Simple Reinitialization** exactly matches "initial" mole fraction at designated point, but **gradient** is poorly predicted
- Approach does not account for unmeasured reactive intermediates **so overall reactivity is TOO LOW** – see CH_4, H_2O, CO, C_2H_6, C_2H_4

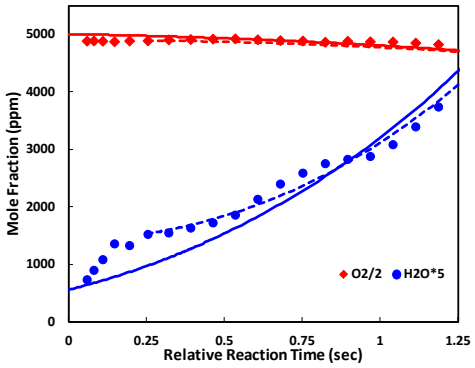
Final Analysis

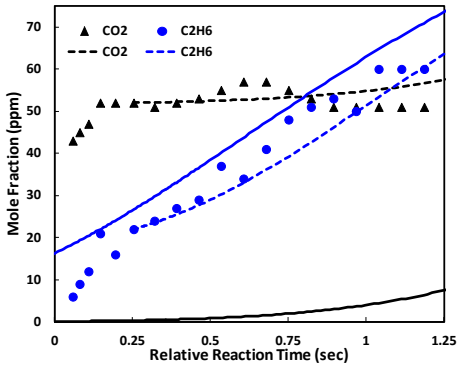
• **For long time constant species unaffected by initialization perturbations (here , coordinate (time)-shift** is adequate for matching profiles and assessing relative reactivity

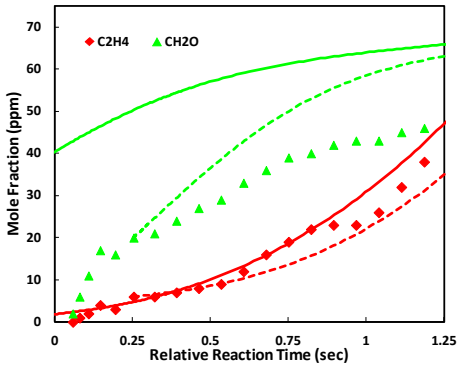
• **Simple Reinitialization** exactly matches “initial” mole fraction at designated point, but **gradient** is poorly predicted

• Approach does not account for unmeasured reactive intermediates **so overall reactivity is TOO LOW** – see CH_4, H_2O, CO, C_2H_6, C_2H_4





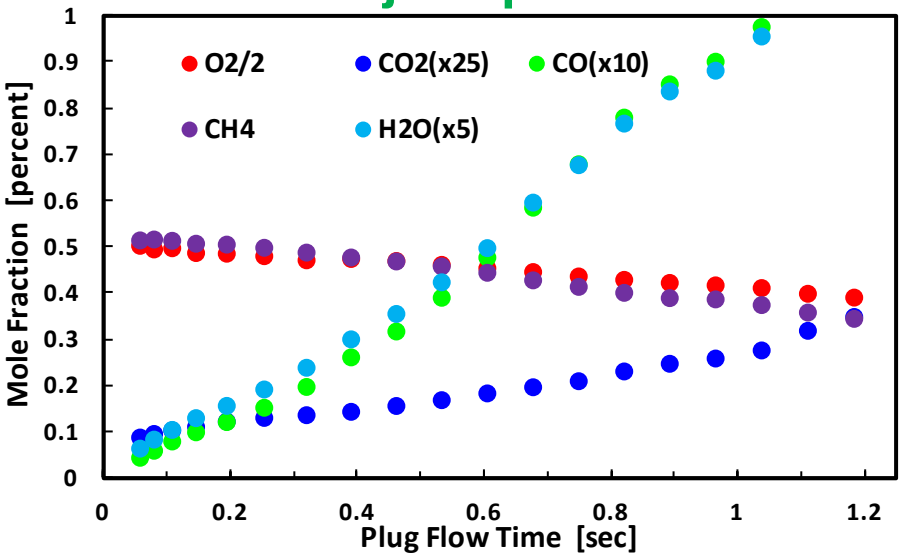




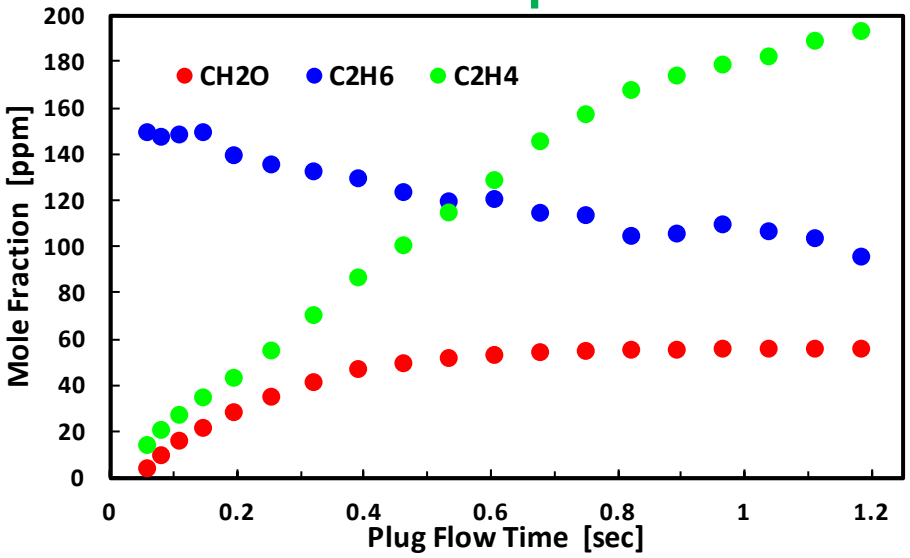
CH_4/C_2H_6 Blend Oxidation in VPFR

Run 4: CH_4 + 4% C_2H_6 Fuel, P = 10 atm, T_0 = 1050 K, φ ~ 1.0

Major Species



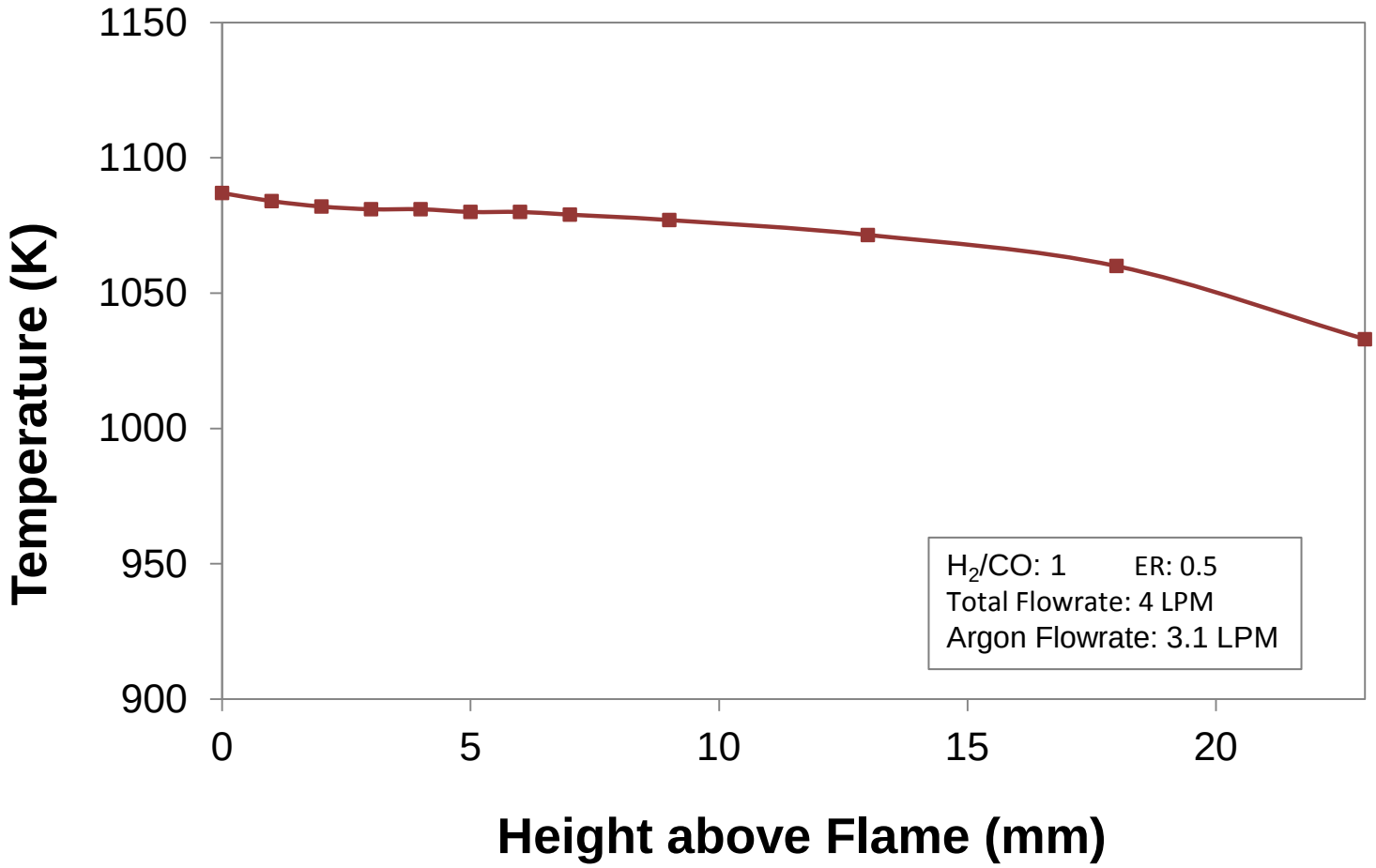
Minor Species



- Here, initial CO_2 generated is a facility effect; later CO_2 is formed by CO oxidation (primarily CO+OH →CO_2+H)
- For all of Runs 1-4, CO to H_2O ratio is between 2-2.5 over most of the reaction profile – provides insight into comparative rates of OH abstraction reactions of $CH_4/C_2H_6/C_2H_4/CH_2O$ and the relatively slow CO+OH bottleneck

Vertical Temperature Profile

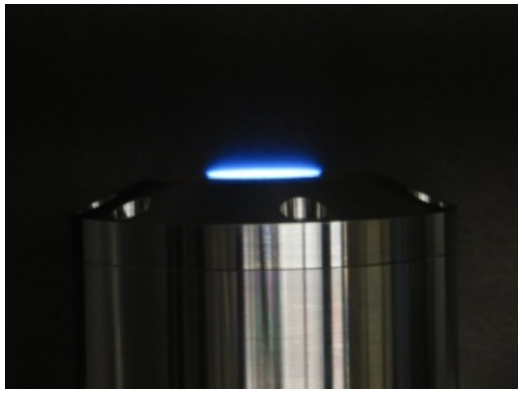
Vertical temperature profile in the burner center



Burner Tests

Changing the flow rate (4-6LPM) at constant φ:1 and H_2/CO:1

4 LPM



5 LPM

6 LPM

- Increasing flow rate ⇒ increased flame height