DNS of Pressure Effects and NO/N_2O Formation in Turbulent Ammonia/Hydrogen/Nitrogen-Air Flames

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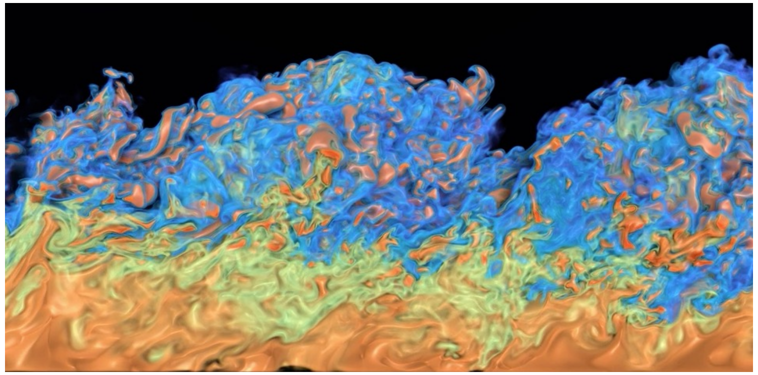
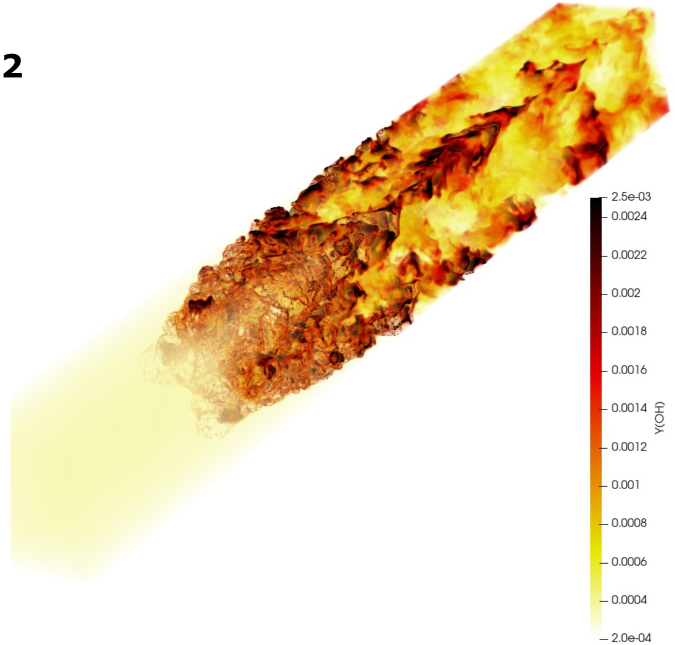
Ammonia Combustion Technology
Group Meeting
January 9, 2024


Temperature in a NH3/H2/N2-Air Premixed Flame in a Shear Layer at 10 atm



OH radical from second stage of a NH3-Air RQL jets-in-crossflow at 20 bar

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Exascale Computing Project: Pele DNS Combustion Suite

PI: Jackie Chen (Sandia), Co-PI: Marc Day (NREL)

Pele is ECP's application suite for high-fidelity detailed simulations of turbulent combustion in open and confined domains

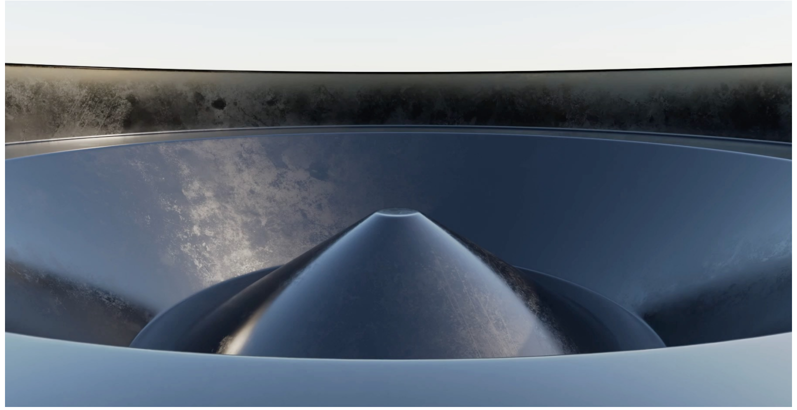
- Detailed physics and geometrical flexibility to evaluate design and operational characteristics of clean, efficient combustors for automotive, industrial, and aviation applications

❖ Code base:

- Compressible flows **PeleC**
- Low Mach number flows **PeleLMeX**
- Thermo-kinetic library **PelePhysics**
- Multiphysics library **PeleMP**
    - Spray, soot and radiation, and soon plasma

❖ Features:

- Adaptive Mesh Refinement (AMR)
- Support modern heterogeneous platform
- Embedded Boundaries for complex geometries
- Advanced algorithms (implicit advance, SDC)



Reactivity controlled compression Ignition IC engine – ndodecane + methane

Scramjet flame holding

❖ Highlights:

- Demonstrated weak scaling on DOE's heterogeneous platforms (Summit, Frontier)
- Simulation of RCCI using up to 60B cells on 50,000+ AMD GPUs on Frontier

Open-source code developed under the DOE ASCR Exascale Computing Project: https://github.com/AMReX-Combustion

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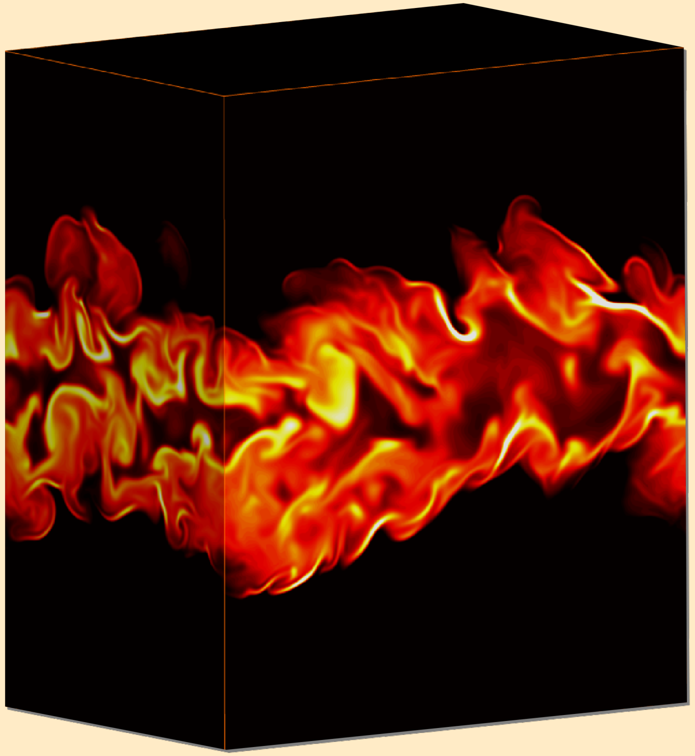
ECP EXASCALE COMPUTING PROJECT

Sandia S3D DNS code for compressible reacting flows

- Solves compressible reacting Navier-Stokes, total energy and species continuity equations
- High-order finite-difference methods
- Detailed reaction kinetics and molecular transport models
- Lagrangian particle tracking (tracers, spray, soot)
- Shock capturing
- Multi-temperature method (nonequilibrium)
- *In situ* analytics and visualization
- Geometry using immersed boundary method and multi-block approach
- Refactored for heterogeneous architectures using dynamic task based programming model (Legion/Regent)

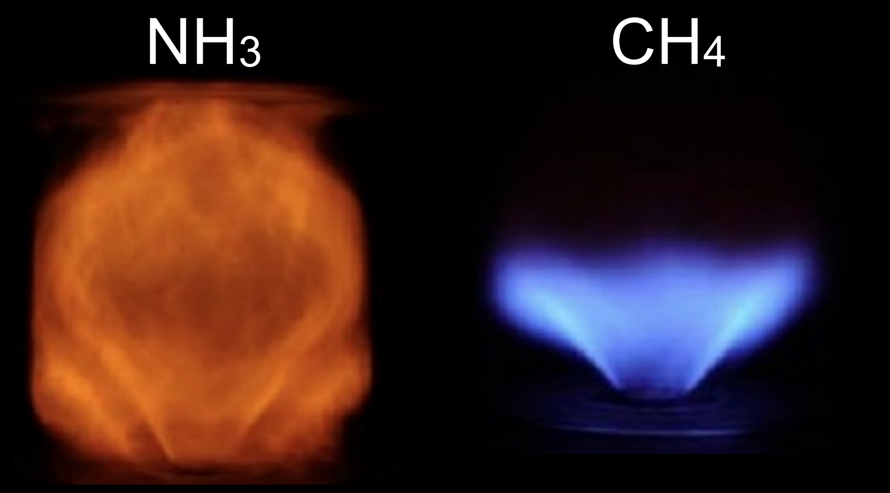


DNS provides unique fundamental insight into the chemistry-turbulence interaction

Chen *et al.*, *Comp. Sci. Disc.*, 2009

Ammonia/hydrogen has the potential to be an alternative **zero-carbon** fuel for hard-to-electrify sectors: long-haul shipping, power generation and agriculture

However, neat ammonia has a low reactivity compared to, e.g., natural gas (CH_4)



Valera-Medina et al., PECS, 2018.

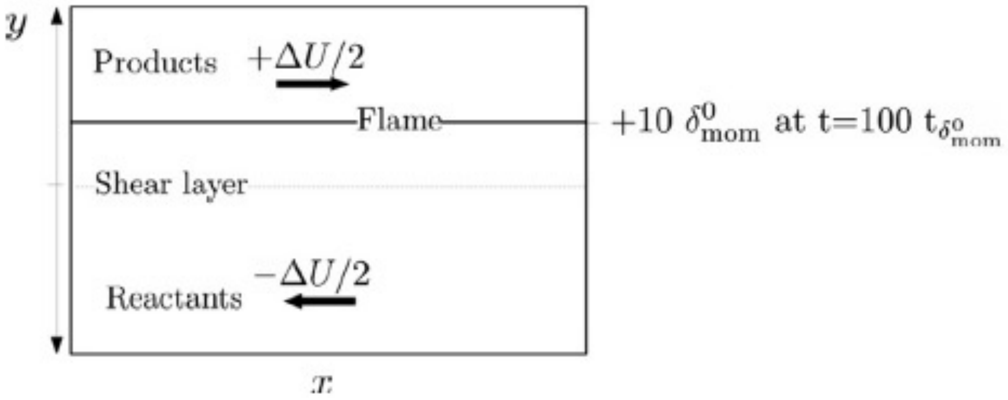
The reactivity can be improved by adding hydrogen or partial cracking of ammonia to hydrogen and nitrogen with laminar flame properties matched to natural gas

Turbulent combustion behavior of ammonia/hydrogen blends is different from laminar flames and poorly understood. DNS can elucidate combustion and emissions characteristics in lab-scale combustors, especially at high pressure

Pressure effects on ammonia/hydrogen premixed flames: DNS in a temporal planar shear layer

Simulation Parameters

- Lean premixed $NH_3/H_2/N_2$-air (40%/45%/15% vol) with equivalence ratio of 0.45 at **1 and 10 atm**
- Reactants are preheated to 750 K
- Flames are nominally in the broken reaction zone regime (Karlovitz # (Ka)>600, turbulent Re # (Re_t)>1000)
- Simulations are designed such that normalized parameters (Re_t, Ka, Damköhler #) are consistent between 1 and 10 atm
- Growing grid size with 11 B grid points on the final grid
- 19 species chemical mechanism (Jiang et al., 2020)
- DNS using S3D-Legion run on OLCF's 200 Pflop Summit supercomputer on ~1000 nodes using a DOE INCITE allocation



Rieth et al. Comb. Flame, 2022

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ASCR Leadership Computing Challenge

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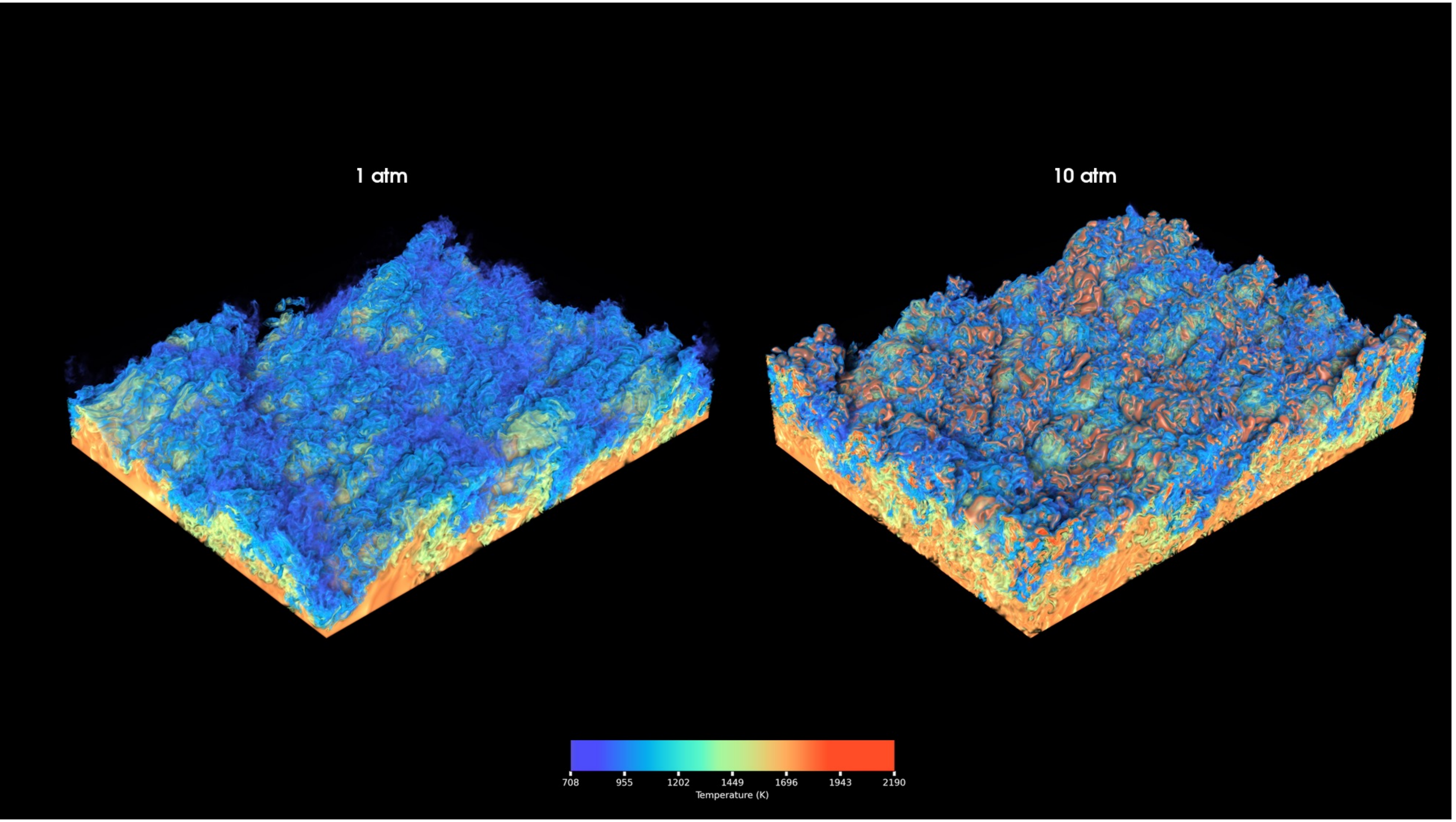
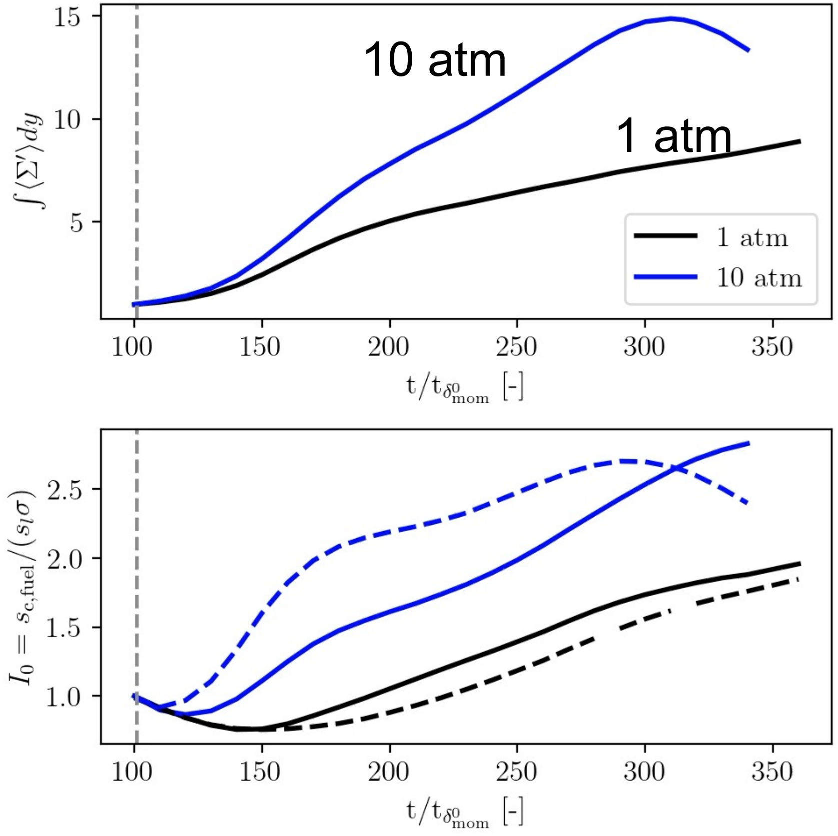
Pressure Effect on $NH_3/H_2/N_2$-air Premixed Flame Turbulent Burning Velocity

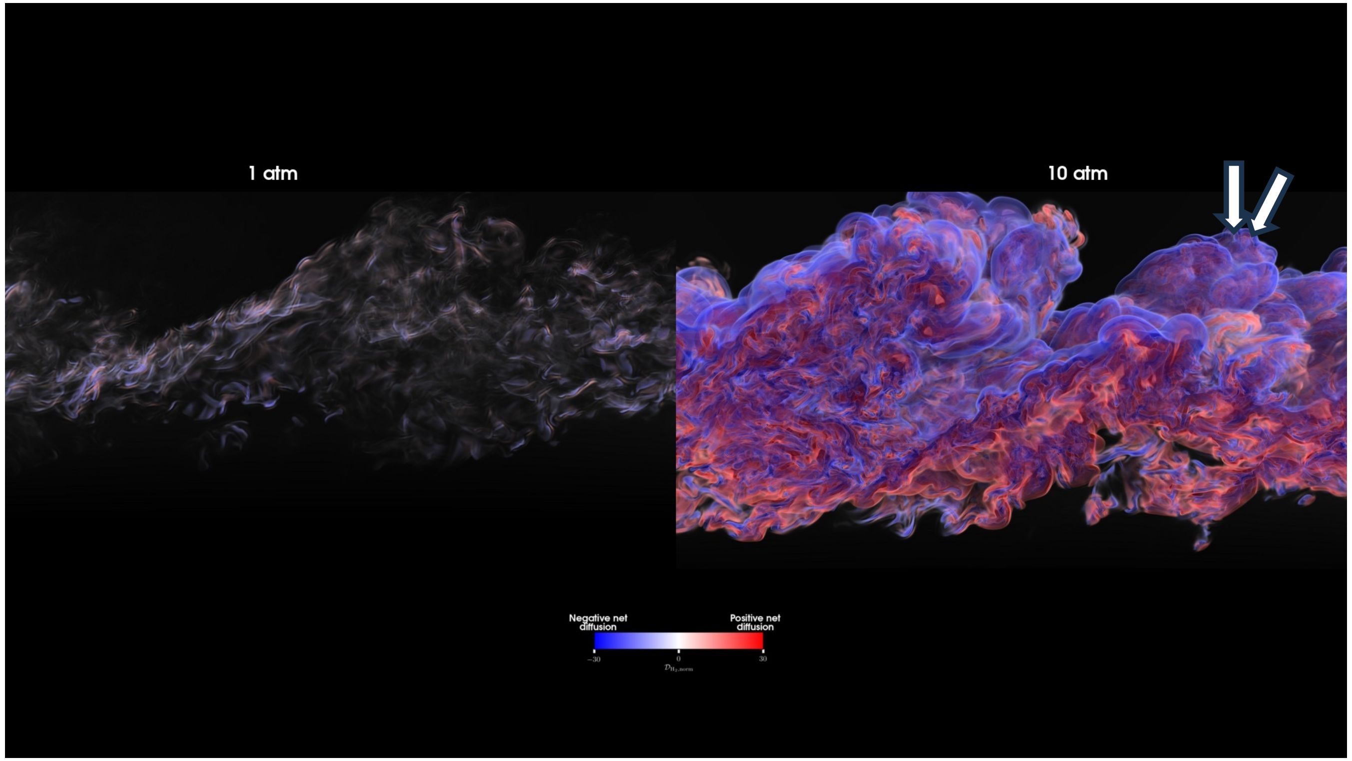


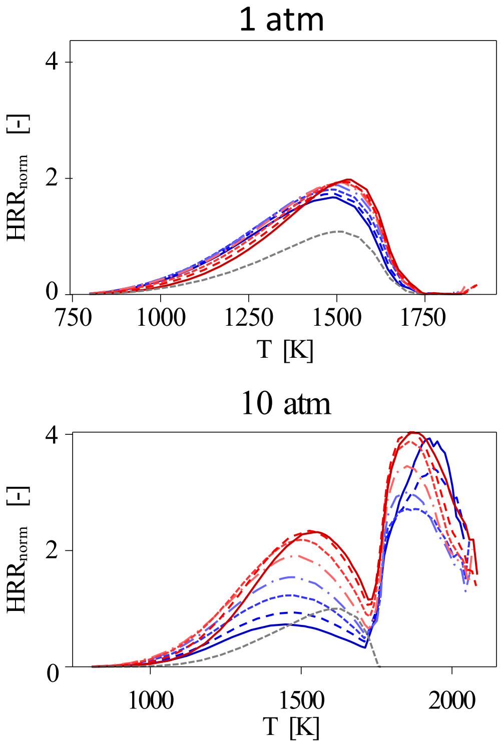


- Faster increase in flame surface density, fuel consumption rate, burning intensity (*I_o*)
- Weaker thinner flames at high pressure, greater preferential diffusion and thermodiffusive effect

Preferential Diffusion Amplified with Pressure (local enrichment at cusps)

Diffusion rates of H_2





convex concave towards reactants

- Preferential diffusion rates are amplified at elevated pressure
- Flame at 10 atm responds more strongly to preferential diffusion, i.e., is more equivalence ratio sensitive (corroborated by simplified 2D studies)

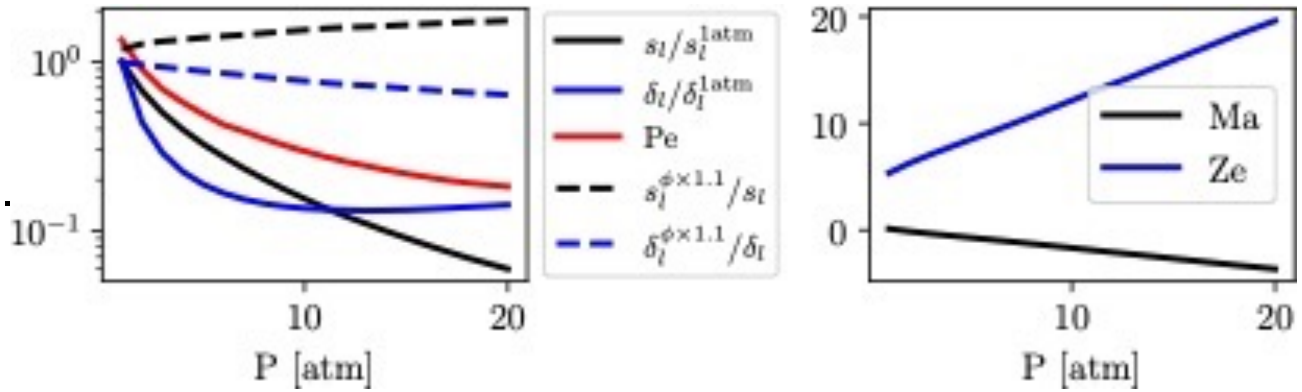
Rieth et al., Comb. Flame 2022

Pressure scaling in a freely propagating lean 1D H2/air flame

Observations with increasing pressure:

- 3-body recombination rate increase leads to decrease in flame speed and smaller decrease in flame thickness
- Ze increases due to 'weakening' of the flame as T_o approaches adiab. flame temp.

H_2/air 1D flame at equiv. ratio of 0.3, reactants at 750K



Zel'dovich number

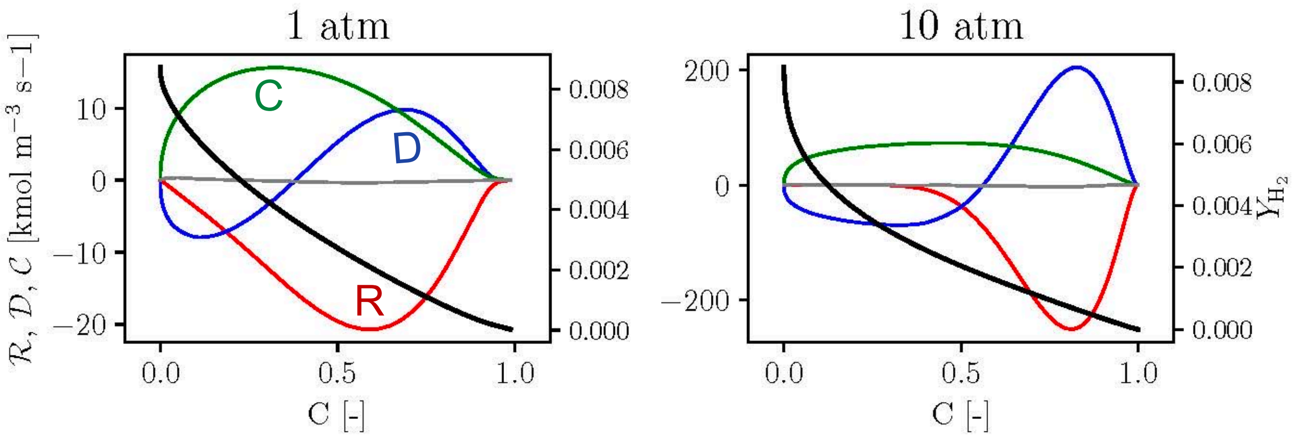
Ze = 4 $(T_b-T_u)/(T_b-T_o)$

where T_o is crossover temperature

Peclet Number

$$\mathrm{Pe}_{\mathrm{orig.}} = |\mathcal{C}_{H_2}|_{1D,\max}/|\mathcal{D}_{H_2}|_{1D,\max} = \frac{|\frac{\partial Y_{H_2}}{\partial x}u|_{1D,\max}}{|\frac{1}{\rho}\frac{\partial}{\partial x}(\rho\frac{W_{H_2}}{W_m}D_{H_2}\frac{\partial X_{H_2}}{\partial x})|_{1D,\max}}$$

Peclet Number

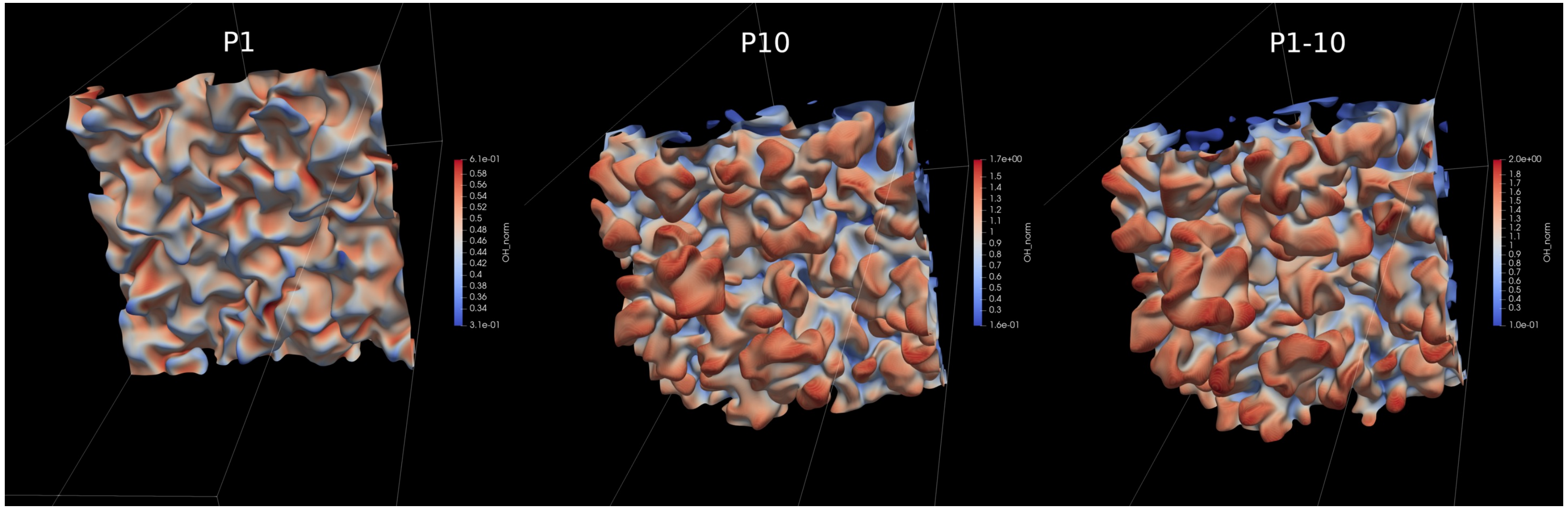


Rieth et al. Combustion and Flame 2023

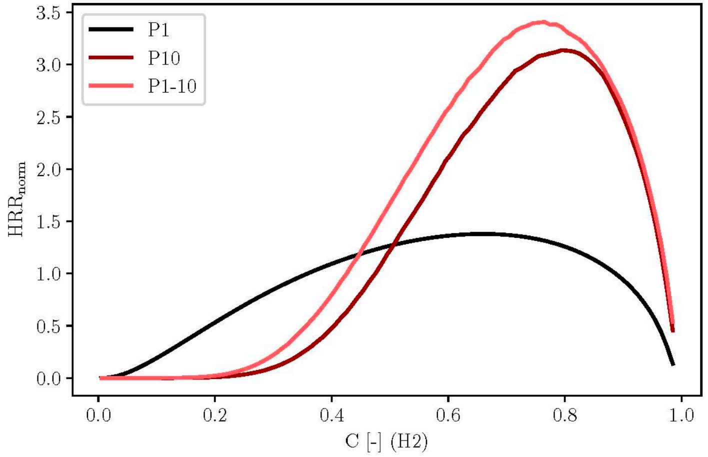
Turbulent burning rate scaling with a mixture’s reactivity and its propensity to become thermo-diffusively unstable

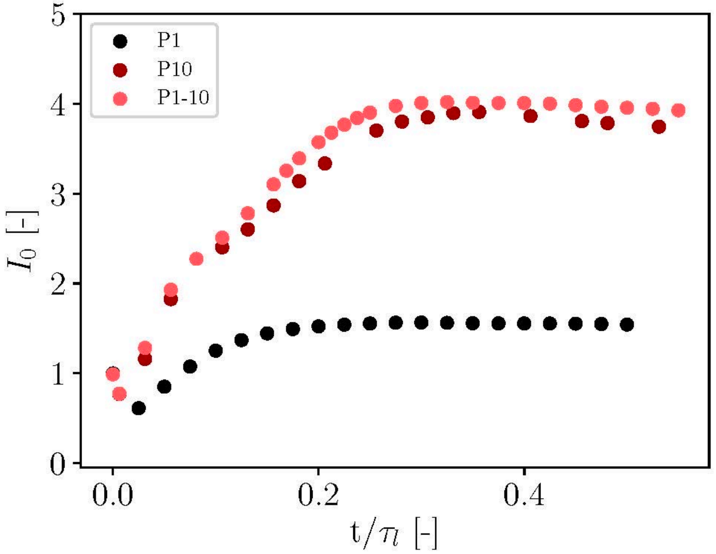
By tuning the reactants’ temperature and equivalence ratio (to mimic weakening of the flame with pressure) to match the thermo-diffusive and reactivity attributes, can high pressure flame structure be reproduced at atmospheric pressure?

Thermo-diffusive (Ma, Le_{eff}, Pe) and Reactivity (Ze)



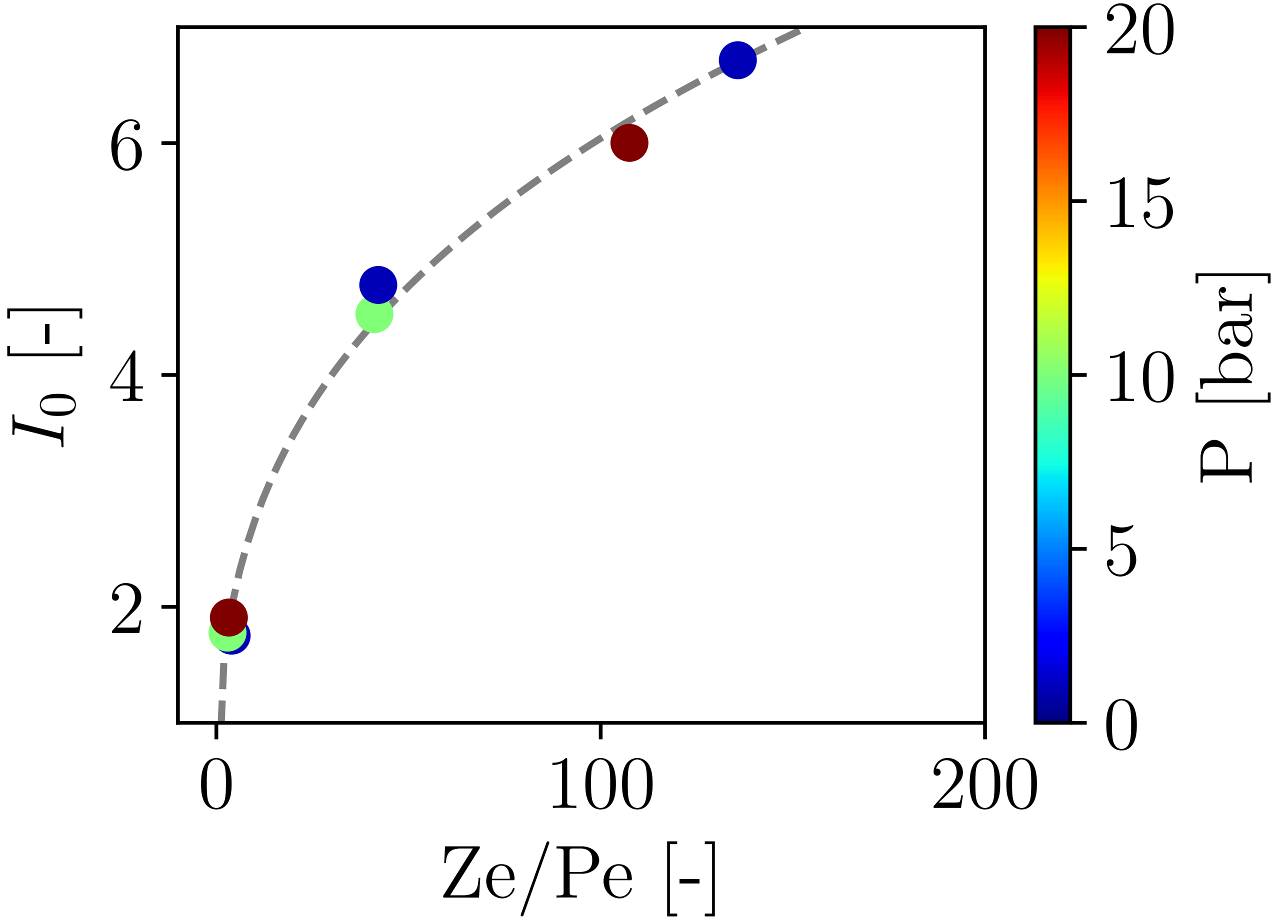
Ka = 300, Re_t = 1000, Da = 0.1 ‘broken reaction zones regime’





Rieth et al. Combustion and Flame 2023

Turbulent burning rate stretch factor scaling for turbulent hydrogen-air flames

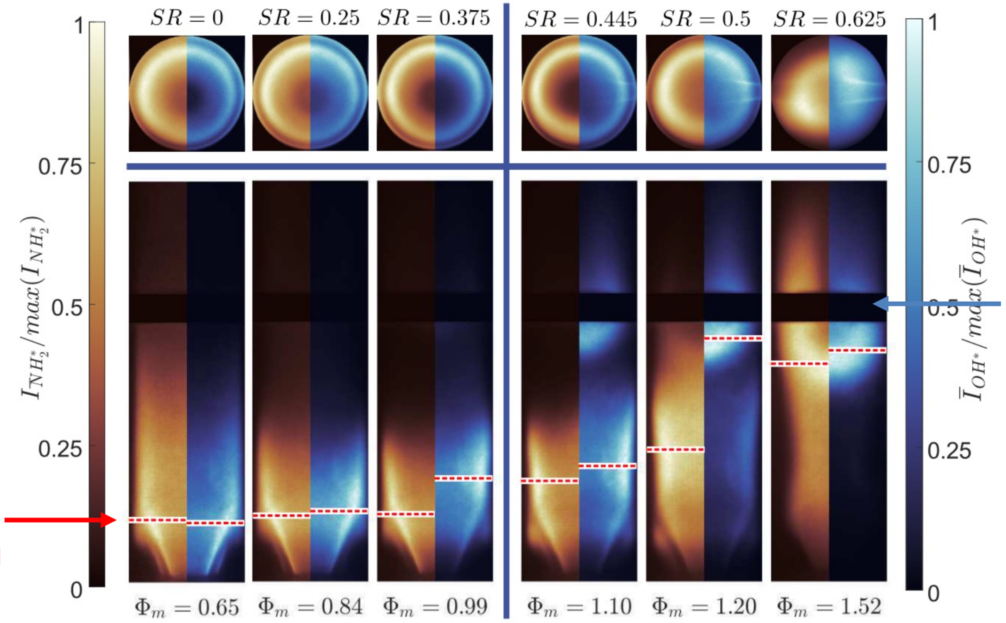


Rieth et al. Comb. Flame 2023

NTNU NH_3/H_2 Axially Staged Lab-Scale Burner

- NTNU (Norway) RQL 2-stage burner configuration: NH_3/H_2 + air [1]
- 1bar, 298 K at inlet
- Experimental data available: OH* and NH_2*
- Detailed study of NO and N_2O formation in realistic configuration

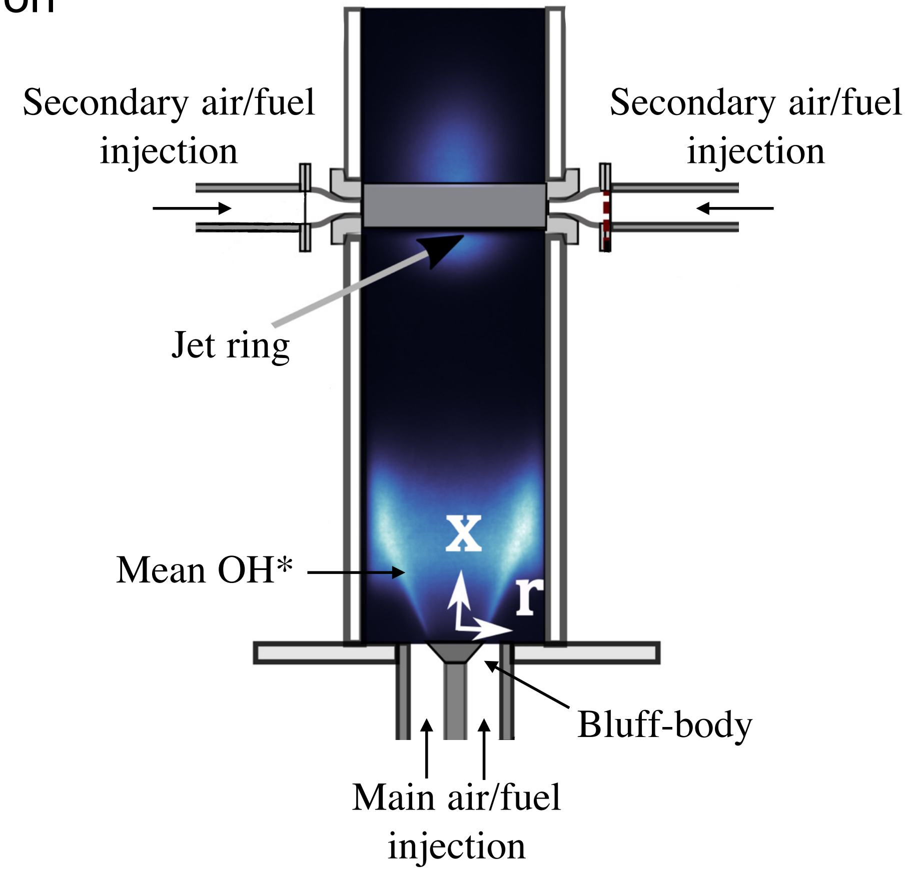
NH_2* (left) and OH* (right) chemiluminescence for 6 different split ratios



Max location

Secondary air injection

NTNU experimental set up

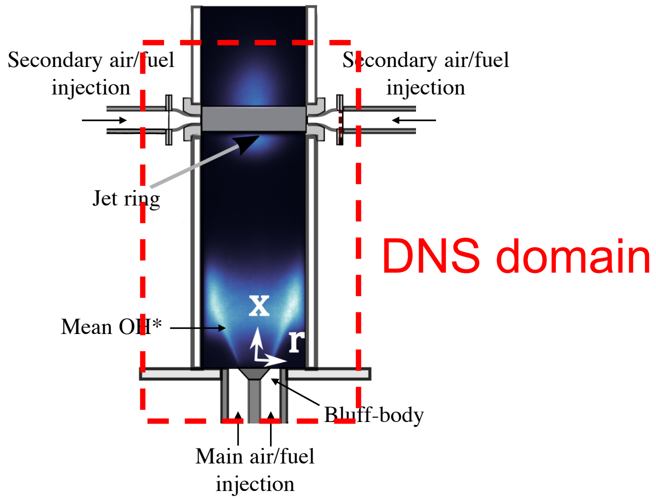


[1] Ånestad, Aksel, et al. "The Structure and Stability of Premixed CH4, H2, and NH3/H2 Flames in an Axially Staged Can Combustor." *Turbo Expo: Power for Land, Sea, and Air*. Vol. 86960. ASME, 2023

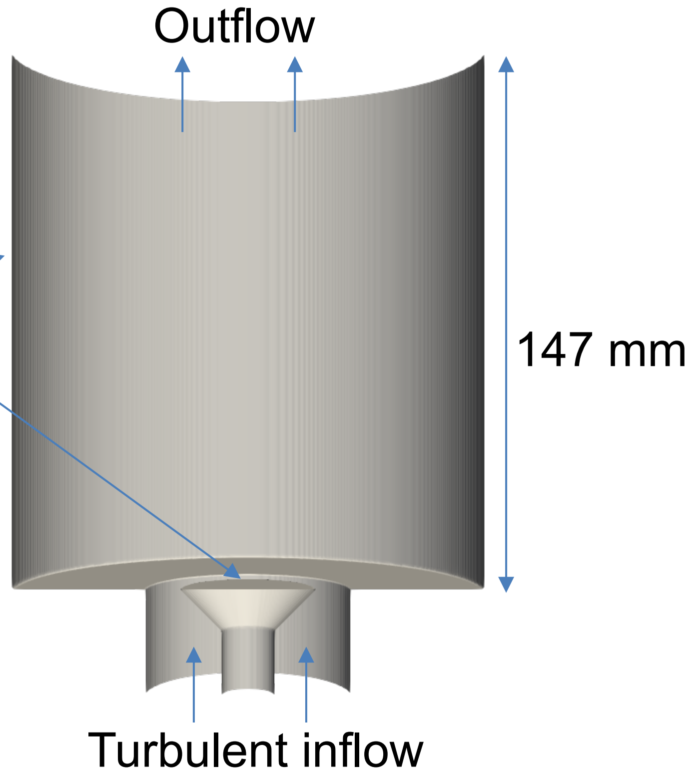
DNS of the Rich-Quench-Lean (RQL) Burner

- DNS simulation of a real combustor using PeleLMeX
- Combustor geometry captured with Embedded Boundary (EB) treatment

Conditions	
Pressure	1 bar
Inlet temperature	298 K
Inlet bulk velocity	15 m/s
Re number	10,540
Kolmogorov length	45 μm
Split ratio	0.0
Equiv. ratio	1.15
Fuel composition	$X_{NH3} = 0.75$; $X_{H2} = 0.25$
Chem. mechanism	Jiang et al



PeleLMeX domain with boundary conditions

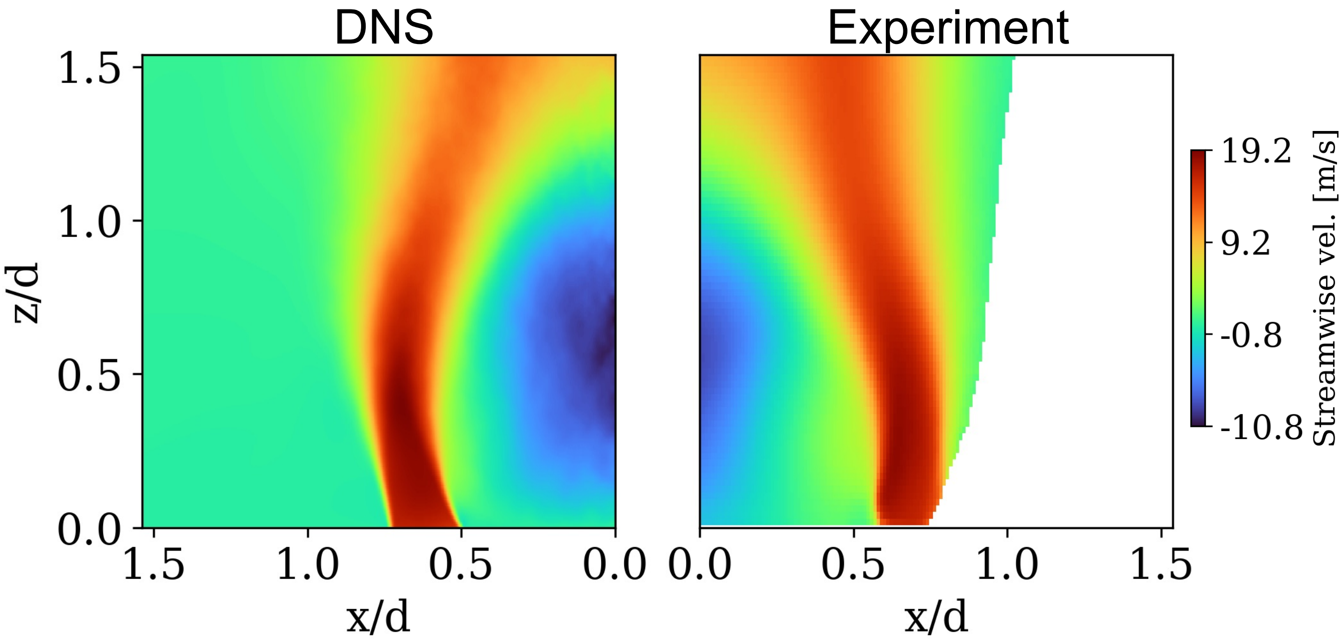


**Length in streamwise direction was reduced for visualization

DNS of the RQL Burner: validation

- Non-reacting and reacting flow field validation using a hydrogen-air configuration with the same bluff-body geometry

Non-reacting

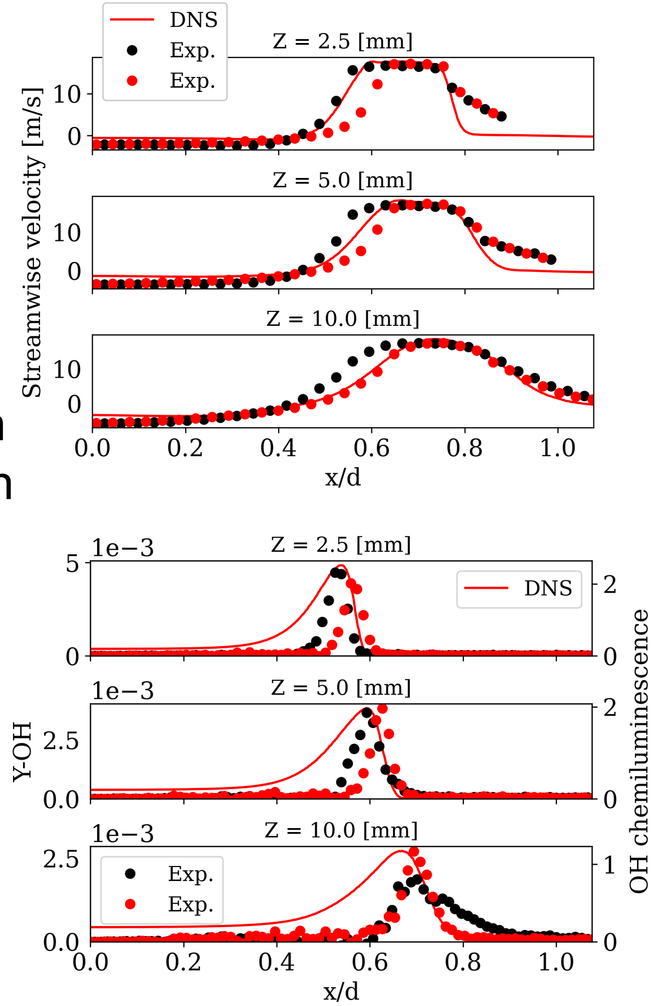


Comparison ensemble average streamwise velocity

- Two experimental means correspond to each side of the combustor along vertical symmetry plane
- Good agreement with streamwise velocity in the reacting flow field

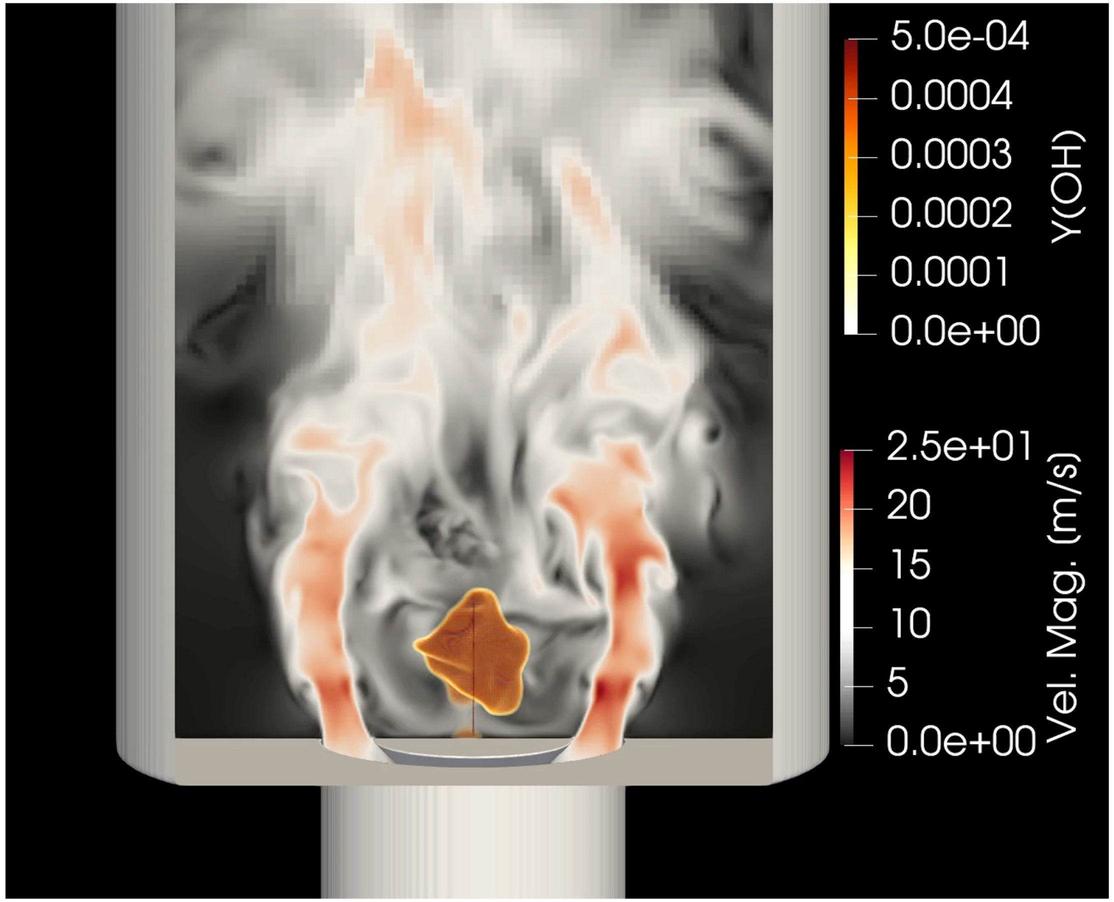
- OH peak correctly captured in the shear layer

Reacting



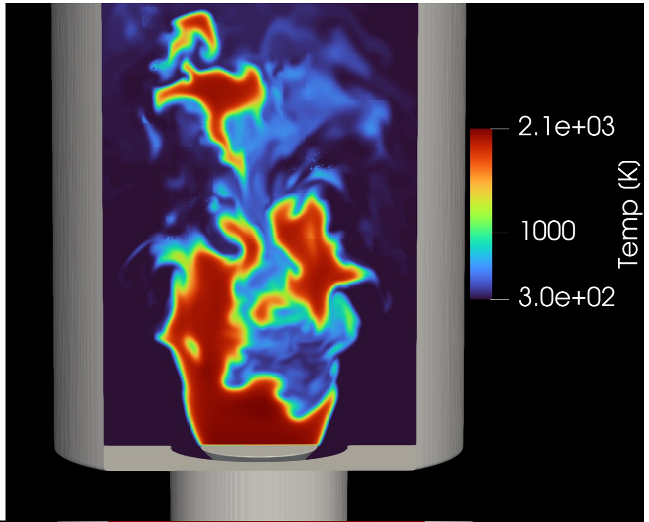
DNS of the RQL Burner: preliminary results

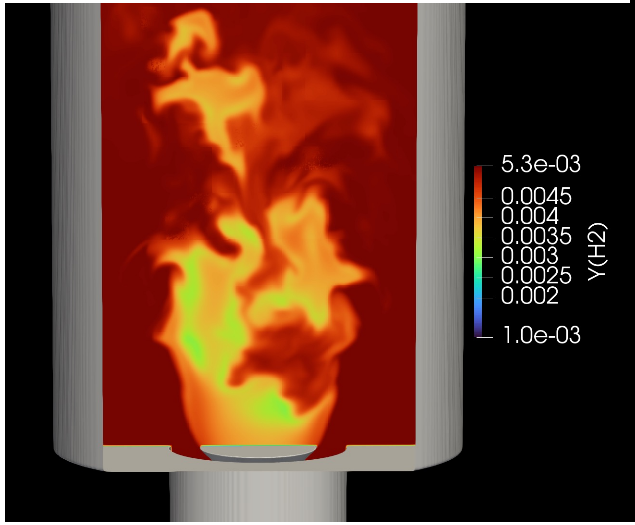
Ignition and early stages of combustion

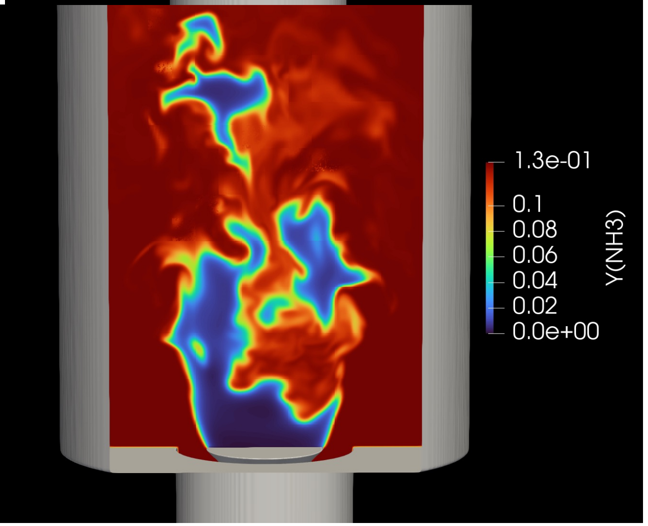


Preliminary instantaneous analysis:

- Ammonia is mostly consumed in the recirculation zone
- H_2 persists in the flame products side
- Potential increase in H_2 contribution to flame stabilization

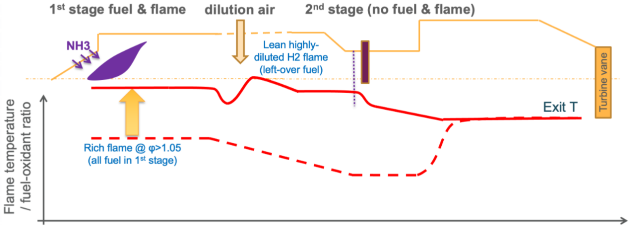






Ammonia Rich-Quench-Lean (RQL) Operation

- Reactivity and NO_x emissions are a challenge for ammonia combustion
- Promising strategy: 2-stage rich-quench-lean (RQL)
  - 1^{st} stage fuel-rich NH_3-air flame (good for NO_x[1])
  - 2^{nd} stage air injection to burn off remaining hydrogen



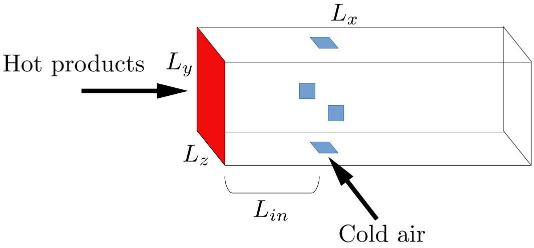
Heggset et al., Proc. of ASME Turbo Expo 2023.

NO_x depends on 2^{nd} stage combustion, mixing & residence times
→ unravel details with DNS

[1] Kobayashi et al, PROCI, 2019.

Numerical Setup

- Cross-flow: hot eq. products of rich ammonia-air flame
- 3 cases for ϕ=1.1, 1.2 and 1.3 + case with ammonia slip
- 4 air jets in cross-flow
- Air/unburned temperature is 780 K
- 25 atm pressure, 1900 K outlet target
- Timescales based on Heggset et al. LES[1], Re scaled down to 20,000
- 19 species NH_3/H_2 mechanism by Jiang et al.[2]



Code:

- PeleLMeX[3] - low-Mach AMR
- >30B grid cells
- Frontier up to 768 nodes

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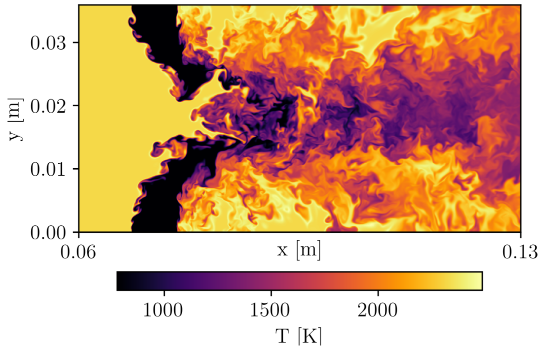
[1] Heggset et al., Proc. of ASME Turbo Expo 2023. [2] Jiang et al., Int. J. Energy Res., 2020.

[3] `https://github.com/AMReX-Combustion/PeleLMeX`

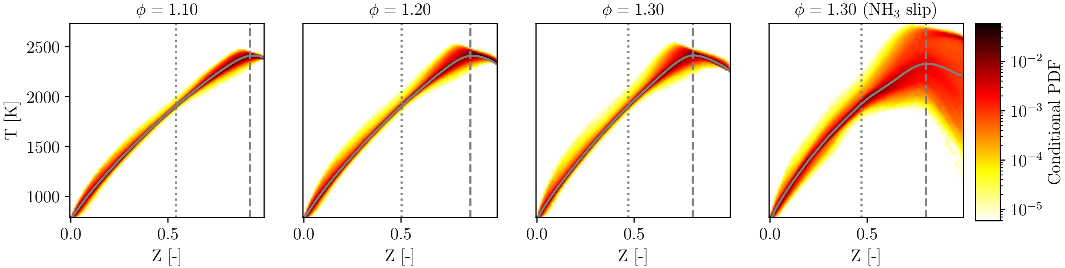
General Observations



- Rapid combustion as rich H_2-containing products come into contact with air jets
- Richer 1st stage → higher amount of H_2 → higher HRR
- Interesting ‘inverted’ diffusion flame (fuel on hot side)

Rieth et al., PROCI, 2024, submitted.

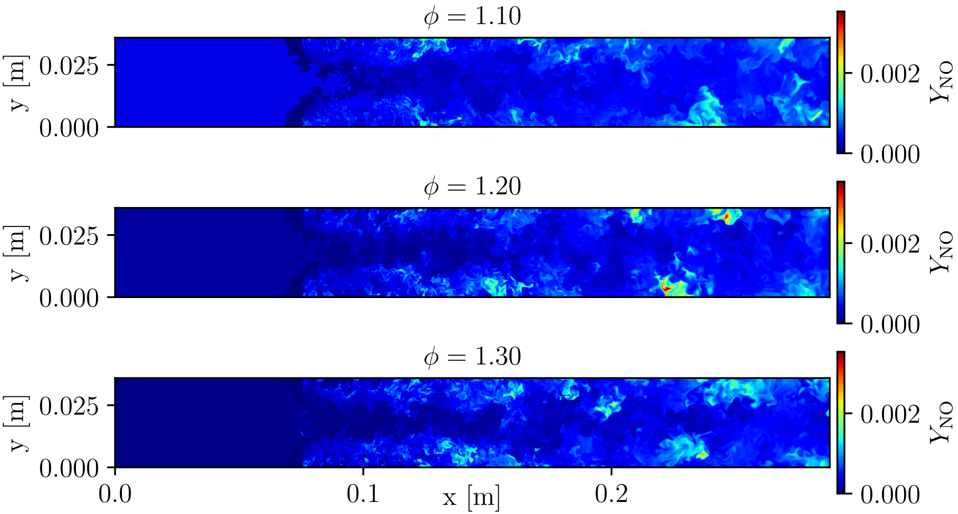
Temperature statistics



- Z=0: cold air, Z=1: cross-stream
- Richer 1st stage: larger temperature variation, especially close to stoichiometric mixture fraction
- Large temperature variation for NH_3 slip case

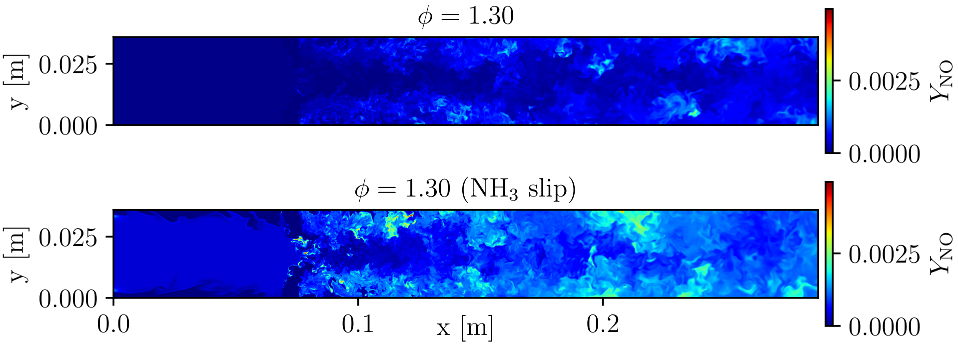
Rieth et al., PROCI, 2024, submitted.

NO formation



- Leaner 1st stage → higher NO at inlet
- NO appears in pockets downstream of air injection

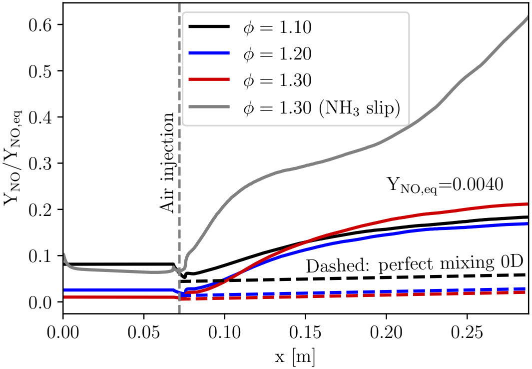
NO formation (with NH_3 slip)



- Additional DNS with ammonia slip in boundary regions of the cross-flow (4% of ammonia at air injection location)
- Significant increase of NO formation, NO produced in flame & downstream

Rieth et al., PROCI, 2024, submitted.

Averaged NO mass fraction profiles

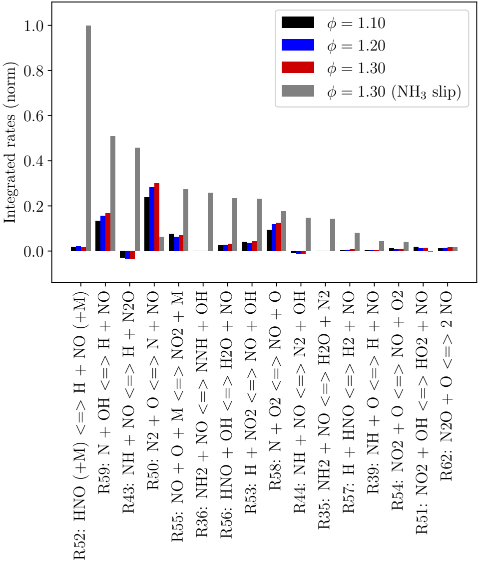


- Outlet NO depends on (1) inlet NO, (2) NO produced post air injection
- Richer 1st stage → lower inlet NO, faster NO production
- Ammonia slip leads to significantly faster NO production
- All cases show NO lower than equilibrium

- Through which pathways is NO produced?

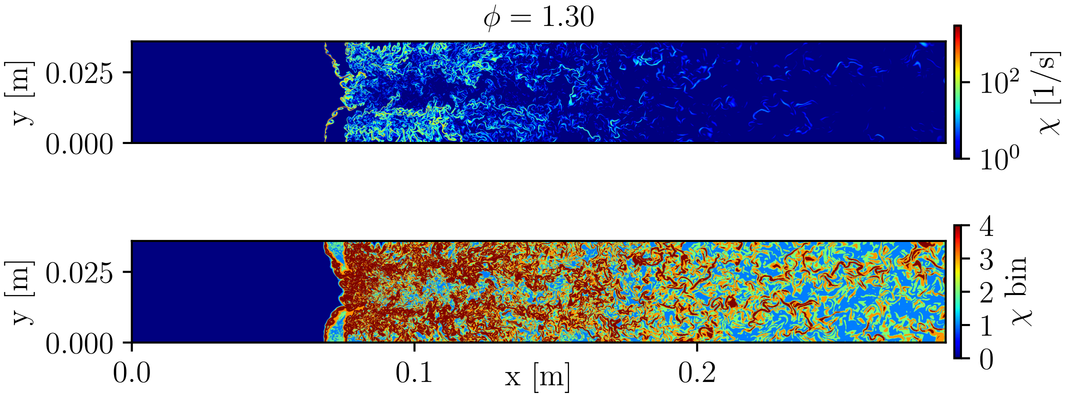
Rieth et al., PROCI, 2024, submitted.

NO formation pathways



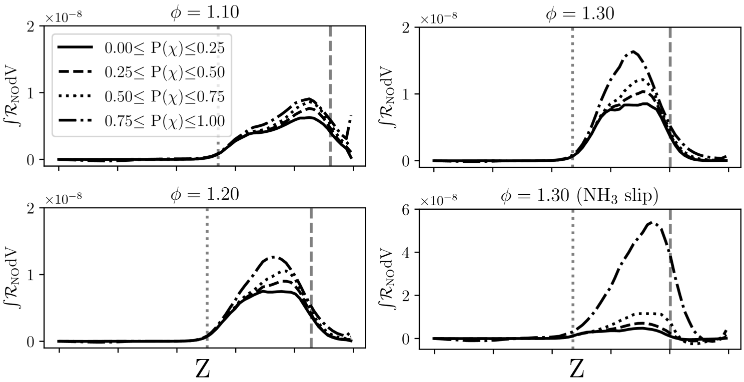
- NO formation for cases without ammonia slip is dominated by thermal pathway
- Higher H_2 content coming from 1^{st} stage leads to larger temperature variation due to heat release and higher probability for hot regions
- Significant NO production through HNO pathway with ammonia slip (mainly: $HNO(+M) = H + NO(+M)$), shifts in thermal production rates

NO formation dependence on mixing rates



- High scalar dissipation rate → high mixing rates
- Scalar dissipation rate binning is based on conditional PDF

NO formation dependence on mixing rates



- Z=0: cold air, Z=1: cross-stream
- Higher first stage equivalence ratio: more NO production in strong mixing regions (due to higher T variation through heat release)
- Significantly higher NO production in ammonia slip case

Conclusions and future work

Conclusions

- Increase in pressure amplifies thermo-diffusive instabilities in lean premixed flames when hydrogen is present
- Flames of hydrogen-enriched fuel blends can be scaled with Pe/Ze (incl. varying pressure and composition)
- Latest bluff-body flame simulations show promising results in simulating flames in more complex geometries
- RQL simulations at high pressure demonstrate NO emission reduction, but also show detrimental impact of ammonia slip

Future work

- Detailed DNS in complex geometries, also to inform DNS of RQL 2^{nd} stage
- Incorporate findings and data in models: emission modeling, burning rate scaling

Acknowledgments

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