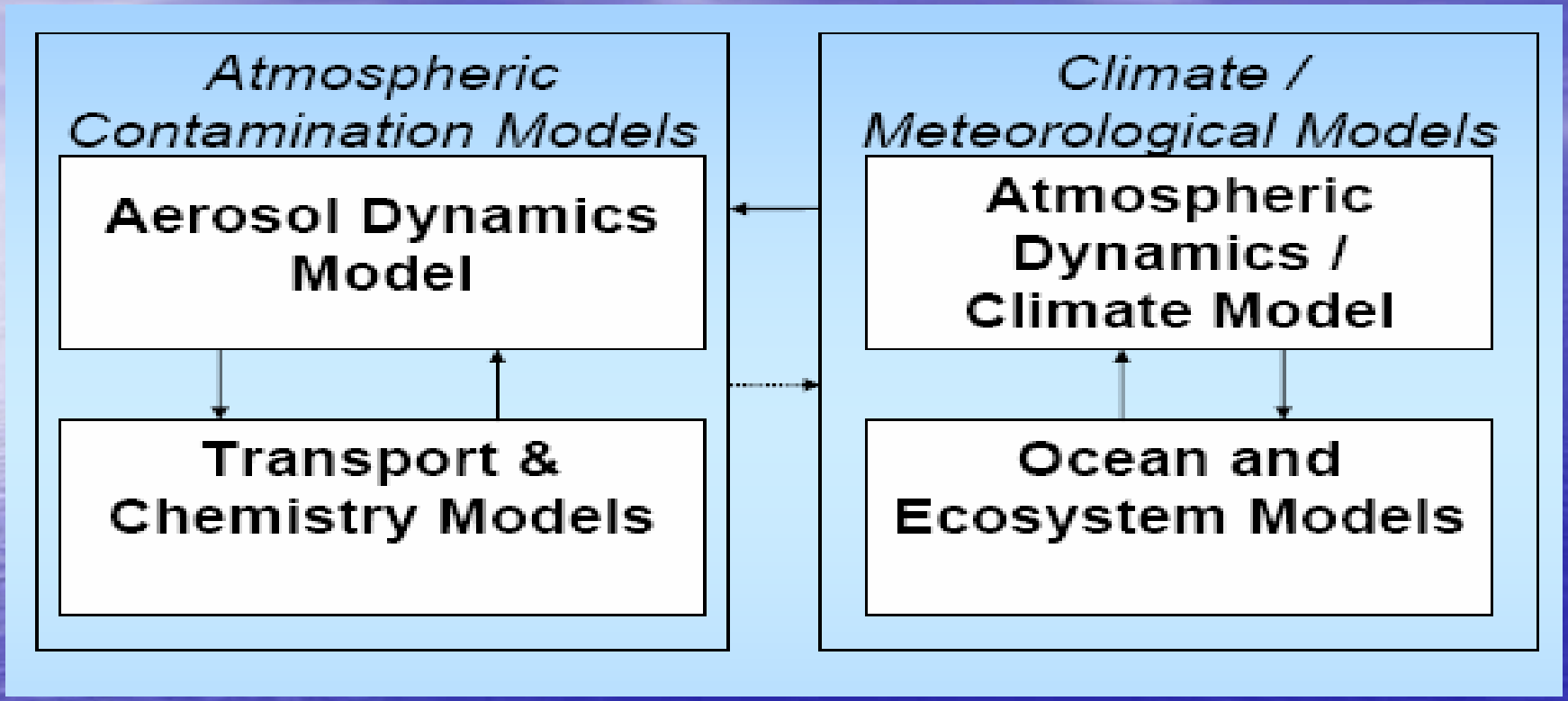
RSHU

Chemical Solvers Benchmark Testing

Sergei Smyshlyaev
Russian State Hydrometeorological University
St.Petersburg, Russia
smyshl@rshu.ru

Integrated Model Structure



- After A.Baklanov "**Integrated systems: on-line and off-line coupling of meteorological and air quality models, advantages and disadvantages**"

Objectives

- The performance of numerical technique in solving stiff differential equations of chemistry is the most important factor in determining the overall computational cost for any integrated model
- Operator splitting technique is the fundamental computational framework of the almost all integrated models
- Chemical transformations may be treated separately and effective chemical solvers may be constructed to use in the atmospheric models of different type

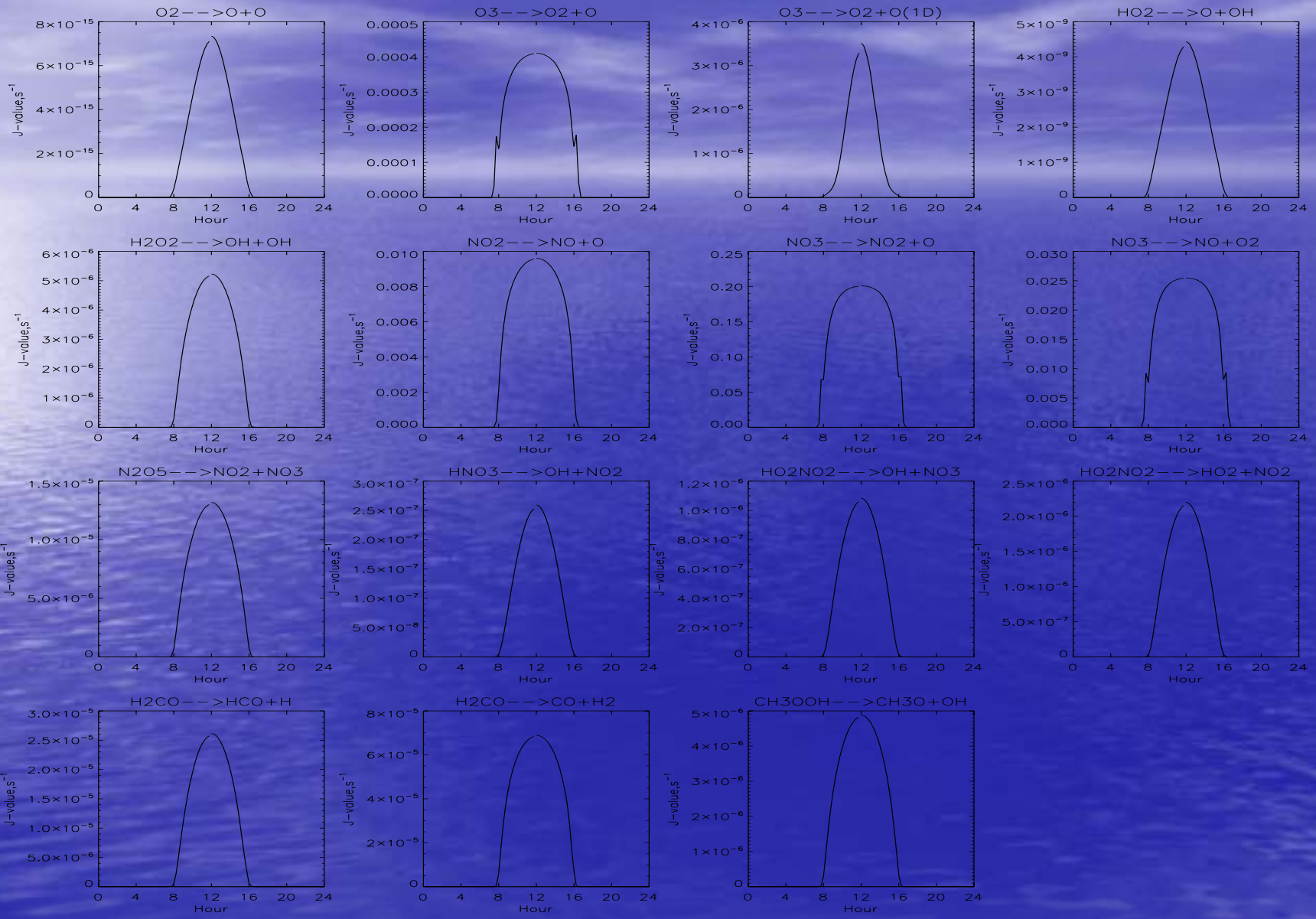
Research Goal

- To study how the different chemical solvers affect the accuracy and computational cost of the box chemistry model based on comparison to benchmark calculation for one spatial point

Benchmark

- Diurnal calculations of short-lived species at 55N, 30E, 10 km altitude for November 15
- 38 gases are calculated, but long-lived species (O3, H2O, CO, CH4 etc) are fixed
- Photolysis rates are precalculated for every 15 min
- Repeating diurnal cycle for 1000 days

Photolysis Rates



Baseline run

- Gear method was used
- Multistep method

$$\mu^{n+1} = \sum_{k=0}^{K} \alpha_k \mu^{n-k} + \Delta t \sum_{k=-1}^{K} \gamma_k S(t^{n-k}, \mu^{n-k})$$

- Backward differentiation formula (BDF)

$$\mu^{n+1} = \sum_{k=0}^{K} \alpha_k \mu^{n-k} + \Delta t \cdot \gamma_{-1} S(t^{n+1}, \mu^{n=1})$$

- Time step and order of approximation are varied depending on tolerance
- Need to solve equations containing large and sparse matrices

Quasi-Steady State Approximation

- Linearization around equilibrium solution

$$\mu_i^{n+1} = \frac{P_i(\mu^n, t_{n+1})}{D(\mu^n, t_{n+1})} + \left[\mu_i^n - \frac{P_i(\mu^n, t_{n+1})}{D(\mu^n, t_{n+1})}\right] \exp\left[-D(\mu^n, t_{n+1})\Delta t\right]$$

- For long-lived species its concentration is calculated with the forward Euler method

- For very short-lived species its concentration is calculated using steady-state equation

Multistep Implicit-Explicit (MIE) Method

- Linearized backward Euler solution – to estimate production and loss terms

$$\mu_{(m+1)}^{n+1} = \frac{\mu^n + \Delta t \cdot P(t_{n+1}, \mu_{(m)}^{n+1})}{1 + D(t_{n+1}, \mu_{(m)}^{n+1})}$$

- Forward Euler appoximation – for final concentration

$$\mu_{(m+1)}^{n+1} = \mu^n + \Delta t \cdot [P(t_{n+1}, \mu_{(m)}^{n+1}) - D(t_{n+1}, \mu_{(m)}^{n+1}) \mu_{(m+1)}^{n+1}]$$

A-Stable Shimazaki Method

- Linearized backward Euler solution

$$\mu_{(m+1)}^{n+1} = \mu^{n} + \alpha \cdot \Delta t \cdot [P(t_{n+1}, \mu_{(m)}^{n+1}) - D(t_{n+1}, \mu_{(m)}^{n+1}) \mu_{(m+1)}^{n+1}] + (1-\alpha) \cdot \mu_{m}^{n+1}$$

- Coefficient α is chosen based on A-stability condition
- Iterative procedure

$$\mu_{(m+1)}^{n+1} = \alpha \frac{\mu^{n} + \Delta t \cdot P(t_{n+1}, \mu_{(m)}^{n+1})}{1 + D(t_{n+1}, \mu_{(m)}^{n+1})} + (1-\alpha) \mu_{m}^{n+1}$$

Computational Time

(SUN Ultra 20 Workstation)

Method	CPU time (sec)
Gear	488
Newton-Raphson	341
QSSA	37
MIE	58
Shimazaki	49

Diurnally Averaged Difference from Gear (Newton-Raphson)

1	OX	3.116E-06	3.116E-06	.00
2	NOY	1.020E-08	1.020E-08	.00
3	CLY	1.778E-09	1.778E-09	.00
4	BRY	1.567E-11	1.567E-11	.00
5	O	5.144E-13	5.134E-13	-.20
6	O(1D)	2.836E-20	2.838E-20	.04
7	O3	3.116E-06	3.116E-06	.00
8	NO	2.656E-11	2.641E-11	-.56
9	NO2	1.791E-10	1.774E-10	-.99
10	NO3	2.041E-12	2.040E-12	-.04
11	N2O5	1.317E-10	1.297E-10	-1.50
12	HNO3	9.082E-09	9.089E-09	.07
13	HO2NO2	8.608E-11	8.365E-11	-2.91
14	H2O	4.046E-06	4.046E-06	.00
15	H	6.307E-21	6.312E-21	.09
16	OH	6.044E-14	6.063E-14	.32
17	HO2	7.072E-13	7.095E-13	.33
18	H2O2	7.931E-12	7.979E-12	.61
19	H2	5.150E-07	5.150E-07	.00
20	CH4	1.214E-06	1.214E-06	.00
21	CH3	5.297E-22	5.313E-22	.29
22	CH3O	6.854E-19	6.874E-19	.28
23	CH3O2	2.195E-13	2.218E-13	1.08
24	CH3OOH	3.237E-12	3.273E-12	1.08
25	HCO	1.701E-23	1.702E-23	.07
26	H2CO	6.074E-12	6.089E-12	.24

Diurnally Averaged Difference from Gear (QSSA)

1	OX	3.116E-06	3.116E-06	.00
2	NOY	1.020E-08	1.020E-08	.00
3	CLY	1.778E-09	1.778E-09	.00
4	BRY	1.567E-11	1.567E-11	.00
5	O	5.144E-13	5.130E-13	-.27
6	O(1D)	2.836E-20	2.838E-20	.04
7	O3	3.116E-06	3.116E-06	.00
8	NO	2.656E-11	1.511E-11	-75.84
9	NO2	1.791E-10	9.959E-11	-79.87
10	NO3	2.041E-12	1.966E-12	-3.82
11	N2O5	1.317E-10	6.597E-11	-99.58
12	HNO3	9.082E-09	9.319E-09	2.54
13	HO2NO2	8.608E-11	6.068E-11	-41.86
14	H2O	4.046E-06	4.046E-06	.00
15	H	6.307E-21	6.738E-21	6.40
16	OH	6.044E-14	6.542E-14	7.61
17	HO2	7.072E-13	8.468E-13	16.49
18	H2O2	7.931E-12	1.116E-11	28.96
19	H2	5.150E-07	5.150E-07	.00
20	CH4	1.214E-06	1.214E-06	.00
21	CH3	5.297E-22	5.182E-22	-2.24
22	CH3O	6.854E-19	6.699E-19	-2.32
23	CH3O2	2.195E-13	4.211E-13	47.89
24	CH3OOH	3.237E-12	6.215E-12	47.91
25	HCO	1.701E-23	1.668E-23	-1.93
26	H2CO	6.074E-12	6.073E-12	-.02

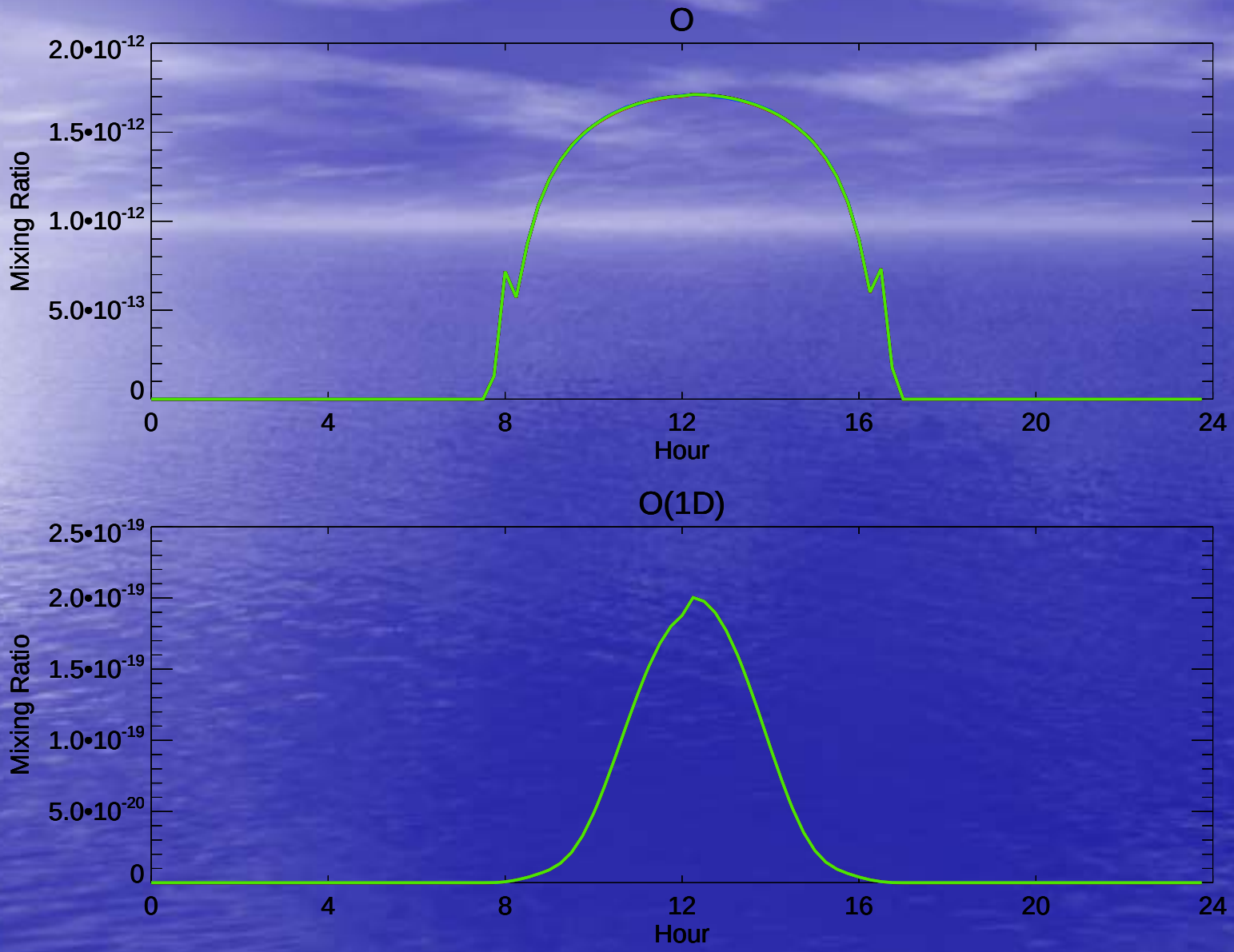
Diurnally Averaged Difference from Gear (MIE)

1	OX	3.116E-06	3.116E-06	.00
2	NOY	1.020E-08	1.020E-08	.00
3	CLY	1.778E-09	1.778E-09	.00
4	BRY	1.567E-11	1.567E-11	.00
5	O	5.144E-13	5.133E-13	-.22
6	O(1D)	2.836E-20	2.838E-20	.04
7	O3	3.116E-06	3.116E-06	.00
8	NO	2.656E-11	2.529E-11	-5.03
9	NO2	1.791E-10	1.731E-10	-3.50
10	NO3	2.041E-12	2.015E-12	-1.29
11	N2O5	1.317E-10	1.271E-10	-3.55
12	HNO3	9.082E-09	9.199E-09	1.28
13	HO2NO2	8.608E-11	8.265E-11	-4.14
14	H2O	4.046E-06	4.046E-06	.00
15	H	6.307E-21	5.962E-21	-5.78
16	OH	6.044E-14	5.978E-14	-1.10
17	HO2	7.072E-13	7.167E-13	1.34
18	H2O2	7.931E-12	8.196E-12	3.23
19	H2	5.150E-07	5.150E-07	.00
20	CH4	1.214E-06	1.214E-06	.00
21	CH3	5.297E-22	4.876E-22	-8.63
22	CH3O	6.854E-19	6.299E-19	-8.81
23	CH3O2	2.195E-13	2.097E-13	-4.65
24	CH3OOH	3.237E-12	3.185E-12	-1.65
25	HCO	1.701E-23	1.559E-23	-9.07
26	H2CO	6.074E-12	5.618E-12	-8.12

Diurnally Averaged Difference from Gear (Shimazaki)

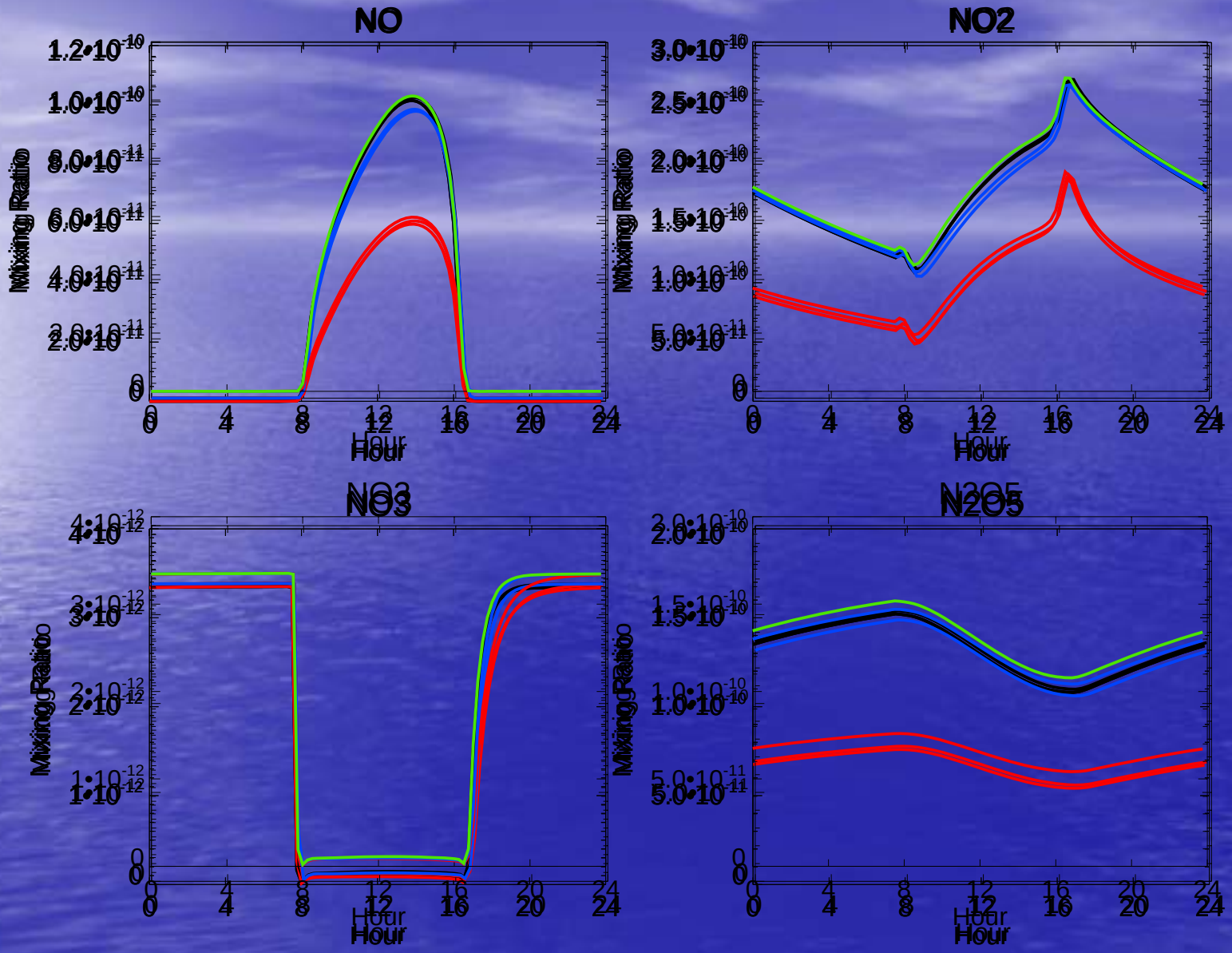
1	OX	3.116E-06	3.116E-06	.00
2	NOY	1.020E-08	1.020E-08	.00
3	CLY	1.778E-09	1.778E-09	.00
4	BRY	1.567E-11	1.567E-11	.00
5	O	5.144E-13	5.134E-13	-.20
6	O(1D)	2.836E-20	2.838E-20	.04
7	O3	3.116E-06	3.116E-06	.00
8	NO	2.656E-11	2.653E-11	-.11
9	NO2	1.791E-10	1.789E-10	-.13
10	NO3	2.041E-12	2.040E-12	-.07
11	N2O5	1.317E-10	1.314E-10	-.21
12	HNO3	9.082E-09	9.082E-09	.00
13	HO2NO2	8.608E-11	8.612E-11	.05
14	H2O	4.046E-06	4.046E-06	.00
15	H	6.307E-21	6.296E-21	-.17
16	OH	6.044E-14	6.049E-14	.08
17	HO2	7.072E-13	7.078E-13	.09
18	H2O2	7.931E-12	7.942E-12	.13
19	H2	5.150E-07	5.150E-07	.00
20	CH4	1.214E-06	1.214E-06	.00
21	CH3	5.297E-22	5.301E-22	.07
22	CH3O	6.854E-19	6.858E-19	.05
23	CH3O2	2.195E-13	2.212E-13	.80
24	CH3OOH	3.237E-12	3.251E-12	.41
25	HCO	1.701E-23	1.698E-23	-.14
26	H2CO	6.074E-12	6.077E-12	.04

Ox gases



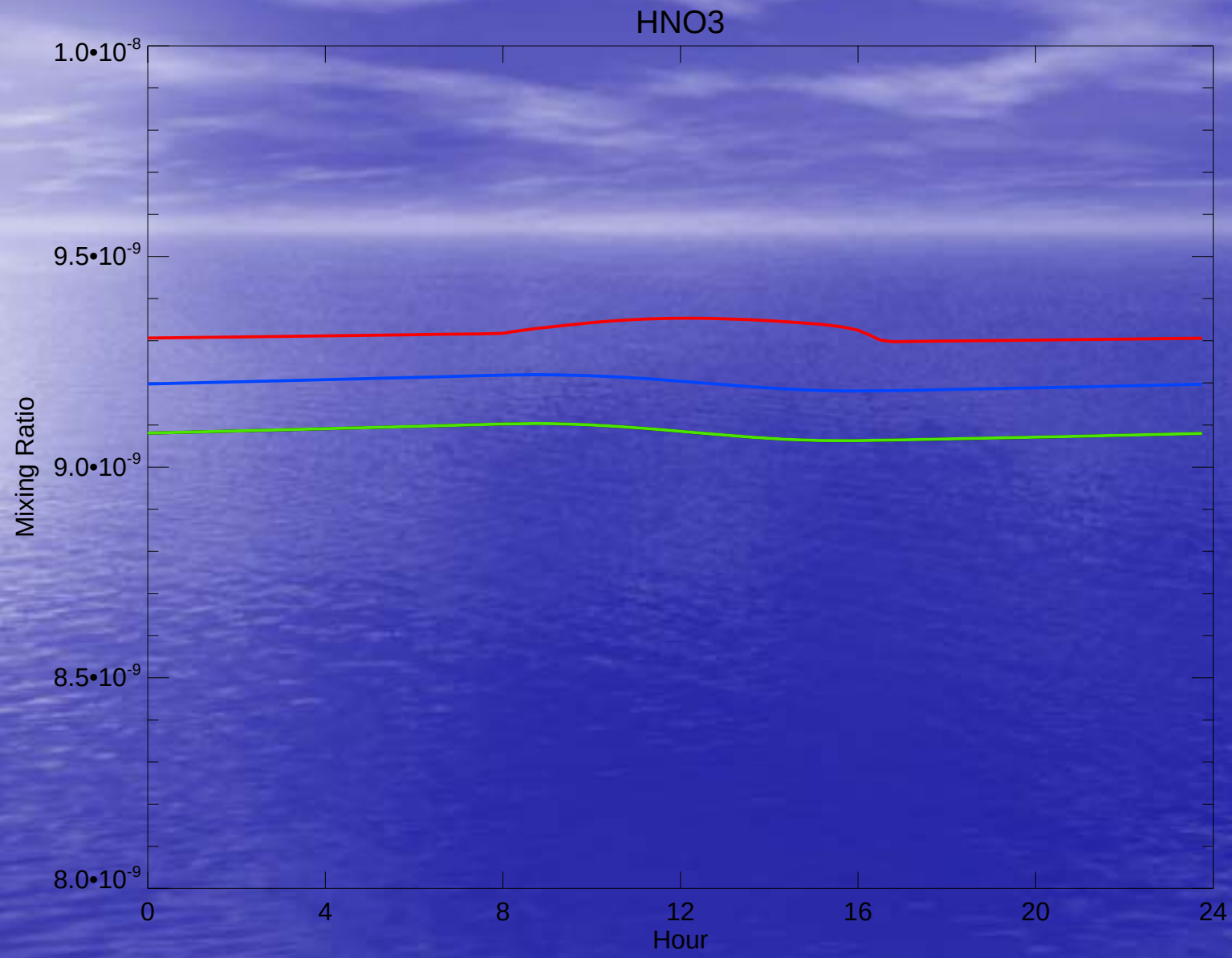
Black – Gear, Red – QSSA, Blue – MIE, Green - Shimazaki

NOx gases



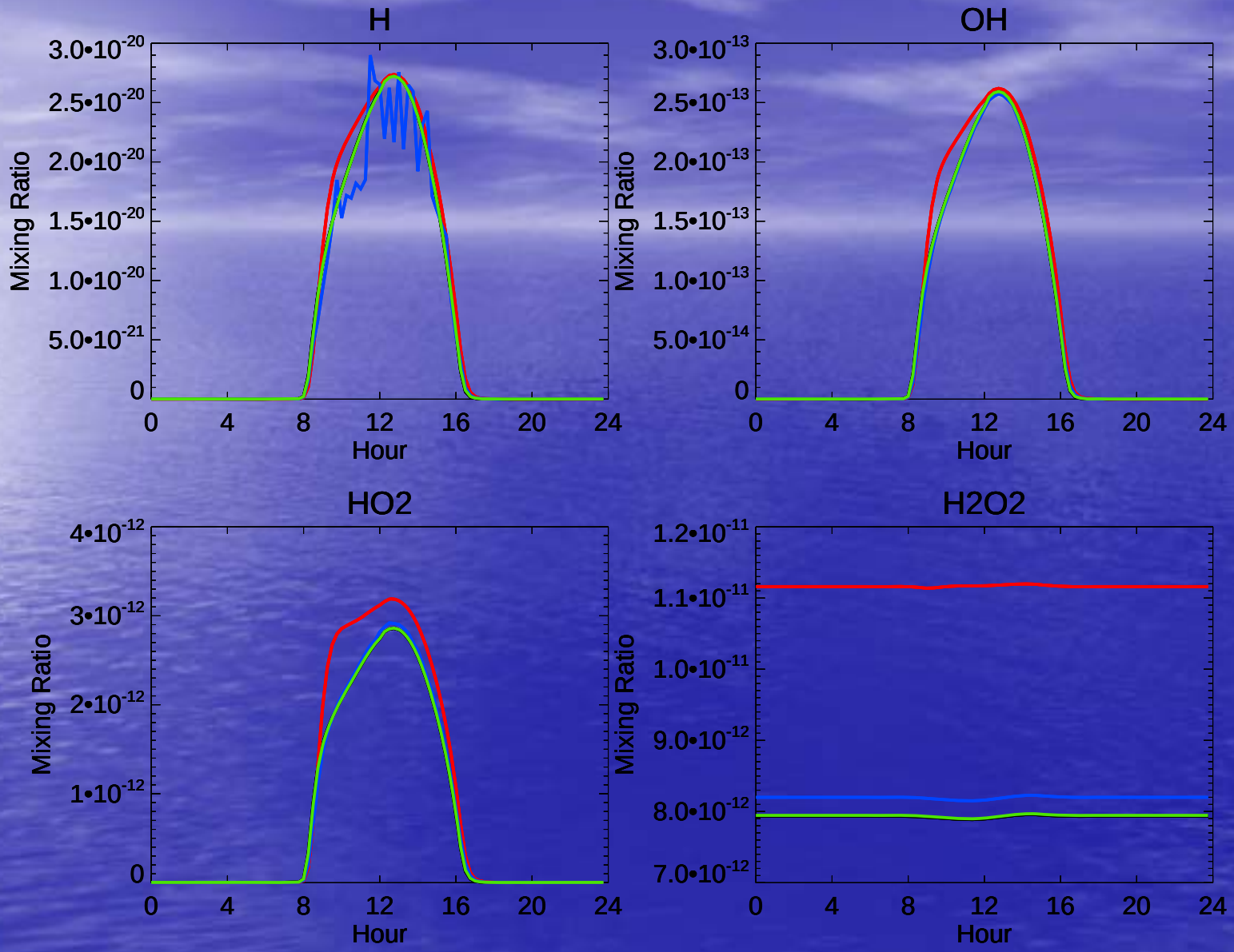
Black – Gear, Red – QSSA, Blue – MIE, Green - Shimazaki

Nitric Acid



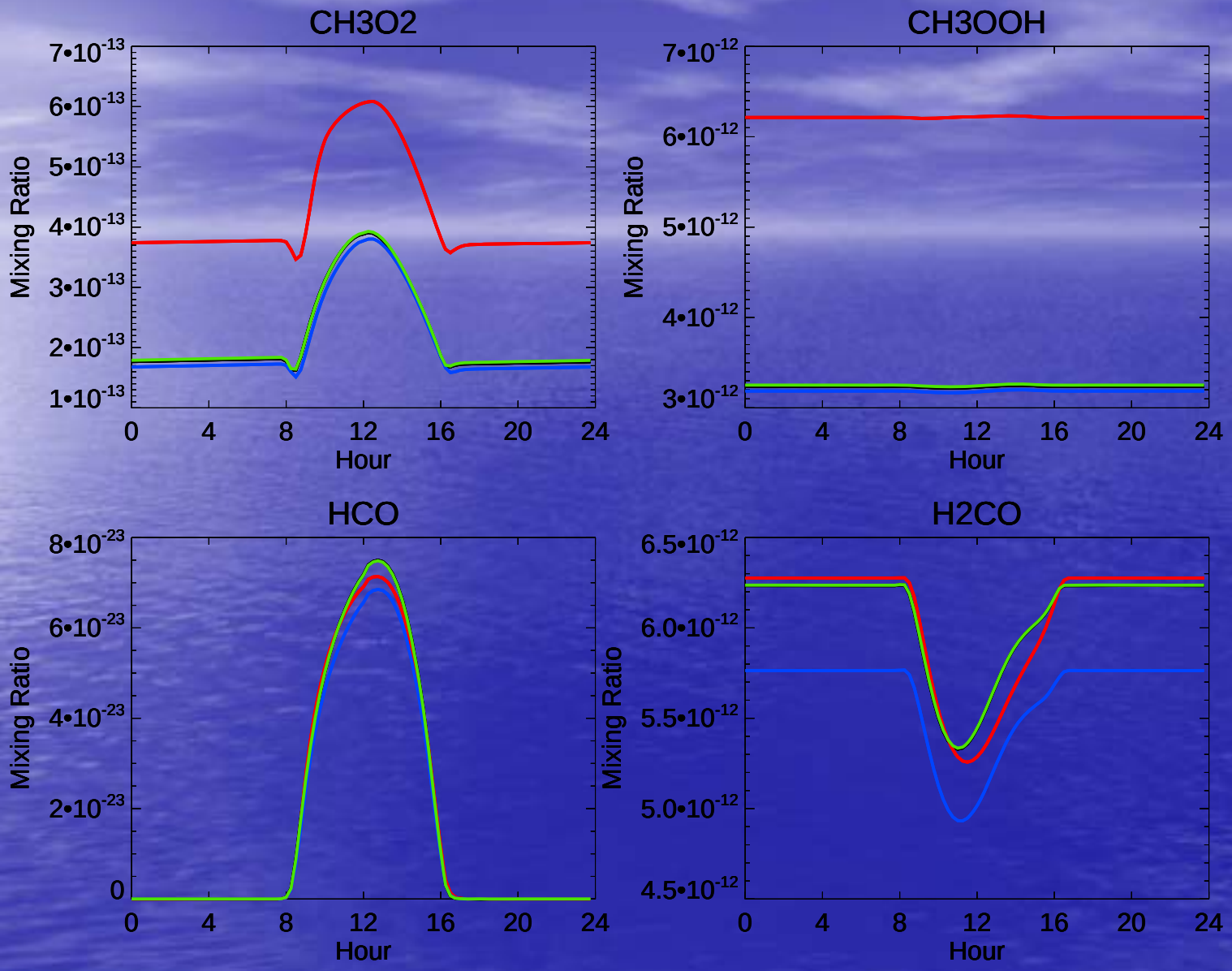
Black – Gear, Red – QSSA, Blue – MIE, Green - Shimazaki

HOx gases



Black – Gear, Red – QSSA, Blue – MIE, Green - Shimazaki

Carbon gases



Black – Gear, Red – QSSA, Blue – MIE, Green - Shimazaki

Additional Runs

- Tried to reach the same accuracy like Shimazaki method and compare CPU time
- For QSSA method – reduced time step and iteration technique
- For MIE method – modified criteria for convergence

QSSA with 10 times reduced time step (CPU time 198 sec)

1	OX	3.116E-06	3.116E-06	.00
2	NOY	1.020E-08	1.020E-08	.00
3	CLY	1.778E-09	1.778E-09	.00
4	BRY	1.567E-11	1.567E-11	.00
5	O	5.144E-13	5.133E-13	-.21
6	O(1D)	2.836E-20	2.838E-20	.04
7	O3	3.116E-06	3.116E-06	.00
8	NO	2.656E-11	2.515E-11	-5.60
9	NO2	1.791E-10	1.707E-10	-4.94
10	NO3	2.041E-12	2.018E-12	-1.13
11	N2O5	1.317E-10	1.236E-10	-6.56
12	HNO3	9.082E-09	9.103E-09	.23
13	HO2NO2	8.608E-11	8.311E-11	-3.57
14	H2O	4.046E-06	4.046E-06	.00
15	H	6.307E-21	6.358E-21	.81
16	OH	6.044E-14	6.112E-14	1.11
17	HO2	7.072E-13	7.188E-13	1.62
18	H2O2	7.931E-12	8.164E-12	2.86
19	H2	5.150E-07	5.150E-07	.00
20	CH4	1.214E-06	1.214E-06	.00
21	CH3	5.297E-22	5.306E-22	.15
22	CH3O	6.854E-19	6.856E-19	.03
23	CH3O2	2.195E-13	2.341E-13	6.28
24	CH3OOH	3.237E-12	3.440E-12	5.90
25	HCO	1.701E-23	1.707E-23	.35
26	H2CO	6.074E-12	6.116E-12	.69

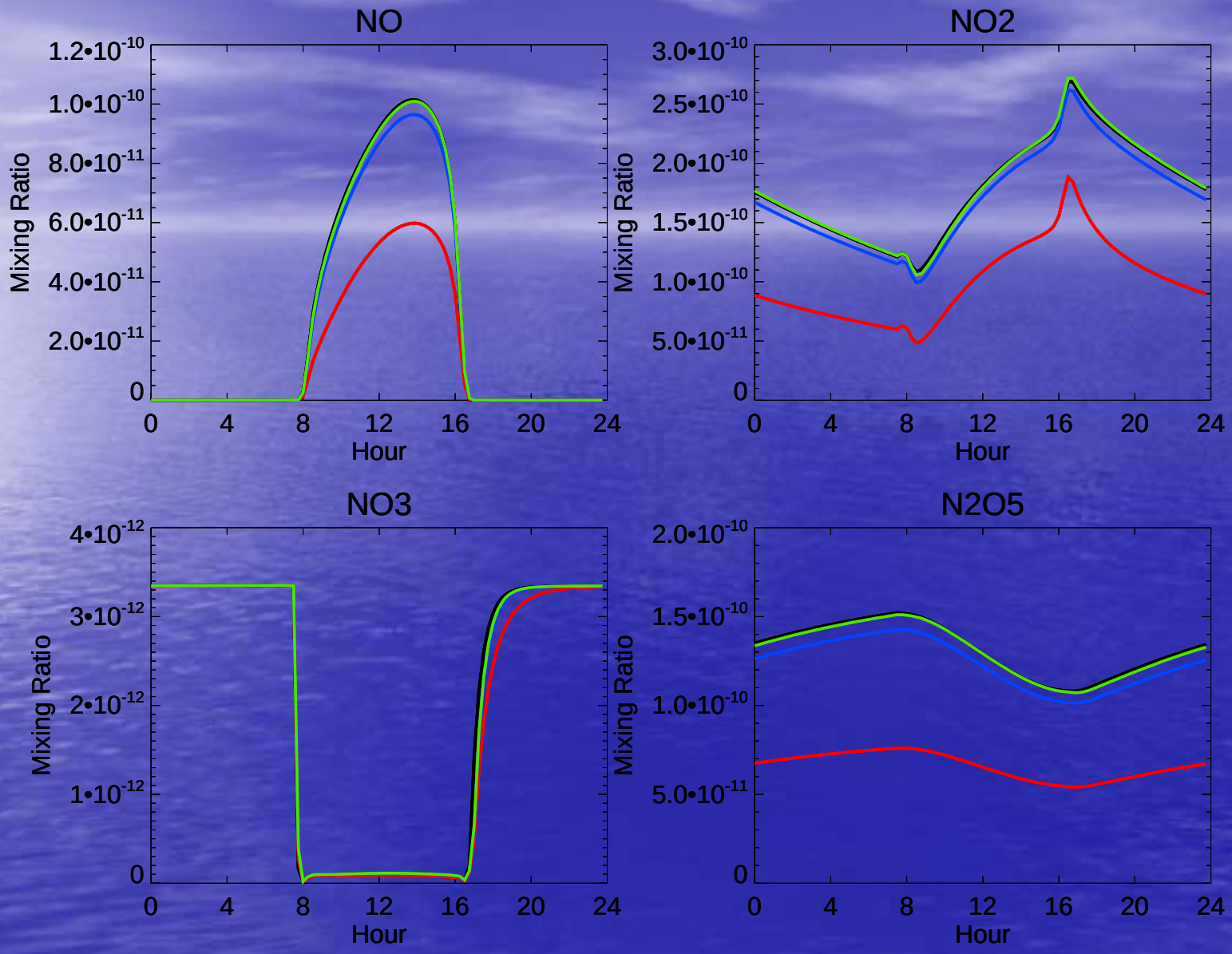
Iterative QSSA
(CPU time 1609 sec)

1	OX	3.116E-06	3.116E-06	.00
2	NOY	1.020E-08	1.020E-08	.00
3	CLY	1.778E-09	1.778E-09	.00
4	BRY	1.567E-11	1.567E-11	.00
5	O	5.144E-13	5.134E-13	-.20
6	O(1D)	2.836E-20	2.838E-20	.04
7	O3	3.116E-06	3.116E-06	.00
8	NO	2.656E-11	2.636E-11	-.78
9	NO2	1.791E-10	1.796E-10	.24
10	NO3	2.041E-12	2.022E-12	-.93
11	N2O5	1.317E-10	1.307E-10	-.74
12	HNO3	9.082E-09	9.073E-09	-.10
13	HO2NO2	8.608E-11	8.492E-11	-1.36
14	H2O	4.046E-06	4.046E-06	.00
15	H	6.307E-21	6.307E-21	.01
16	OH	6.044E-14	6.054E-14	.17
17	HO2	7.072E-13	7.083E-13	.16
18	H2O2	7.931E-12	7.940E-12	.12
19	H2	5.150E-07	5.150E-07	.00
20	CH4	1.214E-06	1.214E-06	.00
21	CH3	5.297E-22	5.324E-22	.50
22	CH3O	6.854E-19	6.880E-19	.37
23	CH3O2	2.195E-13	2.215E-13	.94
24	CH3OOH	3.237E-12	3.267E-12	.90
25	HCO	1.701E-23	1.712E-23	.65
26	H2CO	6.074E-12	6.131E-12	.92

Modified MIE
(CPU time 103 sec)

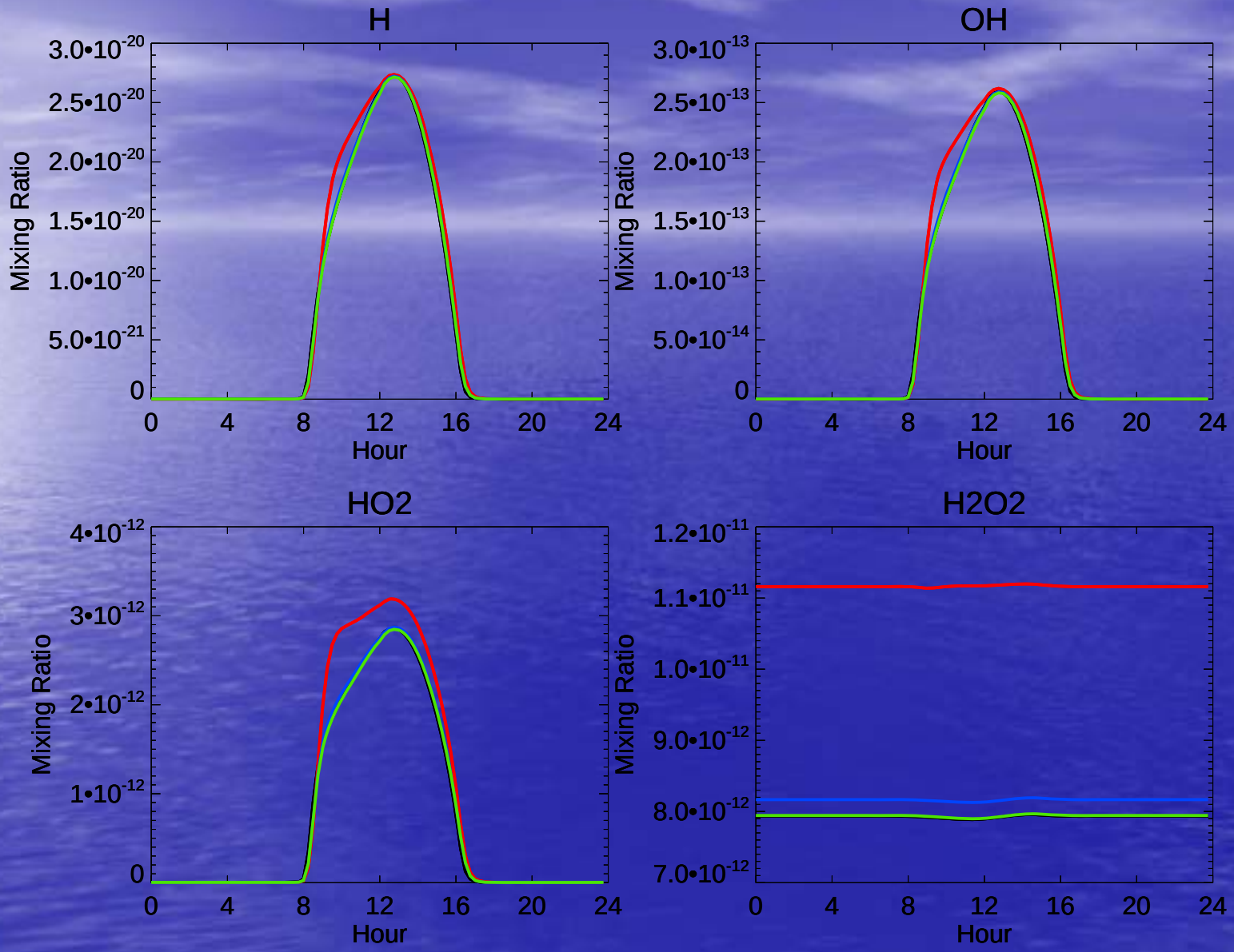
1	OX	3.116E-06	3.116E-06	.00
2	NOY	1.020E-08	1.020E-08	.00
3	CLY	1.778E-09	1.778E-09	.00
4	BRY	1.567E-11	1.567E-11	.00
5	O	5.144E-13	5.133E-13	-.21
6	O(1D)	2.836E-20	2.838E-20	.04
7	O3	3.116E-06	3.116E-06	.00
8	NO	2.656E-11	2.644E-11	-.44
9	NO2	1.791E-10	1.802E-10	.60
10	NO3	2.041E-12	2.022E-12	-.93
11	N2O5	1.317E-10	1.315E-10	-.12
12	HNO3	9.082E-09	9.081E-09	-.01
13	HO2NO2	8.608E-11	8.641E-11	.39
14	H2O	4.046E-06	4.046E-06	.00
15	H	6.307E-21	6.120E-21	-3.05
16	OH	6.044E-14	6.041E-14	-.05
17	HO2	7.072E-13	7.081E-13	.13
18	H2O2	7.931E-12	7.922E-12	-.12
19	H2	5.150E-07	5.150E-07	.00
20	CH4	1.214E-06	1.214E-06	.00
21	CH3	5.297E-22	5.278E-22	-.38
22	CH3O	6.854E-19	6.818E-19	-.53
23	CH3O2	2.195E-13	2.194E-13	-.01
24	CH3OOH	3.237E-12	3.232E-12	-.15
25	HCO	1.701E-23	1.695E-23	-.33
26	H2CO	6.074E-12	6.072E-12	-.03

QSSA NOx gases



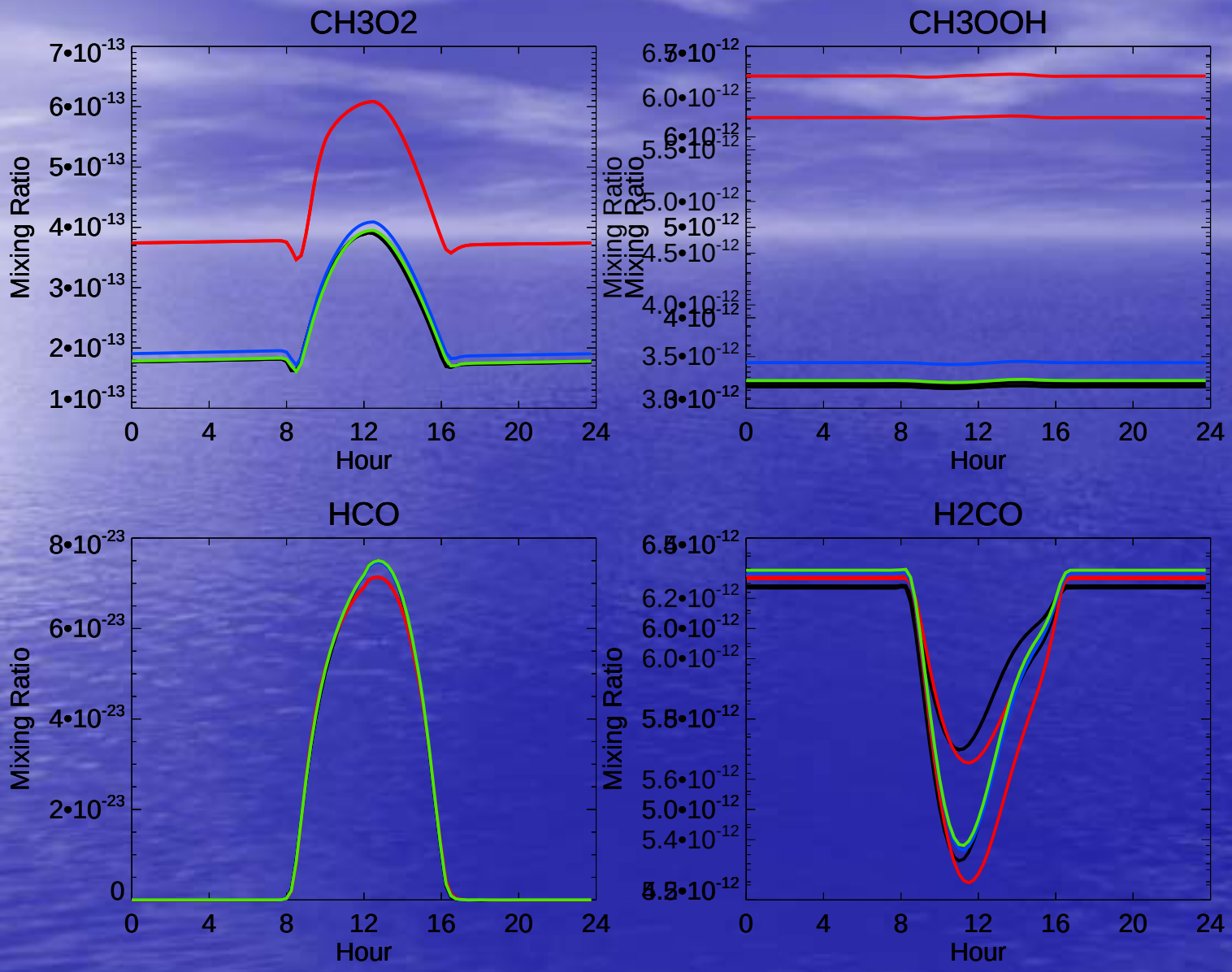
Black – Gear, Red – regQSSA, Blue – iterQSSA, Green - stepQSSA

QSSA HOx gases



Black – Gear, Red – regQSSA, Blue – iterQSSA, Green - stepQSSA

QSSA Carbon gases



Black – Gear, Red – regQSSA, Blue – iterQSSA, Green - stepQSSA

Conclusions

- The chemical solver constructed based on Shimazaki scheme has the best overall performance
- Newton-Raphson solver may need reduced time step that may lead to additional computational costs
- QSSA method needs reduced time step and iteractive technique
- MIE solver needs careful tuning