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

<table>
<thead>
<tr>
<th>Species</th>
<th>Mixing ratio (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>M</td>
<td>1.00E+06</td>
</tr>
<tr>
<td>N₂</td>
<td>7.80E+05</td>
</tr>
<tr>
<td>O₂</td>
<td>2.10E+05</td>
</tr>
<tr>
<td>H₂O</td>
<td>1.00E+04</td>
</tr>
<tr>
<td>CH₄</td>
<td>1.80E+00</td>
</tr>
<tr>
<td>CO</td>
<td>1.00E-01</td>
</tr>
<tr>
<td>H₂</td>
<td>5.00E-01</td>
</tr>
<tr>
<td>O₃</td>
<td>3.00E-02</td>
</tr>
<tr>
<td>H₂O₂</td>
<td>1.00E-03</td>
</tr>
<tr>
<td>NO</td>
<td>1.00E-06</td>
</tr>
<tr>
<td>NO₂</td>
<td>1.00E-05</td>
</tr>
<tr>
<td>HNO₃</td>
<td>5.00E-04</td>
</tr>
<tr>
<td>DMS</td>
<td>2.00E-04</td>
</tr>
<tr>
<td>SO₂</td>
<td>2.00E-05</td>
</tr>
</tbody>
</table>
**Table S2:** Diurnal profile of temperature and boundary layer height

<table>
<thead>
<tr>
<th>Time (h)</th>
<th>Temperature (K)</th>
<th>Boundary Layer Height (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>289.5359</td>
<td>1300</td>
</tr>
<tr>
<td>1</td>
<td>289.1363</td>
<td>1300</td>
</tr>
<tr>
<td>2</td>
<td>289.0000</td>
<td>1350</td>
</tr>
<tr>
<td>3</td>
<td>289.1363</td>
<td>1400</td>
</tr>
<tr>
<td>4</td>
<td>289.5359</td>
<td>1450</td>
</tr>
<tr>
<td>5</td>
<td>290.1716</td>
<td>1500</td>
</tr>
<tr>
<td>6</td>
<td>291.0000</td>
<td>1550</td>
</tr>
<tr>
<td>7</td>
<td>291.9647</td>
<td>1450</td>
</tr>
<tr>
<td>8</td>
<td>293.0000</td>
<td>1400</td>
</tr>
<tr>
<td>9</td>
<td>294.0353</td>
<td>1350</td>
</tr>
<tr>
<td>10</td>
<td>295.0000</td>
<td>1300</td>
</tr>
<tr>
<td>11</td>
<td>295.8284</td>
<td>1250</td>
</tr>
<tr>
<td>12</td>
<td>296.4641</td>
<td>1200</td>
</tr>
<tr>
<td>13</td>
<td>296.8637</td>
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<td>14</td>
<td>297.0000</td>
<td>1200</td>
</tr>
<tr>
<td>15</td>
<td>296.8637</td>
<td>1200</td>
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<td>1100</td>
</tr>
<tr>
<td>19</td>
<td>294.0353</td>
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<tr>
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<td>293.0000</td>
<td>1300</td>
</tr>
<tr>
<td>21</td>
<td>291.9647</td>
<td>1400</td>
</tr>
<tr>
<td>22</td>
<td>291.0000</td>
<td>1400</td>
</tr>
<tr>
<td>23</td>
<td>290.1716</td>
<td>1350</td>
</tr>
<tr>
<td>24</td>
<td>289.5359</td>
<td>1300</td>
</tr>
</tbody>
</table>

**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 HO₂ 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 $\text{SO}_2$ (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.

<table>
<thead>
<tr>
<th>No.</th>
<th>Reaction</th>
<th>Rate (cm$^3$ molecule$^{-1}$ s$^{-1}$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>s1</td>
<td>MTMP → OOCH$_2$SCH$_2$OOH</td>
<td>see note$^a$</td>
<td>Veres et al. (2020)</td>
</tr>
<tr>
<td>s2a</td>
<td>OOCH$_2$SCH$_2$OOH → HPMTF + OH</td>
<td>See note$^b$</td>
<td>Veres et al. (2020)</td>
</tr>
<tr>
<td>s2b</td>
<td>OOCH$_2$SCH$_2$OOH + HO2 → HOOCH$_2$SCH$_2$OOH</td>
<td>$1.13 \times 10^{-13} \exp^{(1300/T)}$</td>
<td>Veres et al. (2020)</td>
</tr>
<tr>
<td>s3</td>
<td>HOOCH$_2$SCH$_2$OOH + OH → HPMTF</td>
<td>$7.03 \times 10^{-11}$</td>
<td>this work (like CH$_2$SCH$_2$OOH in MCMv3.3.1)</td>
</tr>
</tbody>
</table>
Table S4: Conditions for sensitivity runs. For the NOx sensitivity run, NO, NO2, and HNO3 were varied to achieve NOx concentrations between 0.4 ppt and 4.4 ppb.

<table>
<thead>
<tr>
<th>Sensitivity run</th>
<th>Temperature (K)</th>
<th>NO (ppt)</th>
<th>NO2 (ppt)</th>
<th>HNO3 (ppb)</th>
<th>O3 (ppb)</th>
<th>Aerosol surface area (μm² cm⁻³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temp</td>
<td>260 - 310</td>
<td>1</td>
<td>10</td>
<td>0.5</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td>NOx</td>
<td>290</td>
<td>0.1 - 1000</td>
<td>1 - 10,000</td>
<td>0.01 - 100</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td>O3</td>
<td>290</td>
<td>1</td>
<td>10</td>
<td>0.5</td>
<td>10 - 80</td>
<td>15</td>
</tr>
<tr>
<td>Aerosol</td>
<td>290</td>
<td>1</td>
<td>10</td>
<td>0.5</td>
<td>30</td>
<td>0 - 100</td>
</tr>
</tbody>
</table>

a \( 2.24 \times 10^{-11} \exp^{-1.9800/T} \exp^{1.03e8/(T\times T\times T)} \)
b \( 6.09 \times 10^{-11} \exp^{-1.9500/T} \exp^{1.1e8/(T\times T\times T)} \)
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 SO2 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 SO2 the maximum difference is 2% in total. In comparison: if OH-oxidation of HPMTF had been removed, mean SO2 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)

\[ \text{DMS + OH} = \text{SO}_2 : 1.20 \times 10^{-13} \exp(-260/\text{TEMP}); \]

\[ \text{DMS + OH} = \text{MSA} + \text{SO}_2 : 3.62 \times 10^{-13} \exp(350/\text{TEMP}) \exp(7460/\text{TEMP}) \times M/(1+1.106\times M); \]

\[ \text{DMS + NO}_3 = \text{SO}_2 : 1.90 \times 10^{-11} \exp(1000/\text{TEMP}); \]

\[ \text{DMS + O}_3\text{P} = \text{SO}_2 : 1.30 \times 10^{-11} \exp(410/\text{TEMP}); \]

S1.4.2 Wollesen de Jonge et al. (2021)

\[ \text{DMS + NO}_3 = \text{CH}_3\text{SCH}_2\text{O}_2 + \text{HNO}_3 : 1.9 \times 10^{-11} \exp(520/\text{TEMP}); \]

\[ \text{DMS + OH} = \text{CH}_3\text{SCH}_2\text{O}_2 : 1.12 \times 10^{-11} \exp(-250/\text{TEMP}); \]

\[ \text{DMS + OH} = \text{CH}_3\text{SOHCH}_3 : 9.5 \times 10^{-12} \exp(520/\text{TEMP})/(1+7.5 \times 10^{-12} \exp(5610/\text{TEMP})); \]

\[ \text{CH}_3\text{SOHCH}_3 = \text{HODMSO}_2 : 8.5 \times 10^{-12} \exp(7460/\text{TEMP})/(1+7.5 \times 10^{-12} \exp(7460/\text{TEMP})); \]

\[ \text{CH}_3\text{SOHCH}_3 = \text{CH}_3\text{SOH} + \text{CH}_3\text{O}_2 : 5\times 10^{-13}; \]

\[ \text{CH}_3\text{SOHCH}_3 + \text{OH} = \text{CH}_3\text{SO} : 5\times 10^{-11}; \]

\[ \text{CH}_3\text{SCH}_2\text{O}_2 + \text{HO}_2 = \text{CH}_3\text{SCH}_2\text{OOH} : K\text{RO}_2\text{HO}_2 \times 0.387; \]

\[ \text{CH}_3\text{SCH}_2\text{O}_2 + \text{NO} = \text{CH}_3\text{SCH}_2\text{O} + \text{NO}_2 : 4.9 \times 10^{-12} \exp(260/\text{TEMP}); \]

\[ \text{CH}_3\text{SCH}_2\text{O}_2 + \text{NO}_3 = \text{CH}_3\text{SCH}_2\text{O} + \text{NO}_2 : K\text{RO}_2\text{NO}_3; \]

\[ \text{CH}_3\text{SCH}_2\text{O}_2 + \text{O}_3\text{P} = \text{CH}_3\text{SCH}_2\text{O} : 2\times (K\text{298CH}_3\text{O}_2 \times 1.0\times 10^{-11})^{0.5} \times \text{RO}_2 \times 0.8; \]

\[ \text{CH}_3\text{SCH}_2\text{O}_2 + \text{SO}_2 = \text{CH}_3\text{SCH}_2\text{O} : 2\times (K\text{298CH}_3\text{O}_2 \times 1.0\times 10^{-11})^{0.5} \times \text{RO}_2 \times 0.1; \]

\[ \text{CH}_3\text{SCH}_2\text{O}_2 + \text{OH} = \text{CH}_3\text{SCHO} + \text{OH} : (2.048 \times 10^{-11} \exp(2674/\text{TEMP})/(1+5.5 \times 10^{-12} \exp(7640/\text{TEMP}))); \]

\[ \text{CH}_3\text{SCH}_2\text{O} + \text{OH} = \text{CH}_3\text{SCHO} : 5\times 10^{-11}; \]

\[ \text{CH}_3\text{SCH}_2\text{O} = \text{CH}_3\text{S} + \text{HCHO} : K\text{DEC}; \]

\[ \text{CH}_3\text{SCH}_2\text{OOH} + \text{OH} = \text{CH}_3\text{SCHO} + \text{HO}_2 : 2.78 \times 10^{-11}; \]

\[ \text{CH}_3\text{SCHO} + \text{OH} = \text{CH}_3\text{S} + \text{CO} + \text{HCHO} : (1.99 \times 10^{-11} \exp(1000/\text{TEMP})); \]

\[ \text{CH}_3\text{SCH}_2\text{O}_2 = \text{OOCH}_2\text{SCH}_2\text{OOH} : 2.2433 \times 10^{-11} \exp(-9.8016\times 10^3/\text{TEMP}) \exp(1.0348\times 10^8/(\text{TEMP}^2 \times \text{TEMP}^2)); \]

\[ \text{OOCH}_2\text{SCH}_2\text{OOH} + \text{NO} = \text{HOOCH}_2\text{S} + \text{NO}_2 + \text{HCHO} : 4.9 \times 10^{-12} \exp(260/\text{TEMP}); \]

\[ \text{OOCH}_2\text{SCH}_2\text{OOH} + \text{HO}_2 = \text{HOOCH}_2\text{SCH}_2\text{OOH} : 1.13 \times 10^{-13} \exp(1300/\text{TEMP}); \]

\[ \text{HPMTF} + \text{OH} = \text{HOOCH}_2\text{SCO} : 1.4 \times 10^{-12}; \]

\[ \text{HOOCH}_2\text{SCO} = \text{HOOCH}_2\text{S} + \text{CO} : 9.2 \times 10^{-9} \exp(-505.4/\text{TEMP}); \]

\[ \text{HOOCH}_2\text{SCO} = \text{HCHO} + \text{OH} + \text{OCS} : 1.6 \times 10^{-7} \exp(-1468.6/\text{TEMP}); \]

\[ \text{HOOCH}_2\text{S} + \text{O}_3 = \text{HOOCH}_2\text{SO} : 1.15 \times 10^{-12} \exp(430/\text{TEMP}); \]
HOOCH$_2$S + NO$_2$ = HOOCH$_2$SO + NO : 6.00D-11*exp(240/TEMP) ;
HOOCH$_2$SO + O$_3$ = SO$_2$ + HCHO + OH : 4.00D-13 ;
HOOCH$_2$SO + NO$_2$ = SO$_2$ + HCHO + OH + NO : 1.20D-11 ;
CH$_3$S + NO$_2$ = CH$_3$SO + NO : 6.00D-11*exp(240/TEMP) ;
CH$_3$S + O$_3$ = CH$_3$SO : 1.15D-12*exp(430/TEMP) ;
CH$_3$S = CH$_3$SO : 1.20D-16*exp(1580/TEMP)*O$_2$ ;
CH$_3$SO + NO$_2$ = CH$_3$O$_2$ + SO$_2$ + NO : 1.20D-11*0.25 ;
CH$_3$SO + NO$_2$ = CH$_3$SO$_2$ + NO : 1.20D-11*0.75 ;
CH$_3$SO + O$_3$ = CH$_3$O$_2$ + SO$_2$ : 4.00D-13 ;
CH$_3$SO = CH$_3$SO$_2$ : 3.12D-16*exp(1580/TEMP)*O$_2$ ;
CH$_3$SO + NO = CH$_3$SO + NO$_2$ : 1.1D-11 ;
CH$_3$SO + NO$_2$ = CH$_3$SO + NO$_3$ : 2.2D-11 ;
CH$_3$SO + O$_3$ = CH$_3$O$_2$ + SO$_2$ : 5.60D+16*exp(-10870/TEMP) ;
CH$_3$SO = CH$_3$S : 3.50D+10*exp(-3560/TEMP) ;
CH$_3$SO + HO$_2$ = CH$_3$SOOH : 4D-12 ;
CH$_3$SO = CH$_3$SO$_2$ : 1.0 ;
CH$_3$SO + HO$_2$ = CH$_3$SO$_2$ + OH : KAPHO$_2$*0.44 ;
CH$_3$SO + HO$_2$ = CH$_3$SOOH : KAPHO$_2$*0.41 ;
CH$_3$SO + O$_3$ = MSIA + O$_3$ : KAPHO$_2$*0.15 ;
CH$_3$SO + NO = CH$_3$SO + NO$_2$ : 1.00D-11 ;
CH$_3$SO + NO$_2$ = CH$_3$SO$_2$ + NO$_2$ : 1.20D-12*(TEMP/300)**(-0.9) ;
CH$_3$SO + NO$_3$ = CH$_3$O$_2$ + NO$_2$ : KRO$_2$NO$_3$*1.74 ;
CH$_3$SO = CH$_3$S : 9.10E+10*exp(-3560/TEMP) ;
CH$_3$SO + NO$_2$ = CH$_3$SO$_2$ + NO : 5.40D+16*exp(-13112/TEMP) ;
CH$_3$SO + O$_3$ = CH$_3$SO$_2$ + NO$_2$ : 1.00D-11*RO$_2$*0.7 ;
CH$_3$SO = MSIA : 1.00D-11*RO$_2$*0.3 ;
CH$_3$SO + HO$_2$ = MSA : 5D-11 ;
CH$_3$SO + NO = CH$_3$SO + NO$_2$ : 1.00D-12*(TEMP/300)**(-0.9) ;
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) ;
CH₃SO₂O₂OH + OH = CH₃SO₂O₂ : 3.60D-12 ;
CH₃SO₂O₂OH = 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 = MSA + 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 = DMSO₂O₂ + NO₂ : KRO₂NO ;
DMSO₂O₂ + NO₃ = DMSO₂O₂ + 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 ;
DMSO₂O₂OH + OH = CH₃SO₂CHO + OH : 1.26D-12 ;
DMSO₂O₂OH + OH = DMSO₂O : 1.40D-12 ;
DMSO₂O₂OH = 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 + HN0₃ : 1D-13 ;
MSA + OH = CH₃SO₃ : 2.24D-14 ;

**S1.4.3 Fung et al. (2021)**
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) ;


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(-1090/TEMP) ;
CH3SOO = CH3SO2 : 1.00 ;
CH3SO2 + O3 = CH3O2 + SO2 : 5.00D+13*EXP(-10870/TEMP) ;
CH3SO3 + HO2 = MSA + O2 : 5.0d-11 ;
CH3SO3 = CH3SO2 + H2SO4 : 5.00D+13*EXP(-1580/TEMP) ;
CH3SO3 + HO2 = CH3SO2 + SO2 : 5.00D13*EXP(-10870/TEMP) ;
CH3SO3 + HO2 = MSA + O2 : 5.0d-11 ;
CH3SO3 = CH3SO2 + H2SO4 : 5.00D+13*EXP(-1580/TEMP) ;

S1.4.4 Khan et al. (2021)
DMS + OH = MTMP+ H2O : 1.2D-11*EXP(-280/TEMP) ;
DMS + OH = MHTM+ H2O : 1.2D-11*EXP(-280/TEMP) ;
DMS + OH = DMSO + H2O : 9.5D-39*O2*EXP(5270/TEMP)/(1+7.5D-5*(O2/M)*EXP(3644/TEMP)) ;
DMS + OH = DMSO + H2O : 9.5D-39*O2*EXP(5270/TEMP)/(1+7.5D-5*(O2/M)*EXP(3644/TEMP)) ;
DMS + NO3 = MTMP + HNO3 : 1.90D-13*EXP(580/TEMP) ;
DMS + NO3 = MTMP + HNO3 : 1.90D-13*EXP(580/TEMP) ;
DMS + OH = MSIA : 8.94D-11*EXP(800/TEMP)*0.95 ;
DMS + OH = SO2 : 8.94D-11*EXP(800/TEMP)*0.95 ;
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 ;

CH3SCH2OOH = CH3SCH2OOH + OH = CH3S + HCHO + OH : (5.78677D-6)*SUN ; {J41}
CH3SCH2OOH + OH = CH3SCH2OOH + OH = CH3S + HCHO + OH : (5.78677D-6)*SUN ; {J41}

CH3SCH2OOH + OH = CH3SCH2OOH + OH = CH3S + HCHO + OH : (5.78677D-6)*SUN ; {J41}
CH3SCH2OOH + OH = CH3SCH2OOH + OH = CH3S + HCHO + OH : (5.78677D-6)*SUN ; {J41}
CH$_3$SCHO + OH = CH$_3$S + CO : 1.1D-11 ;
CH$_3$SCHO = CH$_3$S + CO + HO$_2$ : (1.99124D-5)*SUN ;
CH$_3$S + O$_3$ = CH$_3$SO : 1.15D-12*EXP(430/TEMP) ;
CH$_3$S + NO$_2$ = CH$_3$SO + NO : 3.00D-12*EXP(210/TEMP) ;
CH$_3$SO + NO$_2$ = CH$_3$SO$_2$ + NO : 1.2d-11*0.82 ;
CH$_3$SO + NO$_2$ = SO$_2$ + CH$_3$O$_2$ + NO : 1.2d-11*0.18 ;
CH$_3$SO + O$_3$ = CH$_3$SO$_2$ : 1.15D-12*EXP(430/TEMP) ;
CH$_3$SO$_2$ + NO$_2$ = CH$_3$SO$_3$ + NO : 3.00D-12*EXP(210/TEMP) ;
CH$_3$SO$_2$ + NO$_2$ = SO$_2$ + CH$_3$O$_2$ + NO : 1.2d-11*0.82 ;
CH$_3$SO$_2$ + NO$_2$ = SO$_2$ + CH$_3$O$_2$ + NO : 1.2d-11*0.18 ;
CH$_3$SO$_2$ + O$_3$ = CH$_3$SO$_3$ : 3.00D-13 ;
CH$_3$SO$_3$ + HO$_2$ = MSA : 5.0d-11 ;
CH$_3$SO$_3$ = CH$_3$O$_2$ + H$_2$SO$_4$ : 1.36D14*EXP(-11071/TEMP) ;
DMSO + OH = MSIA + CH$_3$O$_2$ : 8.7d-11*0.95 ;
MSIA + OH = CH$_3$SO$_2$ + H$_2$O : 9.d-11*0.95 ;
MSIA + OH = MSA + HO$_2$ + H$_2$O : 9.d-11*0.05 ;
MSIA + NO$_3$ = CH$_3$SO$_3$ + NO : 1.20D-11 ;
MSIA + NO$_3$ = CH$_3$SO$_3$ + NO : 1.20D-11 ;
MTMP = OOCH$_2$SCH$_2$OOH + O$_2$: 2.74D+7*EXP(-5950/TEMP) ;
OOCH$_2$SCH$_2$OOH = HPMTF + OH: 4.2D+7*EXP(-5390/TEMP) ;
OOCH$_2$SCH$_2$OOH + NO = HOOCH$_2$S + NO$_2$ + HCHO : 4.9D-12*exp(260/TEMP) ;
OOCH$_2$SCH$_2$OOH + HO$_2$ = HOOCH$_2$S + OCS : 1.13D-13*exp(1300/TEMP) ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
HOOCH$_2$SCH$_2$OOH = HOOC + CH$_3$O$_2$ + HCHO + OH : (5.78677D-6)*SUN ;
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

<table>
<thead>
<tr>
<th></th>
<th>CS2</th>
<th>ST</th>
<th>CS2-HPMTF</th>
<th>CS2-HPMTF-FL</th>
<th>CS2-HPMTF-FP</th>
</tr>
</thead>
<tbody>
<tr>
<td>DMS</td>
<td>35.59 h</td>
<td>36.34 h</td>
<td>34.63 h</td>
<td>34.68 h</td>
<td>34.37 h</td>
</tr>
<tr>
<td>MTMP</td>
<td>26.61 min</td>
<td>-</td>
<td>0.99 min</td>
<td>0.98 min</td>
<td>0.25 min</td>
</tr>
<tr>
<td>DMSO</td>
<td>6.17 h</td>
<td>-</td>
<td>6.15 h</td>
<td>6.16 h</td>
<td>6.09 h</td>
</tr>
<tr>
<td>MSA</td>
<td>-</td>
<td>-</td>
<td>6.08 d</td>
<td>6.08 d</td>
<td>6.07 d</td>
</tr>
<tr>
<td>MSIA</td>
<td>6.38 h</td>
<td>-</td>
<td>4.81 h</td>
<td>4.81 h</td>
<td>4.74 h</td>
</tr>
<tr>
<td>HPMTF</td>
<td>-</td>
<td>-</td>
<td>25.76 h</td>
<td>9.66 h</td>
<td>25.79 h</td>
</tr>
</tbody>
</table>
Table S6: Sulfate in the different aerosol modes by weight.

<table>
<thead>
<tr>
<th></th>
<th>Nucleation</th>
<th>Aitken</th>
<th>Accumulation</th>
<th>Coarse</th>
</tr>
</thead>
<tbody>
<tr>
<td>CS2</td>
<td>2.0%</td>
<td>26.2%</td>
<td>69.8%</td>
<td>2.0%</td>
</tr>
<tr>
<td>ST</td>
<td>1.7%</td>
<td>27.8%</td>
<td>69.2%</td>
<td>1.4%</td>
</tr>
<tr>
<td>CS2-HPMTF</td>
<td>2.1%</td>
<td>26.6%</td>
<td>69.5%</td>
<td>1.8%</td>
</tr>
<tr>
<td>CS2-HPMTF-CLD</td>
<td>2.0%</td>
<td>25.5%</td>
<td>70.0%</td>
<td>2.5%</td>
</tr>
</tbody>
</table>