Supporting Information: Development, intercomparison and evaluation of an improved mechanism for the oxidation of dimethyl sulfide in the UKCA model

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S1 Box model data

S1.1 BOXMOX input files

Table S1: Initial conditions in the box and background air

Species	Mixing ratio (ppm)
M	1.00E+06
N_2	7.80E+05
O_2	2.10E+05
H_2O	1.00E+04
CH_4	1.80E+00
CO	1.00E-01
H_2	5.00E-01
O_3	3.00E-02
H_2O_2	1.00E-03
NO	1.00E-06
NO_2	1.00E-05
HNO_3	5.00E-04
DMS	2.00E-04
SO_2	2.00E-05

Table S2: Diurnal profile of temperature and boundary layer height

Time (h)	Temperature (K)	Boundary Layer Height (m)
0	289.5359	1300
1	289.1363	1300
2	289.0000	1350
3	289.1363	1400
4	289.5359	1450
5	290.1716	1500
6	291.0000	1550
7	291.9647	1450
8	293.0000	1400
9	294.0353	1350
10	295.0000	1300
11	295.8284	1250
12	296.4641	1200
13	296.8637	1200
14	297.0000	1200
15	296.8637	1200
16	296.4641	1150
17	295.8284	1150
18	295.0000	1100
19	294.0353	1200
20	293.0000	1300
21	291.9647	1400
22	291.0000	1400
23	290.1716	1350
24	289.5359	1300

S1.2 Reduction of the HPMTF pathway

HPMTF can have more side reactions than are described in the main paper. Originally, a range of possible reactions were considered based on the literature and reactions that similar molecules undergo (Figure S1, Table S3). Reactions that were assessed include photolysis reactions and aqueous loss of HPMTF as well as reactions of the intermediate formed after the first isomerization step and the oxidation of this by HO_2 and NO, as described by Veres et al. (2020). In the SI, this scheme is referred to as CS2-HPMTF-compl.

To reduce computational time, as few reactions as possible should be included in a mechanism. At the same time, enough reactions need to be included so that the mechanism can faithfully reproduce the time evolution of the species it models. The reactions which are removed from the mechanism should not play a significant role under the majority of atmospheric conditions. In this section, it is tested whether some of the reactions proposed can be removed without changing the concentration of key species. In this case the key species chosen were HPMTF and SO_2 (i.e. the mechanism was optimized to these species) because observational measurements exist for those species.

The BOXMOX runs were set up as described in Section 2.1.1 of the main paper. In the sensitivity runs, temperature, aerosol surface area, and O3 and NO concentrations are varied to represent a diverse set of possible atmospheric conditions (Table S4). Figure S2 shows how the mean values of SO2 and HPMTF concentration respond to the changing conditions in the CS2-HPMTF-compl scheme. Similar trends can be observed with minimum and maximum concentrations, although they are not explicitly shown here. When reactions are removed from the scheme, the response of SO2 and HPMTF concentration to changing conditions should be maintained. The reduced CS2-HPMTF scheme will be referred to as CS2-HPMTF-red.

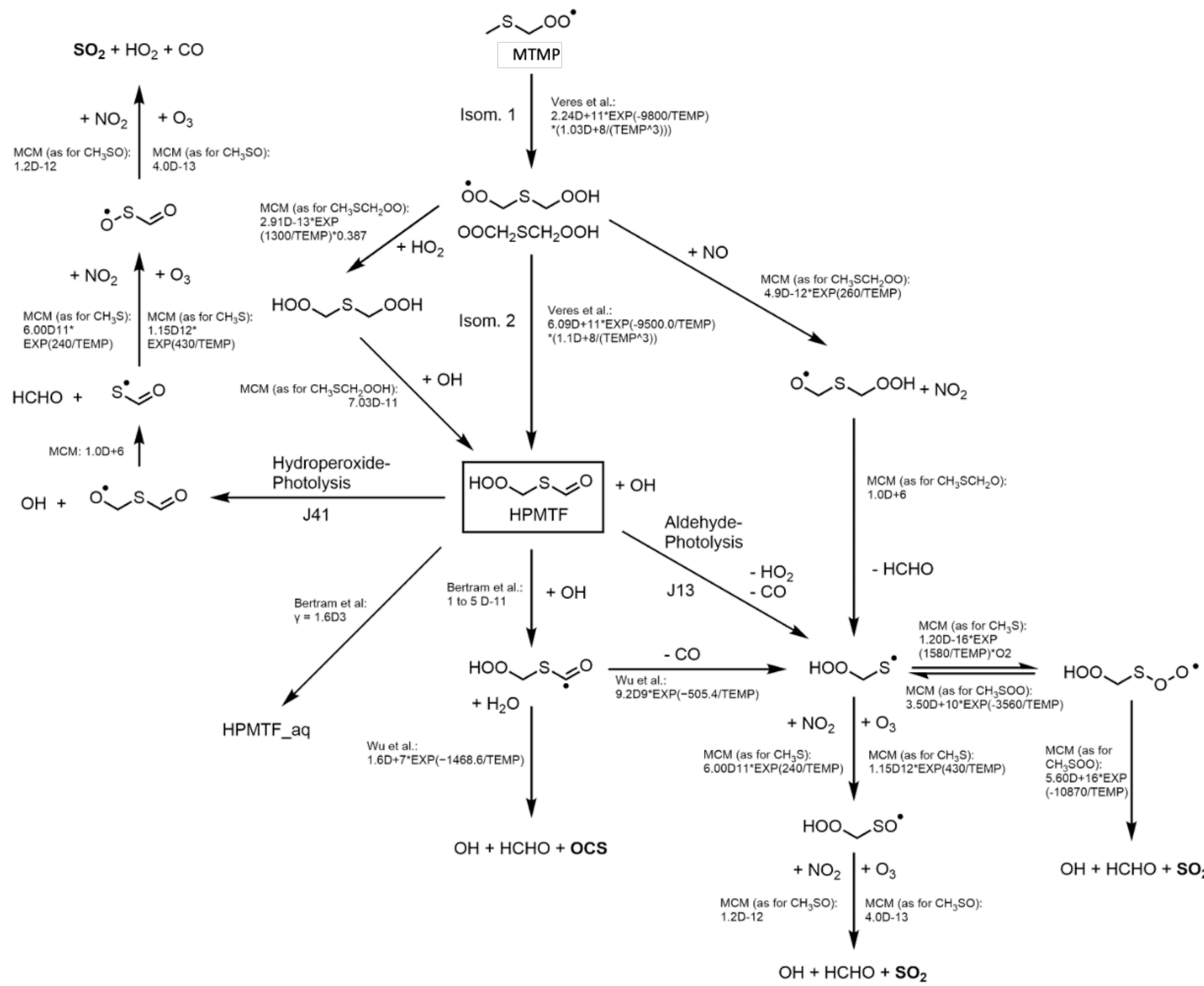

Figure S1: Complete isomerization pathway of HPMTF that was considered before reducing the mechanism.

Table S3: The complete isomerization pathway, referred to as CS2-HPMTF-compl.

No.	Reaction	Rate (cm^3 $molecule^{-1}$ s^{-1})	Reference
s1	MTMP → $OOCH_2SCH_2OOH$	see note[a]	Veres et al. (2020)
s2a	$OOCH_2SCH_2OOH$ → HPMTF + OH	See note[b]	Veres et al. (2020)
s2b	$OOCH_2SCH_2OOH$ + HO2 → $HOOCH_2SCH_2OOH$	$1.13\times10^{-13}\exp^{(1300/T)}$	Veres et al. (2020)
s3	$HOOCH_2SCH_2OOH$ + OH → HPMTF	7.03×10^{-11}	this work (like CH_2SCH_2OOH in MCMv3.3.1)

s2c	$OOCH_2SCH_2OOH + NO \rightarrow OCH2SCH2OOH + NO_2$	$4.90\times10^{-12}\exp^{(260/T)}$	Veres et al. (2020)
s4	$OCH2SCH2OOH \rightarrow HOOCH2S + HCHO$	$1.0\times10^{+6}$	Veres et al. (2020)
s5a	$HPMTF + OH \rightarrow HOOCH_2S + H_2O + CO$	$1.0\times10^{-11}\times 0.9$	this work
s5b	$HPMTF + OH \rightarrow OCS + OH + HCHO + H_2O$	$1.0\times10^{-11}\times 0.1$	this work
s5c	$HPMTF \rightarrow HOOCH_2S + HO_2 + CO$	*J13*	this work
s5d	$HPMTF \rightarrow OCH_2SCHO + OH$	*J41*	this work
s5e	$HPMTF \rightarrow$ loss	gamma=0.1	this work
s6a	$HOOCH_2S + O_3 \rightarrow HOOCH_2SO$	$1.15\times10^{-12}\exp^{(430/T)}$	Wu et al. (2015)
s6b	$HOOCH_2S + NO_2 \rightarrow HOOCH_2SO + NO$	$6.00\times10^{-11}\exp^{(240/T)}$	Wu et al. (2015)
s6c	$HOOCH_2S + O_2 \rightarrow HOOCH_2SOO$	$1.20\times10^{-16}\exp^{(1580/T)}\times[O_2]$	this work (like CH_3S)
s7a	$HOOCH_2SOO \rightarrow HOOCH_2S + O_2$	$3.50\times10^{+10}\exp^{(-3560/T)}$	this work (like CH_3SOO)
s7b	$HOOCH_2SOO \rightarrow HCHO + OH + SO_2$	$5.60\times10^{+16}\exp^{(-10870/T)}$	this work (like CH_3SOO)
s8a	$HOOCH_2SO + O_3 \rightarrow HCHO + OH + SO_2$	4×10^{-13}	Wu et al. (2015)
s8b	$HOOCH_2SO + NO_2 \rightarrow HCHO + OH + NO + SO_2$	1.2×10^{-11}	Wu et al. (2015)
s10	$OCH2SCHO = HCHO + SCHO$	$1.0\times10^{+6}$	this work (like s4)
s11a	$SCHO + O_3 = OSCHO$	$1.15\times10^{-12}\exp^{(430/T)}$	this work (like s6a)
s11b	$SCHO + NO_2 = OSCHO + NO$	$6.00\times10^{-11}\exp^{(240/T)}$	this work (like s6b)
s12a	$OSCHO + O_3 = SO_2 + HO_2 + CO$	4×10^{-13}	this work (like s8a)
s12b	$OSCHO + NO_2 = SO_2 + HO_2 + CO + NO$	1.2×10^{-11}	this work (like s8b)

[a] $2.24\times10^{+11}\exp^{(-9800/T)}\exp^{(1.03e8/(T\times T\times T))}$

[b] $6.09\times10^{+11}\exp^{(-9500/T)}\exp^{(1.1e8/(T\times T\times T))}$

Table S4: Conditions for sensitivity runs. For the NOx sensitivity run, NO, NO_2, and HNO_3 were varied to achieve NOx concentrations between 0.4 ppt and 4.4 ppb.

Sensitivity run	Temperature (K)	NO (ppt)	NO_2 (ppt)	HNO_3 (ppb)	O_3 (ppb)	Aerosol surface area ($\mu m^2\ cm^{-3}$)
Temp	260 - 310	1	10	0.5	30	15
NOx	290	0.1 - 1000	1 - 10,000	0.01 - 100	30	15
O_3	290	1	10	0.5	10 - 80	15
Aerosol	290	1	10	0.5	30	0 - 100

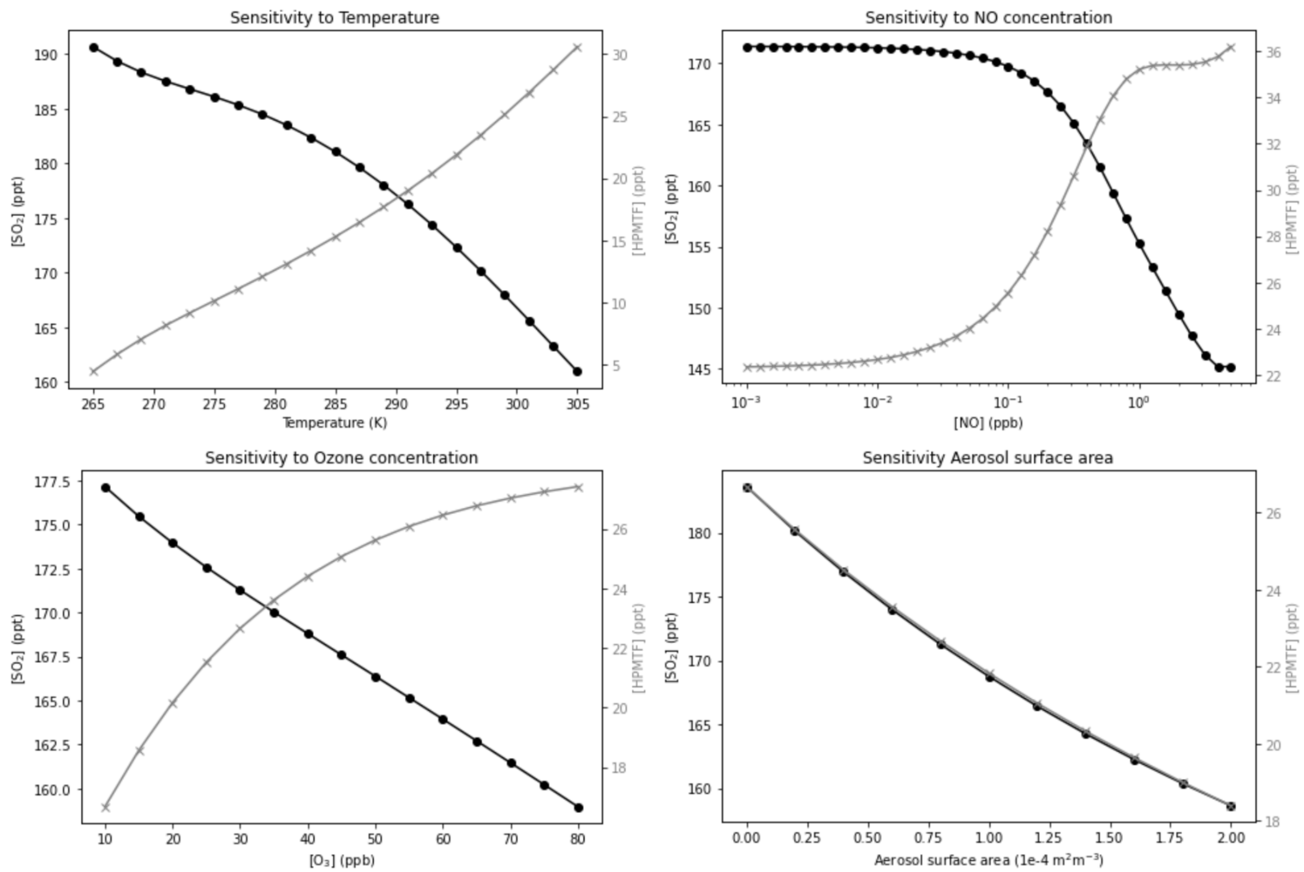

Figure S2: Sensitivity of mean SO2 (black) and HPMTF (grey) concentration with the complete mechanism to the variation of temperature, NO and O3 concentration, and aerosol surface area.

Removal of aldehyde and hydroperoxide photolysis of HPMTF (reactions s5c and s5d) has a negligible influence on HPMTF and SO_2 concentration (Figure S3) under the conditions simulated in the BOXMOX runs. This result is slightly at odds with the work of Khan et al. (2022), who found that photolysis was a dominant loss process. But in line with the work of Novak et al. (2022). The relative difference in HPMTF between CS2-HPMTF-compl and CS2-HPMTF-red is never higher than 6%. For SO_2 the maximum difference is 2% in total. In comparison: if OH-oxidation of HPMTF had been removed, mean SO_2 concentration at high temperatures would have dropped by more than 25% compared to CS2-HPMTF-red. Additionally, the large uncertainty regarding the rate of OH-oxidation is expected to have a much higher influence than the (non)inclusion of the photolysis reactions. Therefore, reactions s5c and s5d and their plausible follow-up reactions are not included in the final CS2-HPMTF mechanism.

The effect of the removal of the two side reactions s2b and s3c after the first isomerization step of MSP, can be observed in Figure S4. At no point does SO_2 concentration change by more than 1% and HPMTF concentration does not vary by more than 5%. The minor role of both pathways was expected, since the second isomerization step s3 was calculated to be approximately a magnitude faster than the first isomerization step s2a (Wu et al., 2015, Veres et al., 2020). These side reactions have therefore also been excluded from the final mechanism.

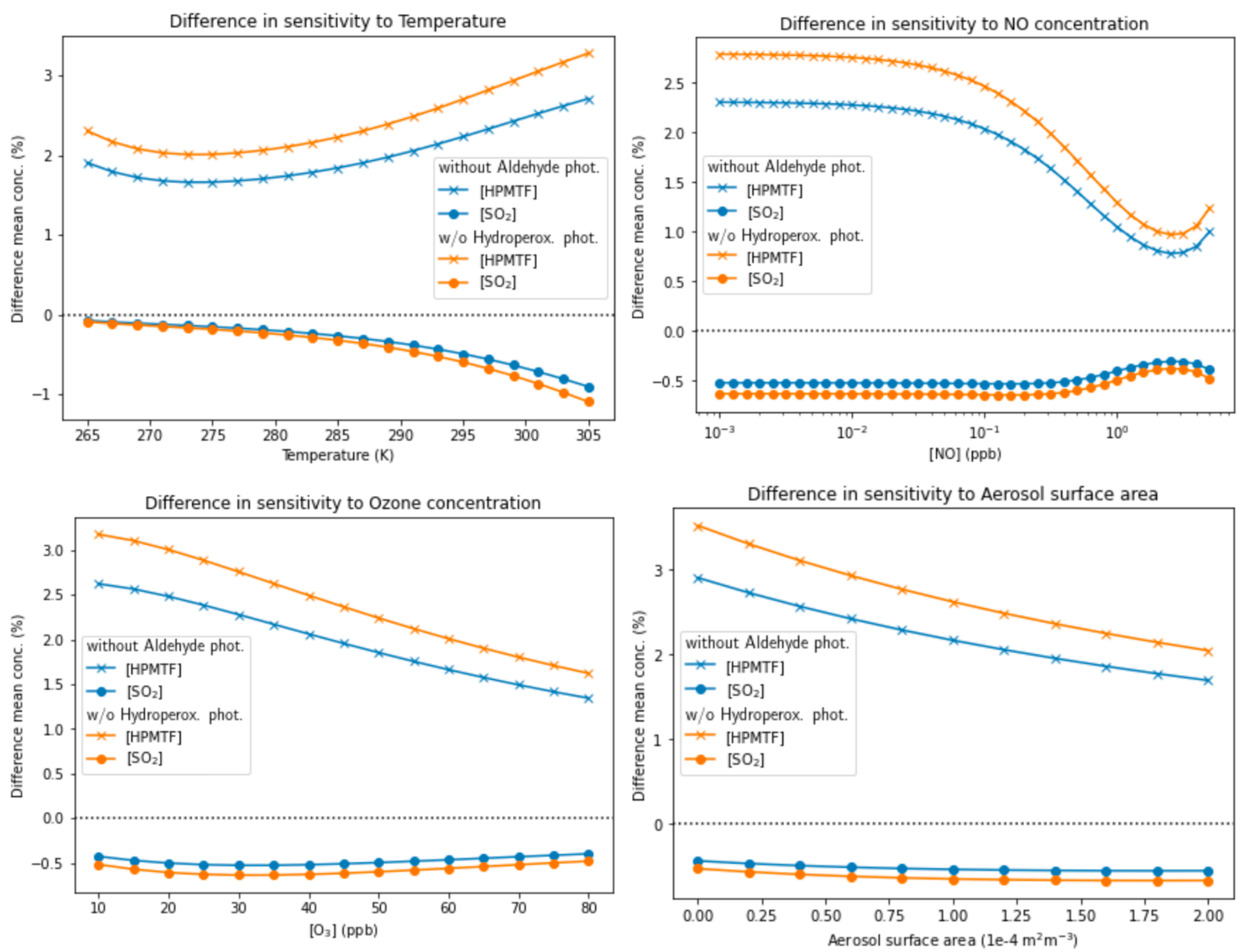

Figure S3: Difference in sensitivity of mean SO_2 and HPMTF concentration after the removal of the photooxidation reactions, compared to the complete mechanism.

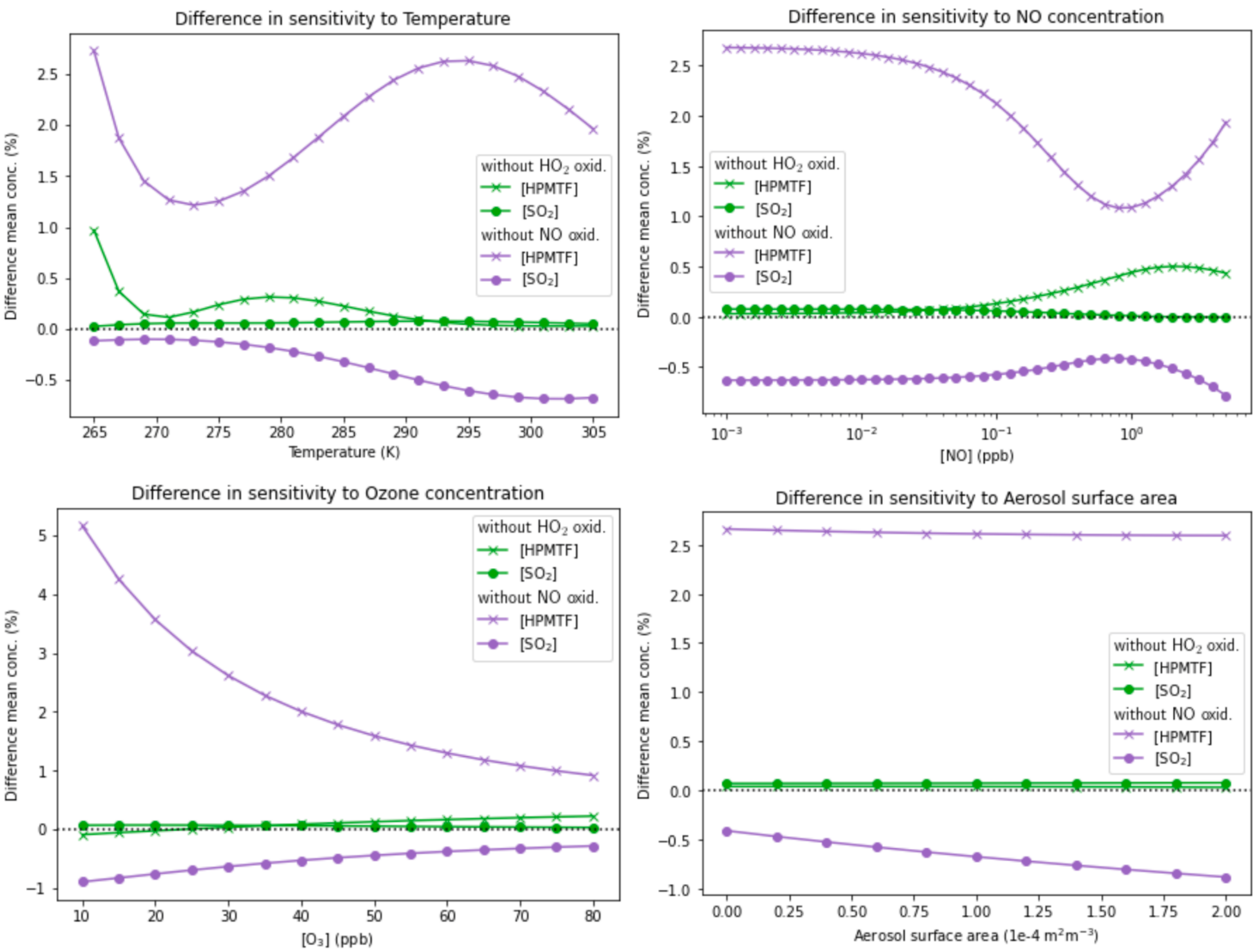

Figure S4: Difference in sensitivity of mean SO_2 and HPMTF concentration after the removal of the side reactions after the first isomerization step during HPMTF formation, compared to the complete mechanism

S1.3 Other plots

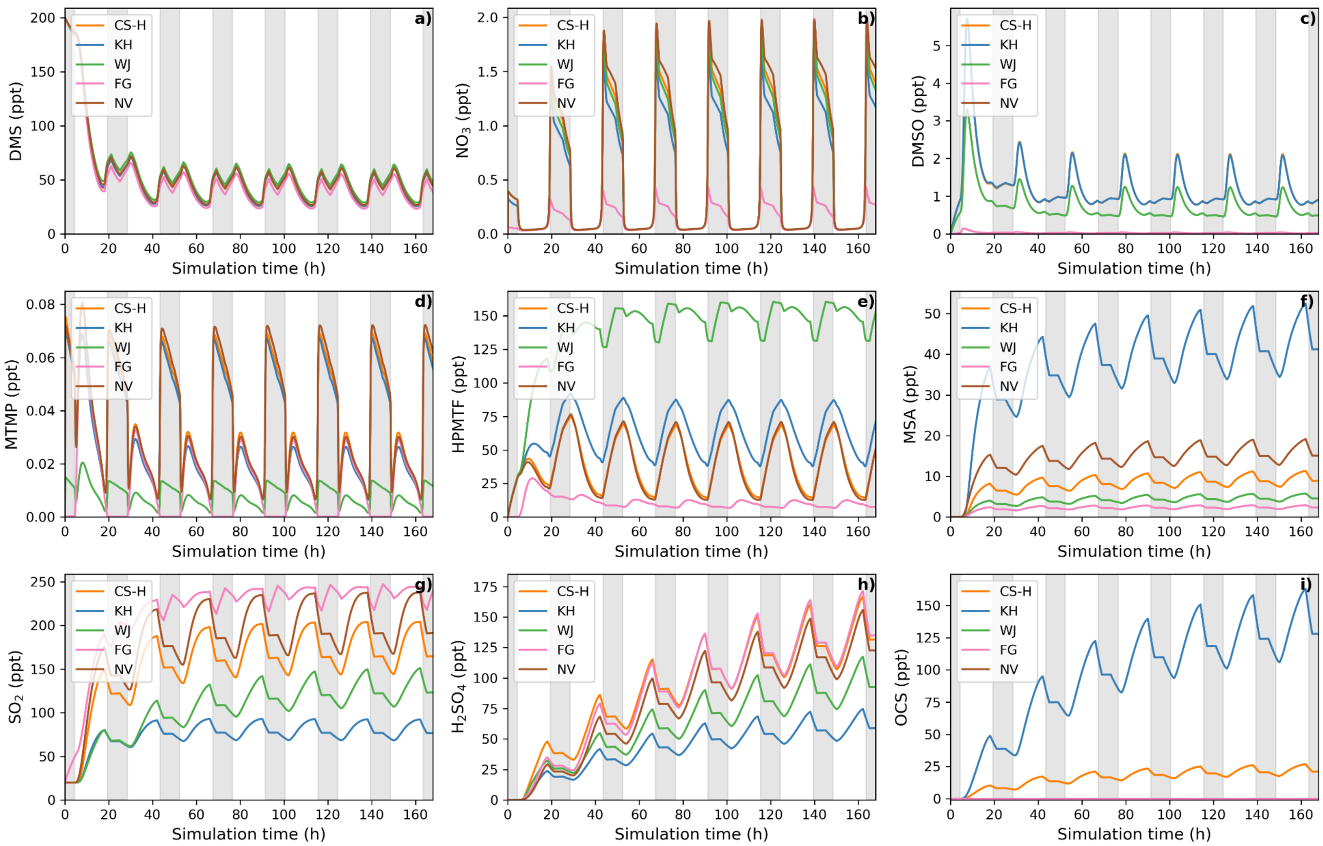

Figure S5: gas-phase concentrations as a function of time for different DMS gas-phase oxidation schemes used in UKCA configurations (oxidation by OH and NO_3). Grey areas denote night- time, when no photolysis reactions are taking place. Average NOx concentration is approximately 100 ppt, with an average temperature of 293 K (range: 289 - 297 K).

S1.4 DMS schemes

S1.4.1 StratTrop (Archibald et al. 2020)

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DMS + OH = SO2 : 1.20D-11*EXP(-260/TEMP) ;
DMS + OH = MSA + SO2 : 3.36224D-43*EXP(350/TEMP)*EXP(7460/TEMP)*M/(1+1.106D-31*EXP(7460/TEMP)*M) ;
DMS + NO3 = SO2 : 1.90D-13*EXP(500/TEMP) ;
DMS + O3P = SO2 : 1.30D-11*EXP(410/TEMP) ;
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S1.4.2 Wollesen de Jonge et al. (2021)

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DMS + NO3 = CH3SCH2O2 + HNO3 : 1.9D-13*exp(520/TEMP)  ;
DMS + OH = CH3SCH2O2 :  1.12D-11*exp(-250/TEMP) ;
DMS + OH = CH3SOHCH3 : 9.5D-39*O2*EXP(5270/TEMP)/(1+7.5D-29*O2*EXP(5610/TEMP)) ;
CH3SOHCH3 = HODMSO2 : 8.5D-13*O2 ;
CH3SOHCH3 = CH3SOH + CH3O2 : 5D+5 ;
CH3SOHCH3 = DMS + OH : (2.048D-14*O2*exp(2674/TEMP)/(1+5.5D-31*O2*exp(7640/TEMP)))/(TEMP) ;
CH3SOH + OH = CH3SO : 5D-11 ;
CH3SCH2O2 + HO2 = CH3SCH2OOH : KRO2HO2*0.387 ;
CH3SCH2O2 + NO = CH3SCH2O + NO2 : 4.9D-12*exp(260/TEMP) ;
CH3SCH2O2 + NO3 = CH3SCH2O + NO2 : KRO2NO3 ;
CH3SCH2O2 = CH3SCH2O : 2*(K298CH3O2*1.0D-11)**0.5*RO2*0.8  ;
CH3SCH2O2 = CH3SCH2OH : 2*(K298CH3O2*1.0D-11)**0.5*RO2*0.1 ;
CH3SCH2O2 = CH3SCHO : 2*(K298CH3O2*1.0D-11)**0.5*RO2*0.1  ;
CH3SCH2OOH + OH = CH3SCHO + OH : 7.03D-11 ;
CH3SCH2OOH = CH3SCH2O + OH : (5.786D-6)*SUN ;
CH3SCH2O = CH3S + HCHO : KDEC  ;
CH3SCH2OH + OH = CH3SCHO + HO2 : 2.78D-11 ;
CH3SCHO + OH = CH3S + CO : 1.11D-11  ;
CH3SCHO = CH3S + CO + HO2 : (1.99D-5)*SUN  ;
CH3SCH2O2 = OOCH2SCH2OOH : 2.2433D11*exp(-9.8016D3/TEMP)*exp(1.0348D8/(TEMP*TEMP*TEMP))*5 ;
OOCH2SCH2OOH = HPMTF + OH : 6.097D11*exp(-9.4892D3/TEMP)*exp(1.102D8/(TEMP*TEMP*TEMP)) ;
OOCH2SCH2OOH + NO = HOOCH2S + NO2 + HCHO : 4.9D-12*exp(260/TEMP) ;
OOCH2SCH2OOH + HO2 = HOOCH2SCH2OOH : 1.13D-13*exp(1300/TEMP) ;
HPMTF + OH = HOOCH2SCO : 1.4D-12 ;
HOOCH2SCO = HOOCH2S + CO : 9.2D9*exp(-505.4/TEMP) ;
HOOCH2SCO = HCHO + OH + OCS : 1.6D7*exp(-1468.6/TEMP) ;
HOOCH2S + O3 = HOOCH2SO : 1.15D-12*exp(430/TEMP) ;
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I56   HOOCH2S + NO2 = HOOCH2SO + NO : 6.00D-11*exp(240/TEMP) ;
I57   HOOCH2SO + O3 = SO2 + HCHO + OH : 4.00D-13 ;
I58   HOOCH2SO + NO2 = SO2 + HCHO + OH + NO : 1.20D-11 ;
I59   CH3S + NO2 = CH3SO + NO : 6.00D-11*exp(240/TEMP) ;
I60   CH3S + O3 = CH3SO : 1.15D-12*exp(430/TEMP)  ;
I61   CH3S = CH3SOO : 1.20D-16*exp(1580/TEMP)*O2  ;
I62   CH3SO + NO2 = CH3O2 + SO2 + NO : 1.20D-11*0.25  ;
I63   CH3SO + NO2 = CH3SO2 + NO : 1.20D-11*0.75  ;
I64   CH3SO + O3 = CH3O2 + SO2 : 4.00D-13 ;
I65   CH3SO = CH3SOO2 : 3.12D-16*exp(1580/TEMP)*O2 ;
I66   CH3SOO + NO = CH3SO + NO2 : 1.1D-11 ;
I67   CH3SOO + NO2 = CH3SO + NO3 : 2.2D-11 ;
I68   CH3SOO = CH3O2 + SO2 : 5.60D+16*exp(-10870/TEMP) ;
I69   CH3SOO = CH3S : 3.50D+10*exp(-3560/TEMP) ;
I70   CH3SOO + HO2 = CH3SOOH : 4D-12 ;
I71   CH3SOO = CH3SO2 : 1.0 ;
I72   CH3SOO2 + HO2 = CH3SO2 + OH : KAPHO2*0.44  ;
I73   CH3SOO2 + HO2 = CH3SOOOH :KAPHO2*0.41  ;
I74   CH3SOO2 + HO2 = MSIA + O3 : KAPHO2*0.15 ;
I75   CH3SOO2 + NO = CH3SO2 + NO2 : 1.00D-11  ;
I76   CH3SOO2 + NO2 = CH3SOO2NO2 : 1.20D-12*(TEMP/300)**(-0.9) ;
I77   CH3SOO2 + NO3 = CH3SO2 + NO2 : KRO2NO3*1.74 ;
I78   CH3SOO2 = CH3SO : 9.10E+10*exp(-3560/TEMP) ;
I79   CH3SOO2 = CH3SO2 : 1.00D-11*RO2*0.7 ;
I80   CH3SOO2 = MSIA : 1.00D-11*RO2*0.3 ;
I81   CH3SOOOH + OH = CH3SOO2 : 9.00D-11 ;
I82   CH3SOOOH = CH3SO2 + OH : (5.786D-6)*SUN  ;
I83   CH3SOO2NO2 + OH = MSIA + NO2 : 1.00D-11  ;
I84   CH3SOO2NO2 = CH3SOO2 + NO2 : 5.40D+16*exp(-13112/TEMP) ;
I85   CH3SO2 + O3 = CH3SO3 : 3.00D-13 ;
I86   CH3SO2 = CH3O2 + SO2 : 5.00D+13*exp(-9673/TEMP) ;
I87   CH3SO2 = CH3SO2O2 : 1.03D-16*exp(1580/TEMP)*O2 ;
I88   CH3SO2 + OH = MSA : 5D-11 ;
I89   CH3SO2 + NO2 = CH3SO3 + NO : 2.2D-11 ;
I90   CH3SO2O2 + HO2 = CH3SO2OOH : KAPHO2*0.41 ;
I91   CH3SO2O2 + HO2 = CH3SO3 + OH : KAPHO2*0.44 ;
I92   CH3SO2O2 + HO2 = MSA + O3 : KAPHO2*0.15  ;
I93   CH3SO2O2 + NO = CH3SO3 + NO2 : 1.00D-11 ;
I94   CH3SO2O2 + NO2 = CH3SO4NO2 : 1.20D-12*(TEMP/300)**(-0.9) ;
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CH3SO2O2 + NO3 = CH3SO3 + NO2 : KRO2NO3*1.74  ;
CH3SO2O2 = CH3SO2 : 3.01D+10*exp(-3560/TEMP) ;
CH3SO2O2 = CH3SO3 : 1.00D-11*RO2*0.7 ;
CH3SO2O2 = MSA : 1.00D-11*RO2*0.3 ;
CH3SO3 + HO2 = MSA : 5.00D-11  ;
CH3SO3 = CH3O2 + SO3 : 5.00D+13*exp(-9946/TEMP) ;
CH3SO2OOH + OH = CH3SO2O2 : 3.60D-12  ;
CH3SO2OOH = CH3SO3 + OH : (5.786D-6)*SUN ;
CH3SO4NO2 + OH = CH3SO2O2 + HNO3 : 3.60D-13  ;
CH3SO4NO2 = CH3SO2O2 + NO2 : 5.40D+16*exp(-13112/TEMP) ;
HODMSO2 + NO = DMSO2 + HO2 + NO2 : KRO2NO ;
HODMSO2 = DMSO + HO2 : 8.90E+10*exp(-6040/TEMP) ;
DMSO + OH = MSIA + CH3O2 : 6.10D-12*exp(800/TEMP) ;
DMSO + NO3 = DMSO2 + NO2 : 2.9D-13 ;
DMSO2 + OH = DMSO2O2 : 4.40D-14 ;
DMSO2O2 + HO2 = DMSO2OOH : KRO2HO2*0.387 ;
DMSO2O2 + NO = DMSO2O + NO2 : KRO2NO  ;
DMSO2O2 + NO3 = DMSO2O + NO2 : KRO2NO3 ;
DMSO2O2 = CH3SO2CHO : 2.00D-12*RO2*0.2 ;
DMSO2O2 = DMSO2O : 2.00D-12*RO2*0.6 ;
DMSO2O2 = DMSO2OH : 2.00D-12*RO2*0.2 ;
DMSO2OOH + OH = CH3SO2CHO + OH : 1.26D-12  ;
DMSO2OOH + OH = DMSO2O2 : 3.60D-12 ;
DMSO2OOH = DMSO2O + OH : (5.786D-6)*SUN  ;
DMSO2O = CH3SO2 + HCHO : KDEC ;
DMSO2OH + OH = CH3SO2CHO + HO2 : 5.23D-13 ;
DMSO2OH + OH = DMSO2O : 1.40D-13 ;
CH3SO2CHO + OH = CH3SO2 + CO : 1.78D-12  ;
CH3SO2CHO = CH3SO2 + CO + HO2 : (1.99D-5)*SUN ;
MSIA + OH = CH3SO2 : 1D-10 ;
MSIA + NO3 =CH3SO2 + HNO3 : 1D-13 ;
MSA + OH = CH3SO3 : 2.24D-14  ;
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S1.4.3 Fung et al. (2021)

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DMS + OH = MTMP:  1.12D-11*EXP(-250/TEMP) ;
MTMP + NO = CH3SCH2O + NO2 : 4.90D-12*EXP(260/TEMP) ;
MTMP = CH3SCH2O + O2 : 3.74D-12*RO2 ;
CH3SCH2O = CH3S +HCHO : 1.00D+6 ;
CH3S + O3 = CH3SO + O2 : 1.15D-12*EXP(430/TEMP) ;
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CH3S + O2 = CH3SOO : 1.20D-16*EXP(1580/TEMP)*O2 ;
CH3SO + O3 = CH3O2 + SO2 : 4.0D-13 ;
CH3SOO = CH3O2 + SO2 : 5.60D+16*EXP(-10870/TEMP) ;
CH3SOO = CH3SO2 : 1.00 ;
CH3SO2 + O3 = CH3SO3 + O2 :   3.0d-13 ;
CH3SO2 = CH3O2 + SO2 : 5.00D13*EXP(-9673/TEMP) ;
CH3SO3 + HO2 = MSA + O2 : 5.0d-11 ;
CH3SO3 = CH3O2 + H2SO4 : 5.00D+13*EXP(-9946/TEMP) ;
MTMP= OOCH2SCH2OOH : 2.24D+11*EXP(-9800/TEMP)*EXP(1.03D+8/(TEMP*TEMP*TEMP)) ;
OOCH2SCH2OOH = HPMTF + OH : 6.09D+11*EXP(-9500.0/TEMP)*EXP(1.1D+8/(TEMP*TEMP*TEMP)) ;
OOCH2SCH2OOH + NO = OCH2SCH2OOH + NO2 : 4.9D-12*EXP(260/TEMP) ;
OCH2SCH2OOH = HOOCH2S + HCHO : 1.00D+6 ;
OOCH2SCH2OOH + HO2 = HOOCH2SCH2OOH + O2 : 1.13D-13*EXP(1300/TEMP) ;
HPMTF + OH = HOOCH2SCO + H2O : 1.11D-11 ;
HOOCH2SCO = HOOCH2S + CO : 9.20D+9*EXP(-505.4/TEMP) ;
HOOCH2SCO = OH + HCHO + OCS : 1.60D+7*EXP(-1468.6/TEMP) ;
HOOCH2S + O3 = HOOCH2SO : 1.15D-12*EXP(430/TEMP) ;
HOOCH2S + NO2 = HOOCH2SO + NO : 6.00D-11*EXP(240/TEMP) ;
HOOCH2SO + O3 = HCHO + OH + SO2 : 4.0D-13 ;
HOOCH2SO + NO2 = SO2 + HCHO + OH  + NO :  1.2d-11;
DMS + OH = SO2 + CH3O2 :  8.2D-39*O2*EXP(5376/TEMP)/(1+7.5D-5*(O2/M)*EXP(3644/TEMP))*0.6 ;
DMS + OH = DMSO + CH3O2 : 8.2D-39*O2*EXP(5376/TEMP)/(1+7.5D-5*(O2/M)*EXP(3644/TEMP))*0.4 ;
DMS + NO3 = SO2 + HNO3 + CH3O2 + HCHO : 1.13D-12*EXP(530/TEMP) ;
DMSO + OH = MSIA : 8.94D-11*EXP(800/TEMP)*0.95 ;
DMSO + OH = SO2 : 8.94D-11*EXP(800/TEMP)*0.05 ;
MSIA + OH = SO2 : 9.00D-11*0.9 ;
MSIA + OH = MSA : 9.00D-11*0.1 ;
MSIA + O3 = MSA : 2.00D-18 ;
MSIA + O3 = MSA : 2.00D–18 ;
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S1.4.4 Khan et al. (2021)

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DMS + OH = MTMP+ H2O :  1.2D-11*EXP(-280/TEMP) ;
DMS + OH = DMSO + HO2 :  9.5D-39*O2*EXP(5270/TEMP)/(1+7.5D-29*O2*EXP(5610/TEMP)) ;
DMS + NO3 = MTMP + HNO3 : 1.90D-13*EXP(580/TEMP) ;
MTMP + NO = HCHO + CH3S + NO2 :  4.90D-12*EXP(260/TEMP)   ;
MTMP + MTMP = HCHO + HCHO + CH3S + CH3S : 1.0d-11 ;
MTMP + HO2 = CH3SCH2OOH: 2.9D-13*EXP(1300/TEMP) ;
CH3SCH2OOH = CH3S + HCHO + OH: (5.78677D-6)*SUN ; {J41}
CH3SCH2OOH + OH = CH3SCHO + OH: 7.0D-11 ;
```

```
CH3SCHO + OH = CH3S + CO : 1.1D-11 ;
CH3SCHO = CH3S + CO + HO2 : (1.99124D-5)*SUN ;
CH3S + O3 = CH3SO :  1.15D-12*EXP(430/TEMP) ;
CH3S + NO2 = CH3SO + NO :  3.00D-12*EXP(210/TEMP) ;
CH3SO + NO2 = CH3SO2 + NO :  1.2d-11*0.82 ;
CH3SO + NO2 = SO2 + CH3O2 + NO :  1.2d-11*0.18 ;
CH3SO + O3 = CH3SO2 : 6.0d-13 ;
CH3SO2 = SO2 + CH3O2 :   5.00D13*EXP(-9673/TEMP) ;
CH3SO2 + NO2 = CH3SO3 + NO : 2.2d-12 ;
CH3SO2 + O3 = CH3SO3 :   3.0d-13 ;
CH3SO3 + HO2 = MSA : 5.0d-11 ;
CH3SO3 = CH3O2 + H2SO4 :  1.36D14*EXP(-11071/TEMP) ;
DMSO + OH = MSIA + CH3O2 :   8.7d-11*0.95 ;
MSIA + OH = CH3SO2 + H2O :           9.d-11*0.95 ;
MSIA + OH = MSA + HO2 + H2O :      9.d-11*0.05 ;
MSIA + NO3 = CH3SO2 + HNO3 :   1.0d-13 ;
MTMP = OOCH2SCH2OOH + O2: 2.74D+7*EXP(-5950/TEMP) ;
OOCH2SCH2OOH = HPMTF + OH: 4.2D+7*EXP(-5390/TEMP) ;
OOCH2SCH2OOH + NO = HOOCH2S + NO2 + HCHO : 4.9D-12*exp(260/TEMP) ;
OOCH2SCH2OOH + HO2 = HOOCH2SCH2OOH : 1.13D-13*exp(1300/TEMP) ;
HOOCH2SCH2OOH = HOOCH2S + HCHO + OH : (5.78677D-6)*SUN ; {J41}
HOOCH2SCH2OOH + OH = OOCH2SCH2OOH : 1.36D-11 ;
HPMTF + OH = HOOCH2S + CO : 1.4D-12*(5.6D-3*TEMP - 1.24) ;
HPMTF + OH = OH + HCHO + OCS : 1.4D-12*(1-(5.6D-3*TEMP - 1.24)) ;
HPMTF = HOOCH2S + CO + HO2: (5.78677D-6)*SUN ; {J41}
HPMTF = OCS + HCHO + OH: (2.0126D-5)*SUN; {J14}
HOOCH2S + O3 = HOOCH2SO : 1.15D-12*exp(430/TEMP) ;
HOOCH2S + NO2 = HOOCH2SO + NO : 6.00D-11*exp(240/TEMP) ;
HOOCH2SO + O3 = SO2 + HCHO + OH : 4.00D-13 ;
HOOCH2SO + NO2 = SO2 + HCHO + OH + NO : 1.20D-11 ;
```

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DMS + OH = MTMP + H2O :  1.12D-11*EXP(-250/TEMP) ;
DMS + OH = SO2 :  (8.2D-39*O2*EXP(5376/TEMP) / (1+ 1.05D5*EXP(3644/TEMP)*0.2095))*0.75 ;
DMS + OH = MSA :  (8.2D-39*O2*EXP(5376/TEMP) / (1+ 1.05D-5*EXP(3644/TEMP)*0.2095))*0.25 ;
DMS + NO3 = MTMP + HNO3 : 1.90D-13*EXP(520/TEMP) ;
MTMP + HO2 = SO2 : 1.13D-13*EXP(1300/TEMP) ;
MTMP + RO2 = SO2 : 1.0D-11 ;
MTMP + NO = SO2 + NO2 : 4.9D-12*EXP(260/TEMP) ;
```

```
MTMP + NO3 = SO2 + NO2 : 2D-12 ;
MTMP = HPMTF : 2.24D+11*EXP(-9800/TEMP)*EXP(1.03D+8/(TEMP*TEMP*TEMP)) ;
HPMTF + OH = SO2 : 1.11D-11 ;
```

S2 Global model

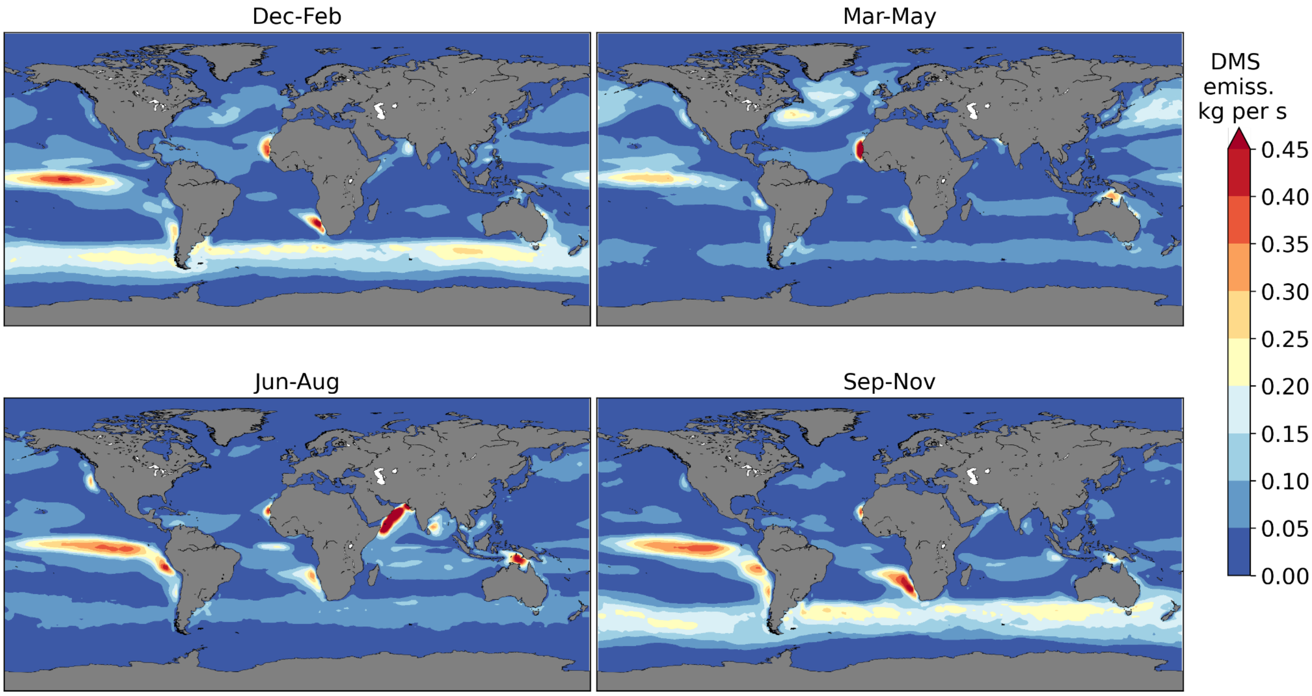

Figure S6: DMS emissions in CS2.

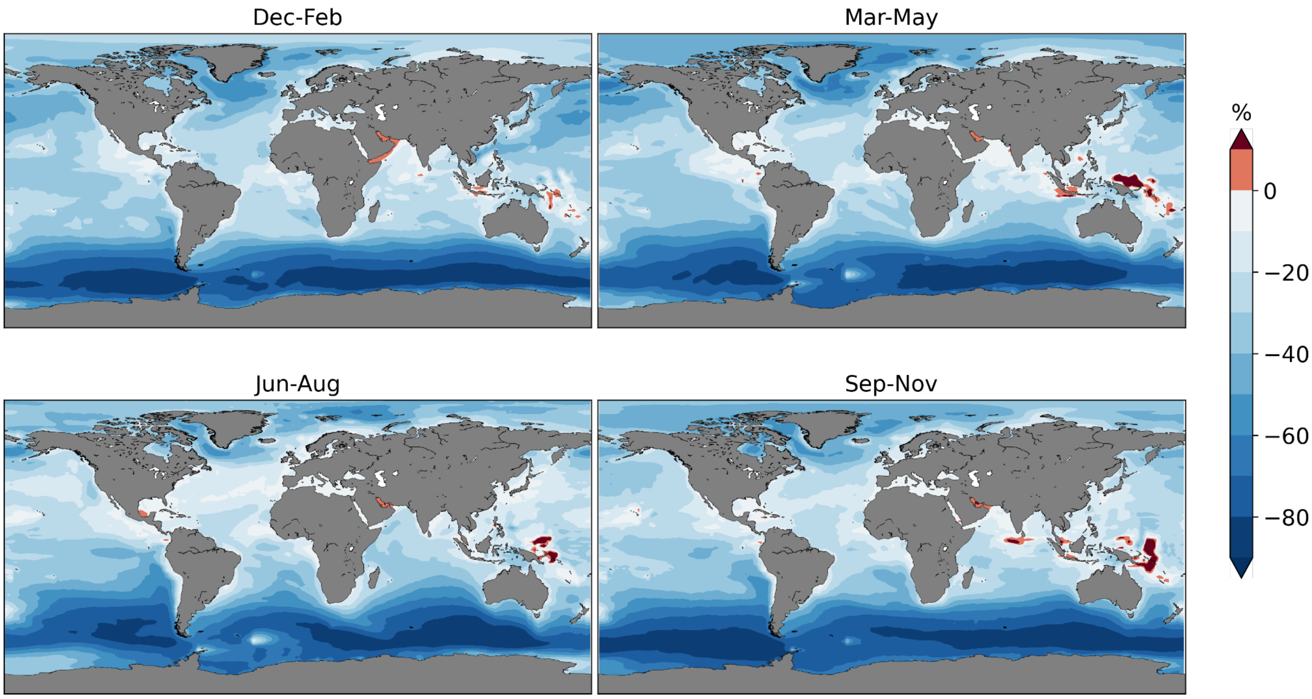

Figure S7: Relative difference in SO_2 mixing ratios in the lower troposphere (< 2 km) between CS2-HPMTF and the StratTrop run ST (CS2-HPMTF - ST). Only values above the ocean are shown.

Table S5: Average tropospheric lifetimes of selected species

	CS2	ST	CS2-HPMTF	CS2-HPMTF-FL	CS2-HPMTF-FP
DMS	35.59 h	36.34 h	34.63 h	34.68 h	34.37 h
MTMP	26.61 min	-	0.99 min	0.98 min	0.25 min
DMSO	6.17 h	-	6.15 h	6.16 h	6.09 h
MSA	-	-	6.08 d	6.08 d	6.07 d
MSIA	6.38 h	-	4.81 h	4.81 h	4.74 h
HPMTF	-	-	25.76 h	9.66 h	25.79 h

SO_2						

Table S6: Sulfate in the different aerosol modes by weight.

	Nucleation	Aitken	Accumulation	Coarse
CS2	2.0%	26.2%	69.8%	2.0%
ST	1.7%	27.8%	69.2%	1.4%
CS2-HPMTF	2.1%	26.6%	69.5%	1.8%
CS2-HPMTF-CLD	2.0%	25.5%	70.0%	2.5%