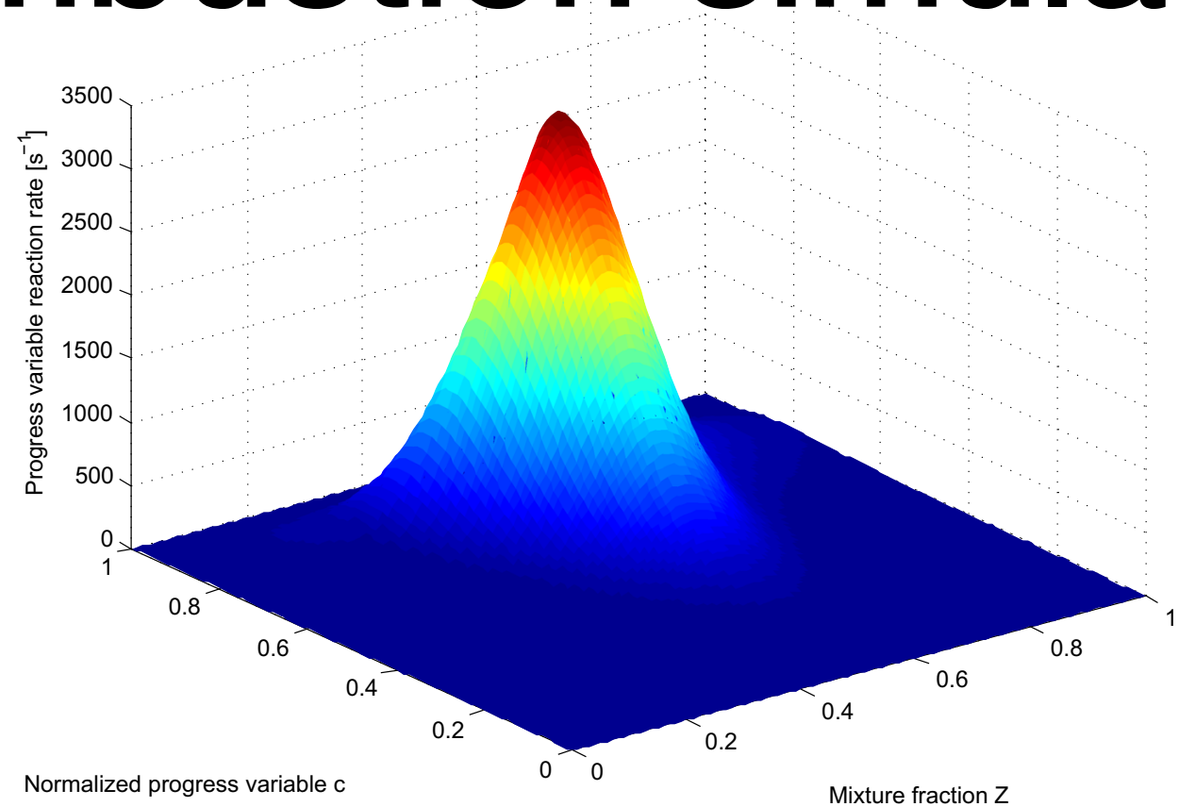
Chemistry reduction for combustion simulations



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Environmental impact of combustion

CO_2

Directly linked to hydrocarbon consumption

- ***related to the overall system efficiency***
- ***weakly sensible to the combustion chamber design***

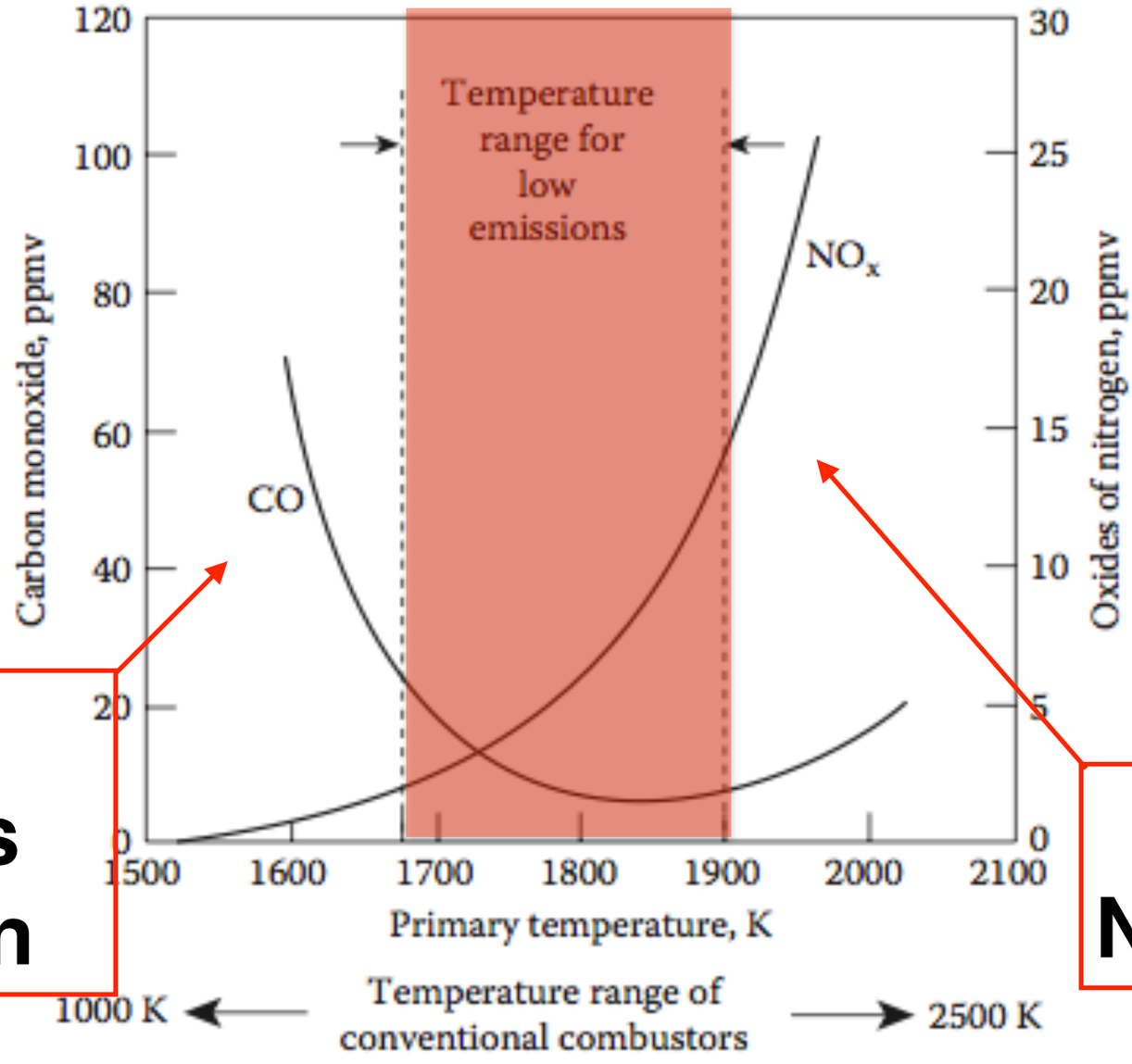
Unburnt hydrocarbons (HC)
Carbon monoxide (CO)
Nitrogen oxides (NOx)
Sulfur oxides (SOx)
Particles…

Combustion challenge

Dangerous in small quantities (ppm to few hundred of ppm)

- *very sensible to the combustion chamber design*

Temperature control is a key to control pollutant emissions



Unburnt hydrocarbons CO production

High NO_X production

From Lefebvre and Ballal, 2010

Order of magnitude of detailed chemical mechanisms sizes

Fuel/Oxidizer	Number of species	Number of reactions
H_2/Air	10	50
CH_4/Air	100	500
Heavy hydrocarbons/ Air (Kerosene, gasoil, …)	1000	5000

TABLE II

Propane-Air Reaction Mechanism Rate Coefficients in the Form $k_f = AT^{\beta} \exp(-E/$
Units are moles, cubic centimeters, seconds, Kelvins and calories/mole.

	REACTION	A	β	E
1.	$O_2 + H \rightleftharpoons OH + O$	2.000E14	0.0	16820.
2.	$H_2 + O \rightleftharpoons OH + H$	5.060E04	2.67	6290.
3.	$H_2 + OH \rightleftharpoons H_2O + H$	1.000E08	1.6	3300.
4.	$OH + OH \rightleftharpoons H_2O + O$	1.500E09	1.14	100.
5.	$H + H + M' \rightleftharpoons H_2 + M'$	1.800E18	-1.0	0.
6.	$H + OH + M' \rightleftharpoons H_2O + M'$	2.200E22	-2.0	0.
7.	$O + O + M' \rightleftharpoons O_2 + M'$	2.900E17	-1.0	0.
8.	$H + O_2 + M' \rightleftharpoons HO_2 + M'$	2.300E18	-0.8	0.
9.	$HO_2 + H \rightleftharpoons OH + OH$	1.500E14	0.0	1000.
10.	$HO_2 + H \rightleftharpoons H_2 + O_2$	2.500E13	0.0	690.
11.	$HO_2 + H \rightleftharpoons H_2O + O$	3.000E13	0.0	1720.
12.	$HO_2 + O \rightleftharpoons OH + O_2$	1.800E13	0.0	-400.
13.	$HO_2 + OH \rightleftharpoons H_2O + O_2$	6.000E13	0.0	0.
14.	$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$	2.500E11	0.0	-1240.
15.	$OH + OH + M' \rightleftharpoons H_2O_2 + M'$	3.256E22	-2.0	0.
16.	$H_2O_2 + H \rightleftharpoons H_2 + HO_2$	1.700E12	0.0	3750.
17.	$H_2O_2 + H \rightleftharpoons H_2O + OH$	1.000E13	0.0	3590.
18.	$H_2O_2 + O \rightleftharpoons OH + HO_2$	2.800E13	0.0	6410.
19.	$H_2O_2 + OH \rightleftharpoons H_2O + HO_2$	5.400E12	0.0	1000.
20.	$CO + OH \rightleftharpoons CO_2 + H$	4.400E06	1.5	-740.
21.	$CO + HO_2 \rightleftharpoons CO_2 + OH$	1.500E14	0.0	23610.
22.	$CO + O$			
23.	$CO + O_2$			
24.	$CH + O$			
25.	$CH + O_2$			
26.	$CH + C$			
27.	$CHO + I$			
28.	$CHO + C$			
29.	$CHO + C$			
30.	$CHO + C$			
31.	$CHO + C$			
32.	$CHO + M \rightleftharpoons CO + H + M$	7.100E14	0.0	16820.

$$k(T) = AT^{b} e^{\frac{-E_a}{RT}}$$

Species for a Propane-Air Flame

C_3H_8	$N*C_3H_7$	$I*C_3H_7$	C_3H_6	C_2H_6	C_2H_5
CH_3CHO	C_2H_4	CH_3CO	C_2H_3	CH_2CO	C_2H_2
C_2HO	C_2H	CH_4	CH_3	CH_2O	CH_2
CHO	CH	CO_2	CO	H_2O_2	H_2O
H_2	HO_2	OH	H	O_2	O
N_2	NO	N			

TABLE II (continued)

Propane-Air Reaction Mechanism Rate Coefficients in the Form $k_f = AT^{\beta} \exp(-E/RT)$
Units are moles, cubic centimeters, seconds, Kelvins and calories/mole.

	REACTION	A	β	E
33.	$CH_2 + H \rightleftharpoons CH + H_2$	4.000E13	0.0	0.
34.	$CH_2 + O \rightarrow CO + H + H$	8.000E13	0.0	0.
35.	$CH_2 + O_2 \rightarrow CO + OH + H$	6.500E12	0.0	1500.
36.	$CH_2 + O_2 \rightarrow CO_2 + H + H$	6.500E12	0.0	1500.
37.	$CH_2O + H \rightleftharpoons CHO + H_2$	2.500E13	0.0	4000.
38.	$CH_2O + O \rightleftharpoons CHO + OH$	3.500E13	0.0	3490.
39.	$CH_2O + OH \rightleftharpoons CHO + H_2O$	3.000E13	0.0	1200.
40.	$CH_2O + HO_2 \rightleftharpoons CHO + H_2O_2$	1.000E12	0.0	8000.
41.	$CH_2O + CH_3 \rightleftharpoons CHO + CH_4$	1.000E11	0.0	6100.
42.	$CH_2O + M' \rightleftharpoons CHO + H + M'$	1.400E17	0.0	76550.
43.	$CH_3 + H \rightleftharpoons CH_2 + H_2$	1.800E14	0.0	15070.
44.	$CH_3 + O \rightleftharpoons CH_2O + H$	7.000E13	0.0	0.
45.	$CH_3 + OH \rightarrow CH_2O + H + H$	9.000E14	0.0	15500.
46.	$CH_3 + OH \rightarrow CH_2O + H_2$	8.000E12	0.0	0.
47.	$CH_3 + O_2 \rightarrow CH_2O + H + O$	1.500E13	0.0	28710.
48.	$CH_3 + CH_3 \rightleftharpoons C_2H_6$	7.470E52	-11.9	19190.
49.	$CH_3 + M \rightarrow CH_2 + H + M$	1.000E16	0.0	90910.
50.	$CH_3 + C_2H_4 \rightarrow C_2H_4 + H_2$	1.000E16	0.0	32060.
51.	$CH_3 + C_2H_4 \rightarrow C_2H_4 + H$	4.000E13	0.0	0.
52.	$CH_3 + H_2 \rightarrow CH_4 + CH_3$	2.200E04	3.0	8760.
53.	$CH_3 + H \rightarrow CH_4 + CH_3$	1.200E07	2.1	7630.
54.	$CH_3 + OH \rightarrow CH_4 + H_2O + CH_3$	1.600E06	2.1	2460.
55.	$CH_3 + HO_2 \rightarrow CH_4 + H_2O_2 + CH_3$	4.000E12	0.0	19425.
56.	$CH_4 + M \rightarrow CH_3 + H$	3.200E34	-6.0	109450.
57.	$CH_4 + CH_3 \rightarrow CH_4 + CH_3$	1.300E13	0.0	9545.
58.	$CH_4 + CH \rightleftharpoons C_2H_4 + H$	3.000E13	0.0	-400.
59.	$C_2H + O \rightleftharpoons CO + CH$	1.000E13	0.0	0.
60.	$C_2H + H_2 \rightleftharpoons C_2H_2 + H$	1.100E13	0.0	2870.
61.	$C_2H + O_2 \rightleftharpoons C_2HO + O$	5.000E13	0.0	1500.
62.	$C_2HO + H \rightleftharpoons CH_2 + CO$	3.000E13	0.0	0.
63.	$C_2HO + O \rightarrow CO + CO + H$	1.000E14	0.0	0.
64.	$C_2H_2 + O \rightleftharpoons CH_2 + CO$	4.100E08	1.5	1700.
65.	$C_2H_2 + O \rightleftharpoons C_2HO + H$	4.300E14	0.0	12130.
66.	$C_2H_2 + OH \rightleftharpoons H_2O + C_2H$	1.000E13	0.0	7000.
67.	$C_2H_2 + M \rightleftharpoons C_2H + H + M$	3.600E16	0.0	106700.

Propane-Air Reaction Mechanism Rate Coefficients in the Form $k_f = AT^{\beta} \exp(-E/RT)$
Units are moles, cubic centimeters, seconds, Kelvins and calories/mole.

	REACTION	A	β	E
68.	$CH_2CO + H \rightleftharpoons CH_3 + CO$	7.000E12	0.0	3000.
69.	$CH_2CO + O \rightleftharpoons CHO + CHO$	1.800E12	0.0	1340.
70.	$CH_2CO + OH \rightleftharpoons CH_2O + CHO$	1.000E13	0.0	0.
71.	$CH_2CO + M' \rightleftharpoons CH_2 + CO + M'$	1.000E16	0.0	59330.
72.	$C_2H_3 + H \rightleftharpoons H_2 + C_2H_2$	2.000E13	0.0	0.
73.	$C_2H_3 + O \rightleftharpoons CH_2CO + H$	3.000E13	0.0	0.
74.	$C_2H_3 + O_2 \rightharpoonup CH_2O + CHO$	1.500E12	0.0	0.
75.	$C_2H_3 \rightleftharpoons C_2H_2 + H$	1.600E32	-5.5	46290.
76.	$CH_3CO + H \rightleftharpoons CH_2CO + H_2$	2.000E13	0.0	0.
77.	$CH_3CO + O \rightleftharpoons CH_3 + CO_2$	2.000E13	0.0	0.
78.	$CH_3CO + CH_3 \rightleftharpoons C_2H_6 + CO$	5.000E13	0.0	0.
79.	$CH_3CO \rightleftharpoons CH_3 + CO$	2.300E26	-5.0	17990.
80.	$C_2H_4 + H \rightleftharpoons C_2H_3 + H_2$	1.500E14	0.0	10215.
81.	$C_2H_4 + O \rightharpoonup CH_3CO + H$	1.600E09	1.2	740.
82.	$C_2H_4 + OH \rightleftharpoons C_2H_3 + H_2O$	3.000E13	0.0	3000.
83.	$C_2H_4 + CH_3 \rightleftharpoons C_2H_3 + CH_4$	4.200E11	0.0	11120.
84.	$C_2H_4 + M' \rightleftharpoons C_2H_2 + H_2 + M'$	2.500E17	0.0	76500.
85.	$CH_3CHO + H \rightleftharpoons CH_3CO + H_2$	4.000E13	0.0	4210.
86.	$CH_3CHO + O \rightleftharpoons CH_3CO + OH$	5.000E12	0.0	1790.
87.	$CH_3CHO + OH \rightleftharpoons CH_3CO + H_2O$	8.000E12	0.0	0.
88.	$CH_3CHO + HO_2 \rightleftharpoons CH_3CO + H_2O_2$	1.700E12	0.0	10720.
89.	$CH_3CHO + CH_2 \rightleftharpoons CH_3CO + CH_3$	2.500E12	0.0	3800.
90.	$CH_3CHO + CH_3 \rightleftharpoons CH_3CO + CH_4$	8.500E10	0.0	6000.
91.	$CH_3CHO \rightleftharpoons CH_3 + CHO$	2.000E15	0.0	79190.
92.	$C_2H_5 + H \rightleftharpoons CH_3 + CH_3$	3.000E13	0.0	0.
93.	$C_2H_5 + O \rightleftharpoons CH_3CHO + H$	5.000E13	0.0	0.
94.	$C_2H_5 + O_2 \rightleftharpoons HO_2 + C_2H_4$	2.000E12	0.0	5000.
95.	$C_2H_5 + CH_3 \rightleftharpoons C_3H_8$	7.000E12	0.0	0.
96.	$C_2H_5 + C_2H_5 \rightleftharpoons C_2H_4 + C_2H_6$	1.400E12	0.0	0.
97.	$C_2H_5 \rightleftharpoons C_2H_4 + H$	1.300E19	-2.0	41480.
98.	$C_2H_6 + H \rightleftharpoons H_2 + C_2H_5$	5.400E02	3.5	5215.
99.	$C_2H_6 + O \rightleftharpoons OH + C_2H_5$	3.000E07	2.0	5120.
100.	$C_2H_6 + OH \rightleftharpoons H_2O + C_2H_5$	6.300E06	2.0	645.
101.	$C_2H_6 + HO_2 \rightleftharpoons H_2O_2 + C_2H_5$	6.000E12	0.0	19420.
102.	$C_2H_6 + CH_3 \rightleftharpoons C_2H_5 + CH_4$	5.500E-01	4.0	8300.
103.	$C_2H_6 + CH_2 \rightleftharpoons CH_3 + C_2H_5$	2.200E13	0.0	8680.
104.	$C_2H_6 + CH \rightleftharpoons H + C_3H_6$	1.100E14	0.0	-260.

Propane-Air Reaction Mechanism Rate Coefficients in the Form $k_f = AT^{\beta} \exp(-E/RT)$
Units are moles, cubic centimeters, seconds, Kelvins and calories/mole.

	REACTION	A	β	E
105.	$C_3H_8 + H \rightleftharpoons N * C_3H_7 + H_2$	1.300E14	0.0	9710.
106.	$C_3H_8 + H \rightleftharpoons I * C_3H_7 + H_2$	1.000E14	0.0	8350.
107.	$C_3H_8 + O \rightleftharpoons N * C_3H_7 + OH$	3.000E13	0.0	5765.
108.	$C_3H_8 + O \rightleftharpoons I * C_3H_7 + OH$	2.600E13	0.0	4470.
109.	$C_3H_8 + OH \rightleftharpoons N * C_3H_7 + H_2O$	6.300E06	2.0	645.
110.	$C_3H_8 + OH \rightleftharpoons I * C_3H_7 + H_2O$	1.200E08	1.46	-190.
111.	$C_3H_8 + HO_2 \rightleftharpoons N * C_3H_7 + H_2O_2$	6.000E12	0.0	19420.
112.	$C_3H_8 + HO_2 \rightleftharpoons I * C_3H_7 + H_2O_2$	2.000E12	0.0	17000.
113.	$C_3H_8 + CH_3 \rightleftharpoons N * C_3H_7 + CH_4$	7.500E12	0.0	14950.
114.	$C_3H_8 + CH_3 \rightleftharpoons I * C_3H_7 + CH_4$	4.300E12	0.0	13280.
115.	$N * C_3H_7 + H \rightleftharpoons C_3H_8$	2.000E13	0.0	0.
116.	$I * C_3H_7 + H \rightleftharpoons C_3H_8$	2.000E13	0.0	0.
117.	$N * C_3H_7 + O_2 \rightleftharpoons C_3H_6 + HO_2$	1.000E12	0.0	5000.
118.	$I * C_3H_7 + O_2 \rightleftharpoons C_3H_6 + HO_2$	1.000E12	0.0	2990.
119.	$N * C_3H_7 \rightleftharpoons C_3H_6 + H$	1.000E14	0.0	37340.
120.	$I * C_3H_7 \rightleftharpoons C_3H_6 + H$	2.000E14	0.0	38730.
121.	$N * C_3H_7 \rightleftharpoons C_2H_4 + CH_3$	3.000E14	0.0	33250.
122.	$C_3H_6 + O \rightleftharpoons CH_3CO + CH_3$	5.000E12	0.0	450.
123.	$C_3H_6 + OH \rightharpoonup C_2H_2 + CH_3 + H_2O$	2.000E13	0.0	3060.
124.	$N_2 + O \rightleftharpoons N + NO$	0.136E15	0.0	76400.
125.	$N + O_2 \rightleftharpoons O + NO$	0.267E11	0.72	7080.
126.	$N + OH \rightleftharpoons H + NO$	0.280E14	0.0	0.

Third body efficiencies for M' : $\alpha(O_2) = 0.4$, $\alpha(N_2) = 0.4$, $\alpha(CO) = 0.75$, $\alpha(CO_2) = 1.5$, $\alpha(H_2O) = 6.5$, $\alpha(C_3H8) = 3$.

Balance equations for CFD with detailed chemistry

Too expensive
for 3-D simulations:

- number of Eqs
- stiffness of reaction rates

$$\frac{\partial \rho E}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i E) = \frac{\partial}{\partial x_i}(\lambda \frac{\partial T}{\partial x_i}) + \frac{\partial}{\partial x_j}(\tau_{ij} u_i)$$

$$\frac{\partial \rho Y_1}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i Y_1) = \frac{\partial}{\partial x_i}(\rho D_k \frac{\partial Y_1}{\partial x_i}) + \dot{\omega}_1$$

$$\cdots$$

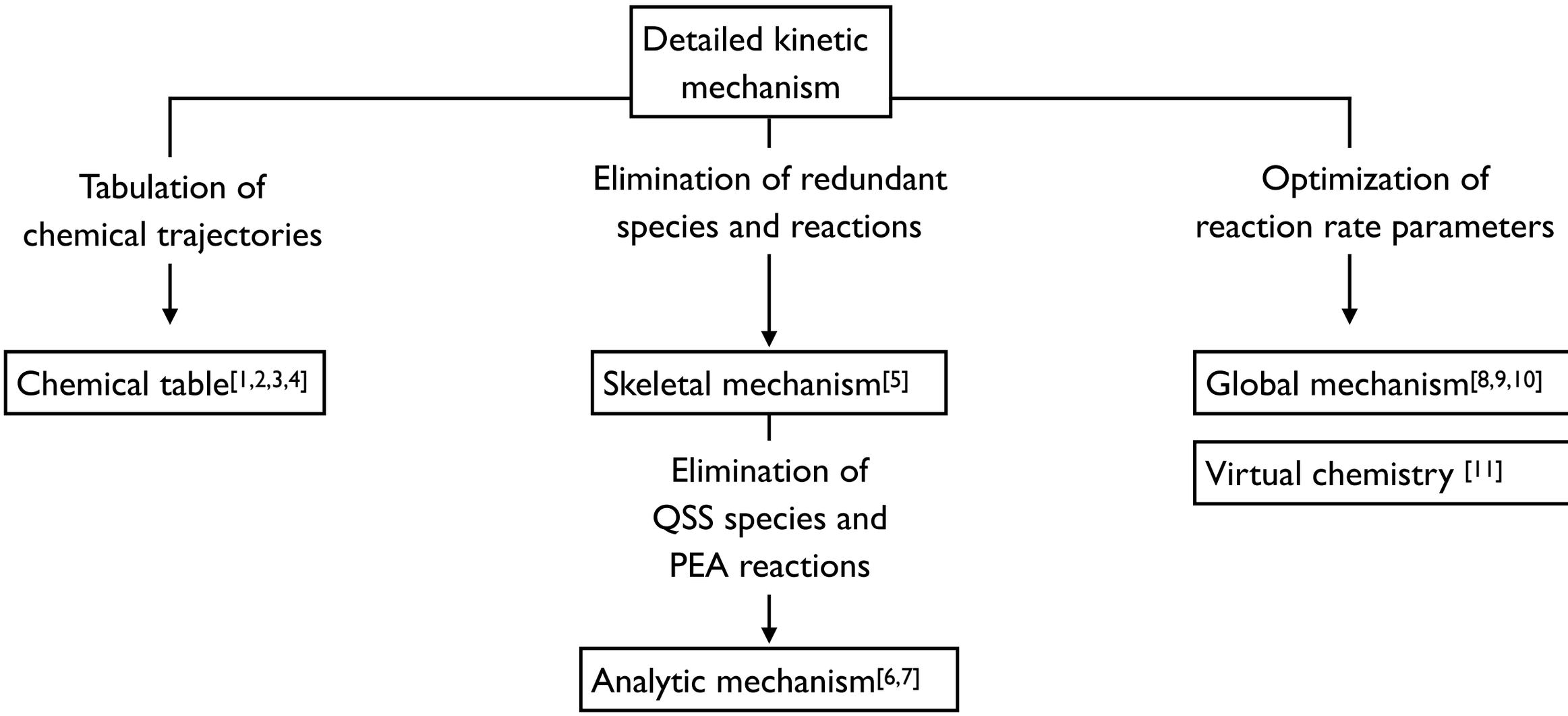
$$\frac{\partial \rho Y_k}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i Y_k) = \frac{\partial}{\partial x_i}(\rho D_k \frac{\partial Y_k}{\partial x_i}) + \dot{\omega}_k$$

$$\cdots$$

$$\frac{\partial \rho Y_n}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i Y_n) = \frac{\partial}{\partial x_i}(\rho D_k \frac{\partial Y_n}{\partial x_i}) + \dot{\omega}_n$$

Kinetic scheme reduction strategies for combustion modeling

B. Fiorina, D. Veynante and S. Candel. Modeling Combustion Chemistry in Large Eddy Simulation of Turbulent Flames. Flow Turb. and Combustion. Vol 94, Issue 1, pp3-42 (2015).



[1] N. Peters (1984)
[2] U. Maas & S. Pope (1992)
[3] O. Gicquel et al., (2000)
[4] J.A. Van Oijien et al., (2001)

[5] J. Luche (2003)
[6] T. Lu et al., (2007)
[7] T. Jaravel et al., (2016)

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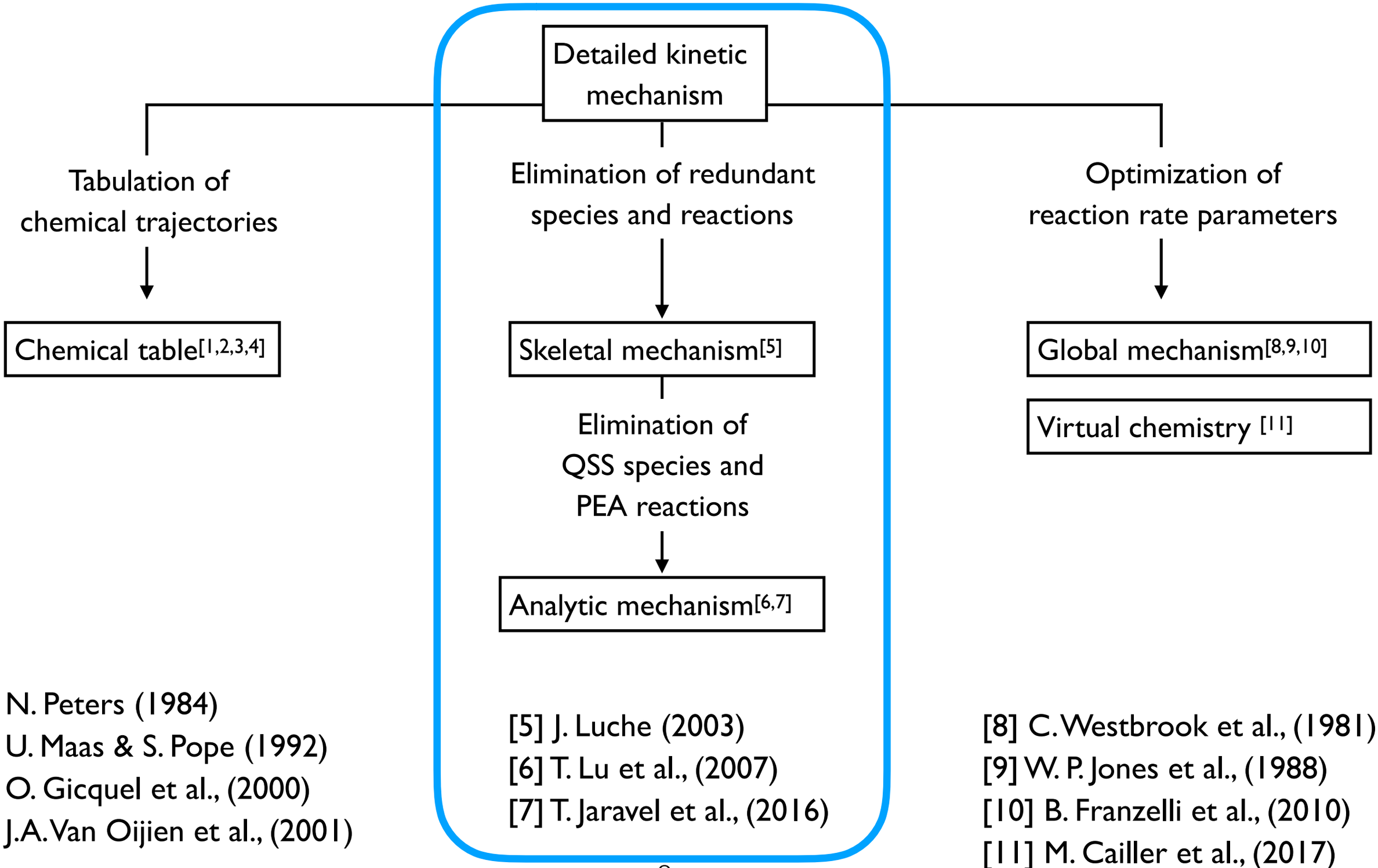

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Two main steps

Detailed mechanism

remove redundant species and reactions

Skeletal mechanism

find analytical relations between species

Analytically reduced mechanism

Two main steps

Detailed mechanism

remove redundant species and reactions

Skeletal mechanism

find analytical relations between species

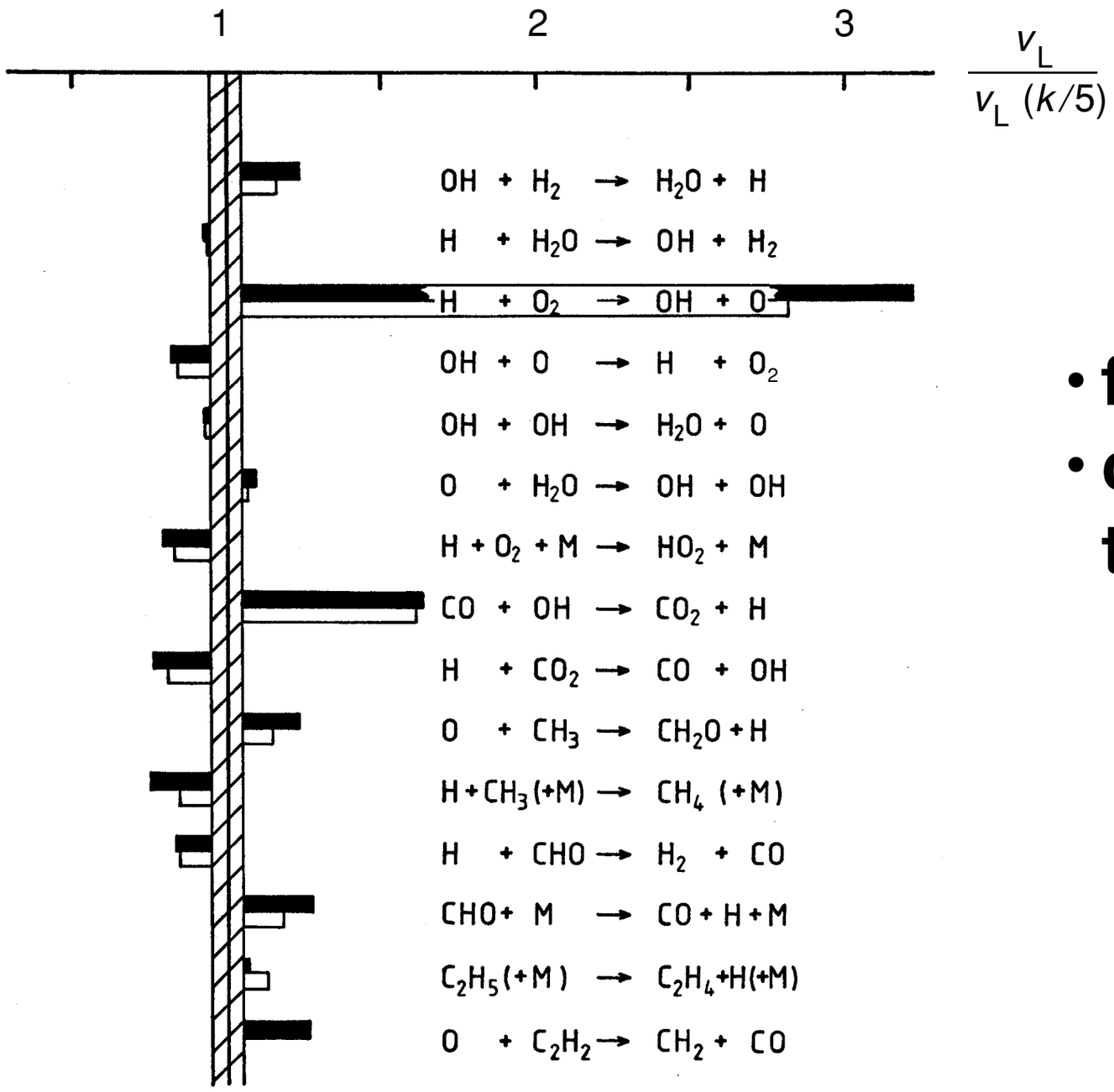
Analytically reduced mechanism

From detailed to skeletal mechanism

T. Turanyi. *Reduction of large reaction mechanisms.* New journal of chemistry. 1990

- **Species can be classified into three classes**
  - *important species*: essential to represent combustion processes
  - *necessary species*: required for the prediction of important species
  - *redundant species*: can be removed without affecting combustion phenomena
- **Reactions are also classified as:**
  - *important reactions*: essential to represent chemical pathway and kinetics rate
  - *necessary reactions*: essential to represent chemical pathway
  - *redundant reactions*: can be removed without altering production of important and necessary species

Steady premixed flame configuration: flame speed targeted



- **few sensitive reactions**
- **quasi-independant of the fuel**

Fig. 7.6. Sensitivity analysis for the flame velocity v_L in premixed stoichiometric CH_4- (black) and C_2H_6-air flames (white) at p = 1 bar, T_u = 298 K (Warnatz 1984).

Self - ignition case: OH concentration target

(1) $2\text{-}C_{10}H_{21} \rightarrow 5\text{-}C_{10}H_{21}$

(2) $5\text{-}C_{10}H_{21} \rightarrow 2\text{-}C_{10}H_{21}$

(3) $2\text{-}C_{10}H_{21} + O_2 \rightarrow 2\text{-}C_{10}H_{21}\text{-}O_2$

(4) $2\text{-}C_{10}H_{21}\text{-}O_2 \rightarrow 2\text{-}C_{10}H_{21} + O_2$

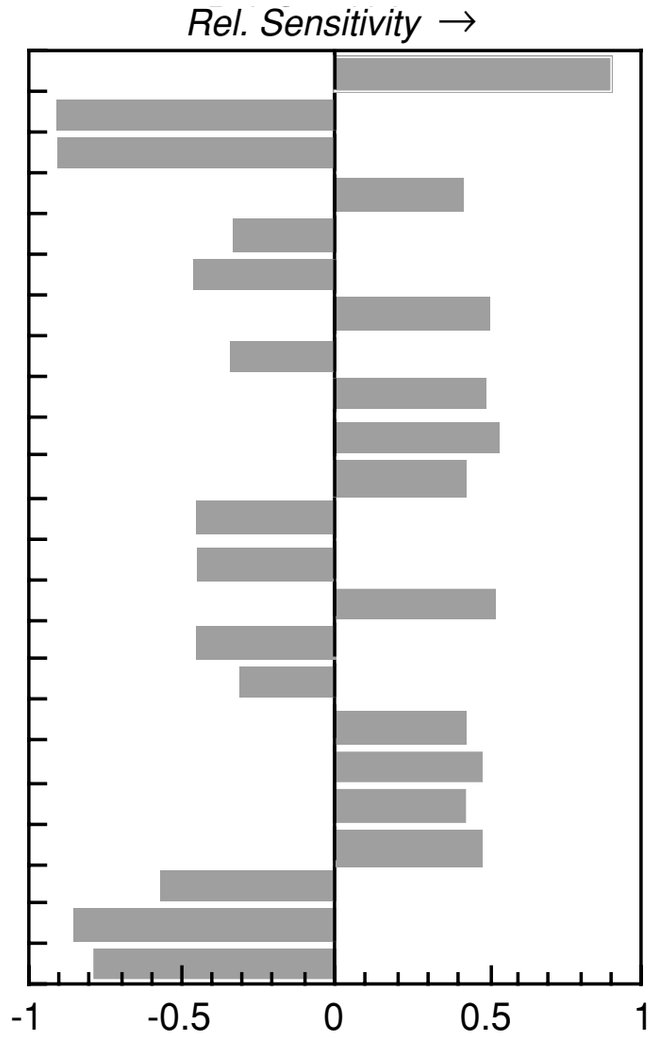
(5) $5\text{-}C_{10}H_{21}\text{-}O_2 \rightarrow 5\text{-}C_{10}H_{21} + O_2$

(6)
(7)
(8) $C_{10}H_{21}\text{-}O_2 \rightarrow C_{10}H_{20}\text{-}OOH$
(9)
(10)

(11)
(12) $C_{10}H_{20}\text{-}OOH \rightarrow C_{10}H_{21}\text{-}O_2$
(13)
(14)

(15)
(16)
(17) $C_{10}H_{20}\text{-}OOH + O_2 \rightarrow O_2\text{-}C_{10}H_{20}\text{-}OOH$
(18)
(19)

(20)
(21) $O_2\text{-}C_{10}H_{20}\text{-}OOH \rightarrow O_2\text{-}C_{10}H_{20}\text{-}O + OH$
(22)
(23)



- **more sensitive reactions than steady cases**

Fig. 7.7. Sensitivity analysis for the OH concentration in an igniting stoichiometric $C_{10}H_{22}$-air mixture at p = 13 bar, T_u = 800 K (Nehse et al. 1996); the numbers in front of the formulae denote the location of the free electron or of O_2 in the chains, respectively. For clearness, no isomeric structures are given in Reactions (6)-(23).

Redundant species and reactions will depend on the configuration targeted !
Wide range of operating conditions targeted —> less redundant species and reactions

Elimination of redundant species and reactions

Direct Relation Graph method (DRG)

T. Lu, C. Law, Proc. Combust. Inst. 30 (2005)

- **measure of the coupling between two species that are directly related**

$$r_{AB}^{\mathrm{DRG}} \equiv \frac{\sum_{i=1,n_{\mathrm{R}}} |\nu_{i,A}\omega_i \delta_B^i|}{\sum_{i=1,n_{\mathrm{R}}} |\nu_{i,A}\omega_i|}$$

$$\omega_i = \omega_{\mathrm{f},i} - \omega_{\mathrm{b},i} \qquad \delta_B^i = \begin{cases} 1, & \text{if the } i\text{th reaction involves species } B \\ 0, & \text{otherwise.} \end{cases}$$

- **measure if A and B are directly related**
- **Target A: if** $r_{AB}^{DRG} < \epsilon$ **then B can be removed**

T. Lu, C. Law, Proc. Combust. Inst. 30 (2005)

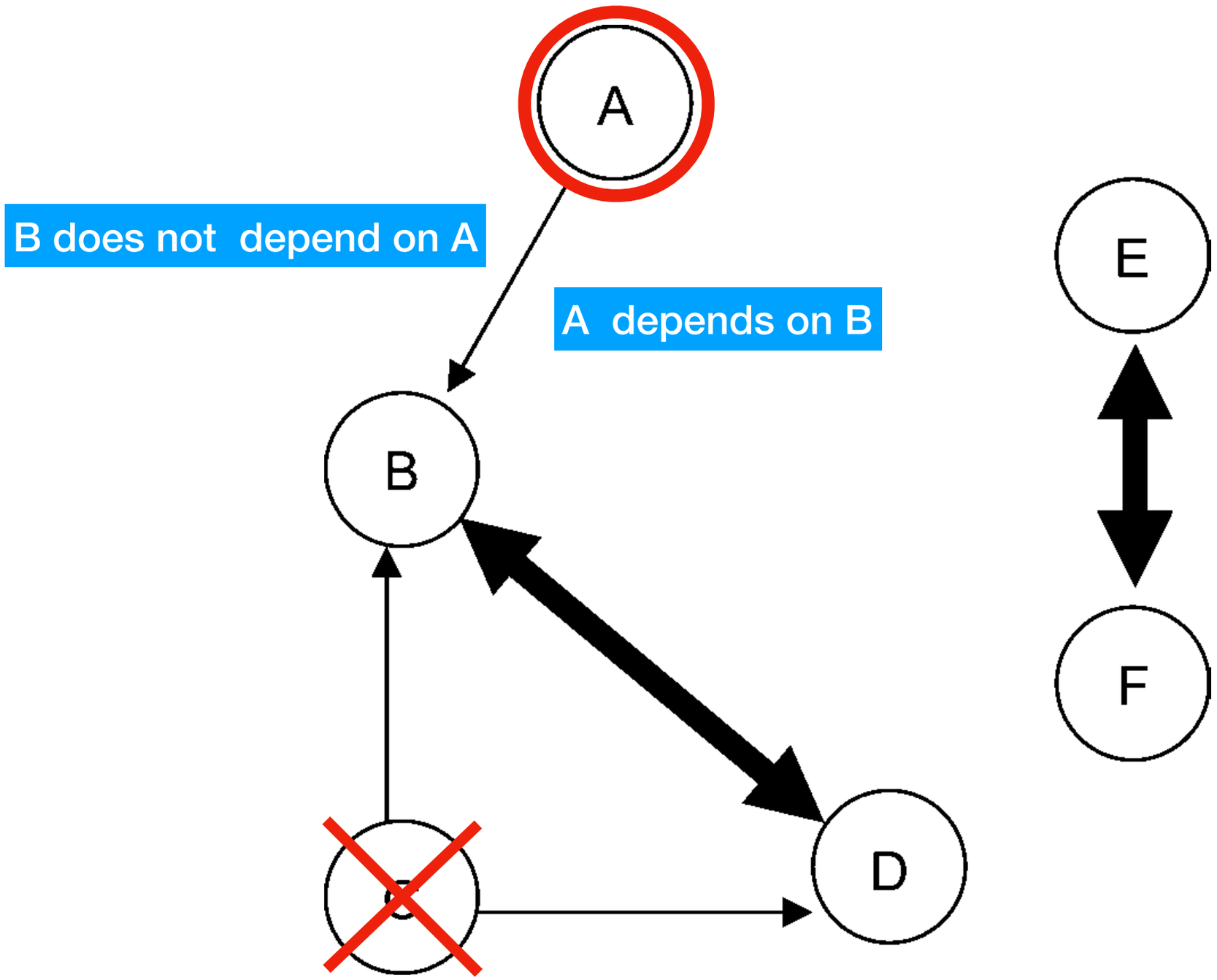


Fig. 1. A directed relation graph showing typical relations of the species.

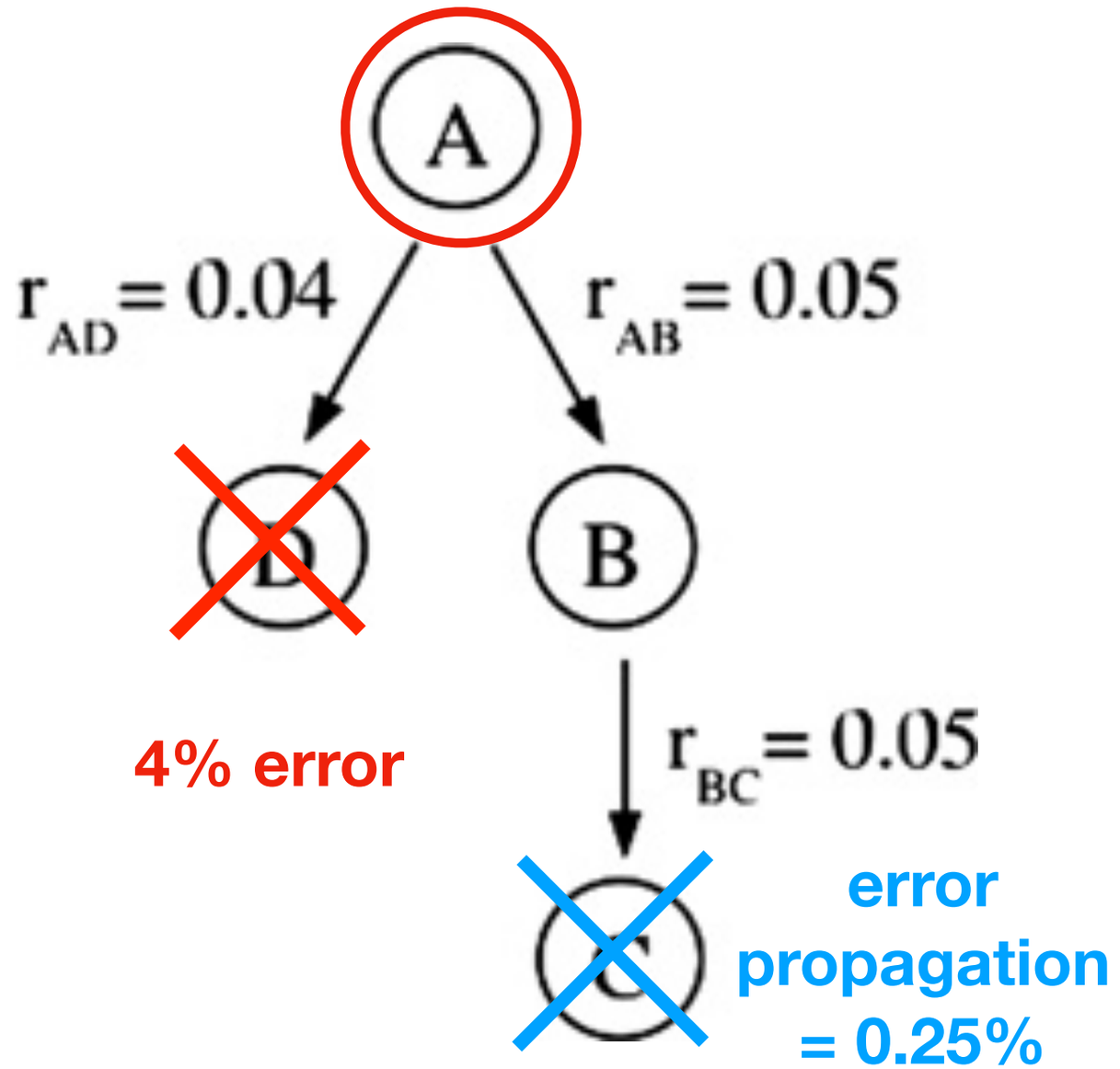
Elimination of redundant species and reactions

Direct Relation Graph with Error Propagation (DRGEP)

P. Pepiot-Desjardin and H. Pitsch. An efficient error-propagation-based reduction method for large chemical kinetic mechanisms. Comb & Flame (2008)

$$r_{AB,p} = \min_{1<i<n-1} r^*_{S_i S_{i+1}}$$

$$r^*_{AB,p} = \prod_{1<i<n-1} r^*_{S_i S_{i+1}}$$



Two main steps

Detailed mechanism

remove redundant species and reactions

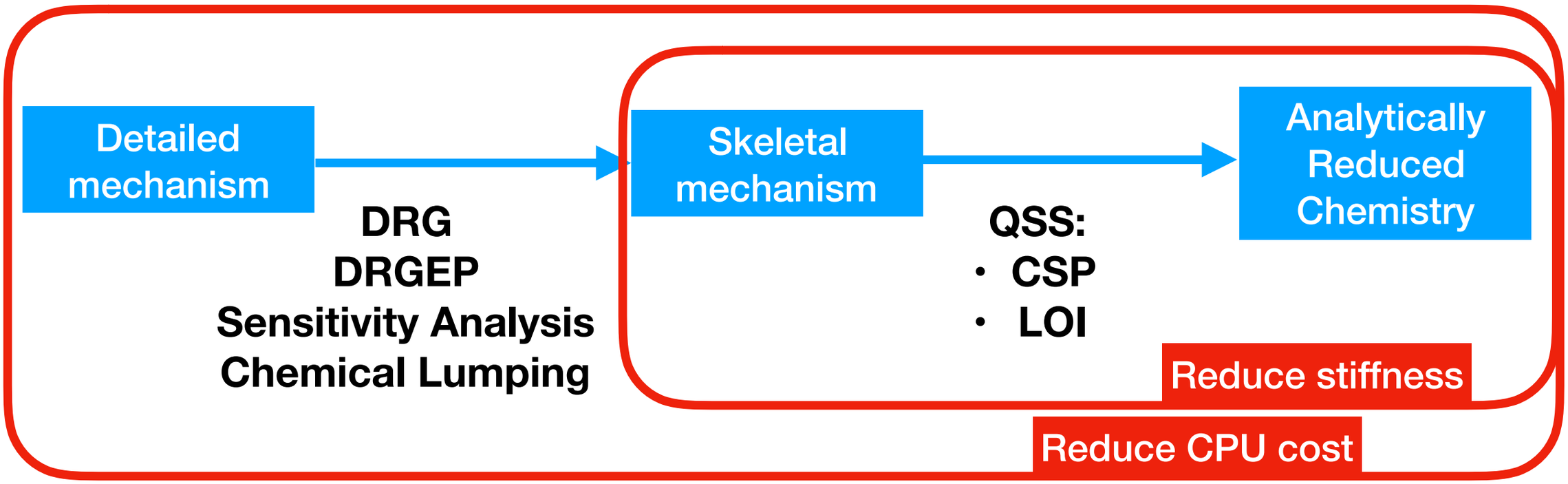
Skeletal mechanism

find analytical relations between species

Analytically reduced mechanism

Automatic ARC generation

Methods are successively applied and combined in practice

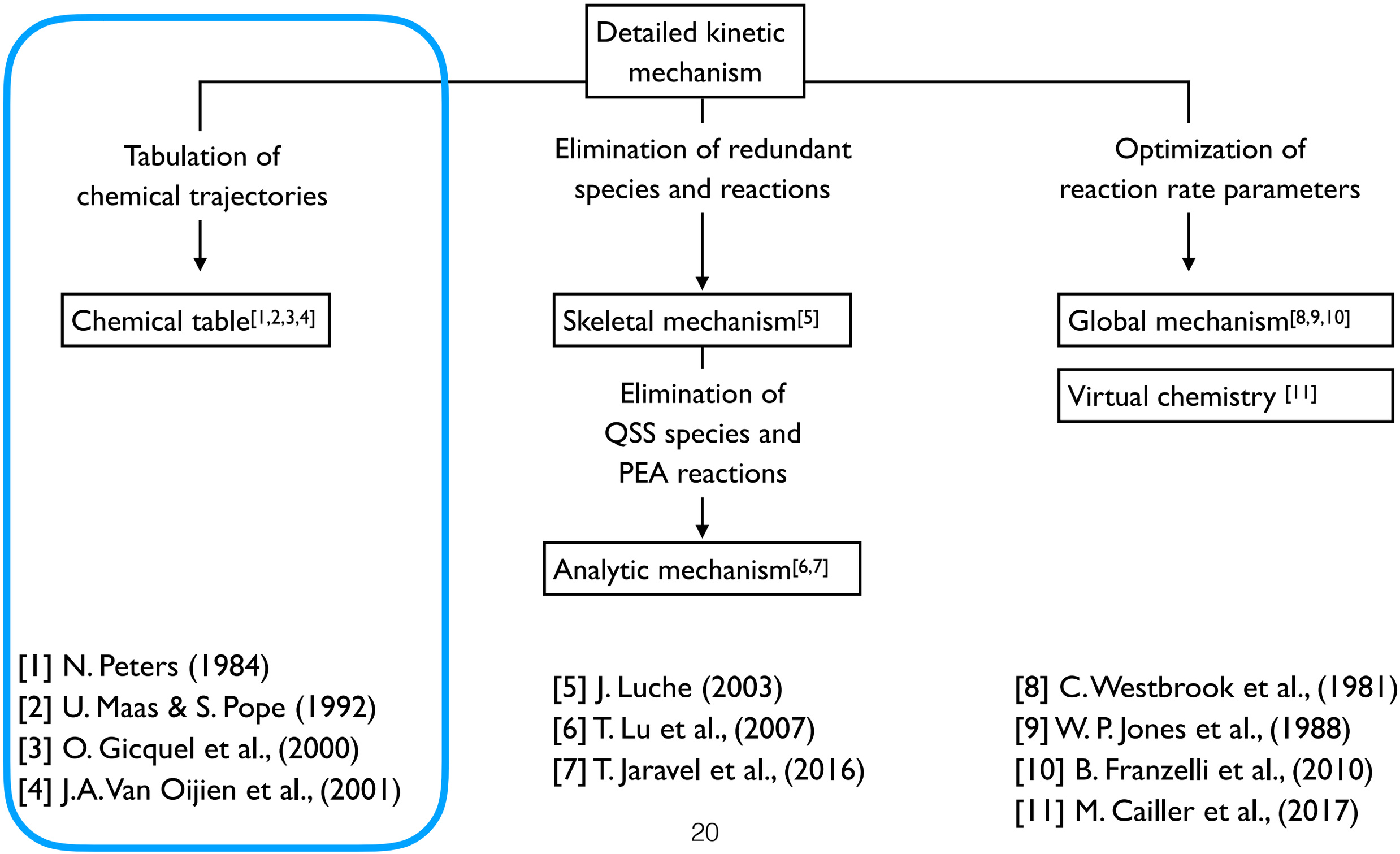


	Strategy I (Lu & Law, 2008*a*)	**Strategy II** : YARC (Pepiot, 2008)	**Strategy III** : KINALC (Turányi, 1990*a*)
STEP I	DRG(X/ASA)	DRGEP Chemical Lumping	Jacobian investigations (remove species) PCA (remove reactions)
STEP II	QSS via CSP pointers	QSS via LOI	QSS via error estimation (Turányi *et al.*, 1993)
Examples	(Lu & Law, 2008*a*,*b*) and more online: (Lu, 2008)	(Pepiot-Desjardins & Pitsch, 2008) (Pepiot & Pitsch, 2008; Jaravel, 2016)	(Tomlin *et al.*, 1992) and more online: (Turányi, 1981)

From Anne Felden PhD thesis (2017)

Kinetic scheme reduction strategies for combustion modeling

B. Fiorina, D. Veynante and S. Candel. Modeling Combustion Chemistry in Large Eddy Simulation of Turbulent Flames. Flow Turb. and Combustion. Vol 94, Issue 1, pp3-42 (2015).



[1] N. Peters (1984)
[2] U. Maas & S. Pope (1992)
[3] O. Gicquel et al., (2000)
[4] J.A. Van Oijien et al., (2001)

[5] J. Luche (2003)
[6] T. Lu et al., (2007)
[7] T. Jaravel et al., (2016)

[8] C. Westbrook et al., (1981)
[9] W. P. Jones et al., (1988)
[10] B. Franzelli et al., (2010)
[11] M. Cailler et al., (2017)

Tabulated chemistry

B. Fiorina, D. Veynante and S. Candel. Modeling Combustion Chemistry in Large Eddy Simulation of Turbulent Flames. Flow Turb. and Combustion. Vol 94, Issue 1, pp3-42 (2015).

Thermochemical state

$$\varphi = \mathcal{F}(p, T, Y_1, Y_2, \ldots, Y_{n_{sp}})$$
$$\varphi = \mathcal{G}(p, h, Y_1, Y_2, \ldots, Y_{n_{sp}})$$
$$\varphi = \mathcal{H}(\rho, e, Y_1, Y_2, \ldots, Y_{n_{sp}})$$

Reduced chemistry

$n_{red} < n_{sp}$

tabulated chemistry

$$\varphi = \mathcal{F}'(p, T, \psi_1, \psi_2, \ldots, \psi_n)$$
$$\varphi = \mathcal{G}'(p, h, \psi_1, \psi_2, \ldots, \psi_n)$$
$$\varphi = \mathcal{H}'(\rho, e, \psi_1, \psi_2, \ldots, \psi_n)$$

$n << n_{sp}$

Tabulated chemistry

B. Fiorina, D. Veynante and S. Candel. Modeling Combustion Chemistry in Large Eddy Simulation of Turbulent Flames. Flow Turb. and Combustion. Vol 94, Issue 1, pp3-42 (2015).

No analytical expression: defined in the discrete form of a database

$$\varphi = \mathcal{F}'(p, T, \psi_1, \psi_2, \ldots, \psi_n)$$
$$\varphi = \mathcal{G}'(p, h, \psi_1, \psi_2, \ldots, \psi_n)$$
$$\varphi = \mathcal{H}'(\rho, e, \psi_1, \psi_2, \ldots, \psi_n)$$

$n << n_{sp}$

- **Tabulate thermochemical states in a look-up table during a pre-processing step**

- **Key issues**

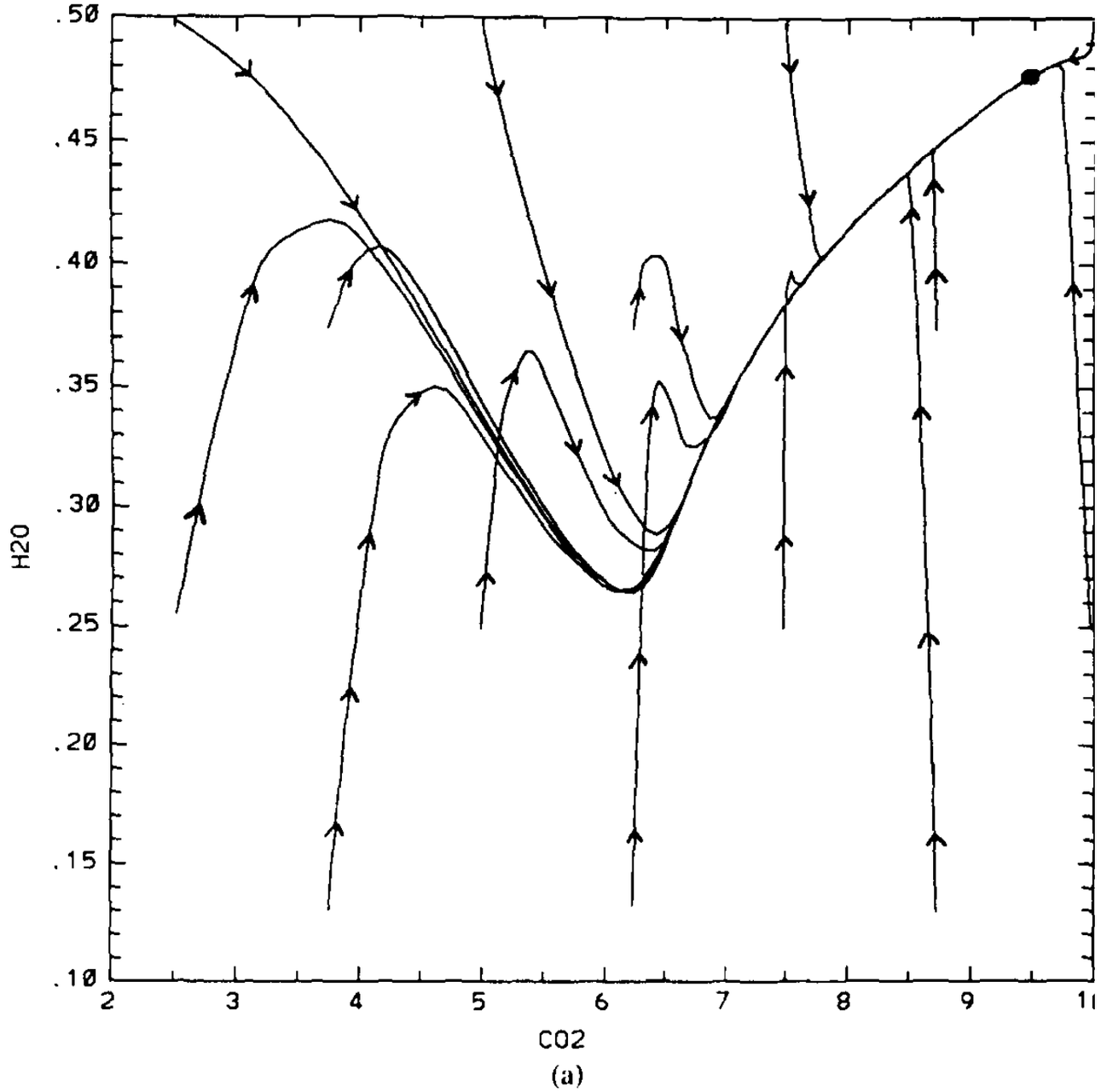
  - identification of the reduced subspace

  - building up of the chemical table (or manifold)

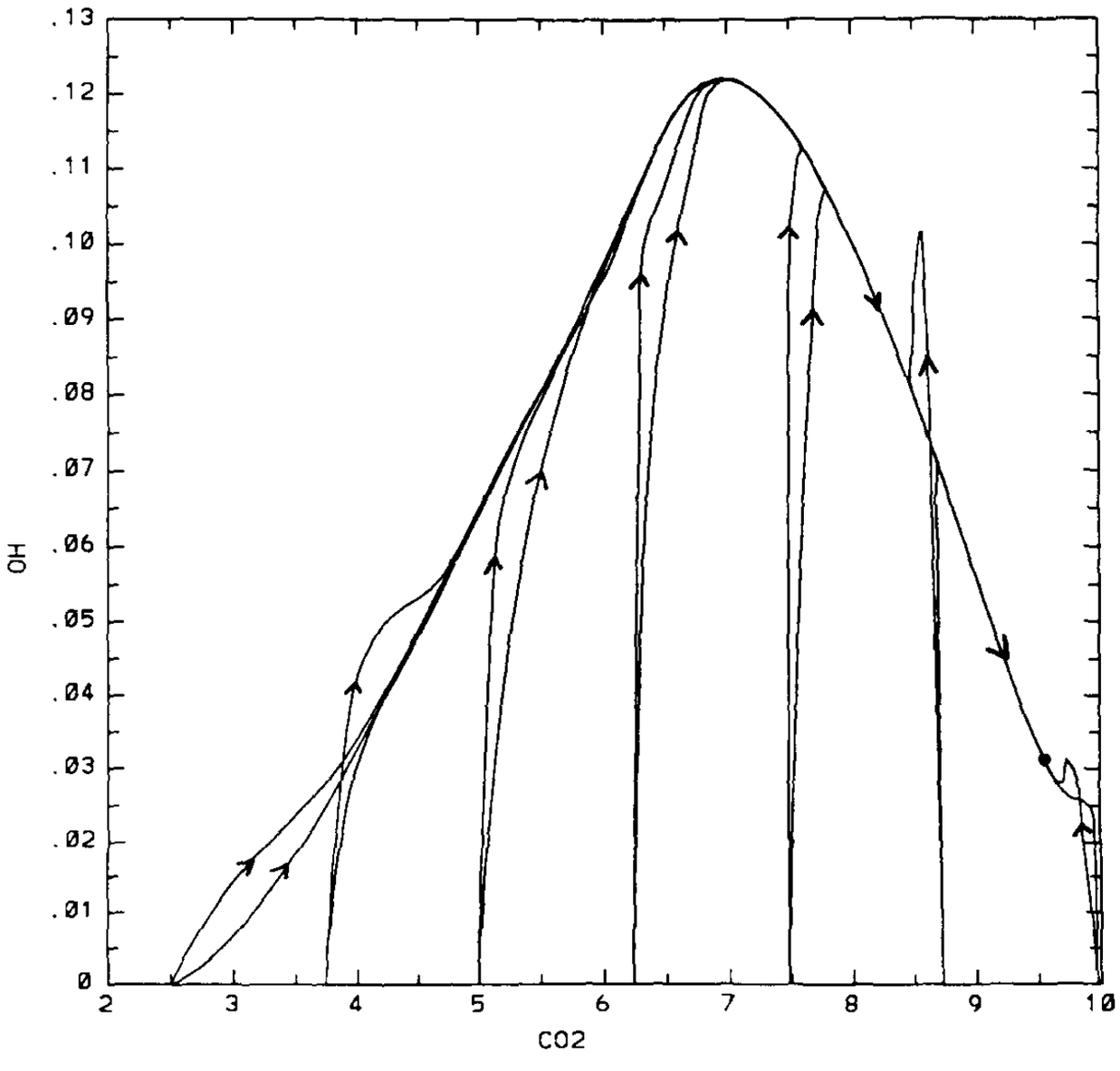
  - size of the table

Tabulated chemistry
ILDM method

Maas U. and Pope S. (1992) " Simplifying chemical-kinetics - Intrinsic Low-Dimensional Manifolds in composition space" in Combustion and Flame, 88-3-4, pp 239-264



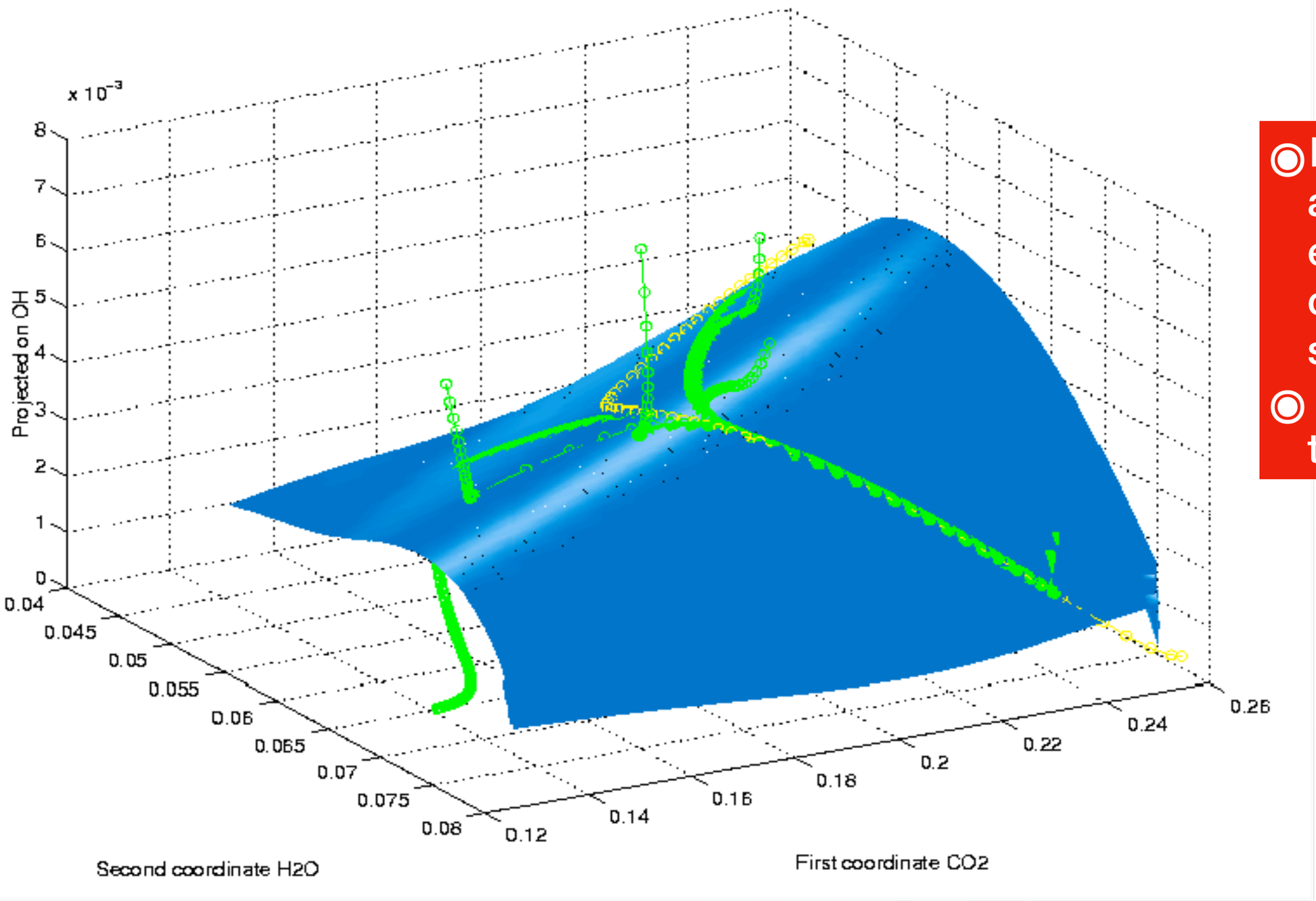
(a)



(b)

Tabulated chemistry
ILDM method

Maas U. and Pope S. (1992) " Simplifying chemical-kinetics - Intrinsic Low-Dimensional Manifolds in composition space" in Combustion and Flame, 88-3-4, pp 239-264



- Reduced attractive surface exist in the chemical phase subspace
- How to identify these surfaces ?

Tabulated chemistry
Flamelet-based methods

Physical approach: build up the chemical database from simple combustion elements computed with detailed chemistry

- Using non-premixed flamelets

  Flamelet model:

  Peters, N.: Prog. Energy Combustion. Sci. 10, 319 (1984)

  Flamelet Progress Variable (FPV):

  Pierce, C., Moin, P.: J. Fluid Mech. 504, 73 (2004)

- Using premixed flamelets

  Flamelet Prolongation of ILDM (FPI):

  Gicquel, O., Darabiha, N., Thévenin, D.: Proc. Combust. Inst. 28, 1901 (2000)

  Flamelet Generated Manifold (FGM):

  van Oijen, J.A., Lammers, F.A., de Goey, L.P.H.: Combust. Flame 127(3), 2124 (2001)

Anticipated by Bradley et al. Bradley al. Combust. Flame 71, 109 (1988)

- Using 0-D reactors

  Tabulated Kinetic of Ignition:

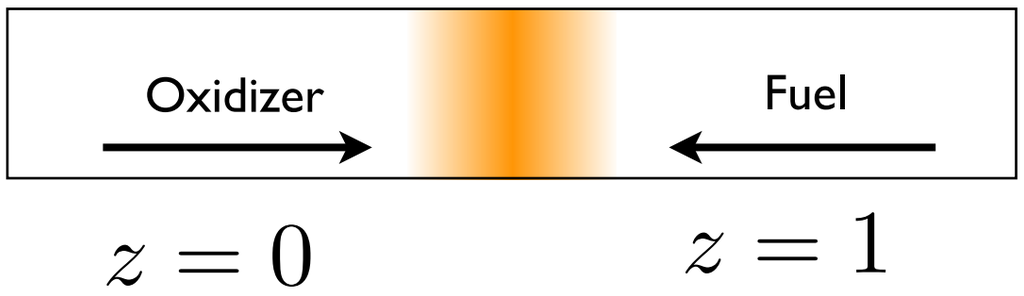
  Colin, O., da Cruz, A.P., Jay, S.: Proc. Combust. Inst. 30(2), 2649 (2005)

Tabulated chemistry
Flamelet based methods
Main variables

- <u>Non-premixed flame</u>

  Mixture fraction z

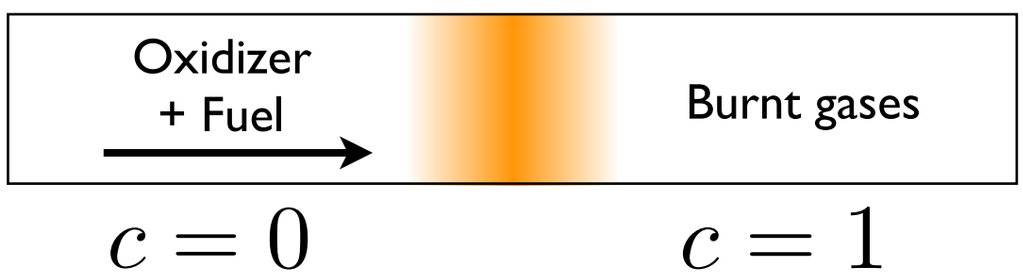
$$z = \frac{Y_z - Y_{z_o}}{Y_{z_f} - Y_{z_o}}$$



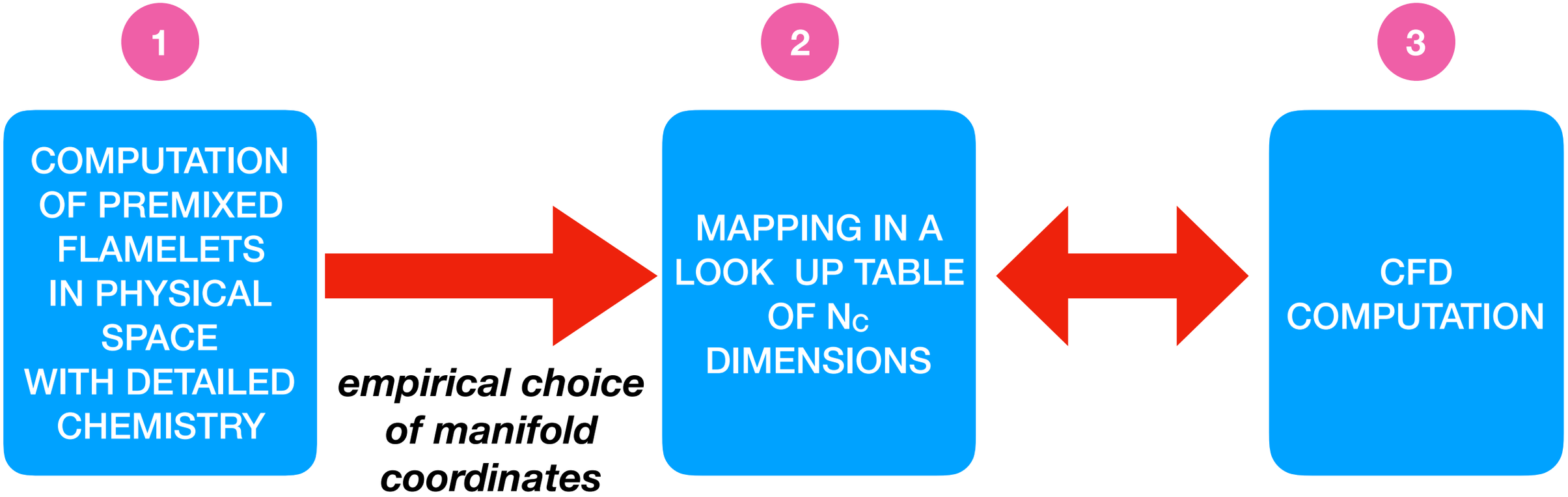
- <u>Premixed flame</u>

  Progress variable c

$$c = \frac{Y_c - Y_{c_f}(Y_z)}{Y_{c_b}(Y_z) - Y_{c_f}(Y_z)}$$

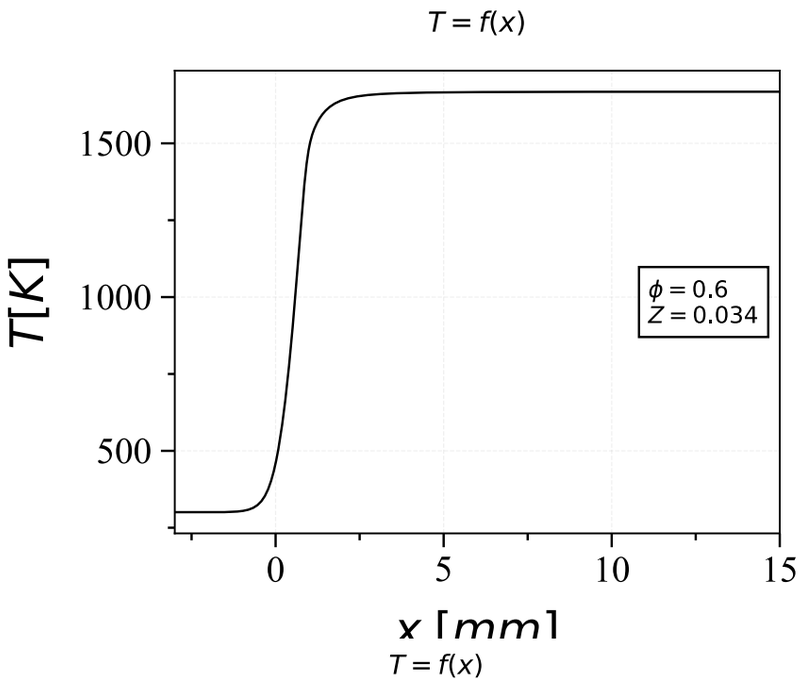


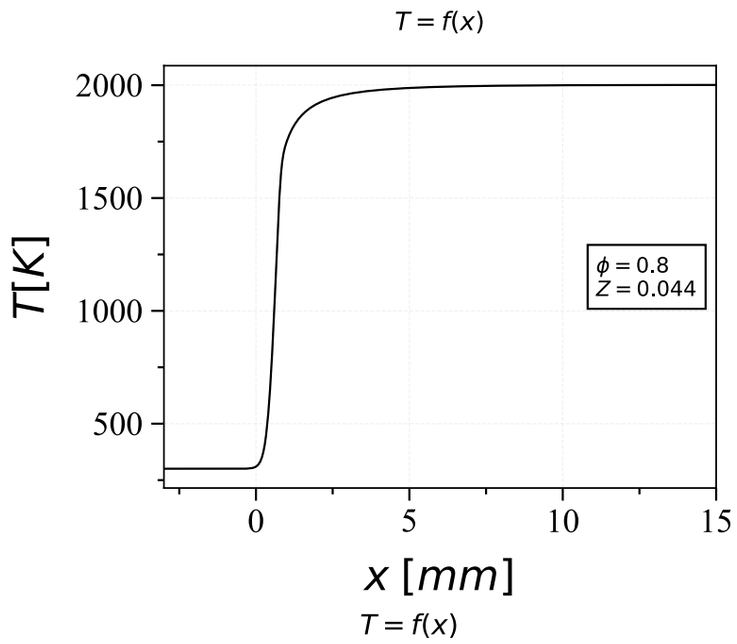
Tabulated chemistry
Example: FPI & FGM

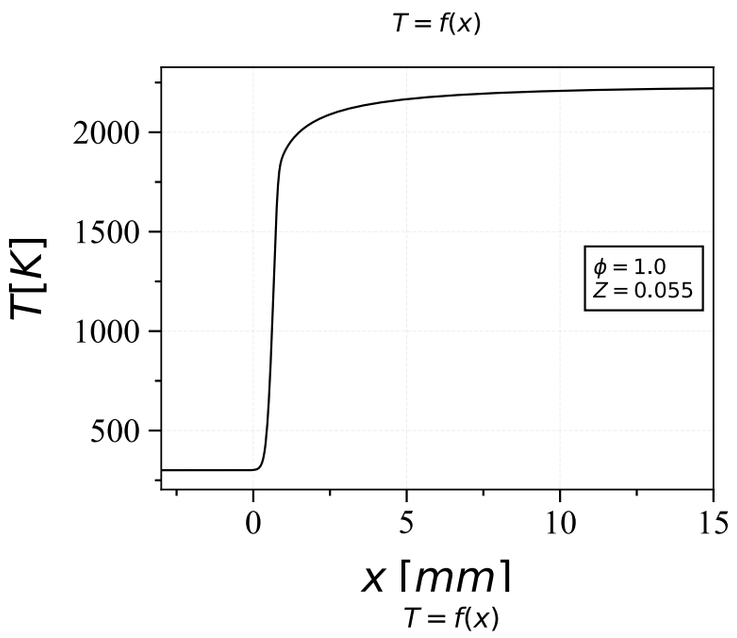


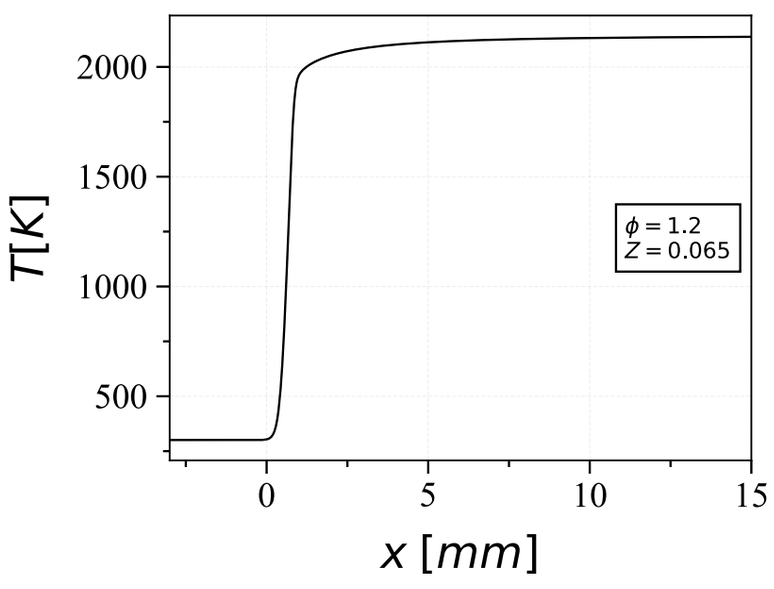
1

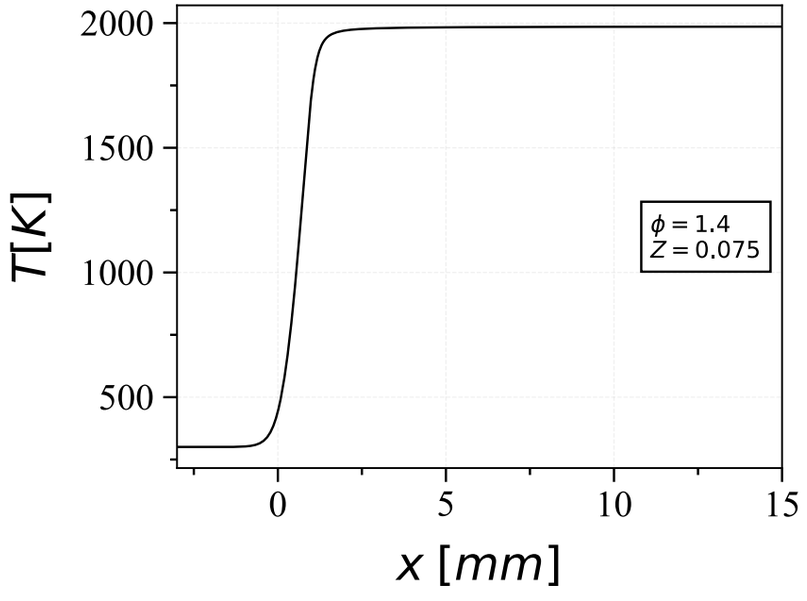
COMPUTATION OF PREMIXED FLAMELETS IN PHYSICAL SPACE WITH DETAILED CHEMISTRY

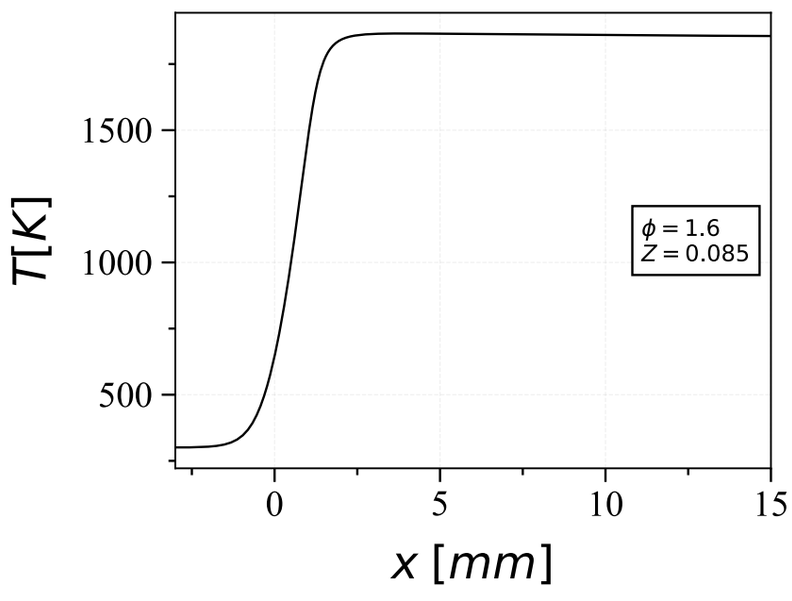








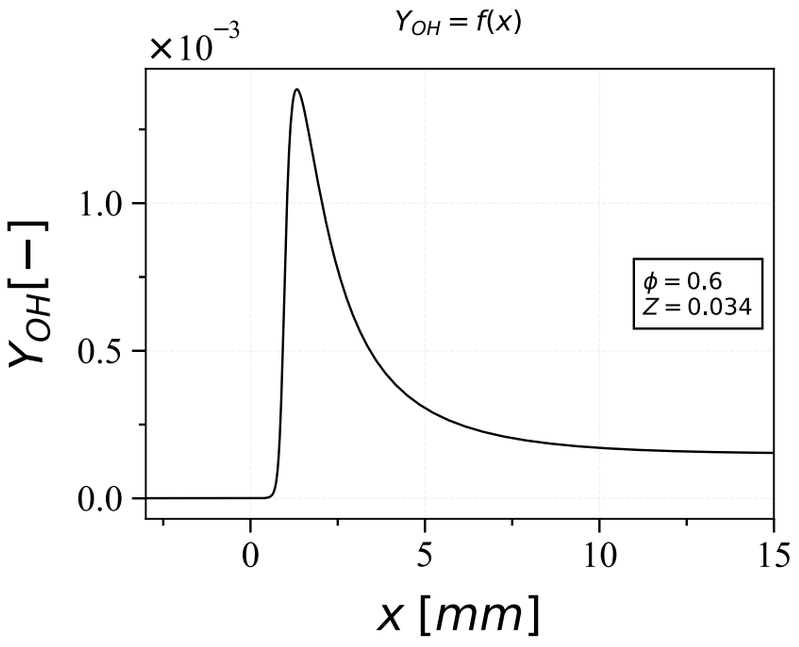


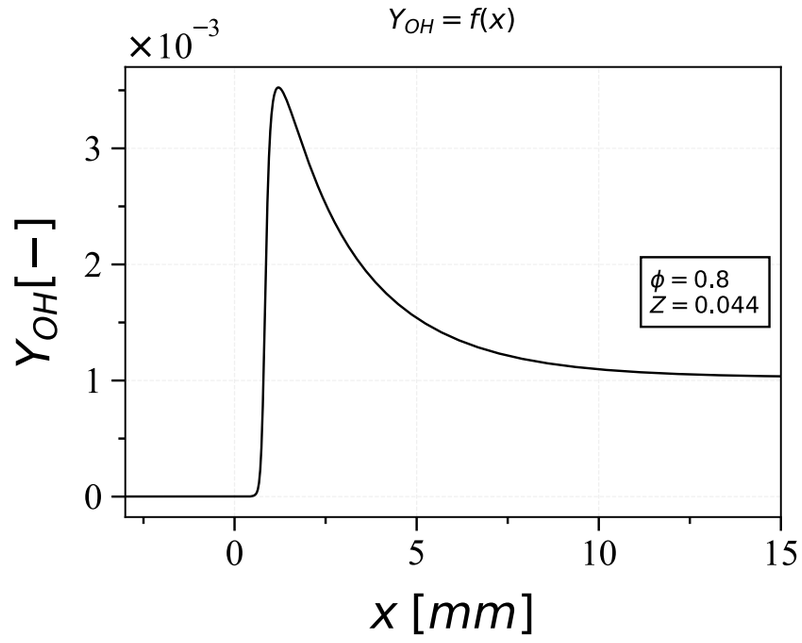


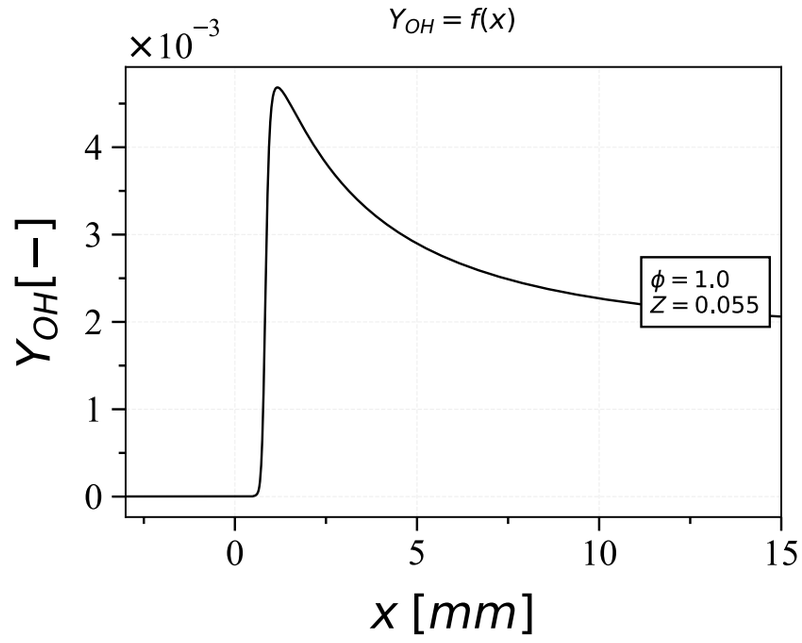
REGATH Solver (N. Darabiha 1992) + GRI Mechanism (53 species, 323 reactions

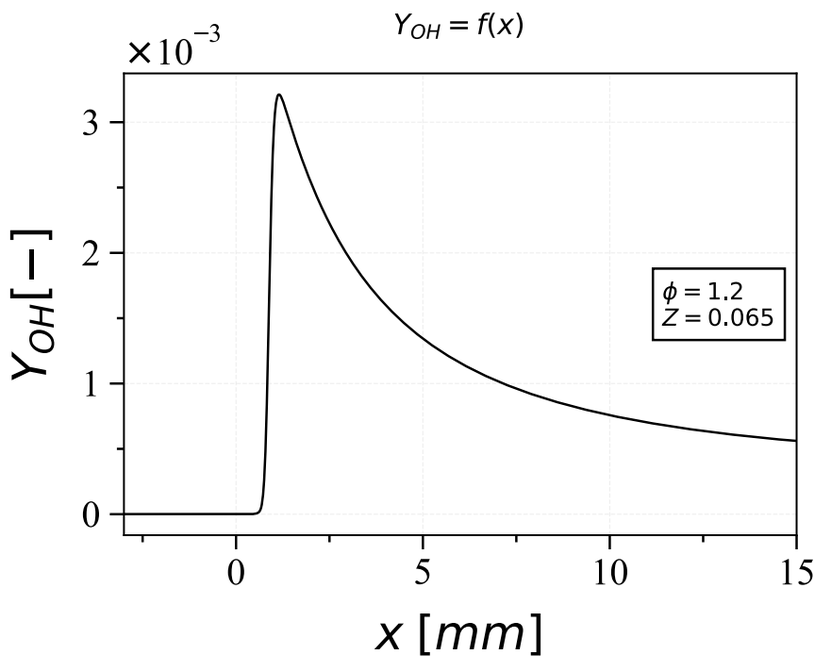
1

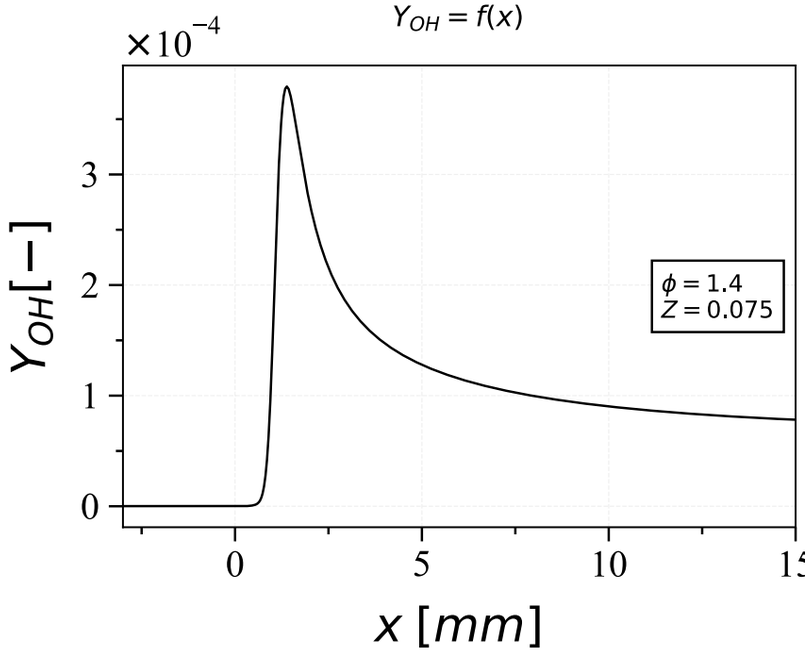
COMPUTATION OF PREMIXED FLAMELETS IN PHYSICAL SPACE WITH DETAILED CHEMISTRY

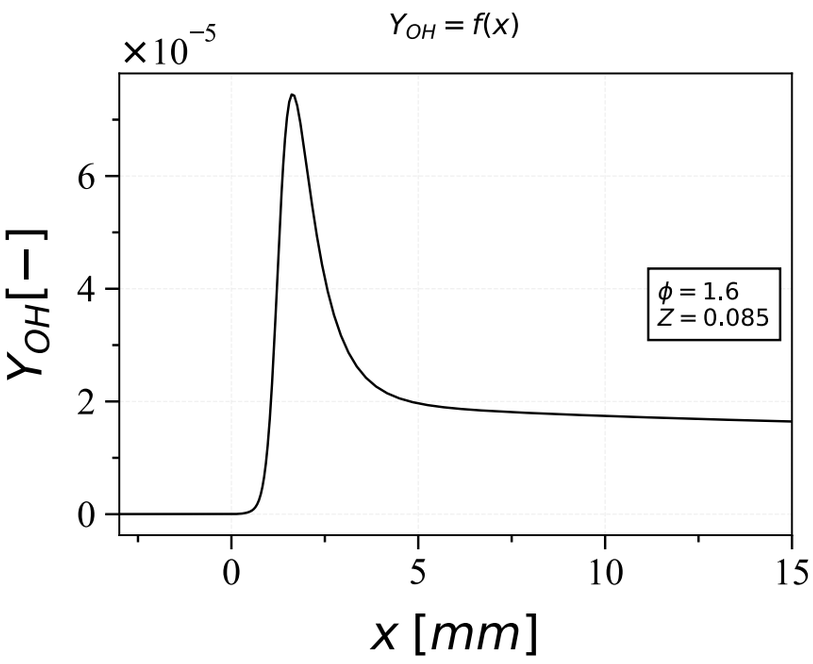








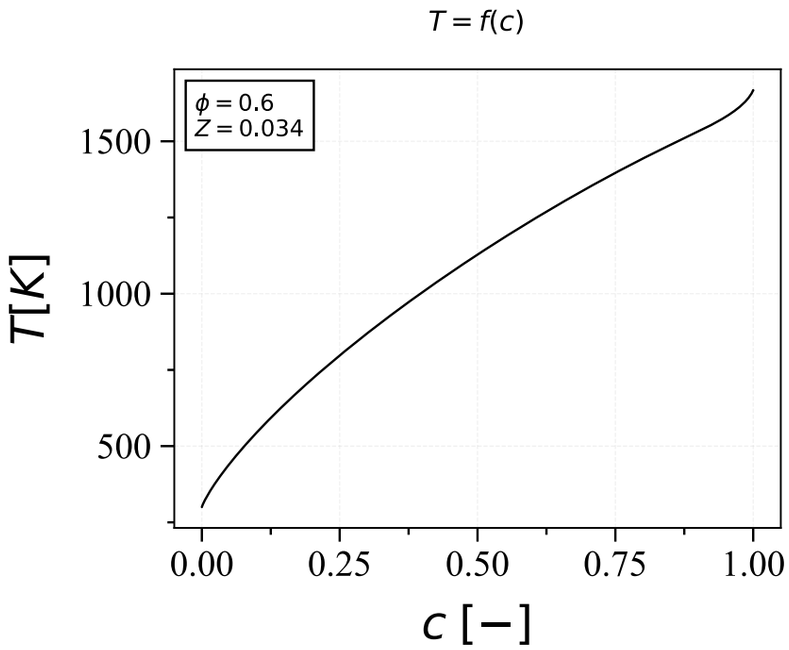


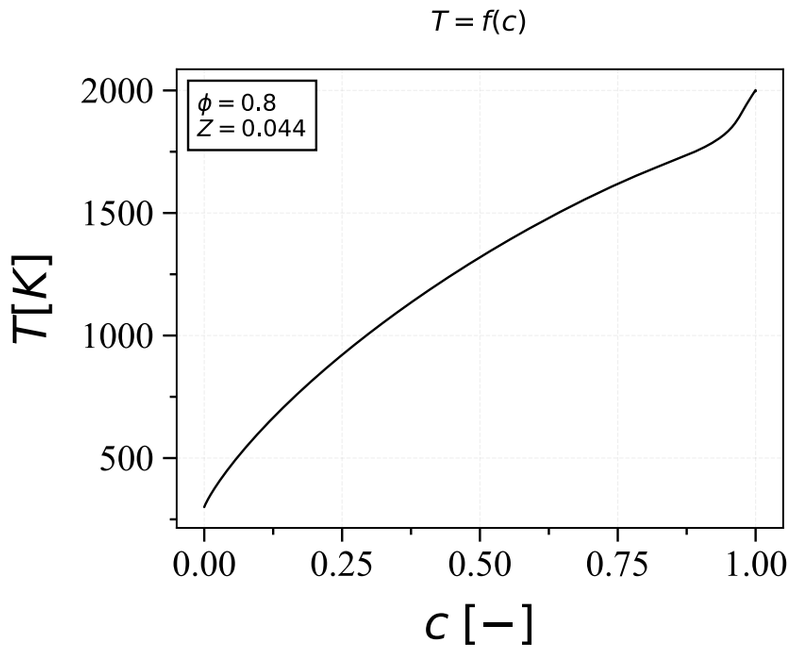


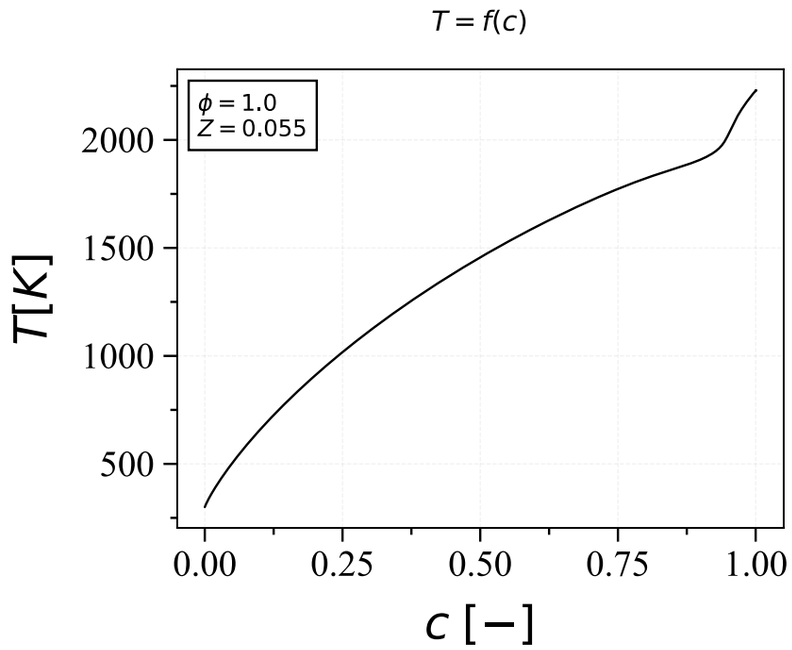
REGATH Solver (N. Darabiha 1992) + GRI Mechanism (53 species, 323 reactions

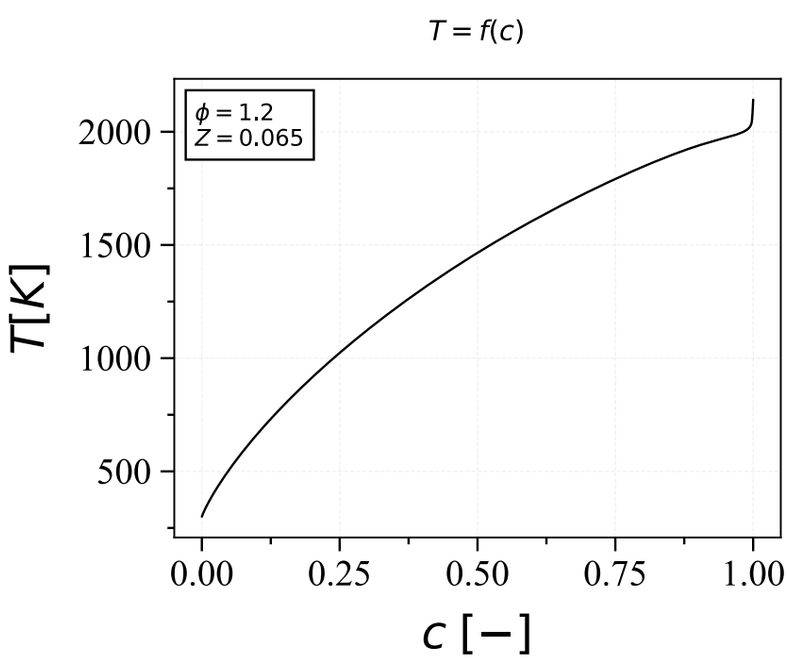
2

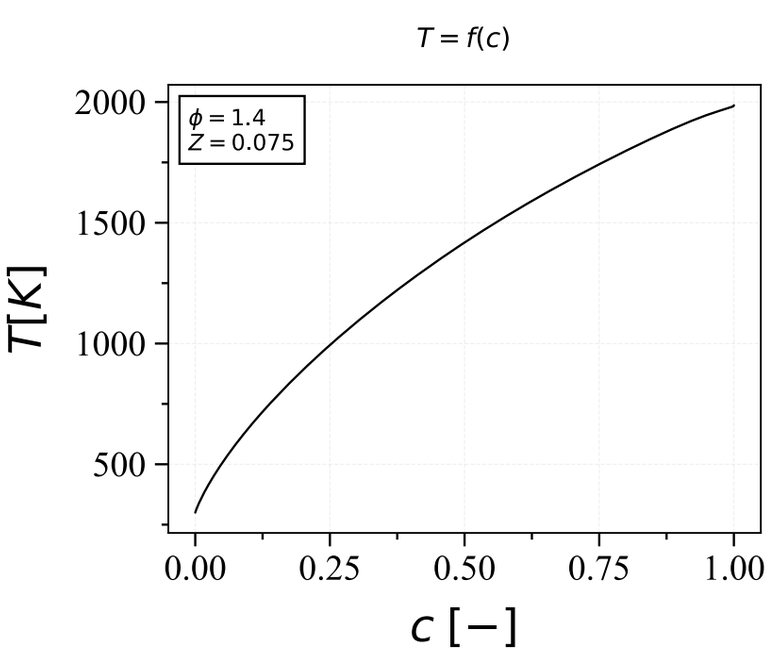
MAPPING IN A LOOK UP TABLE OF N_C DIMENSIONS

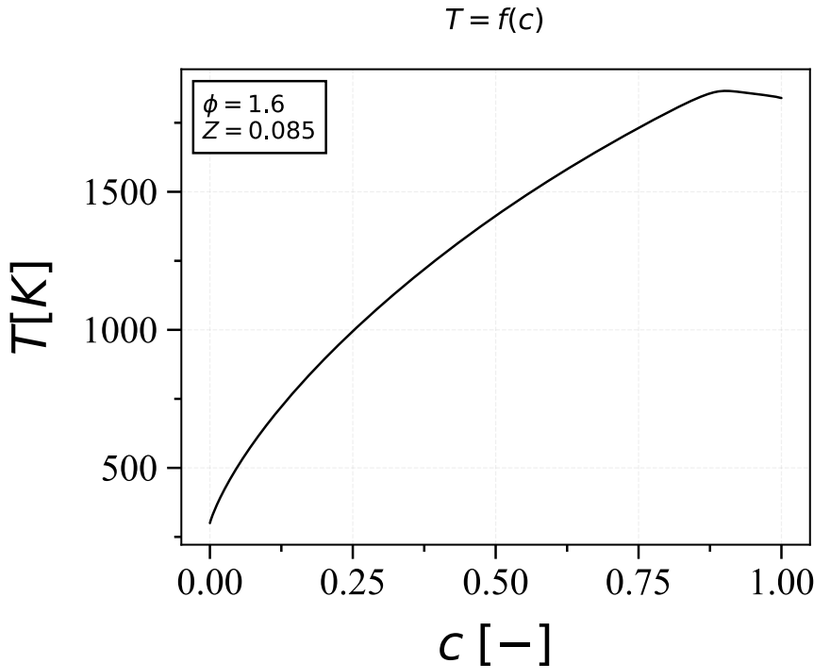






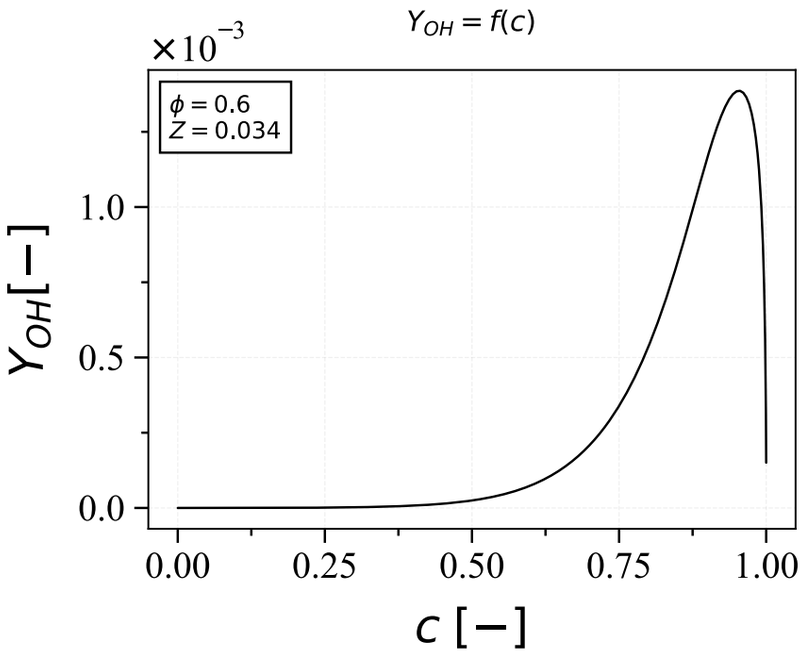


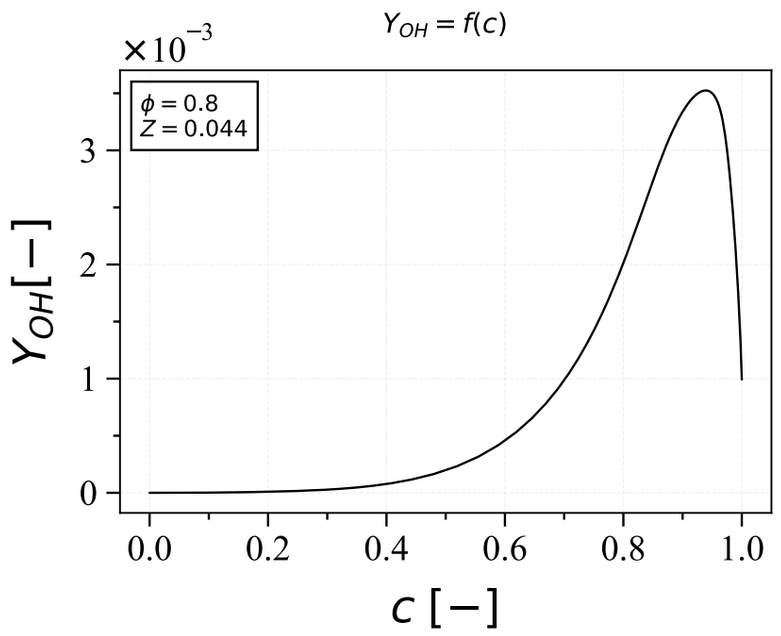


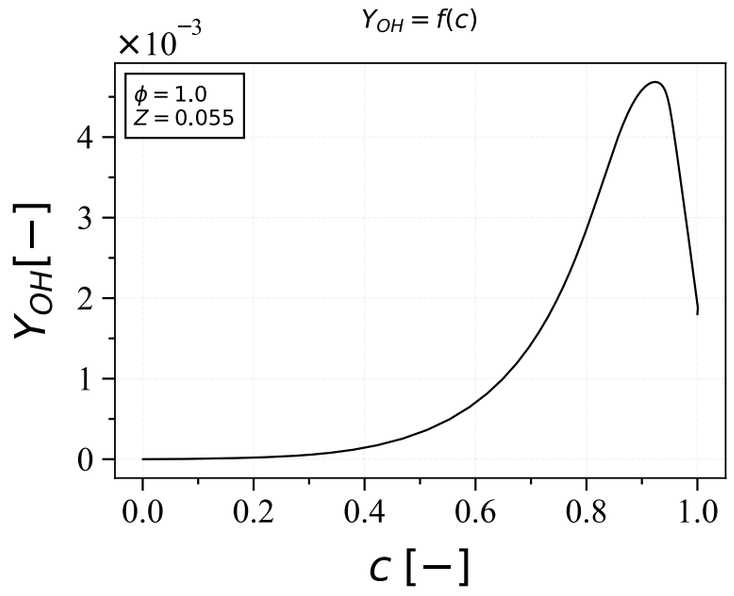


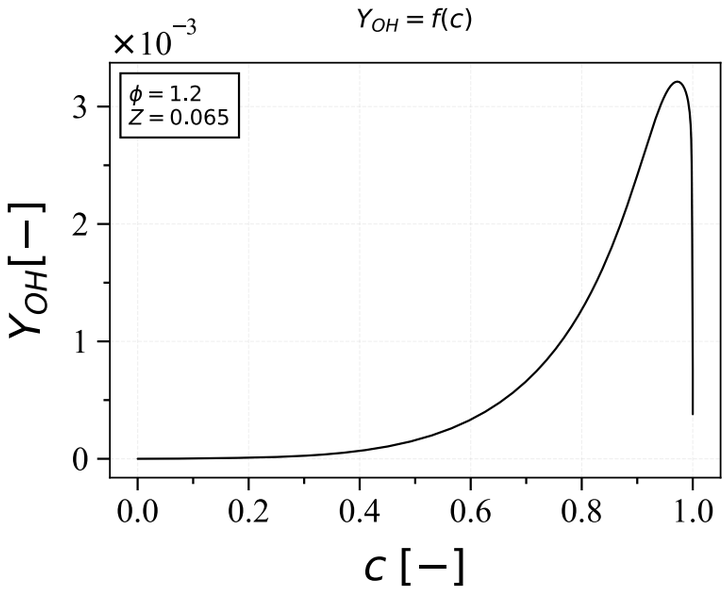
2

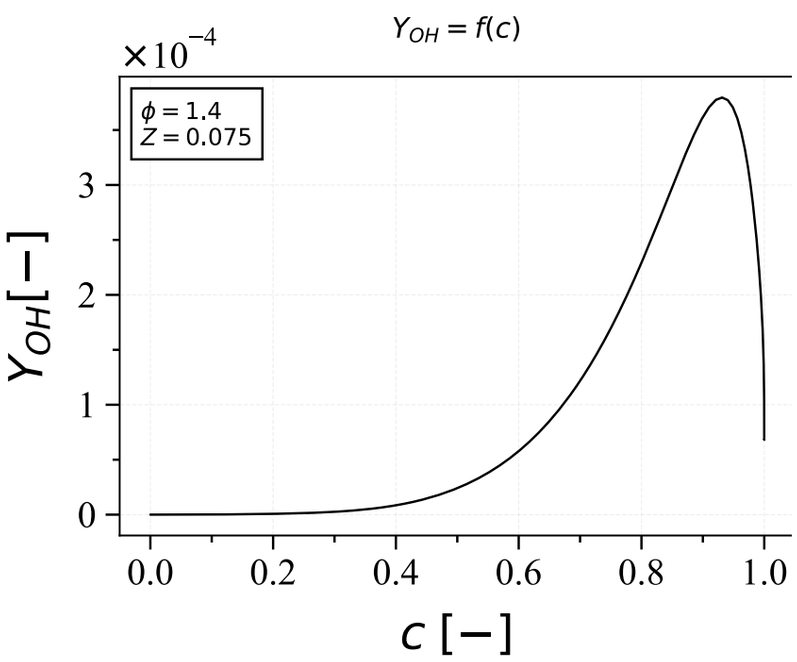
MAPPING IN A LOOK UP TABLE OF N_C DIMENSIONS

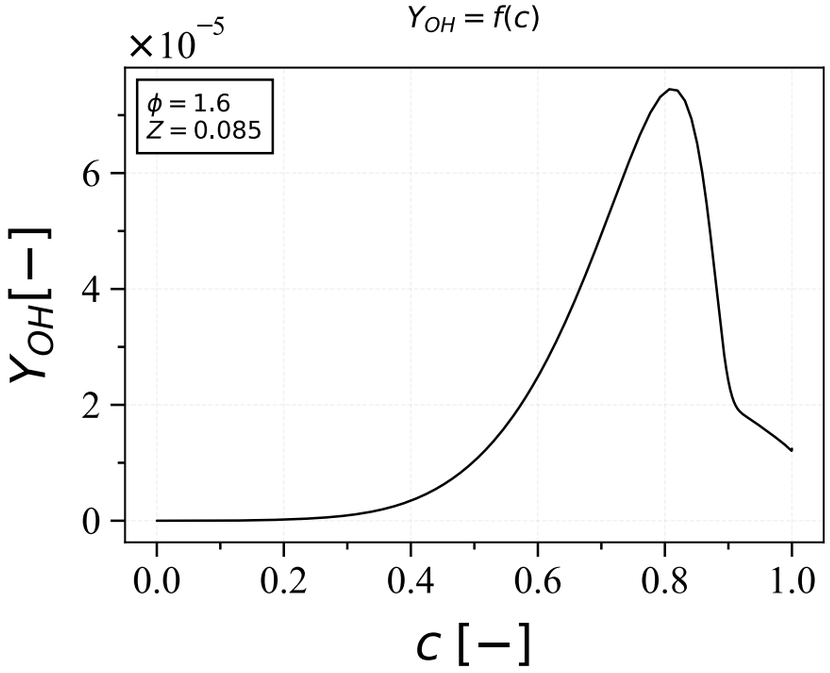












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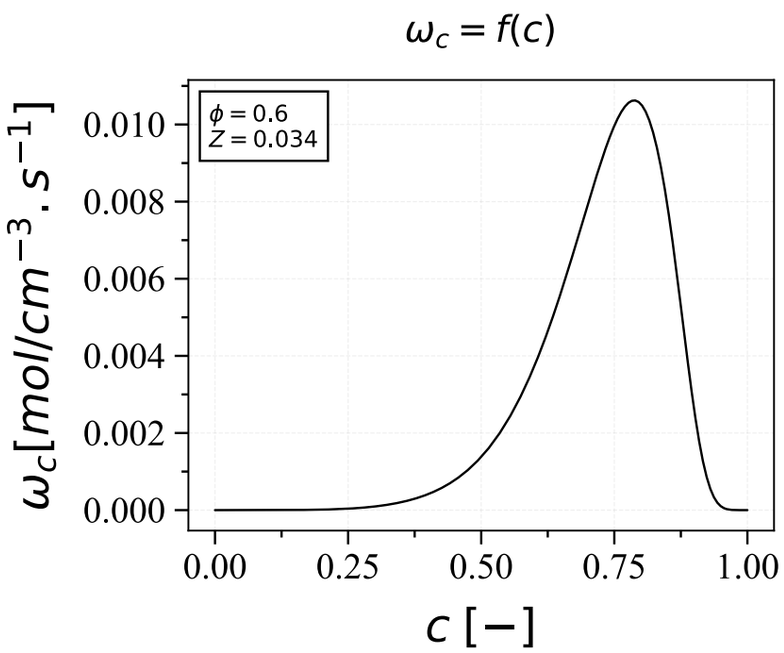

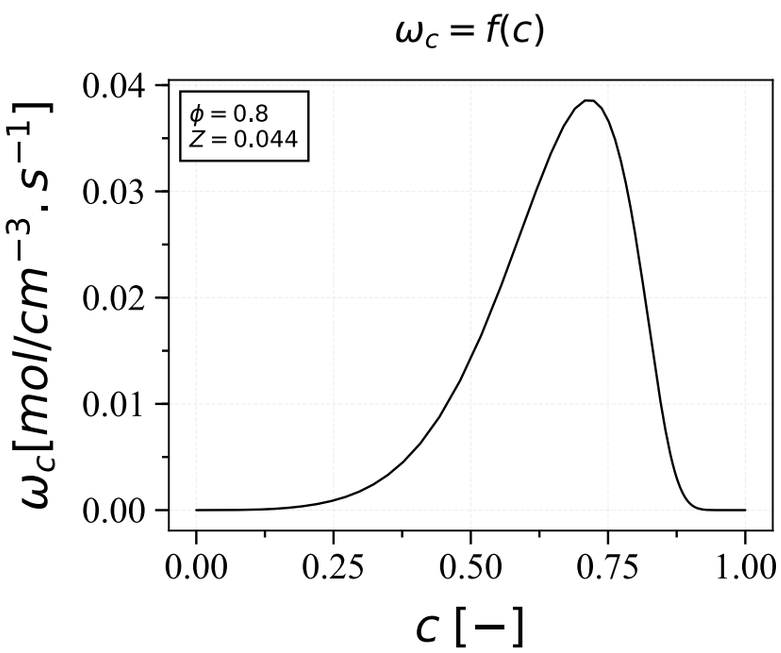

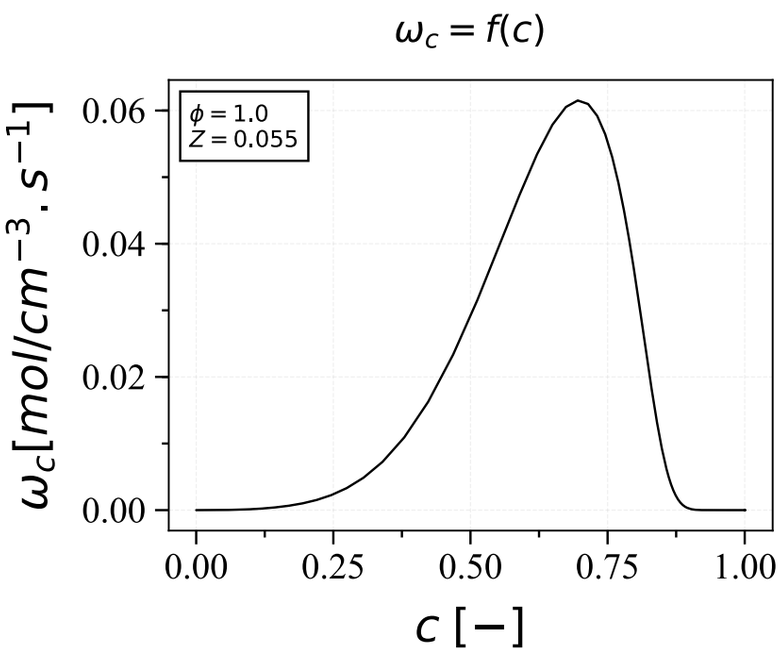

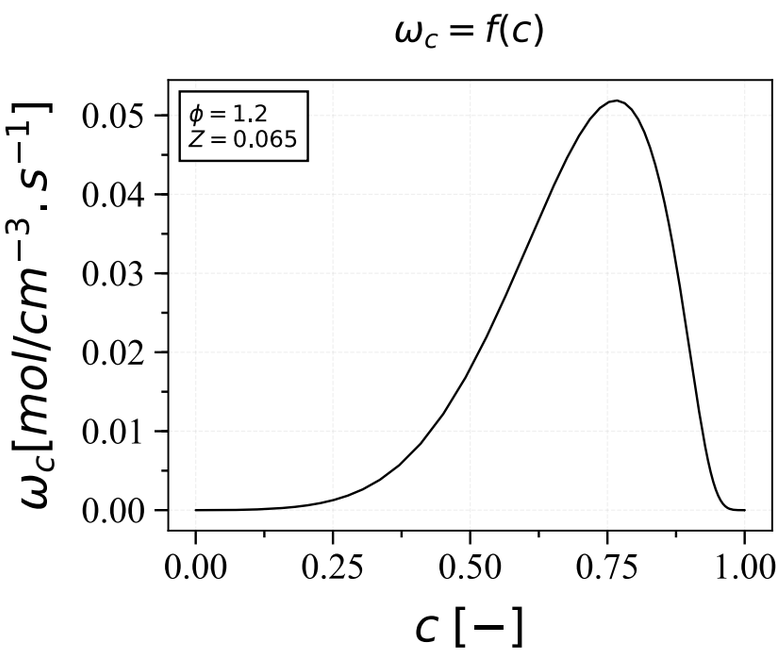

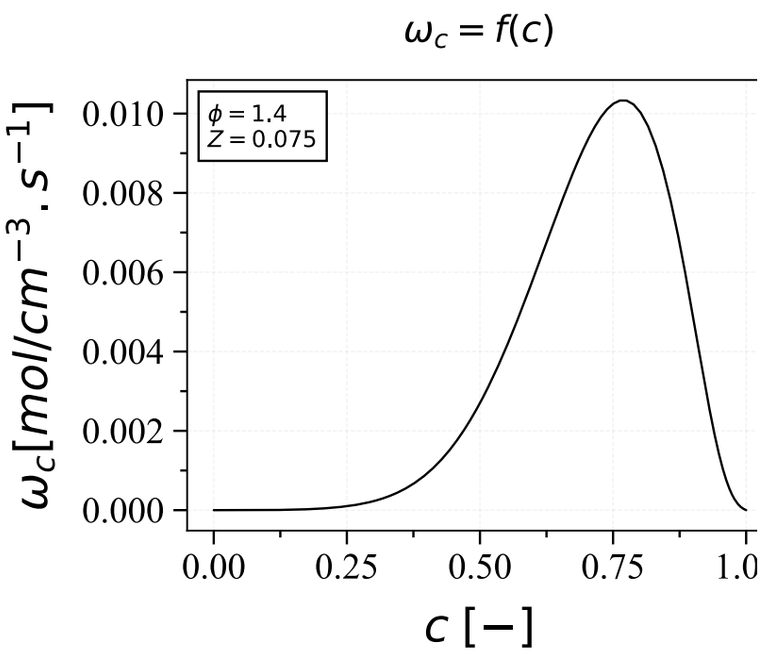

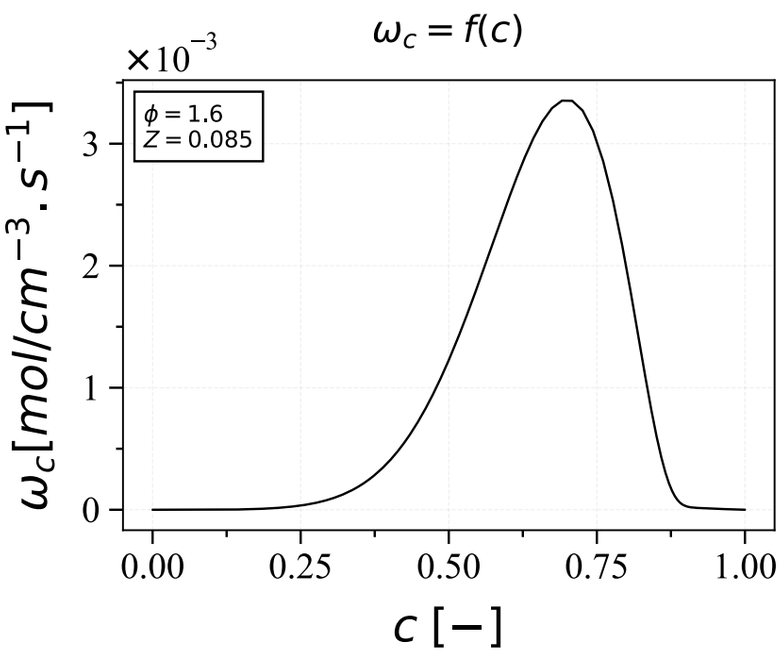

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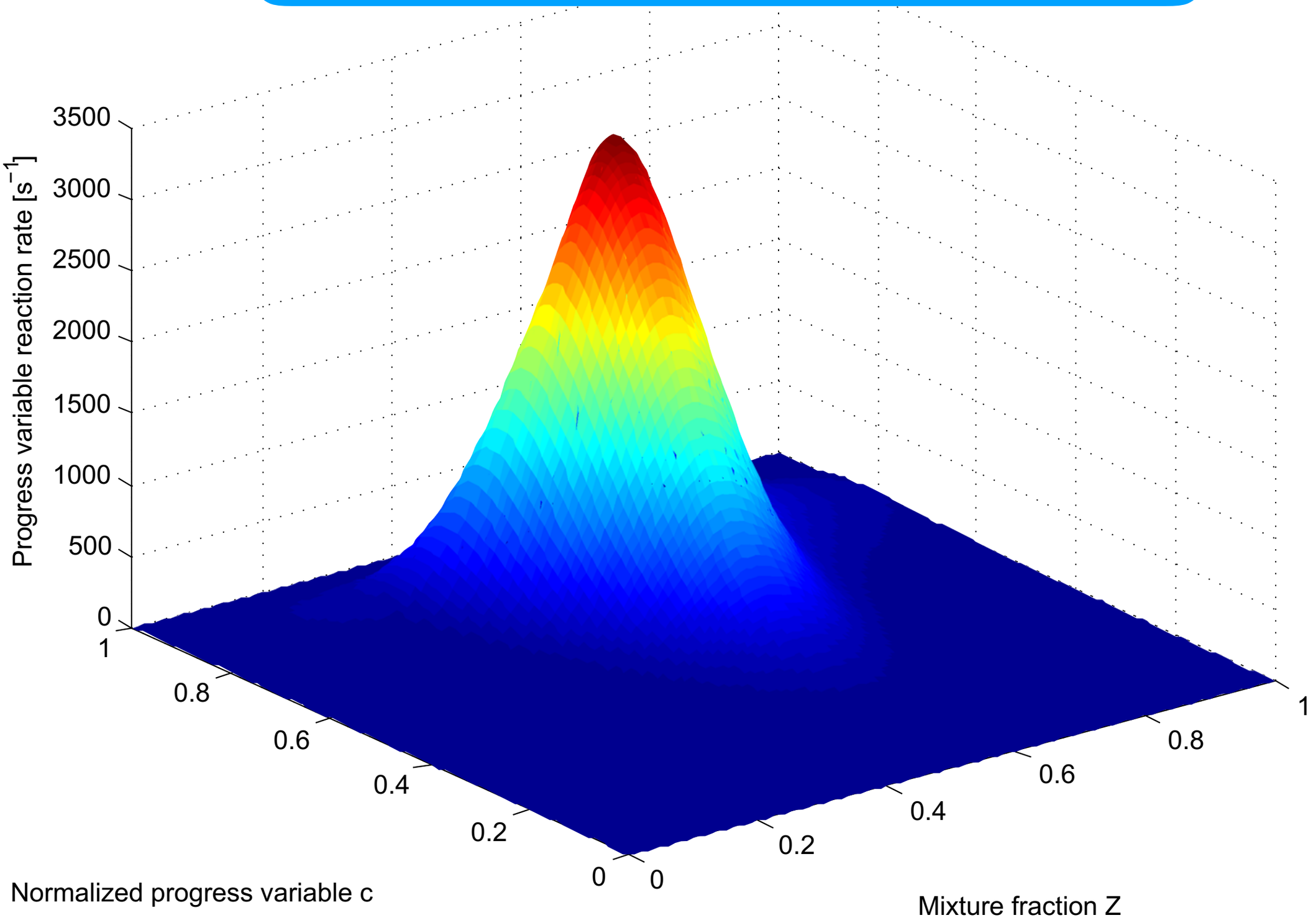
MAPPING IN A LOOK UP TABLE OF N_C DIMENSIONS



3

CFD COMPUTATION

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i) = 0$$

$$\frac{\partial \rho u_j}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i u_j) = -\frac{\partial P}{\partial x_j} + \frac{\partial \tau_{ij}}{\partial x_i} \qquad P/\rho = rT$$

$$\frac{\partial \rho E}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i E) = \dot{\omega}_T - \frac{\partial q_i}{\partial x_i} - \frac{\partial}{\partial x_i}(P u_i) + \frac{\partial}{\partial x_j}(\tau_{ij} u_i)$$

$$\frac{\partial \rho Y_1}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i Y_1) = \frac{\partial}{\partial x_i}(\rho D_k \frac{\partial Y_1}{\partial x_i}) + \dot{\omega}_1$$

$$\cdots$$

$$\frac{\partial \rho Y_k}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i Y_k) = \frac{\partial}{\partial x_i}(\rho D_k \frac{\partial Y_k}{\partial x_i}) + \dot{\omega}_k$$

$$\cdots$$

$$\frac{\partial \rho Y_n}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i Y_n) = \frac{\partial}{\partial x_i}(\rho D_k \frac{\partial Y_n}{\partial x_i}) + \dot{\omega}_n$$

3

CFD COMPUTATION

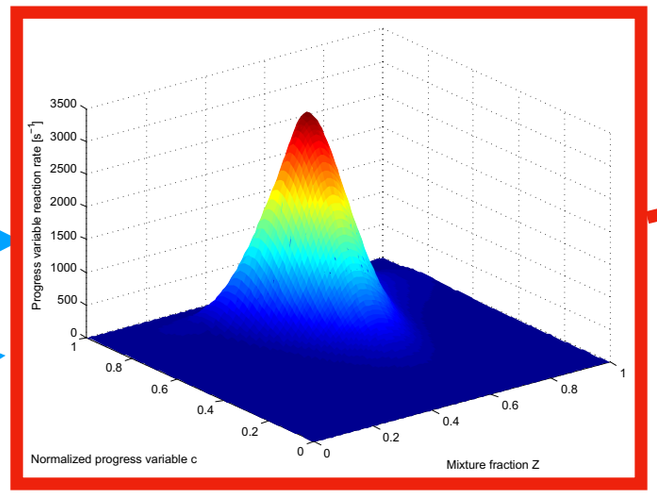
$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i) = 0$$

$$\frac{\partial \rho u_j}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i u_j) = -\frac{\partial P}{\partial x_j} + \frac{\partial \tau_{ij}}{\partial x_i} \qquad P/\rho = rT$$

$$\frac{\partial \rho E}{\partial t} + \frac{\partial}{\partial x_i}(\rho u_i E) = \dot{\omega}_T - \frac{\partial q_i}{\partial x_i} - \frac{\partial}{\partial x_i}(P u_i) + \frac{\partial}{\partial x_j}(\tau_{ij} u_i)$$

$$\frac{\partial \rho z}{\partial t} + \nabla.(\rho \mathbf{u} z) = \nabla.(\rho D \nabla z)$$

$$\frac{\partial \rho Y_c}{\partial t} + \nabla.(\rho \mathbf{u} Y_c) = \nabla.(\rho D \nabla Y_c) + W_c \dot{\omega}_c$$

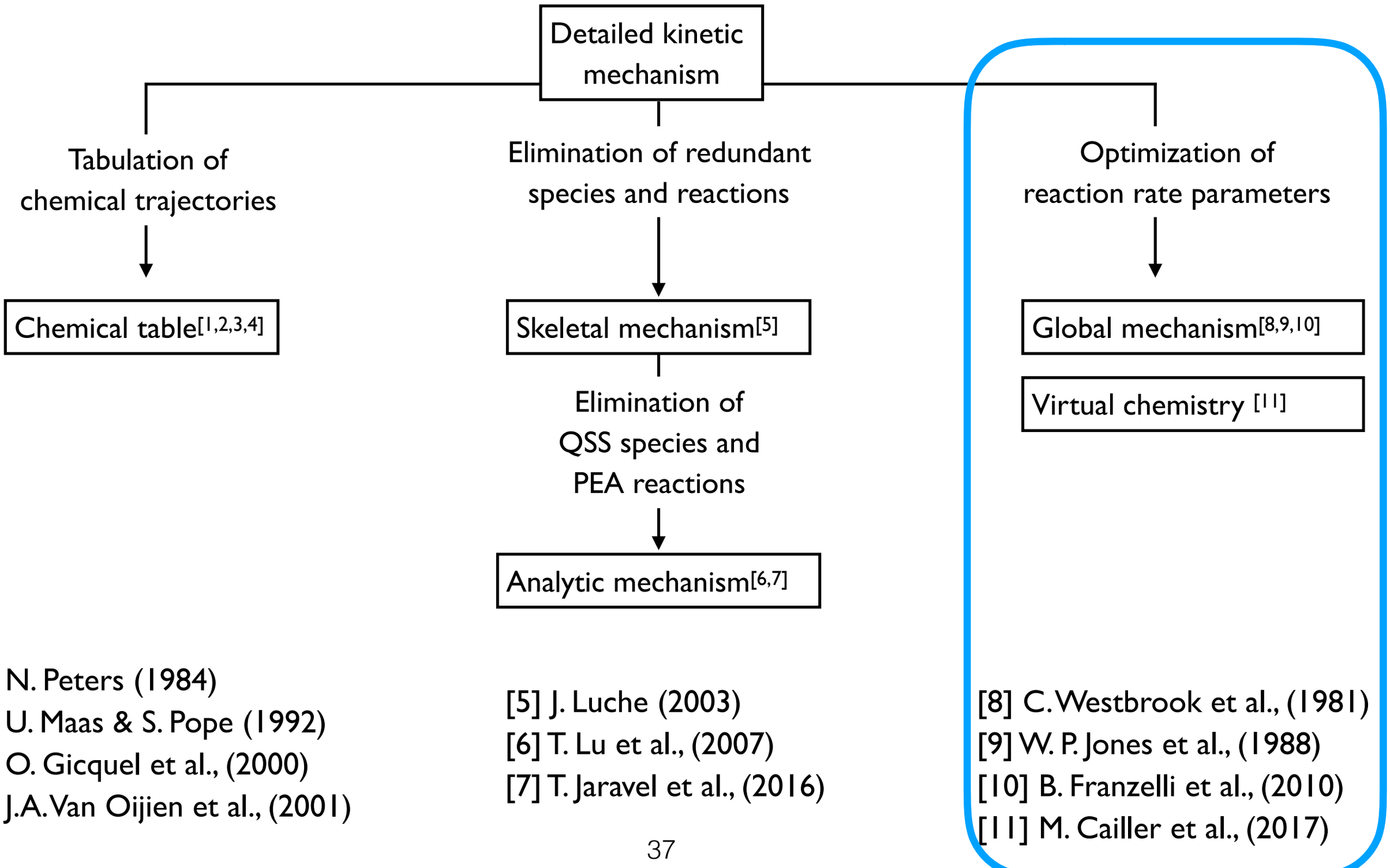


Chemistry tabulation

- **Chemistry tabulation is accurate and efficient**
  - compare well with detailed chemistry solutions
  - dramatic decrease of dimensions
- **But there is no universal solution for tabulation**
  - depends on the flame regime: premixed, partially-premixed or non premixed
  - Number of coordinates increase with operating conditions: pressure, heat exchanges, dilution with burnt gases, initial temperature ...

Kinetic scheme reduction strategies for combustion modeling

B. Fiorina, D. Veynante and S. Candel. Modeling Combustion Chemistry in Large Eddy Simulation of Turbulent Flames. Flow Turb. and Combustion. Vol 94, Issue 1, pp3-42 (2015).



[1] N. Peters (1984)
[2] U. Maas & S. Pope (1992)
[3] O. Gicquel et al., (2000)
[4] J.A. Van Oijien et al., (2001)

[5] J. Luche (2003)
[6] T. Lu et al., (2007)
[7] T. Jaravel et al., (2016)

[8] C. Westbrook et al., (1981)
[9] W. P. Jones et al., (1988)
[10] B. Franzelli et al., (2010)
[11] M. Cailler et al., (2017)

Global empirical chemistry

1) Selection of species and reactions
2) Optimization of reaction rate parameters to retrieve target flame properties

$$C_nH_m + \left(\frac{n}{2} + \frac{m}{4}\right)O_2 = nCO + \frac{m}{2}H_2O$$

$$CO + 1/2O_2 = CO_2.$$

Westbrook, C., Dryer, F.: Combust. Sci. Technol. 27, 31 (1981)

$$KERO + 10O_2 \Rightarrow 10CO + 10H_2($$

$$CO + 0.5O_2 \Longleftrightarrow CO_2.$$

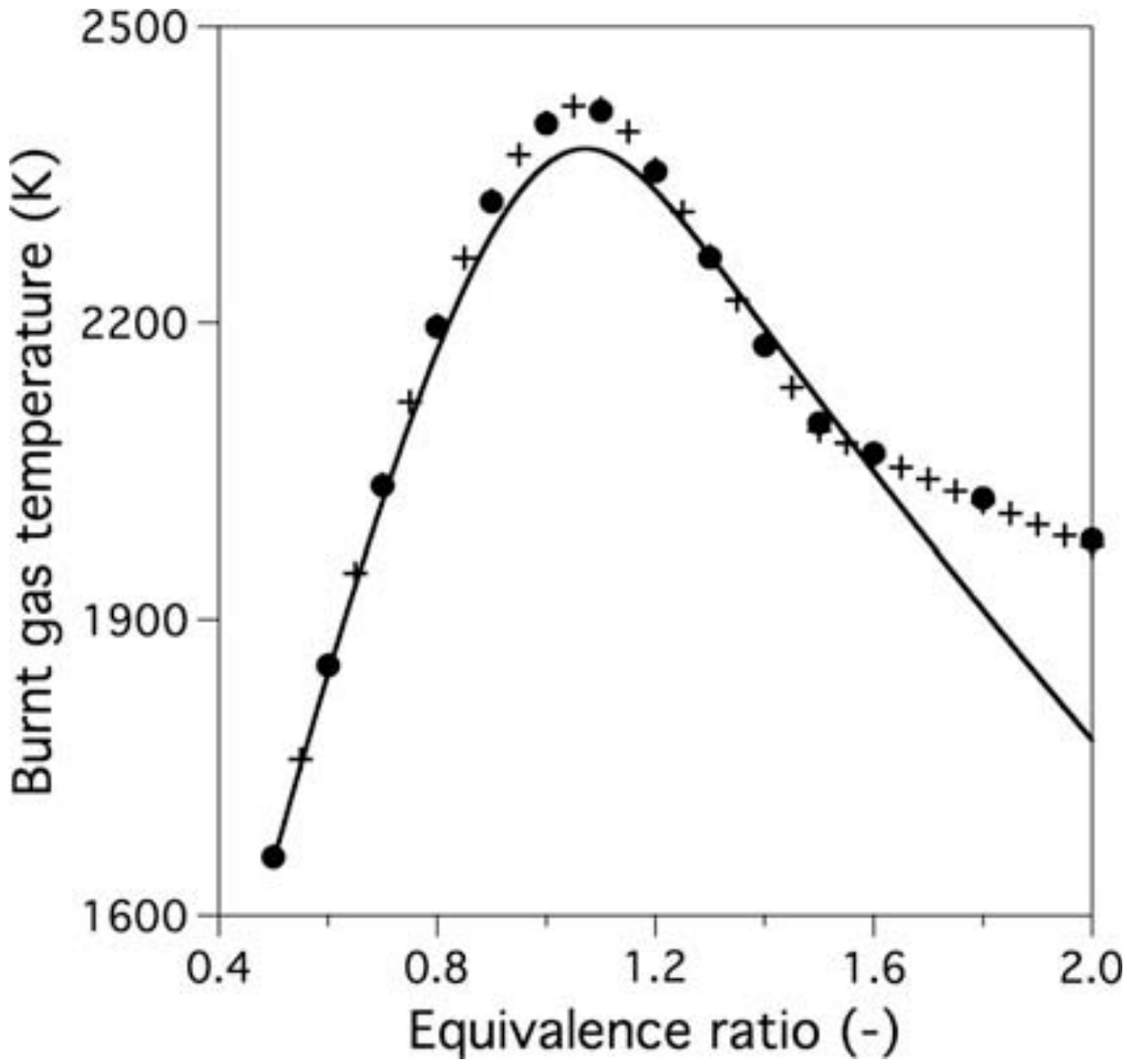
$$k_{f,1} = A_1 f_1(\phi) e^{(-E_{a,1}/RT)} [KERO]^{n_{KERO}} [O_2]^{n_C}$$

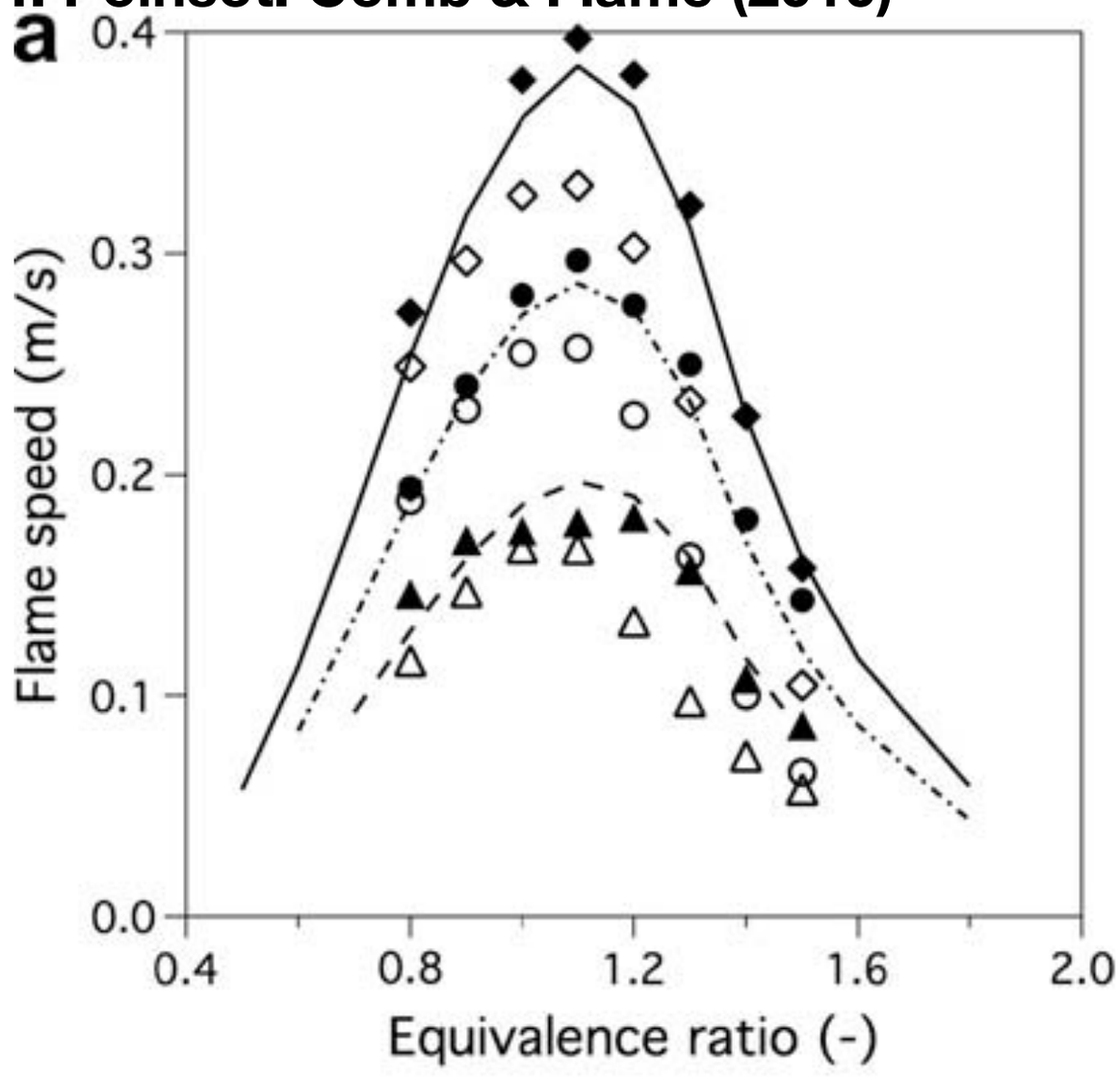
$$k_{f,2} = A_2 f_2(\phi) e^{(-E_{a,2}/RT)} [CO]^{n_{CO}} [O_2]^{n_{O_2,2}},$$

B. Franzelli, E. Riber, M. Sanjosé, T. Poinsot. Comb & Flame (2010)

Global empirical chemistry
Kerosen - Air combustion

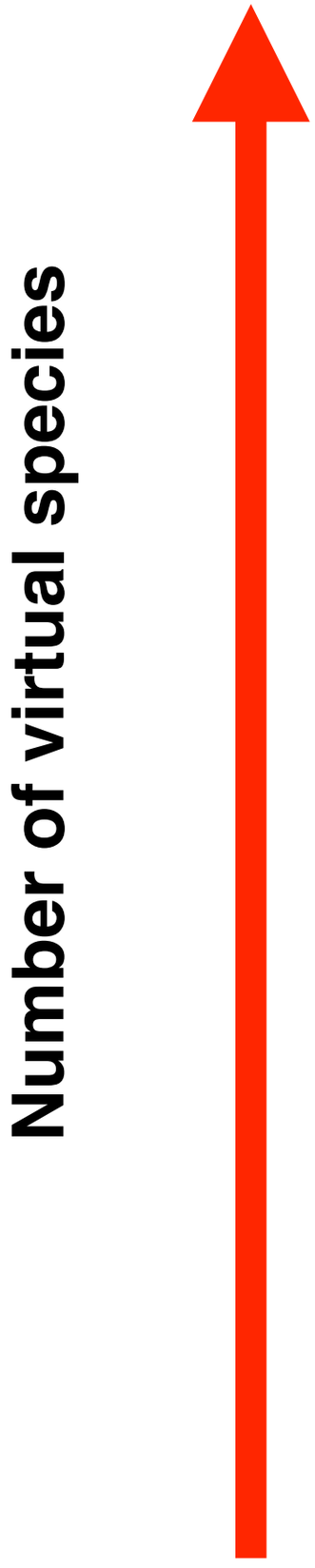
B. Franzelli, E. Riber, M. Sanjosé, T. Poinsot. Comb & Flame (2010)





- Flame speed is well retrieved
- not enough species to get the flame temperature in rich regions
- Pollutant are not captured … CO is not CO

Virtual chemistry



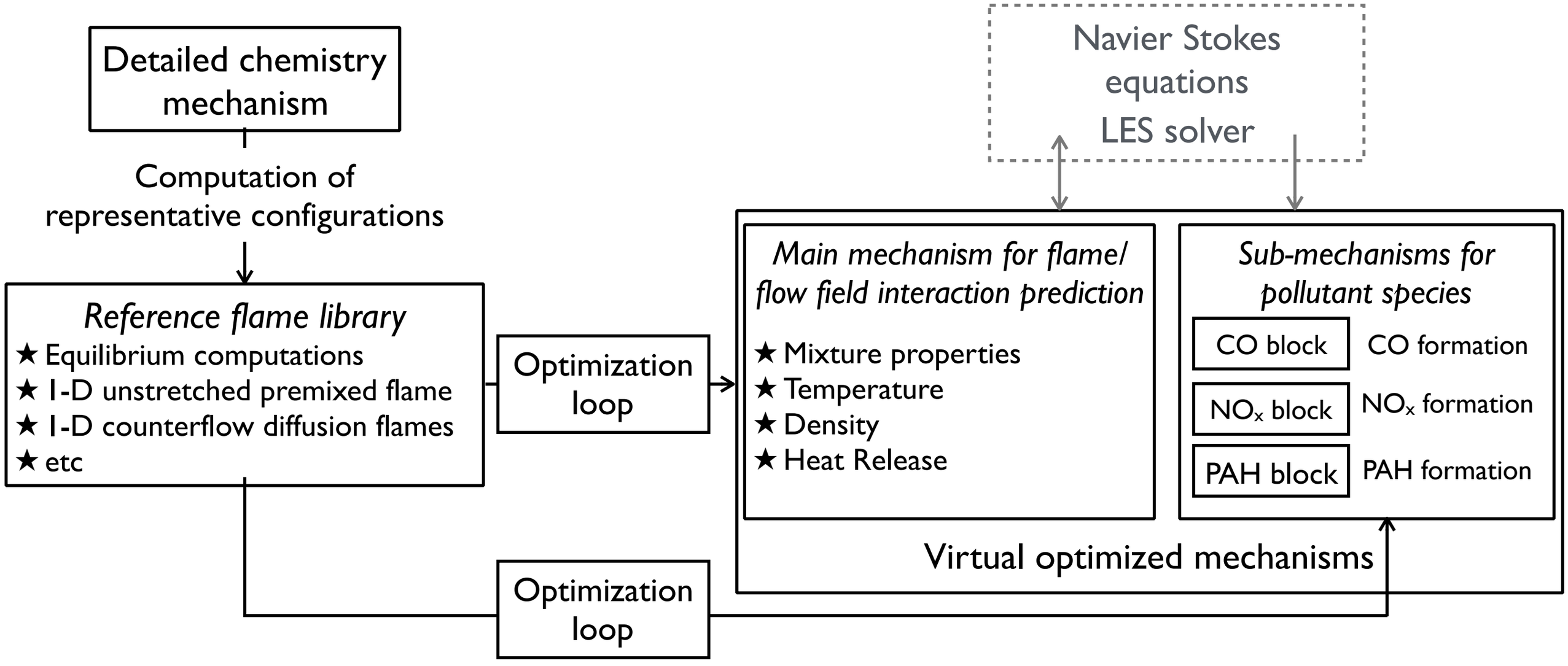
1) building-up a reduced chemical mechanism from scratch

2) using virtual species and reactions whose chemical rate constants and thermo-chemical properties are optimized

M. Cailler, N. Darabiha, D. Veynante and B. Fiorina. Building-up virtual optimized mechanism for flame modeling. Proceeding of the Combustion Institute (2017)

Virtual optimized mechanisms architecture

- Virtual optimized chemistry approach relies on
  - The optimization of reaction rate parameters and virtual species physical properties
  - The use of sub-mechanisms for dedicated flame property prediction



Sub-mechanism for CO prediction

- Optimization of kinetic scheme and reaction rate parameters
  - Three-step mechanism

$$\alpha_F F + \alpha_{Ox} Ox \rightarrow \alpha CO + (1-\alpha) V_1$$
$$F + V_1 \rightarrow F + CO$$
$$CO \leftrightarrow V_2$$

- Rate of progress:

$$q_3 = A_3 f_3\left(Y_D^v\right) \exp\left(\frac{-E_{a,3}}{RT}\right) [F]^{F_F^3} [Ox]^{F_{Ox}^3}$$

$$q_4 = A_4 f_4\left(Y_D^v\right) \exp\left(\frac{-E_{a,4}}{RT}\right) [F]^{F_F^4} [V_1]^{F_{V_1}^4}$$

$$q_5 = A_5 f_5\left(Y_D^v\right) \exp\left(\frac{-E_{a,5}}{RT}\right) \left([CO]^{F_{CO}^5} [V_1]^{F_{V_1}^5} - \frac{[CO]^{R_{CO}^5} [V_1]^{R_{V_1}^5}}{\exp\left(\frac{-\Delta G_5^0\left(Y_D^v\right)}{RT}\right)} \right)$$

Pre-tabulated to capture Y_{CO} at equilibrium

M. Cailler, N. Darabiha and B. Fiorina. Virtual chemistry for pollutant emissions prediction (submitted)

Sub-mechanism for CO prediction

- Optimization of kinetic scheme and reaction rate parameters
  - Three-step mechanism

$$\alpha_F F + \alpha_{Ox} Ox \rightarrow \alpha CO + (1-\alpha) V_1$$
$$F + V_1 \rightarrow F + CO$$
$$CO \leftrightarrow V_2$$

- Rate of progress:

$$q_3 = A_3 f_3\left(Y_D^v\right) \exp\left(\frac{-E_{a,3}}{RT}\right) [F]^{F_F^3} [Ox]^{F_{Ox}^3}$$

$$q_4 = A_4 f_4\left(Y_D^v\right) \exp\left(\frac{-E_{a,4}}{RT}\right) [F]^{F_F^4} [V_1]^{F_{V_1}^4}$$

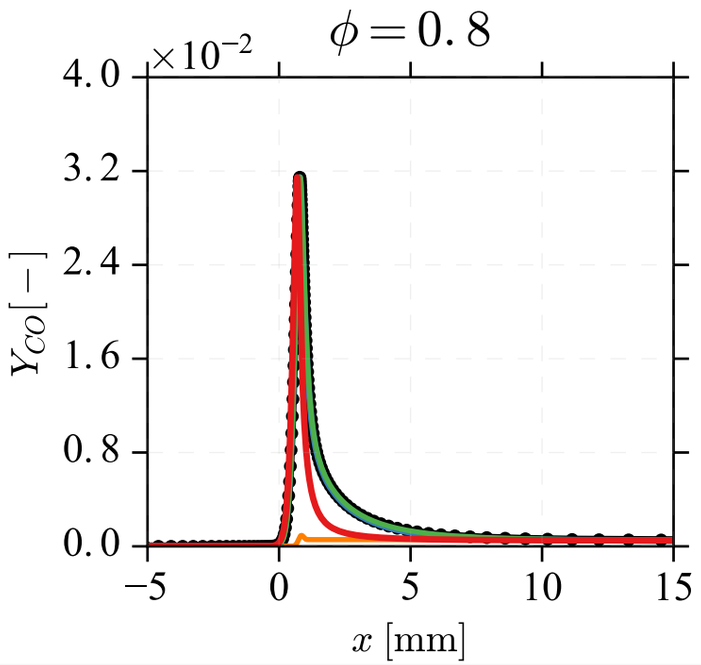
$$q_5 = A_5 f_5\left(Y_D^v\right) \exp\left(\frac{-E_{a,5}}{RT}\right) \left([CO]^{F_{CO}^5} [V_1]^{F_{V_1}^5} - \frac{[CO]^{R_{CO}^5} [V_1]^{R_{V_1}^5}}{\exp\left(\frac{-\Delta G_5^0\left(Y_D^v\right)}{RT}\right)} \right)$$

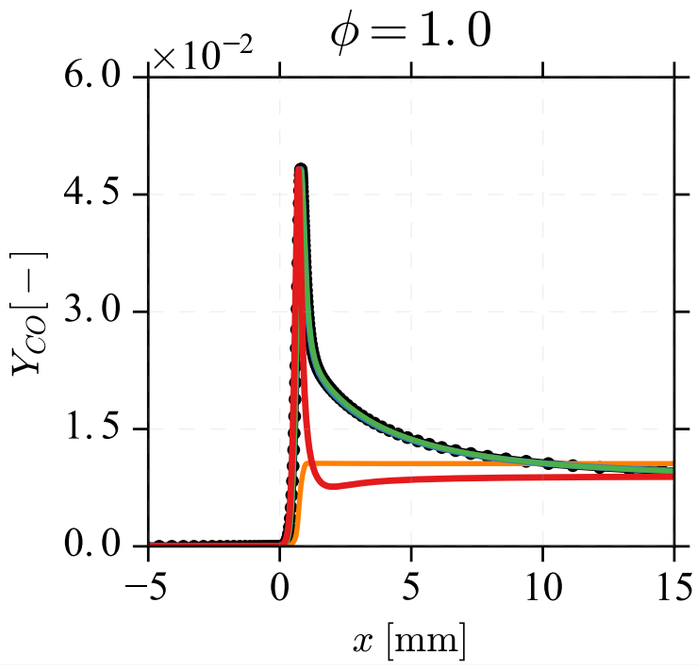
Pre-tabulated to capture Y_{CO} at equilibrium

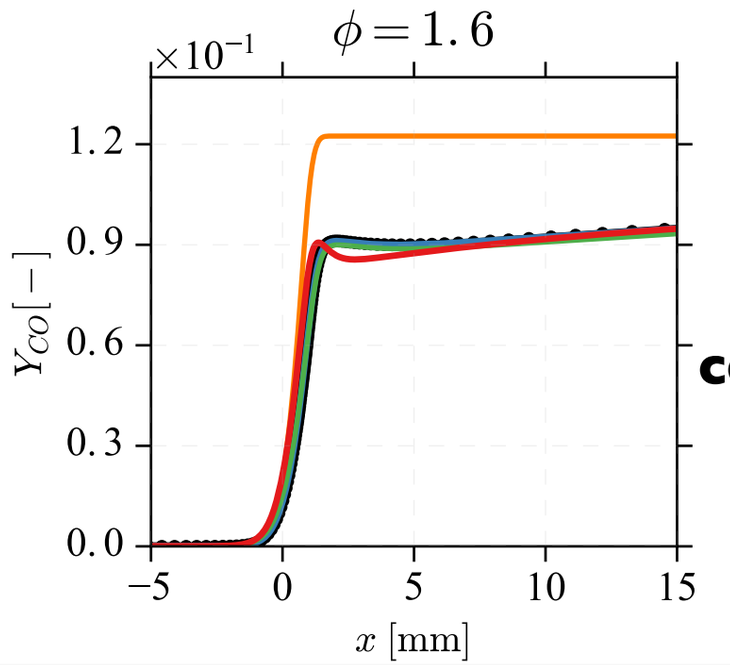
- A_i, $E_{a,i}$, $F_{k,i}$, f_4 and f_5 are optimized through an in-house genetic algorithm

M. Cailler, N. Darabiha and B. Fiorina. Virtual chemistry for pollutant emissions prediction (submitted)

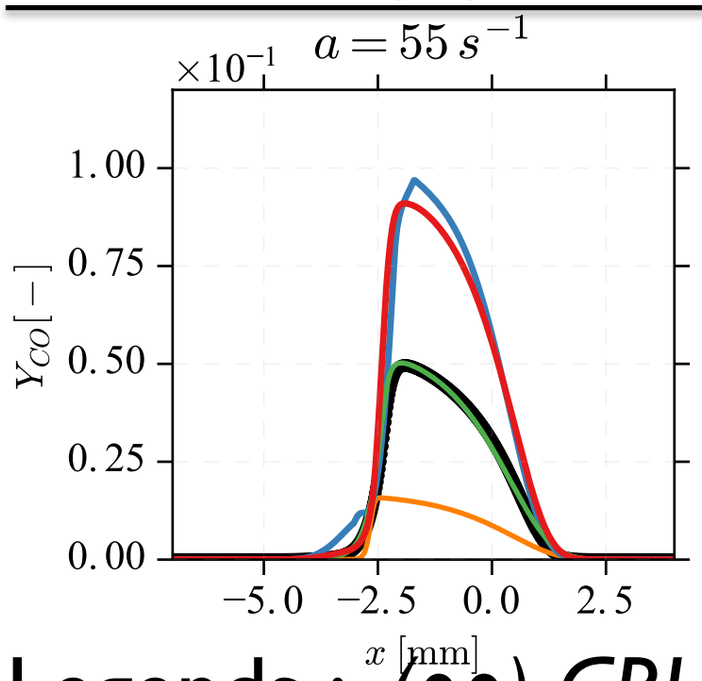
Y_{CO} profiles: Reference library = {Premixed}

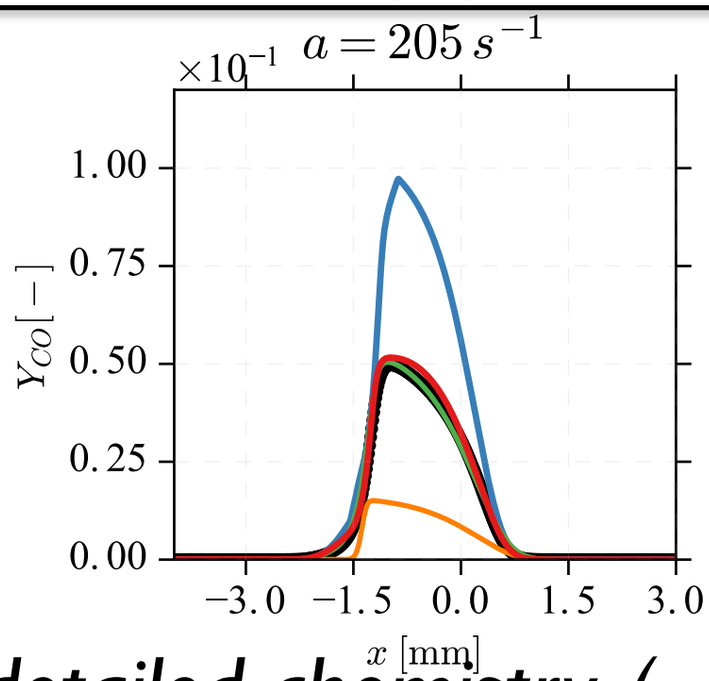


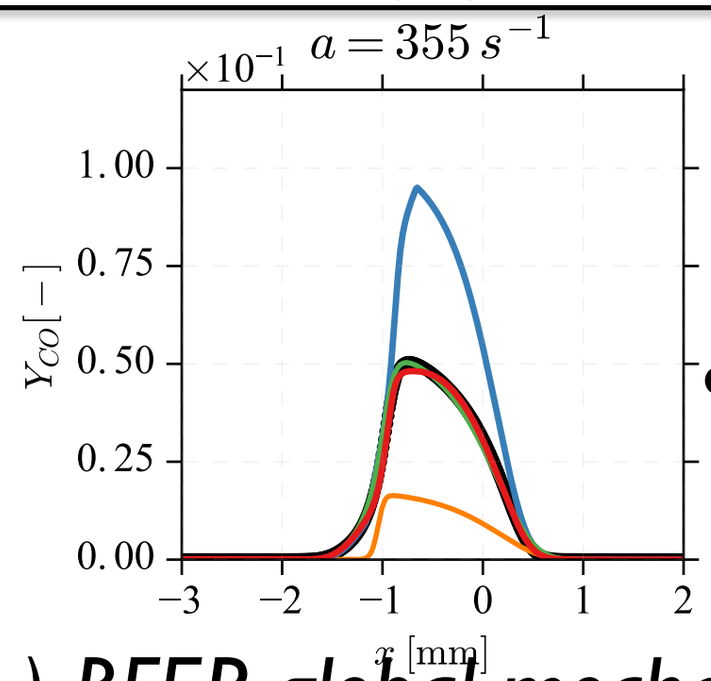




Premixed configuration



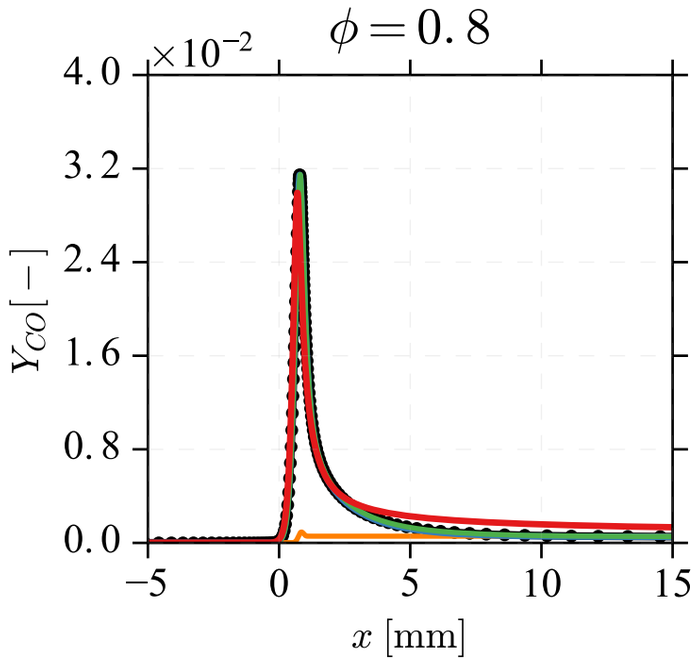


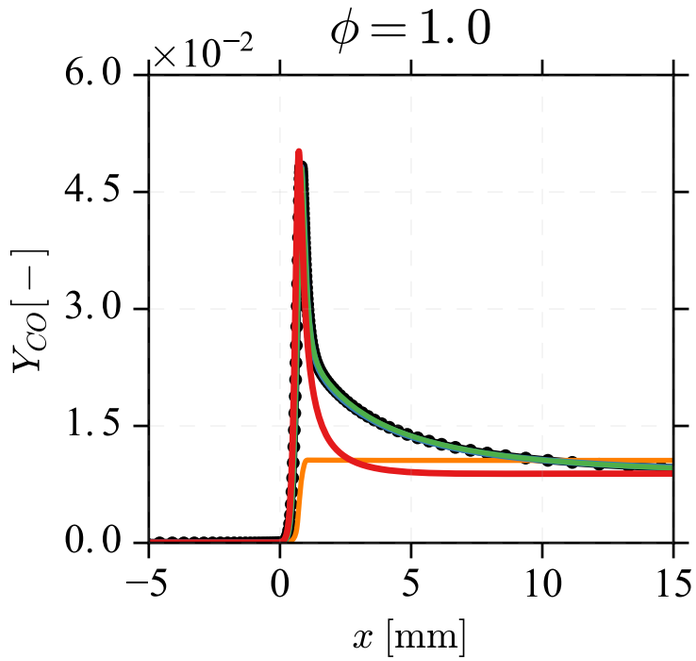


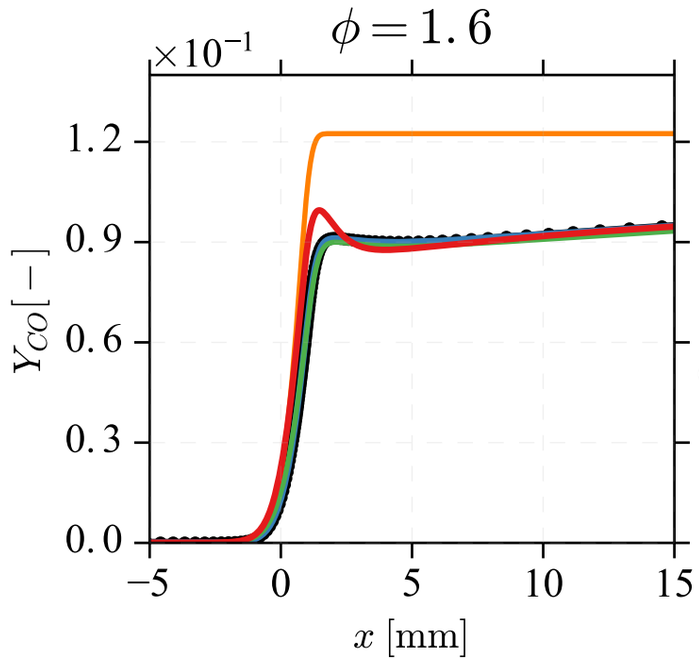
Counterflow configuration

Legends : (●●) *GRI detailed chemistry,* (—) *BFER global mechanism,* (—) *FPI premixed tabulated,* (—) *LU19 analytic scheme,* (—) *CO-PREM virtual mechanism*

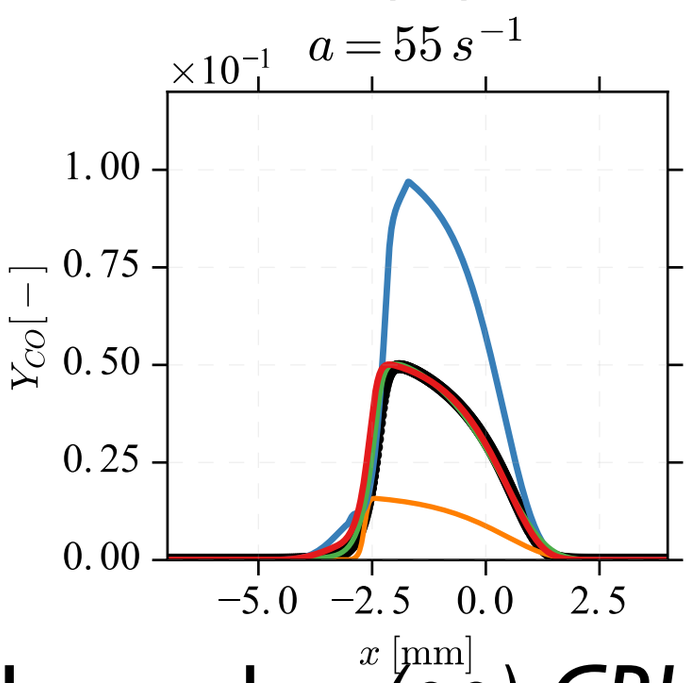
Y_{CO} profiles: Reference library = {Premixed + Diffusion}

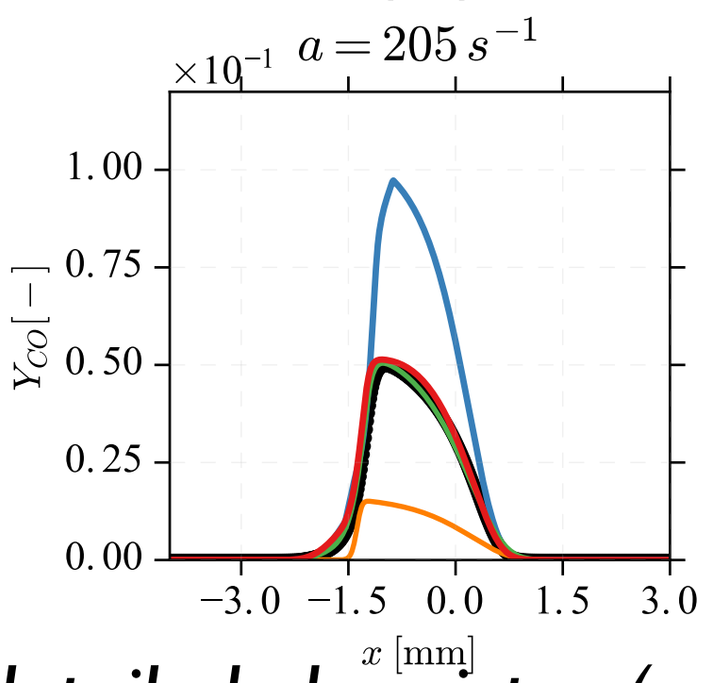


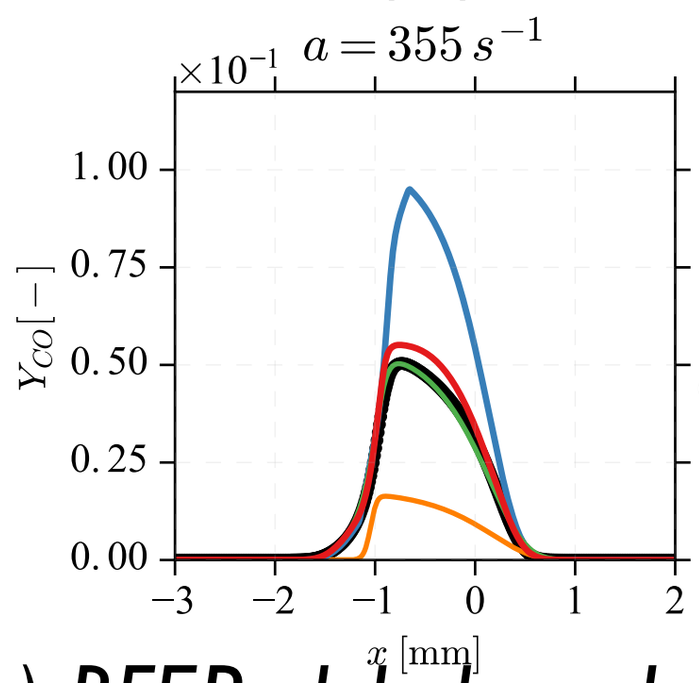




Premixed configuration



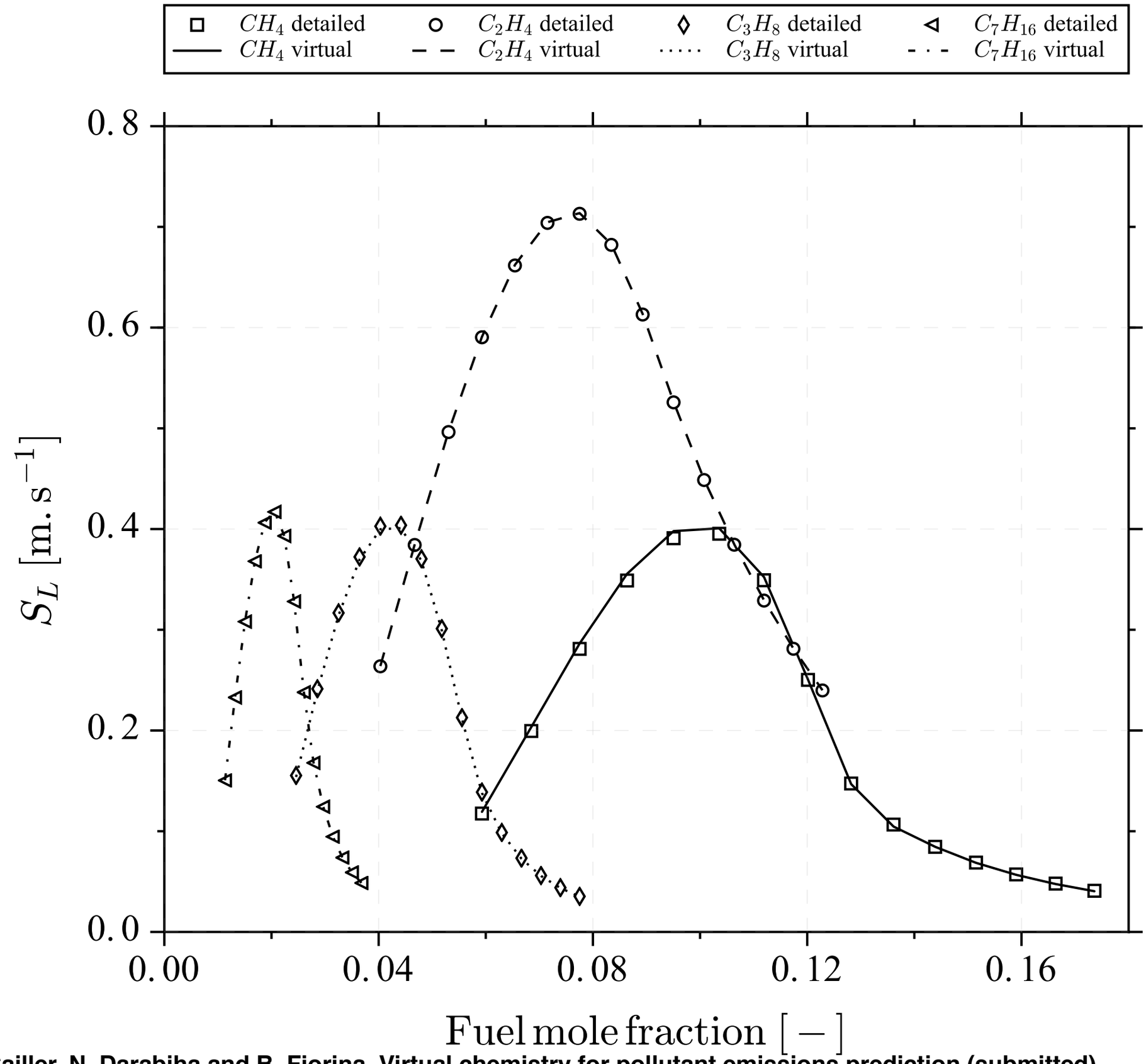




Counterflow configuration

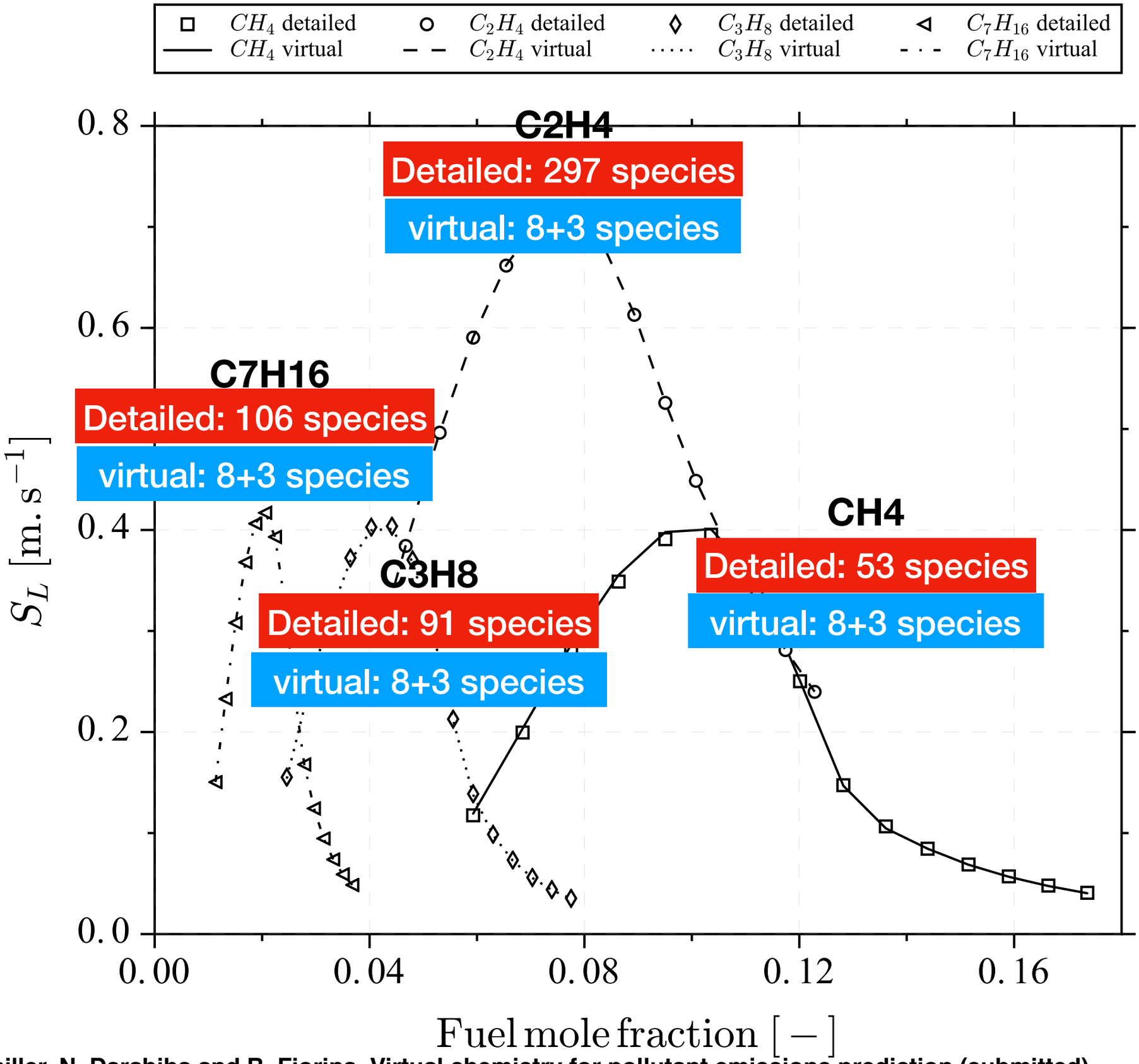
Legends : *(••) GRI detailed chemistry, (—) BFER global mechanism, (—) FPI premixed tabulated, (—) LU19 analytic scheme, (—) CO-PREM-DIFF virtual mechanism*

Heavier fuels



M. Cailler, N. Darabiha and B. Fiorina. Virtual chemistry for pollutant emissions prediction (submitted)

Heavier fuels



M. Cailler, N. Darabiha and B. Fiorina. Virtual chemistry for pollutant emissions prediction (submitted)

REDUCED OR TABULATED OR OPTIMIZED ?

premixed flames

non premixed flames

auto-ignition

pressure variation

non-adiabatic

burnt gases dilution

Detailed scheme

premixed flamelet

non premixed flamelets

Need target flames to identify redundant species and apply QSS

ARC scheme

premixed flamelets

non premixed flamelets

Need flame library to build the look-up table

tabulated

premixed flamelets

non premixed flamelets

Need target flames to satisfy objective functions

optimized scheme

Conclusion

Tabulated chemistry

✓ very efficient on canonical configuration

- less suitable to « complex » flame structure where many flame regimes are encountered

- A single flamelet is non sufficient to build the table:
  - Which flamelet archetypes to tabulate ?
  - How to map a database with different archetypes?
  - New coordinates are needed. How to close the look-up table coordinates balance equations ?

Conclusion

Analytically Reduced Chemistry

✓ Progress have been made to automatize the generation of ARC mechanisms

✓ Massively computing facilities make possible their use in CFD

- still expensive as many species remains to be transported

- stiffness of reaction rates

Conclusion

Virtual Chemistry

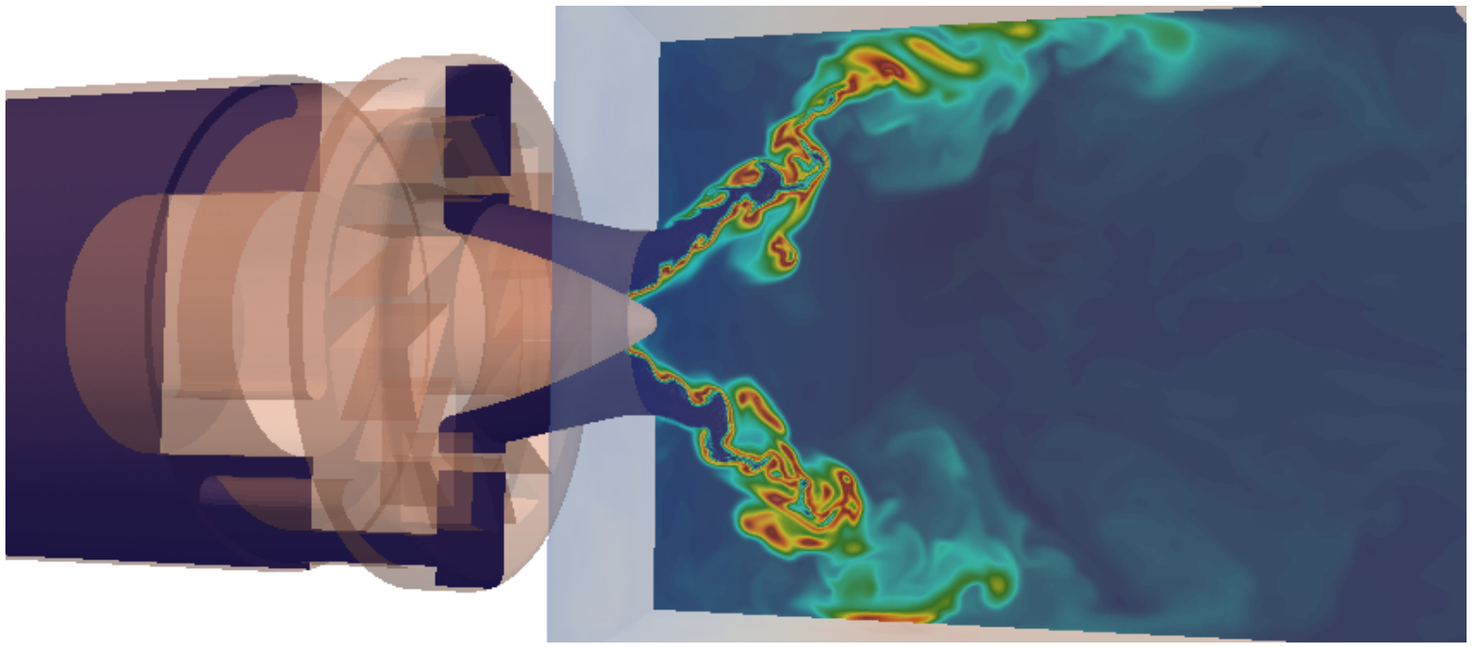
✓Enables the prediction of pollutant with a few reactions

✓Size does not depends on the fuel

- mechanims and sub-mechanisms structure remains to be automatized

- intuitive identification of the degree of freedoms

Chemistry is reduced to compute turbulent flames



Preccinsta burner simulation with virtual chemistry (G. Maio PhD)

… a few examples

Joined comparative study, TNF 2012 and 2014

CentraleSupélec

TECHNISCHE UNIVERSITÄT DARMSTADT

UNIVERSITÄT DUISBURG ESSEN

RWTH AACHEN UNIVERSITY

Research groups have developed their own computational strategies

- different turbulent combustion models
- different codes with different numerics
- different meshes
- different computational domains
- different phd students who run and analyse the computations

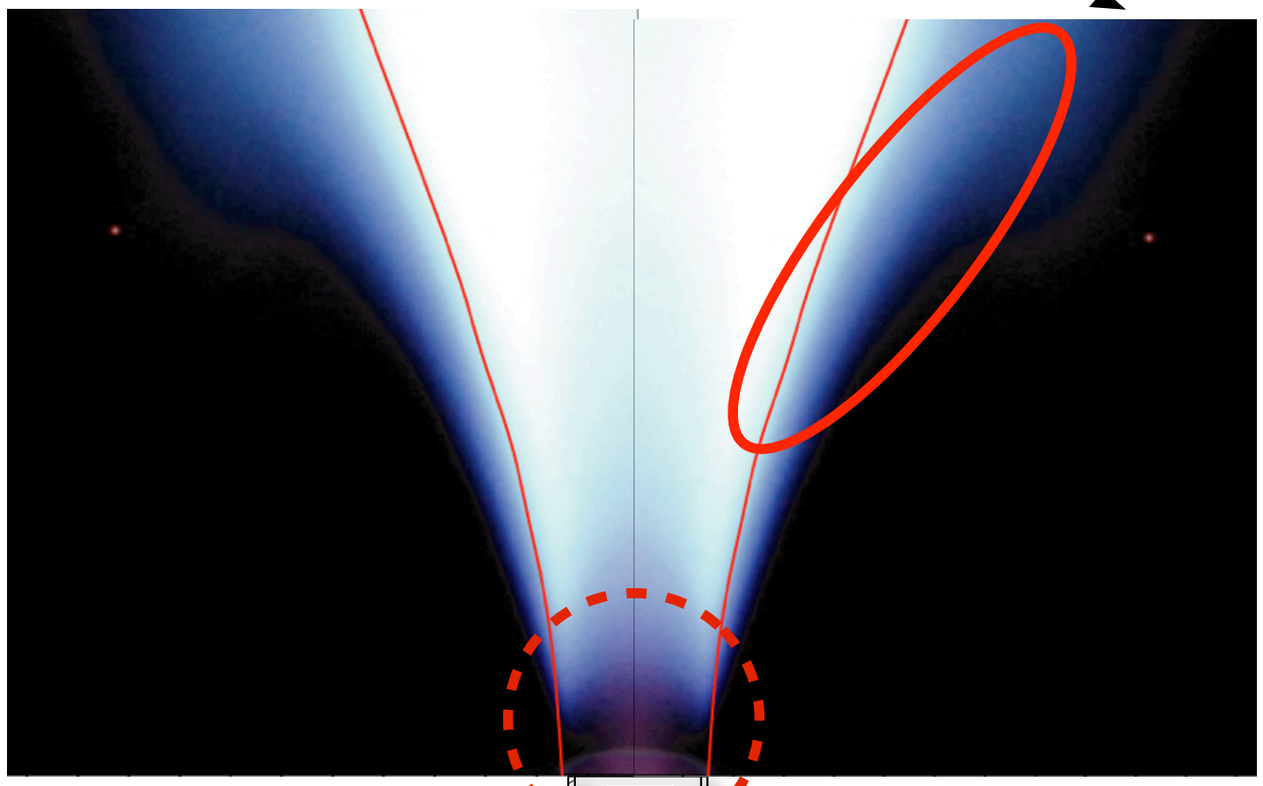
We cannot conclude on specific submodel perfomances with this exercice !

But aim is to illustrate the state of the art

B. Fiorina, R. Mercier, G. Kuenne, A. Ketelheun, A. Avdic, J. Janicka, D. Geyer, A. Dreizler, E. Alenius, C. Duwig, P. Trisjono, K. Kleinheinz, S. Kang, H. Pitsch, F. Proch, F. Cavallo Marincola, A. Kempf . Challenging modeling strategies for LES of non-adiabatic turbulent stratified combustion. Comb & Flame (2015)

TSFA: no shear layer / stratification

stratification zones



TECHNISCHE UNIVERSITÄT DARMSTADT

LDV measurements:

- Axial/radial velocity

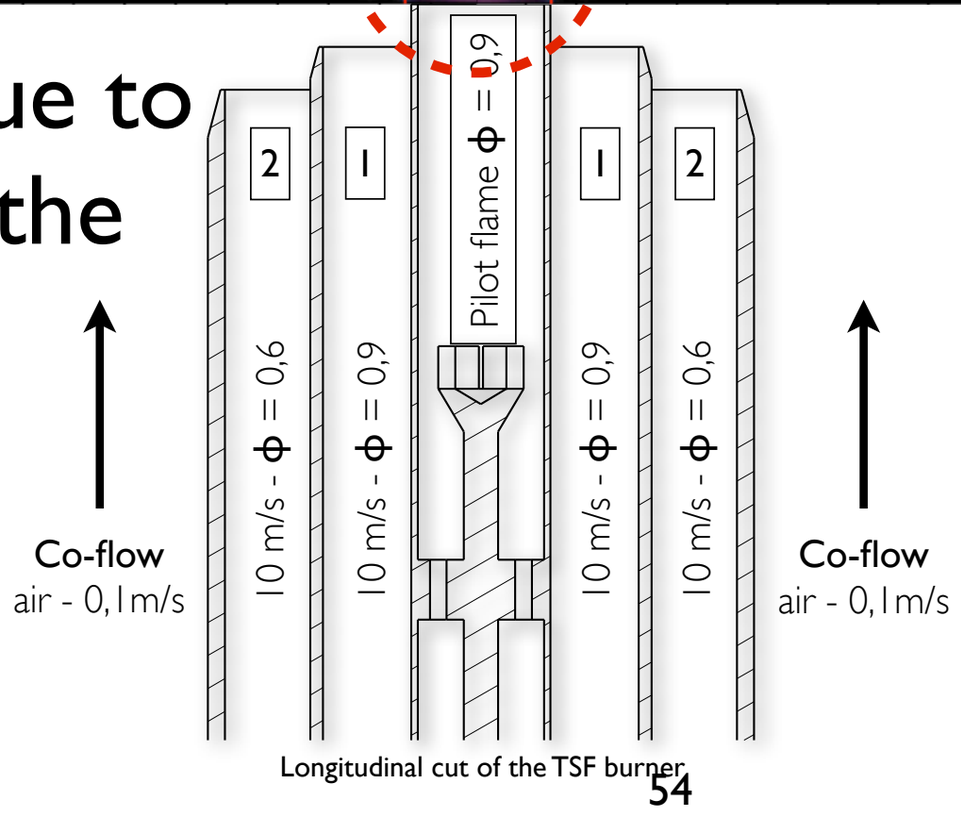
Raman/Rayleigh measurements:

- $T - Y_{O2} - Y_{CH4} - Y_{CO2} - Y_{H2O} - Y_{N2}$

B. Böhm et al., Proc. Comb. Inst. (2010)

F. Seffrin et al., Comb. & Flame 157. (2010)

Flame lifting due to heat losses at the burner lips

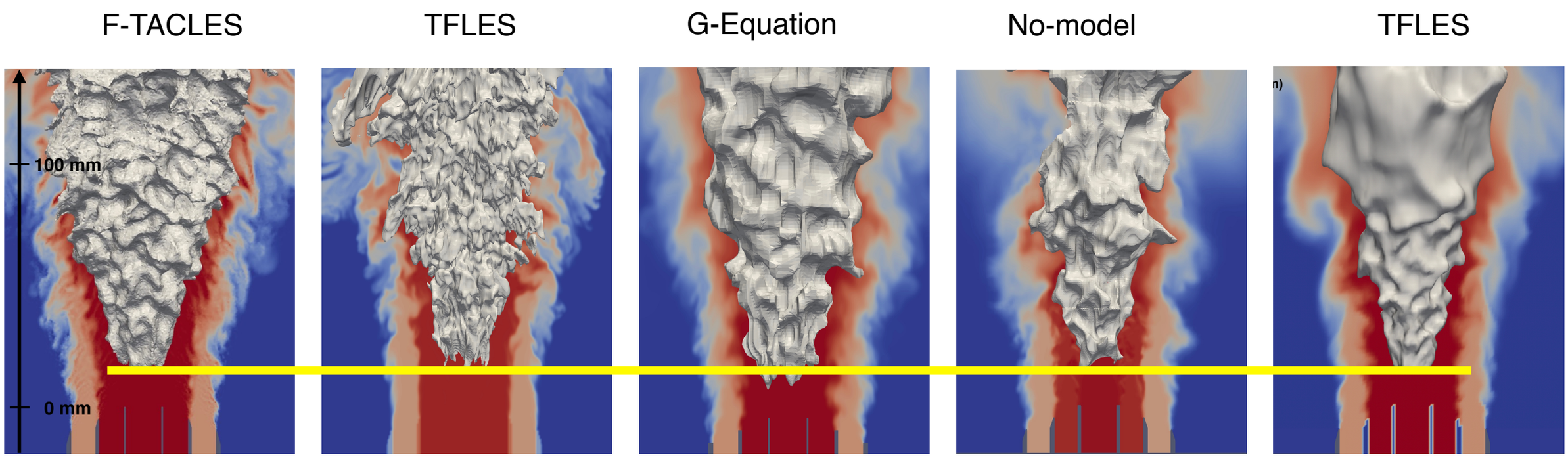


Longitudinal cut of the TSF burner

Turbulent Combustion model	Chemistry modeling	Kinetic scheme	Flame resolution management	Flame wrinkling model
ATF-FGM (TUD)	Tabulated chemistry (premixed flamelets)	GRI3.0 Lewis number unity [25]	Thickening	Charlette *et al.* [21]
F-TACLES (EM2C)	Tabulated chemistry (premixed flamelets)	Lindstedt [34]	Filtering	Charlette *et al.* [21]
Coupled G-C (ITV)	Tabulated chemistry (premixed flamelets)	Peters & Rogg [35]	Level-set	Pitsch [15]
NCM (LUND)	Semi-global scheme	Jones & Lindstedt [36]	Resolved	None
FSD (UDE)	Flame speed tabulation	GRI3.0 Lewis number unity [25]	None (see Sec. 2.6.2)	Fureby [27]

4 « Tabulated chemistry »

1 « No turbulent combustion model »



(a) EM2C (b) UDE (c) ITV (d) LUND (e) TUD

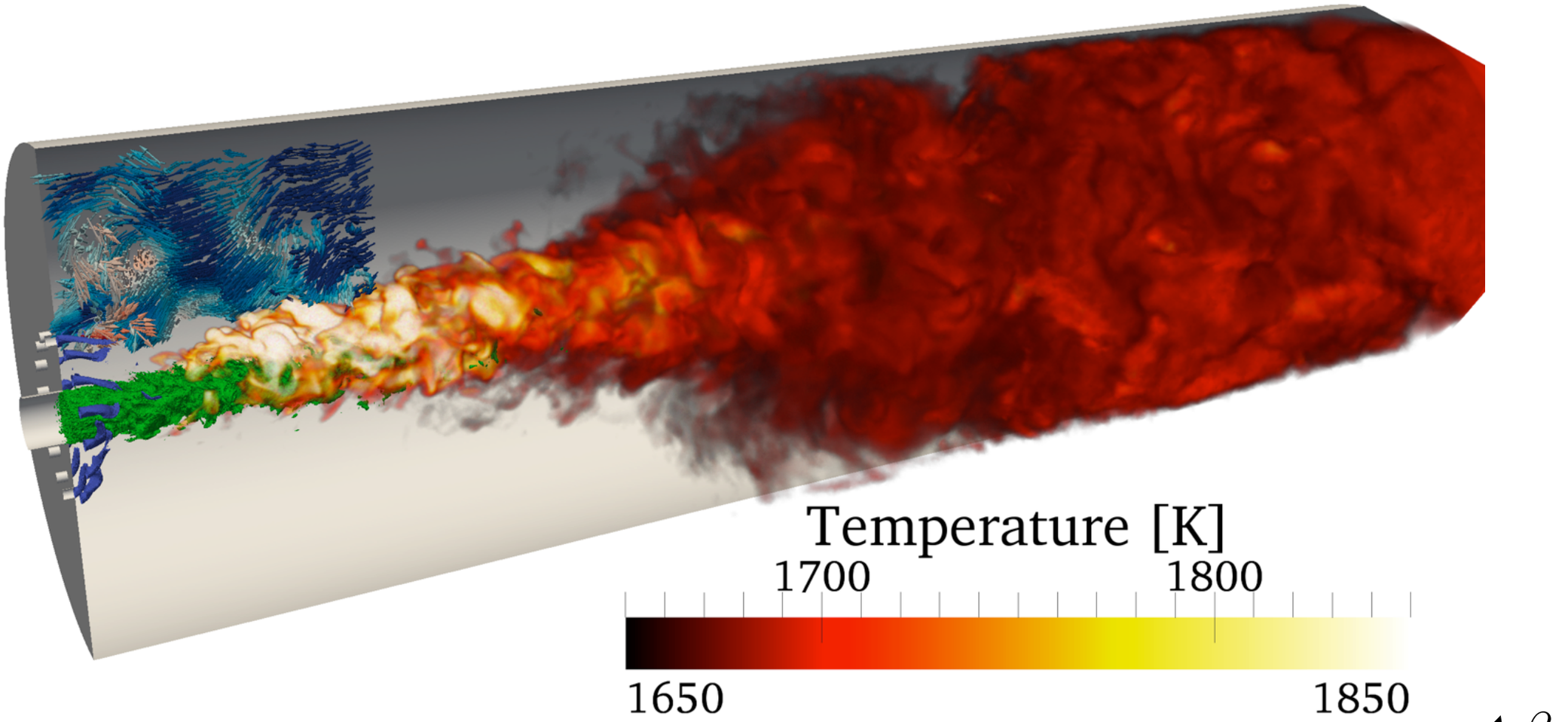
Relative difference of 18% between LES

➡ Good prediction of the flame lift-off

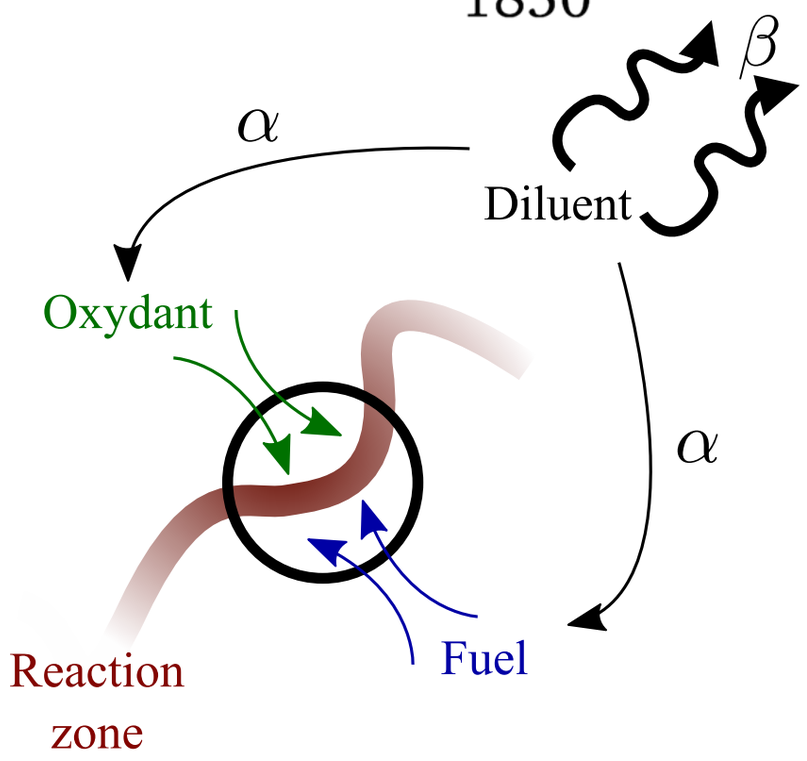
➡ In general a fair agreement has been observed between LES and experiments

B. Fiorina, R. Mercier, G. Kuenne, A. Ketelheun, A. Avdic, J. Janicka, D. Geyer, A. Dreizler, E. Alenius, C. Duwig, P. Trisjono, K. Kleinheinz, S. Kang, H. Pitsch, F. Proch, F. Cavallo Marincola, A. Kempf . Challenging modeling strategies for LES of non-adiabatic turbulent stratified combustion. Comb & Flame (2015)

Large Eddy Simulation (LES) of turbulent combustion



$$\phi = \phi(Z, Y_C, \alpha, \beta)$$

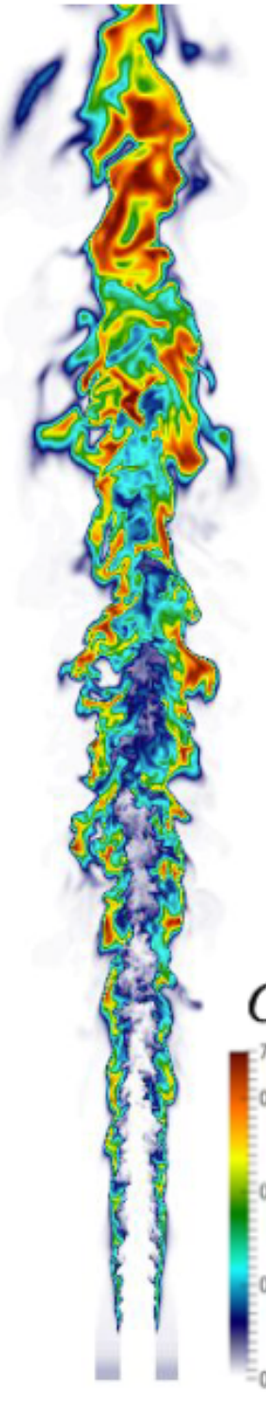


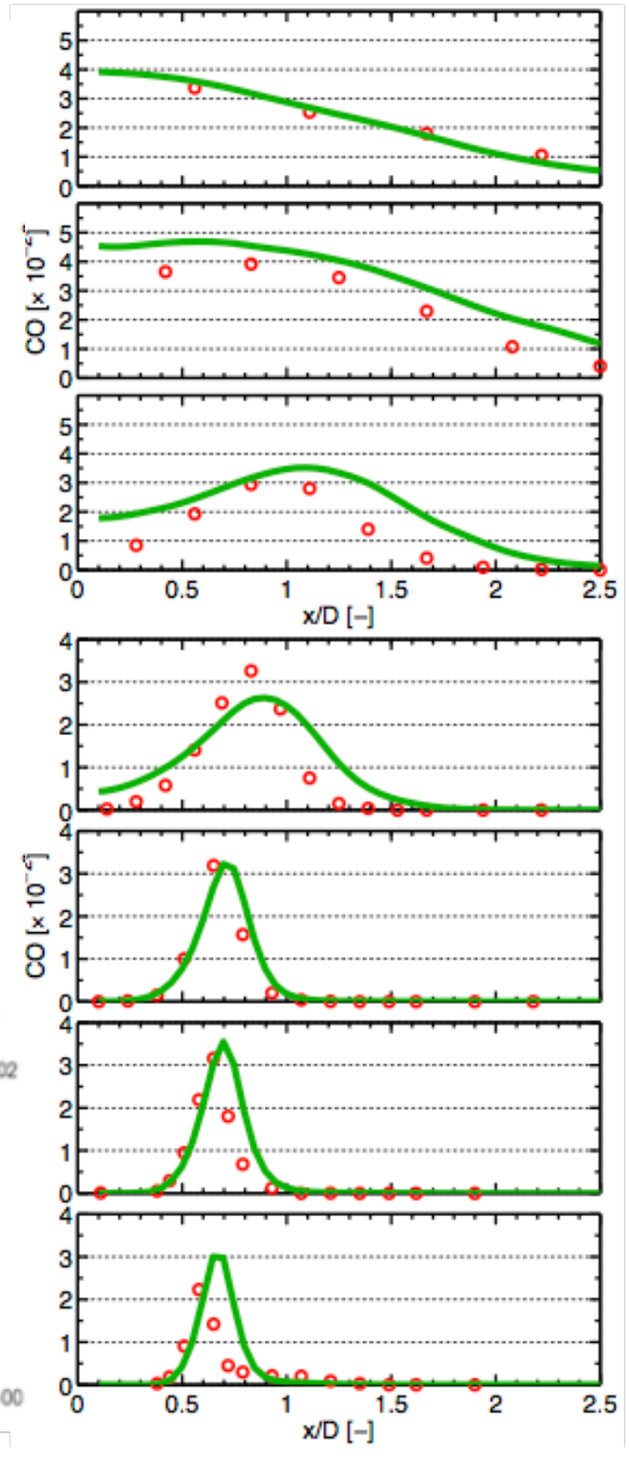
J. Lamouroux, M. Ihme, B. Fiorina and O. Gicquel. Tabulated chemistry approach for diluted. combustion regimes with internal recirculation and heat losses. Comb. Flame. (2014)

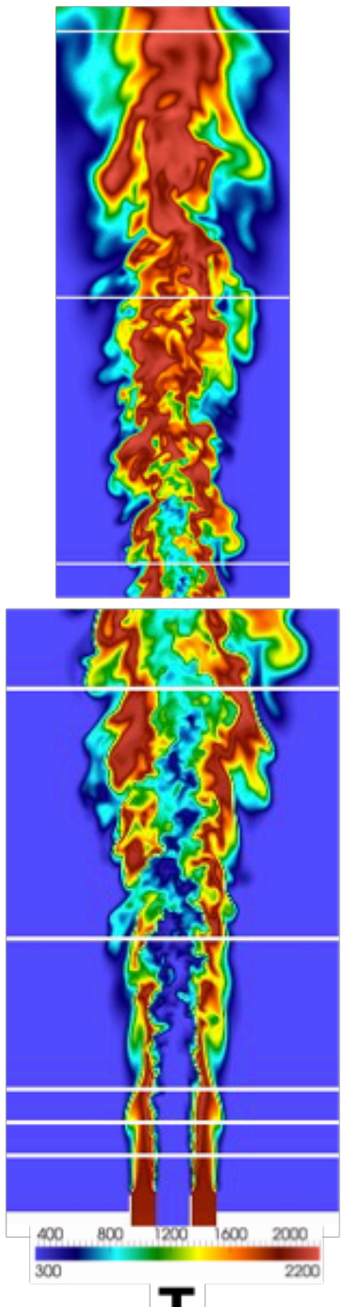
Example of ARC in LES: SANDIA D Flame

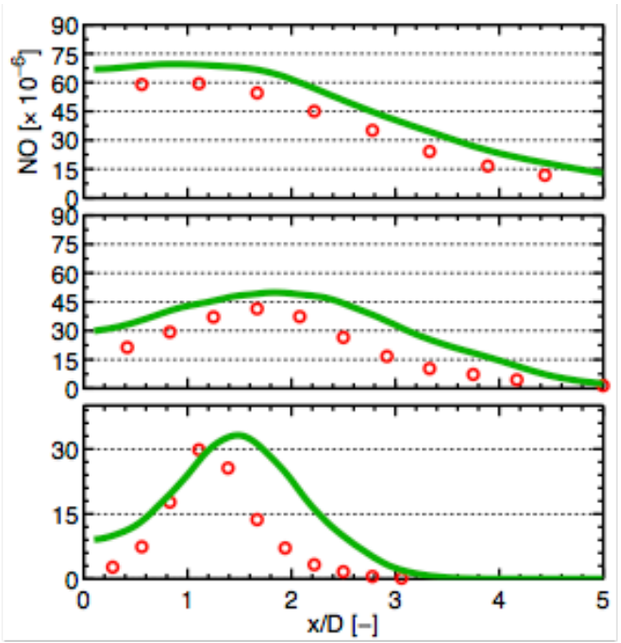
(PhD of T. Jaravel)

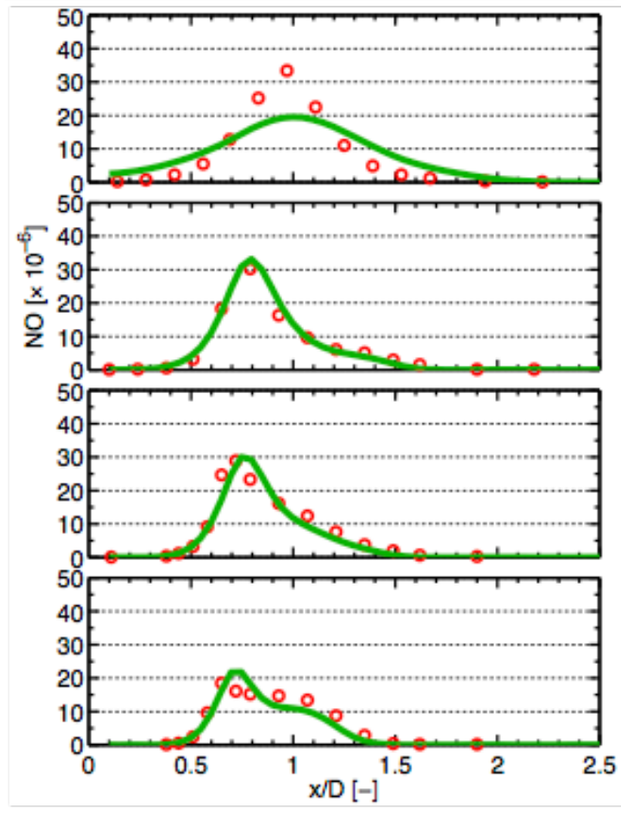
No thickening: flame front resolution











O Exp

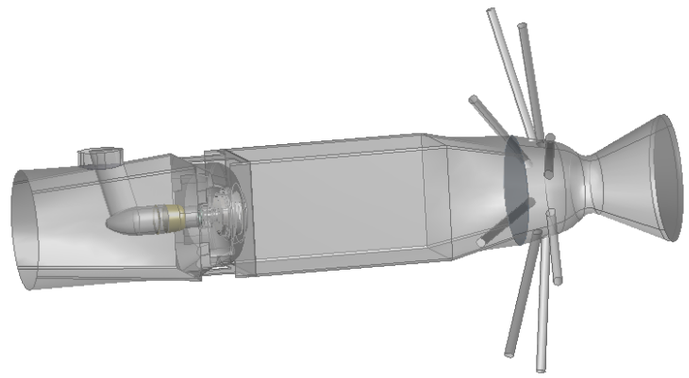
- ARC_22_GRI211

Example of Virtual chemistry in LES: HERON combustor

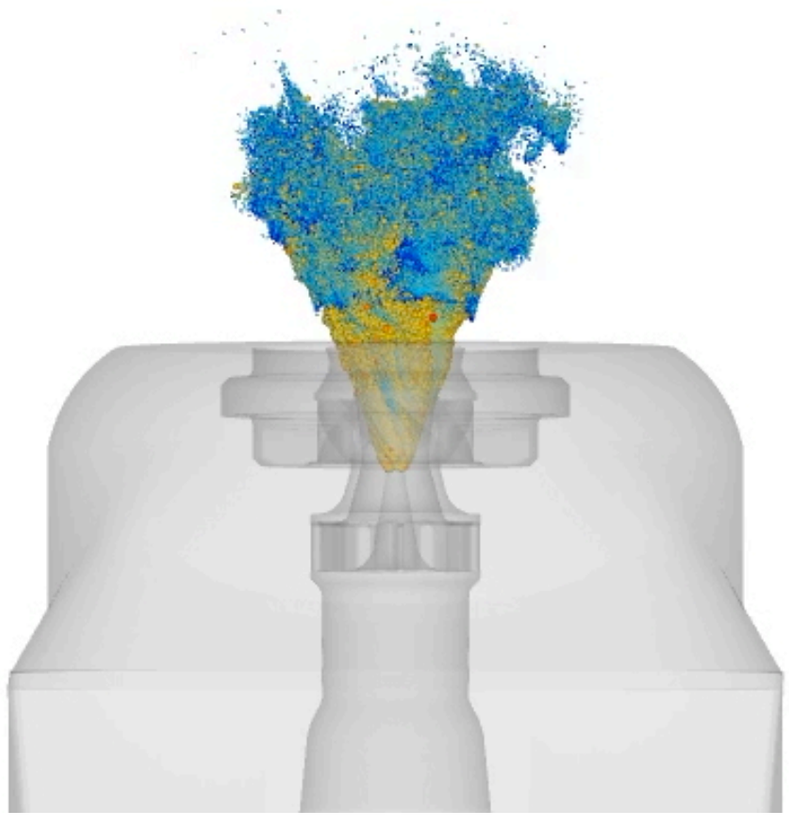
(PhD of M. Cailler)

Flame front is thickened by TFLES, Subgrid flame wrinkling is modeled

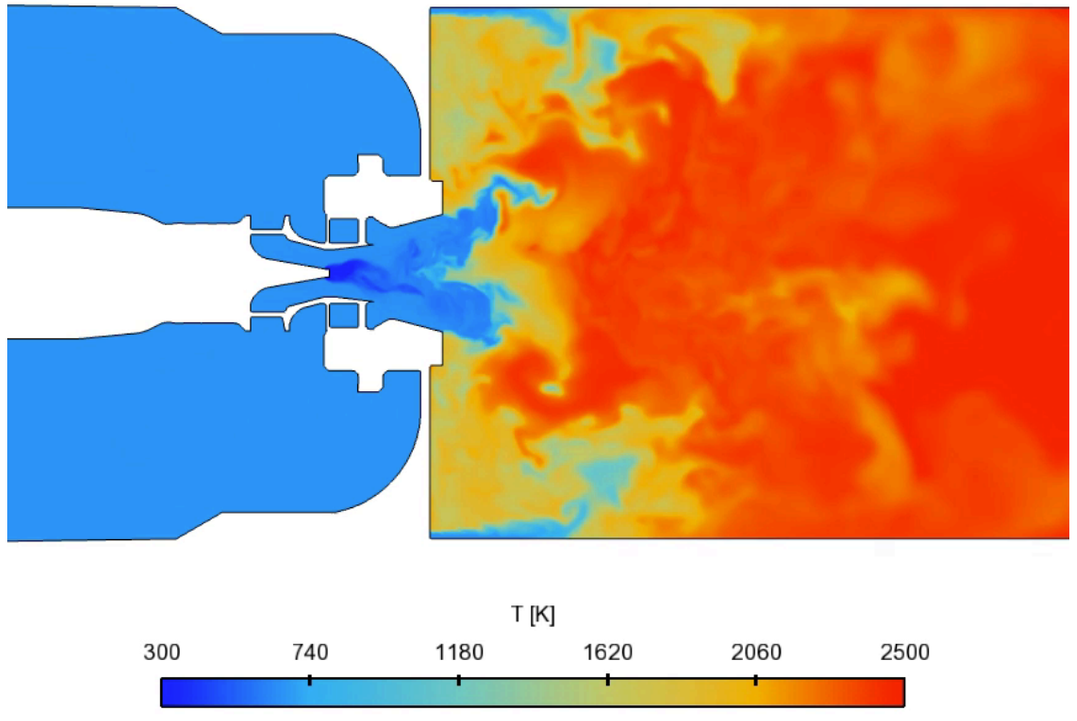
- High pressure test-rig HERON equipped with a Lean-Premixed injection system (Malbois, Proceedings of ASME Turbo expo, 2017)



- Designed to investigate the impact of pressure and equivalence ratio on the flame structure and polluant formation



Lagrangian particles colored by their diameter and gaseous kerosene mass fraction



Instantaneous field of temperature