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*Supplement of*

**Evaluating simplified chemical mechanisms within present-day simulations of the Community Earth System Model version 1.2 with CAM4 (CESM1.2 CAM-chem): MOZART-4 vs. Reduced Hydrocarbon vs. Super-Fast chemistry**

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	CO	NO	NO2	SO2	C2H4	CH2O	CH3CHO	TOLUENE	NH3	ISOP	C10H16	OLE	PAR
CO	1												
NO		1											
NO2			1										
SO2				1									
BIGALK													5
BIGENE												1	2
C2H4					1								
C2H5OH													
C2H6													0.4
C3H6												1	1
C3H8													1.5
CH2O						1							
CH3CHO							1						
CH3COCH3													2
CH3OH													1
MEK													3
TOLUENE								1					
NH3									1				
ISOP										1			
C10H16											1		

**Supplemental Table S1:** Mapping of MOZART species (rows) to the lumped Reduced Hydrocarbon mechanism species

5 (columns)

<b>Photolysis Reactions</b>			
<b>Reactant</b>	<b>Products</b>		<b>Relation to Emmons et al. (2010)</b>
(i)	O <sub>3</sub> + hv	→ 2·OH	simplified, includes reaction of O( <sup>1</sup> D) with H <sub>2</sub> O
(ii)	H <sub>2</sub> O <sub>2</sub> + hv	→ 2·OH	identical
(iii)	NO <sub>2</sub> + hv	→ NO + O <sub>3</sub>	simplified, O → O <sub>3</sub>
(iv)	CH <sub>2</sub> O + hv	→ CO + 2·HO <sub>2</sub>	identical
(v)	CH <sub>2</sub> O + hv	→ CO	simplified, no H <sub>2</sub>
(vi)	CH <sub>3</sub> OOH + hv	→ CH <sub>2</sub> O + HO <sub>2</sub> + OH	simplified, H → HO <sub>2</sub>

  

<b>Gas-Phase Reactions</b>				
<b>Reactants</b>	<b>Products</b>	<b>Rate</b>	<b>Relation to Emmons et al. (2010)</b>	
(1)	O <sub>3</sub> + OH	→ HO <sub>2</sub> + O <sub>2</sub>	1.70E-12·exp(-940/T)	identical
(2)	HO <sub>2</sub> + O <sub>3</sub>	→ 2·O <sub>2</sub> + OH	1.00E-14·exp(-490/T)	identical
(3)	HO <sub>2</sub> + OH	→ H <sub>2</sub> O + O <sub>2</sub>	4.80E-11·exp(250/T)	identical
(4)	HO <sub>2</sub> + HO <sub>2</sub>	→ H <sub>2</sub> O <sub>2</sub> + O <sub>2</sub>	(2.3E-13·exp(600/T)+1.7E-33·[M]·exp(1000/T))·(1 + 1.4E-21·[H <sub>2</sub> O]·exp(2200/T))	identical
(5)	H <sub>2</sub> O <sub>2</sub> + OH	→ H <sub>2</sub> O + HO <sub>2</sub>	1.80E-12	identical
(6)	NO + O <sub>3</sub>	→ NO <sub>2</sub> + O <sub>2</sub>	3.00E-12·exp(-1500/T)	identical
(7)	HO <sub>2</sub> + NO	→ NO <sub>2</sub> + OH	3.50E-12·exp(250/T)	different rates
(8)	NO <sub>2</sub> + OH + M	→ HNO <sub>3</sub>	ko=1.80E-30(300/T) <sup>3.00</sup> ; ki=2.80E-11; f=0.60	identical
(9)	CH <sub>4</sub> + OH	→ CH <sub>3</sub> O <sub>2</sub> + H <sub>2</sub> O	2.45E-12·exp(-1775/T)	identical
(10)	CO + OH	→ HO <sub>2</sub>	ko_m/(1+(ko_m/k_c))·0.6·exp((1/(1+log(ko_m/k_c))) <sup>2</sup> )+(k_0/(1+(k_0/k_c_m)))-0.6·exp((1/(1+log(k_0/k_c_m))) <sup>2</sup> ))	simplified: no CO <sub>2</sub> , H → HO <sub>2</sub> , see note A
(11)	CH <sub>2</sub> O + OH	→ CO + H <sub>2</sub> O + HO <sub>2</sub>	5.50E-12·exp(125/T)	rates identical, simplified: H → HO <sub>2</sub>
(12)	CH <sub>3</sub> O <sub>2</sub> + HO <sub>2</sub>	→ CH <sub>3</sub> OOH + O <sub>2</sub>	4.10E-13·exp(750/T)	identical
(13a)	CH <sub>3</sub> OOH + OH	→ CH <sub>3</sub> O <sub>2</sub> + H <sub>2</sub> O	2.70E-12·exp(200/T)	in combination, equivalent
(13b)	CH <sub>3</sub> OOH + OH	→ CH <sub>3</sub> O + H <sub>2</sub> O + OH	1.10E-12·exp(200/T)	
(14)	CH <sub>3</sub> O <sub>2</sub> + NO	→ CH <sub>2</sub> O + HO <sub>2</sub> + NO <sub>2</sub>	2.80E-12·exp(300/T)	identical
(15)	CH <sub>3</sub> O <sub>2</sub> + CH <sub>3</sub> O <sub>2</sub>	→ 2·CH <sub>2</sub> O + 0.80·HO <sub>2</sub>	9.50E-14·exp(390/T)	different rates, simplified: 1 reaction instead of 2
(16)	H <sub>2</sub> O + NO <sub>2</sub>	→ 0.50·HNO <sub>3</sub>	4.00E-24	no equivalent in MOZART, see note B
(17a)	DMS + OH	→ SO <sub>2</sub>	1.100E-11·exp(-240/T)	different, see note C
(17b)	DMS + OH	→ 0.75·SO <sub>2</sub>	2.00E-10·exp(5820·[M])/((2.00E29/[O <sub>2</sub> ]) + exp(6280·[M]))	
(18)	OH + SO <sub>2</sub> + M	→ SO <sub>4</sub>	ko=3.30E-31(300/T) <sup>4.30</sup> ; ki=1.60E-12; f=0.60	different, see note C
(19)	H <sub>2</sub> O <sub>2</sub> + SO <sub>2</sub>	→ SO <sub>4</sub>	aqueous chemistry (see note D)	no equivalent, see note C
(20)	O <sub>3</sub> + SO <sub>2</sub>	→ SO <sub>4</sub>	aqueous chemistry (see note D)	no equivalent, see note C
(21a)	ISOP + OH	→ 2·CH <sub>3</sub> O <sub>2</sub>	2.70E-11·exp(390/T)	different, see note E
(21b)	ISOP + OH	→ ISOP	2.70E-11·exp(390/T)	
(21c)	ISOP + OH	→ ISOP + 0.5·OH	2.70E-11·exp(390/T)	different, see note E
(22)	ISOP + O <sub>3</sub>	→ .87·CH <sub>2</sub> O + 1.86·CH <sub>3</sub> O <sub>2</sub> + 0.06·HO <sub>2</sub> + 0.05·CO	5.59E-15·exp(-1814/T)	

**NOTES:**

A: For rate: ko = 5.90E-33·(300/T)<sup>1.4</sup>; k<sub>c</sub> = 1.10E-12·(T/300)<sup>1.3</sup>; ko\_m = ko·[M]; k<sub>0</sub> = 1.50E-13·(T/300)<sup>0.6</sup>; k<sub>c\_m</sub> = (2.10E9·(T/300)<sup>6.1</sup>)/[M]

B: HNO<sub>3</sub> chemistry included only as reaction 8 and 16, with reaction 16 involving heterogeneous chemistry parameterization

C: DMS chemistry limited only to reaction with OH (reaction 17), SO<sub>4</sub> production simplified to reactions 18 with OH and 19 and 20 with aqueous chemistry (with a fixed pH in the cloud droplets)

D: Rate equations are included within the aerosol routines adapted from the MOZART-4 mechanism implementation within CAM-chem

E: Isoprene chemistry parameterized from UCI for ISOP + OH and from LLNL-IMPACT for ISOP + O<sub>3</sub>, see text for full details

**Table S2:** Full description of Super-Fast chemical mechanism as compared to the MOZART-4 mechanism of Emmons et al. (2010). Reaction rates are written out if they are of the Arrhenius form, or otherwise formulated. If the reaction rates are of the Troe form, they list the ko and ki parameters, as in Emmons et al. (2010). The simplifications made in the SF are noted by indicating what species is missing or modified when compared to Emmons et al. (2010). Chemical species are the same as in Emmons et al. (2010).

		[ppbv]	[ppbv]	[ppbv]	[%]	[ppbv]	[ppbv]	[ppbv]	[%]
		Mean	Median	Standard Deviation	Variability	90th Percentile	99th Percentile	99th - 90th Percentile	
<b>W US</b>	<b>CASTNET</b>	49.9	49.6	4.82	9.66	56.4	61.3	4.87	109
	<b>MO</b>	54.9	54.9	3.37	6.14	59.4	62.4	2.99	105
	<b>RH</b>	55.9	55.8	3.58	6.41	60.6	64.0	3.47	106
	<b>SF</b>	50.5	50.5	3.22	6.38	54.7	57.8	3.15	106
<b>MW US</b>	<b>CASTNET</b>	57.3	57.0	7.57	13.2	67.1	76.9	9.77	115
	<b>MO</b>	71.0	70.7	11.03	15.5	85.5	96.0	10.6	112
	<b>RH</b>	72.6	72.1	11.26	15.5	87.4	98.8	11.4	113
	<b>SF</b>	70.6	69.8	13.44	19.0	88.7	104	15.1	117
<b>MW US single grid cell</b>	<b>CASTNET</b>	62.4	60.6	14.5	23.2	81.7	104	22.7	128
	<b>MO</b>	84.6	83.8	16.2	19.2	106	126	19.4	118
	<b>RH</b>	86.2	85.3	15.8	18.4	107	126	19.5	118
	<b>SF</b>	85.4	82.5	22.4	26.2	116	149	32.8	128
<b>SE US</b>	<b>CASTNET</b>	51.5	51.3	8.18	15.9	62.3	71.9	9.64	115
	<b>MO</b>	60.2	59.5	9.22	15.3	72.5	83.7	11.2	115
	<b>RH</b>	61.3	60.5	10.1	16.6	74.9	87.9	12.9	117
	<b>SF</b>	58.6	57.8	11.4	19.4	73.9	87.1	13.2	118
<b>SE US single grid cell</b>	<b>CASTNET</b>	53.8	53.1	12.8	23.8	68.7	93.5	24.8	136
	<b>MO</b>	83.4	84.7	19.1	22.9	107	124	17.3	116
	<b>RH</b>	85.2	86.1	20.4	24.0	111	131	20.1	118
	<b>SF</b>	83.6	82.1	27.0	32.3	120	153	33.1	128

**Table S3:** Summary Statistics for the Daily Maximum 8-Hour (MDA8) O<sub>3</sub> over the globe other regions, accompanying Table 2. The last two columns indicate the difference between the 99<sup>th</sup> percentile and the 90<sup>th</sup> percentile, expressed both in absolute values (ppb) and as a percent.

## Supplemental Description of the Super-Fast Chemical Mechanism

The SF mechanism is in the CESM code archive as an unsupported chemical mechanism, which can be activated using the option ‘-chem super\_fast\_llnl’. The SF mechanism has been included in several model inter-comparison projects, including the ACCMIP (e.g. Lamarque et al., 2013a), a comparison of stratospheric dynamics and ozone production (Hsu et al., 2013), a comparison of isoprene mechanisms and ozone changes (Squire et al., 2015), and a multi-model assessment of surface ozone and observations (Schnell et al., 2015). Here we briefly review the findings of these four model inter-comparison projects.

The SF only simulates sulfate (SO<sub>4</sub>) and not the other aerosols, so the SF mechanism was not included in many of the ACCMIP aerosol comparisons (Lamarque et al., 2013a). While the inclusion of non-sulfate aerosols within the CESM can be easily accomplished, there are two aerosol modules (either bulk or modal) to which aerosols could be added, which was beyond the scope of this project, so aerosol model capabilities are not examined in the present study.

We now summarize the ACCMIP results as they pertain to the SF mechanism. Within the ACCMIP, the SF mechanism has lower rates of ozone production and loss compared to the ACCENT models (biases of -24% and -22% respectively), as well as low ozone deposition (bias of -38%) (Young et al., 2013). In this comparison, natural emissions were not prescribed and different treatments of meteorology were used, which may account for some of the noted differences. This results in a high bias for the ozone lifetime (+3 days, or +14%), as well as a low ozone burden bias (-34 Tg, or -10%) (Young et al., 2013). In addition, the models that showed similarly low ozone production and loss rates have lower emissions of VOCs. The SF mechanism falls within the ACCMIP range for human health results due to ozone exposure (Silva et al., 2013). The SF mechanism simulated the historical and future changes in the tropospheric ozone column and radiative forcing within the range of the other ACCMIP models (Stevenson et al., 2013), with SF being near the end of the range that simulated larger changes. The SF mechanism also has a high bias for global-mean OH (+16% compared to the ACCMIP mean) and a low bias for the calculation of the methane lifetime due to OH oxidation (-14%) (Voulgarakis et al., 2013).

The SF mechanism was tested against MOZART by Hsu et al. (2013) who concluded that the selection of a chemical mechanism was only a secondary influence on the stratospheric chemistry since they used a linearized scheme. However, the SF mechanism did produce a less stratified tropopause and a warmer troposphere due largely to the impact of ozone forcings on the simulated dynamics and thermodynamics. Unfortunately, the Hsu et al. (2013) analysis had a bug with their SF simulations, which resulted in the aerosols not being communicated to the cloud nucleation routines, but this didn’t affect their conclusions on the sensitivity of the stratosphere to uncertainty in the O<sub>2</sub> photolysis cross-section.

Squire et al. (2015) compared the SF isoprene scheme with three other schemes of much greater complexity. They concluded that the “1-species, 2-reaction” isoprene scheme from the SF mechanism, as simple as it is, is preferable to neglecting biogenic chemistry entirely, although the SF mechanism shows the highest biases in regions where isoprene chemistry is important for simulating accurate ozone concentrations. They also explored some of the other biases within the SF mechanism scheme, which include: (1) under high-isoprene conditions, the SF mechanism overestimates O<sub>3</sub>; (2) under

low-isoprene and low-NO<sub>x</sub> conditions, the SF mechanism overestimates O<sub>3</sub>; (3) due to the simplicity of SF mechanism, HO<sub>x</sub> is sequestered into the organic hydroperoxides, and methyl hydroperoxide (CH<sub>3</sub>OOH) has low reactivity, which results in high levels of the peroxy radicals, an enhanced rate of CH<sub>3</sub>O<sub>2</sub> + NO, and therefore a high bias (up to +80%) for ozone; and (4) the NO<sub>x</sub> lifetime is too short, except in high-NO<sub>x</sub> emission regions. They conclude that the addition of a PAN formation scheme would significantly improve the O<sub>3</sub> distribution. Finally, they find that many of the errors described above largely cancel each other out, which results in the globally averaged O<sub>3</sub> bias for SF mechanism to be small (-2.6% compared to the Master Chemical Mechanism).

The SF mechanism has a known anomalous annual cycle (see Schnell et al., 2015), in which peak ozone occurs in March/April rather than May. In the main article we show that this anomaly exists at global scales, but not within all regions. In addition, the size and extent of ozone pollution episodes is anomalously high, and these large events occur mainly in the springtime (Schnell et al., 2015). Interestingly, the SF mechanism outperforms many of the more sophisticated mechanisms in simulating the observed summertime diurnal cycle for ozone (Schnell et al., 2015).

Other studies that have used SF, or intercompare its results with other atmospheric chemistry models, include: ozone impacts in the Coupled Model Intercomparison Project Phase 5 (CMIP5) (Eyring, et al., 2013); air pollution and mortality (Silva et al., 2016, 2017); the role of dimethyl sulphide (DMS) in climate (Xu et al., 2016; Wang et al., 2018a, 2018b), and other ACCMIP related papers (Fiore et al., 2012; Bowman, et al., 2013; Naik, et al., 2013, Lamarque, et al., 2013b; Kirschke, et al., 2013). Overall, SF produced simulations that were generally acceptable.

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