

*Supplementary Information*

**Global-scale analysis of the effect of gas-phase Criegee intermediates (CIs) to the sulphate aerosol formation: General trend and the importance of hydroxy radical decomposed from vibrationally-excited CIs**

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## Text S1 □ Additional information of “Methodology” section

Table S1: Latitudes and longitudes of the five sites analysed in this study

Site	Country	City	Latitude	Longitude
CHI	China	Wuhan	30.6	114.3
IND	India	Raipur	21.3	81.6
ZAF	South Africa	Johannesburg	-26.2	28.0
AUS	Australia	Sydney	-33.9	151.2
BOL	Bolivia	Riberalta	-10.8	-66.1

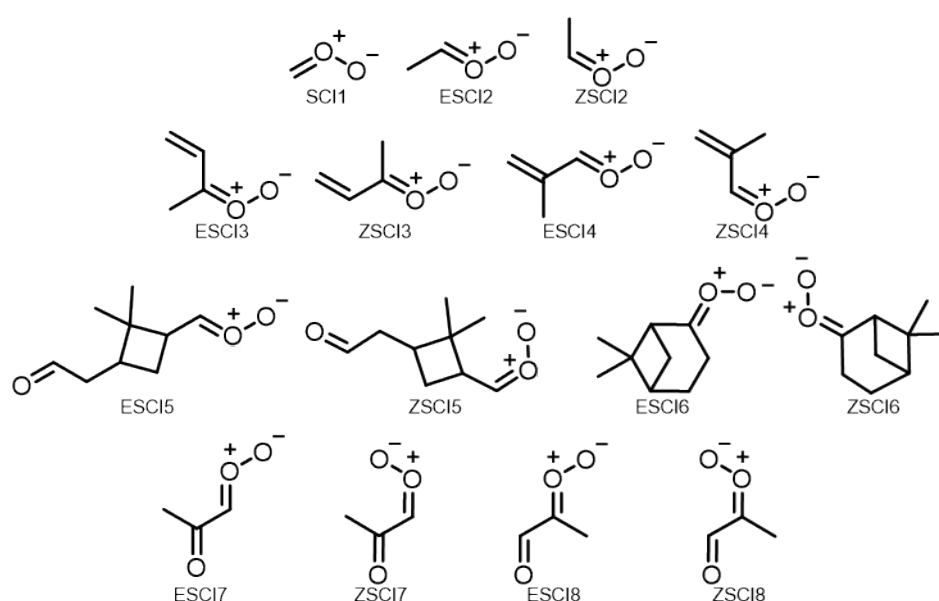


Figure S1: Detailed structures of the Stabilised Criegee intermediates (sCIs) considered in this study

Table S2: Kinetic parameters of the unimolecular reactions of Stabilised Criegee intermediates (sCIs). The kinetic expression is defined by the modified Arrhenius equation [ $k = A(T/300)^B \exp(-E_a/T)$ ], where  $k$  is the rate constant ( $s^{-1}$ ) and  $T$  is the temperature (K). RCHO is an aldehyde with more than C3-chain.

sCIs	Type	Products	$A$ ( $s^{-1}$ )	$B$	$E_a$ (K)
SCI1	13ring	HCOOH	$1.345 \times 10^{11}$	4.00	8024
ESCI2	13ring	ACTA	$3.733 \times 10^{12}$	1.35	7445
ZSCI2	14Hmig	CO + 2OH + CH <sub>2</sub> O	$2.635 \times 10^{-2}$	25.00	-2685
	13ring	ACTA	$4.360 \times 10^{11}$	2.98	10755
ESCI3	13ring	MEK	$5.517 \times 10^{12}$	0.48	10385

	14Hmig	MVK + OH	$6.265 \times 10^{-3}$	25.90	-2737
ZSCI3	15ring	RCHO	$1.198 \times 10^{12}$	0.65	5617
	13ring	MEK	$2.076 \times 10^{12}$	1.12	8291
ESCI4	13ring	RCOOH	$2.453 \times 10^{12}$	1.46	7832
ZSCI4	15ring	RCHO	$1.956 \times 10^{12}$	0.44	6102
	13ring	RCOOH	$8.008 \times 10^{11}$	1.68	8476
ESCI5	13ring	MEK	$4.036 \times 10^{11}$	2.43	6732
ZSCI5	14Hmig	MEK + OH	$6.373 \times 10^{-1}$	23.00	-2099
	13ring	MEK	$2.802 \times 10^{11}$	3.45	9906
ESCI6	13ring	MEK	$2.295 \times 10^{12}$	1.23	10358
	14Hmig	MEK + OH	$2.358 \times 10^{-1}$	23.27	-2251
ZSCI6	13ring	MEK	$3.744 \times 10^{12}$	1.33	8425
ESCI7	13ring	RCOOH	$1.176 \times 10^{12}$	2.30	9127
ZSCI7	13ring	RCOOH	$5.178 \times 10^{11}$	2.41	7141
ESCI8	14Hmig	RCHO + OH	$2.375 \times 10^{-5}$	30.08	-3644
	13ring	RCHO	$1.439 \times 10^{12}$	2.34	12328
ZSCI8	13ring	RCHO	$2.361 \times 10^{12}$	0.78	5162

Table S3: Kinetic parameters for the reactions of Stabilised Criegee intermediates (sCIs) with H<sub>2</sub>O. The kinetic expression is defined by the modified Arrhenius equation [ $k = A(T/300)^B \exp(-E_a/T)$ ], where  $k$  is the rate constant (cm<sup>3</sup> molec.<sup>-1</sup> s<sup>-1</sup>) and  $T$  is the temperature (K). HMHP, hydroxymethyl hydroperoxide; PP, propyl peroxide; MVKHP, hydroxyhydroperoxide derived from MVK; MACR1OOH, hydroxy hydroperoxide derived from MACR; GLYX, glyoxal.

SCIs	Products	$A$ (cm <sup>3</sup> molec. <sup>-1</sup> s <sup>-1</sup> )	$B$	$E_a$ (K)
SCI1	0.4CH <sub>2</sub> O + 0.55HMHP + 0.05HCOOH	$9.056 \times 10^{-15}$	1.23	698
ESCI2	0.4ALD <sub>2</sub> + 0.55PP + 0.05ACTA	$6.045 \times 10^{-15}$	1.27	-405
ZSCI2	0.4ALD <sub>2</sub> + 0.55PP + 0.05ACTA	$3.177 \times 10^{-15}$	1.68	2513
ESCI3	0.4MVK + 0.55MVKHP	$2.924 \times 10^{-15}$	1.46	3132
ZSCI3	0.4MVK + 0.55MVKHP	$9.278 \times 10^{-16}$	2.27	1858
ESCI4	0.4MACR + 0.55MACR1OOH + 0.05RCOOH	$4.351 \times 10^{-15}$	1.74	929
ZSCI4	0.4MACR + 0.55MACR1OOH + 0.05RCOOH	$3.425 \times 10^{-15}$	1.52	2977
ESCI5	0.4MEK + 0.55PP	$1.295 \times 10^{-14}$	0.17	-71
ZSCI5	0.4MEK + 0.55PP	$5.283 \times 10^{-17}$	3.83	1023
ESCI6	0.4MEK + 0.55PP	$3.030 \times 10^{-16}$	2.37	1525
ZSCI6	0.4MEK + 0.55PP	$2.931 \times 10^{-16}$	2.64	-121

ESCI7	0.4GLYX + 0.55PP + 0.05RCOOH	$9.804 \times 10^{-15}$	1.53	2258
ZSCI7	0.4GLYX + 0.55PP + 0.05RCOOH	$2.435 \times 10^{-15}$	1.03	-131
ESCI8	0.45MGLY + 0.55PP	$4.674 \times 10^{-15}$	1.87	4177
ZSCI8	0.45MGLY + 0.55PP	$1.133 \times 10^{-15}$	1.68	-757

Table S4: Kinetic parameters of the reactions of Stabilised Criegee intermediates (sCIs) with  $(\text{H}_2\text{O})_2$ . The kinetic expression is defined as  $k = A(T/300)^B$ , where  $k$  is the rate constant ( $\text{cm}^3 \text{ molec.}^{-1} \text{ s}^{-1}$ ) and  $T$  is the temperature (K). The products are assumed to be the same as those used in the reactions of the sCIs with  $\text{H}_2\text{O}$ , according to Nakamura et al.

SCIs	Products	$A$ ( $\text{cm}^3 \text{ molec.}^{-1} \text{ s}^{-1}$ )	$B$
SCI1	0.4CH <sub>2</sub> O + 0.55HMHP + 0.05HCOOH	$2.900 \times 10^{-33}$	-11.1
ESCI2	0.4ALD2 + 0.55PP + 0.05ACTA	$4.964 \times 10^{-32}$	-14.7
ZSCI2	0.4ALD2 + 0.55PP + 0.05ACTA	$4.209 \times 10^{-36}$	-4.82
ESCI3	0.4MVK + 0.55MVKHP	$6.525 \times 10^{-37}$	-3.1
ZSCI3	0.4MVK + 0.55MVKHP	$9.914 \times 10^{-36}$	-6.38
ESCI4	0.4MACR + 0.55MACR1OOH + 0.05RCOOH	$6.927 \times 10^{-34}$	-9.88
ZSCI4	0.4MACR + 0.55MACR1OOH + 0.05RCOOH	$1.172 \times 10^{-36}$	-3.51
ESCI5	0.4MEK + 0.55PP	$3.308 \times 10^{-32}$	-14.5
ZSCI5	0.4MEK + 0.55PP	$7.396 \times 10^{-36}$	-7.55
ESCI6	0.4MEK + 0.55PP	$7.385 \times 10^{-36}$	-7.23
ZSCI6	0.4MEK + 0.55PP	$1.145 \times 10^{-33}$	-12.5
ESCI7	0.4GLYX + 0.55PP + 0.05RCOOH	$2.718 \times 10^{-35}$	-5.74
ZSCI7	0.4GLYX + 0.55PP + 0.05RCOOH	$8.339 \times 10^{-33}$	-14
ESCI8	0.45MGLY + 0.55PP	$4.273 \times 10^{-38}$	0.58
ZSCI8	0.45MGLY + 0.55PP	$2.702 \times 10^{-32}$	-15.5

Table S5: Kinetic parameters for the reactions of Stabilised Criegee intermediates (sCIs) with  $\text{SO}_2$ .  $k$ , rate constant;  $\text{SO}_4$ , sulphate in the gas phase.

SCIs	Products	$k$ ( $\text{cm}^3 \text{ molec.}^{-1} \text{ s}^{-1}$ )
SCI1	CH <sub>2</sub> O + SO <sub>4</sub>	$3.700 \times 10^{-11}$
ESCI2	ALD2 + SO <sub>4</sub>	$1.400 \times 10^{-10}$
ZSCI2	ALD2 + SO <sub>4</sub>	$2.700 \times 10^{-11}$
ESCI3	MVK + SO <sub>4</sub>	$1.400 \times 10^{-10}$
ZSCI3	MVK + SO <sub>4</sub>	$2.700 \times 10^{-11}$
ESCI4	MACR + SO <sub>4</sub>	$1.400 \times 10^{-10}$
ZSCI4	MACR + SO <sub>4</sub>	$2.700 \times 10^{-11}$

ESCI5	MEK + SO4	$1.400 \times 10^{-10}$
ZSCI5	MEK + SO4	$2.700 \times 10^{-11}$
ESCI6	MEK + SO4	$1.400 \times 10^{-10}$
ZSCI6	MEK + SO4	$2.700 \times 10^{-11}$
ESCI7	GLYX + SO4	$1.400 \times 10^{-10}$
ZSCI7	GLYX + SO4	$2.700 \times 10^{-11}$
ESCI8	MGLY + SO4	$1.400 \times 10^{-10}$
ZSCI8	MGLY + SO4	$2.700 \times 10^{-11}$

Table S6: Kinetic parameters of the reactions of Stabilised Criegee intermediates (sCIs) with  $\text{NO}_2$ .  $k$ , rate constant;  $\text{NO}_3$ , nitrate in the gas phase; ALD2, acetaldehyde.

SCIs	Products	$k$ ( $\text{cm}^3 \text{ molec.}^{-1} \text{ s}^{-1}$ )
SCI1	0.300CH2O + 0.300NO3	$4.000 \times 10^{-12}$
ESCI2	0.300ALD2 + 0.300NO3	$4.000 \times 10^{-12}$
ZSCI2	0.300ALD2 + 0.300NO3	$4.000 \times 10^{-12}$
ESCI3	0.300MVK + 0.300NO3	$4.000 \times 10^{-12}$
ZSCI3	0.300MVK + 0.300NO3	$4.000 \times 10^{-12}$
ESCI4	0.300MACR + 0.300NO3	$4.000 \times 10^{-12}$
ZSCI4	0.300MACR + 0.300NO3	$4.000 \times 10^{-12}$
ESCI5	0.300MEK + 0.300NO3	$4.000 \times 10^{-12}$
ZSCI5	0.300MEK + 0.300NO3	$4.000 \times 10^{-12}$
ESCI6	0.300MEK + 0.300NO3	$4.000 \times 10^{-12}$
ZSCI6	0.300MEK + 0.300NO3	$4.000 \times 10^{-12}$
ESCI7	0.300GLYX + 0.300NO3	$4.000 \times 10^{-12}$
ZSCI7	0.300GLYX + 0.300NO3	$4.000 \times 10^{-12}$
ESCI8	0.300MGLY + 0.300NO3	$4.000 \times 10^{-12}$
ZSCI8	0.300MGLY + 0.300NO3	$4.000 \times 10^{-12}$

Table S7: Kinetic parameters for the reactions of Stabilised Criegee intermediates (sCIs) with  $\text{HNO}_3$ .  $k$  is a rate constant.

SCIs	Products	$k$ ( $\text{cm}^3 \text{ molec.}^{-1} \text{ s}^{-1}$ )
SCI1	0.21OH	$5.400 \times 10^{-10}$
ESCI2	0.21OH	$5.400 \times 10^{-10}$
ZSCI2	0.21OH	$5.400 \times 10^{-10}$
ESCI3	0.21OH	$5.400 \times 10^{-10}$
ZSCI3	0.21OH	$5.400 \times 10^{-10}$

ESCI4	0.21OH	$5.400 \times 10^{-10}$
ZSCI4	0.21OH	$5.400 \times 10^{-10}$
ESCI5	0.21OH	$5.400 \times 10^{-10}$
ZSCI5	0.21OH	$5.400 \times 10^{-10}$
ESCI6	0.21OH	$5.400 \times 10^{-10}$
ZSCI6	0.21OH	$5.400 \times 10^{-10}$
ESCI7	0.21OH	$5.400 \times 10^{-10}$
ZSCI7	0.21OH	$5.400 \times 10^{-10}$
ESCI8	0.21OH	$5.400 \times 10^{-10}$
ZSCI8	0.21OH	$5.400 \times 10^{-10}$

Table S8: Kinetic parameters for the reactions of Stabilised Criegee intermediates (sCIs) with HCOOH.  $k$ , rate constant; ATOOH, acetone-derived peroxides.

SCIs	Products	$k$ ( $\text{cm}^3 \text{ molec.}^{-1} \text{ s}^{-1}$ )
SCI1	ATOOH	$1.200 \times 10^{-10}$
ESCI2	ATOOH	$3.800 \times 10^{-10}$
ZSCI2	CO + 2.000OH + CH <sub>2</sub> O + HCOOH	$2.100 \times 10^{-10}$
ESCI3	MVK + OH + HCOOH	$3.800 \times 10^{-10}$
ZSCI3	ATOOH	$2.100 \times 10^{-10}$
ESCI4	ATOOH	$3.800 \times 10^{-10}$
ZSCI4	ATOOH	$2.100 \times 10^{-10}$
ESCI5	MEK + HCOOH	$3.800 \times 10^{-10}$
ZSCI5	MEK + OH + HCOOH	$2.100 \times 10^{-10}$
ESCI6	MEK + OH + HCOOH	$3.800 \times 10^{-10}$
ZSCI6	MEK	$2.100 \times 10^{-10}$
ESCI7	ATOOH	$3.800 \times 10^{-10}$
ZSCI7	ATOOH	$2.100 \times 10^{-10}$
ESCI8	RCHO	$3.800 \times 10^{-10}$
ZSCI8	ATOOH	$2.100 \times 10^{-10}$

Table S9: Kinetic parameters for the reactions of Stabilised Criegee intermediates (sCIs) with CH<sub>3</sub>COOH.  $k$  is a rate constant for which the value of the rate constants was assumed to be the same as that of the reactions of sCIs with HCOOH, according to Nakamura et al.

SCIs	Products	$k$ ( $\text{cm}^3 \text{ molec.}^{-1} \text{ s}^{-1}$ )
SCI1	ATOOH	$1.200 \times 10^{-10}$
ESCI2	ATOOH	$3.800 \times 10^{-10}$

ZSCI2	ACET + OH + ACTA	$2.100 \times 10^{-10}$
ESCI3	MVK + OH + ACTA	$3.800 \times 10^{-10}$
ZSCI3	ATOOH	$2.100 \times 10^{-10}$
ESCI4	ATOOH	$3.800 \times 10^{-10}$
ZSCI4	ATOOH	$2.100 \times 10^{-10}$
ESCI5	MEK + ACTA	$3.800 \times 10^{-10}$
ZSCI5	MEK + OH + ACTA	$2.100 \times 10^{-10}$
ESCI6	MEK + OH + ACTA	$3.800 \times 10^{-10}$
ZSCI6	MEK + ACTA	$2.100 \times 10^{-10}$
ESCI7	ATOOH	$3.800 \times 10^{-10}$
ZSCI7	ATOOH	$2.100 \times 10^{-10}$
ESCI8	RCHO + ACTA	$3.800 \times 10^{-10}$
ZSCI8	ATOOH	$3.800 \times 10^{-10}$

Table S10: Kinetic parameters for the reactions of Stabilised Criegee intermediates (sCIs) with RCOOH.  $k$  is a rate constant for which the value of the rate constants was assumed to be the same as that of the reactions of sCIs with HCOOH and CH<sub>3</sub>COOH, according to Nakamura et al.

SCIs	Products	$k$ (cm <sup>3</sup> molec. <sup>-1</sup> s <sup>-1</sup> )
SCI1	ATOOH	$1.200 \times 10^{-10}$
ESCI2	ATOOH	$3.800 \times 10^{-10}$
ZSCI2	ACET + OH + RCOOH	$2.100 \times 10^{-10}$
ESCI3	MVK + OH + RCOOH	$3.800 \times 10^{-10}$
ZSCI3	ATOOH	$2.100 \times 10^{-10}$
ESCI4	ATOOH	$3.800 \times 10^{-10}$
ZSCI4	ATOOH	$2.100 \times 10^{-10}$
ESCI5	MEK + RCOOH	$3.800 \times 10^{-10}$
ZSCI5	MEK + OH + RCOOH	$2.100 \times 10^{-10}$
ESCI6	MEK + OH + RCOOH	$3.800 \times 10^{-10}$
ZSCI6	MEK + RCOOH	$2.100 \times 10^{-10}$
ESCI7	ATOOH	$3.800 \times 10^{-10}$
ZSCI7	ATOOH	$2.100 \times 10^{-10}$
ESCI8	RCHO + RCOOH	$3.800 \times 10^{-10}$
ZSCI8	ATOOH	$3.800 \times 10^{-10}$

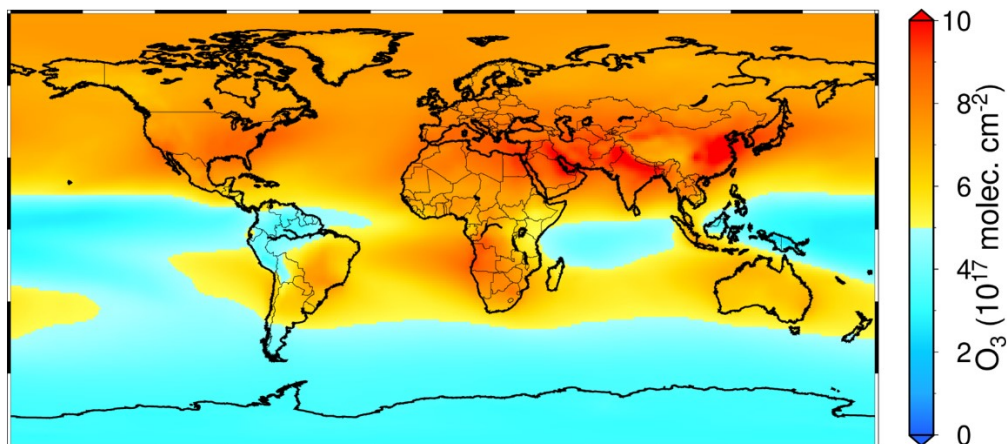
Table S11: Alkene ozonolysis reactions in this study

Alkenes	Products from alkene ozonolysis
Ethylene (C <sub>2</sub> H <sub>4</sub> )	CH <sub>2</sub> O + 0.370SCI <sub>1</sub> + 0.130HO <sub>2</sub> + 0.130OH + 0.630CO
Propene (PRPE)	0.500ACET + 0.500CH <sub>2</sub> O + 0.120SCI <sub>1</sub> + 0.060ESCI <sub>2</sub> + 0.060ZSCI <sub>2</sub> + 0.820HO <sub>2</sub> + 0.360OH + 0.560CO + 0.280MO <sub>2</sub>
Methyl Vinyl Ketone (MVK)	0.600CH <sub>2</sub> O + 0.120SCI <sub>1</sub> + 0.060ESCI <sub>7</sub> + 0.060ZSCI <sub>7</sub> + 0.780HO <sub>2</sub> + 0.360OH + 1.060CO + 0.780MCO <sub>3</sub> + 0.100HO <sub>2</sub> + 0.100PYAC
Methacrolein (MACR)	0.500CH <sub>2</sub> O + 0.090SCI <sub>1</sub> + 0.045ESCI <sub>8</sub> + 0.045ZSCI <sub>8</sub> + 0.910HO <sub>2</sub> + 0.820OH + 1.320CO + 0.910MCO <sub>3</sub>
Isoprene (ISOP)	0.300MACR + 0.200MVK + 0.110SCI <sub>1</sub> + 0.022ESCI <sub>3</sub> + 0.022ZSCI <sub>3</sub> + 0.033ESCI <sub>4</sub> + 0.033ZSCI <sub>4</sub> + 0.271HO <sub>2</sub> + 0.270OH + 0.522CO + 0.709CH <sub>2</sub> O + 0.105MO <sub>2</sub> + 0.128PRPE
Pinene (MTPA)	0.100ESCI <sub>5</sub> + 0.100ZSCI <sub>5</sub> + 0.800OH + 0.200CO + 0.800KO <sub>2</sub> + 0.30CH <sub>2</sub> O + 0.074SCI <sub>1</sub> + 0.028ESCI <sub>6</sub> + 0.028ZSCI <sub>6</sub> + 0.026HO <sub>2</sub>
Other monoterpenes (MTPO)	0.050ESCI <sub>5</sub> + 0.050ZSCI <sub>5</sub> + 0.576OH + 0.226CO + 0.700KO <sub>2</sub> + 0.30CH <sub>2</sub> O + 0.074SCI <sub>1</sub> + 0.028ESCI <sub>6</sub> + 0.028ZSCI <sub>6</sub> + 0.026HO <sub>2</sub>
Limonene (LIMO)	0.050ESCI <sub>5</sub> + 0.050ZSCI <sub>5</sub> + 0.576OH + 0.226CO + 0.700KO <sub>2</sub> + 0.30CH <sub>2</sub> O + 0.074SCI <sub>1</sub> + 0.028ESCI <sub>6</sub> + 0.028ZSCI <sub>6</sub> + 0.026HO <sub>2</sub>

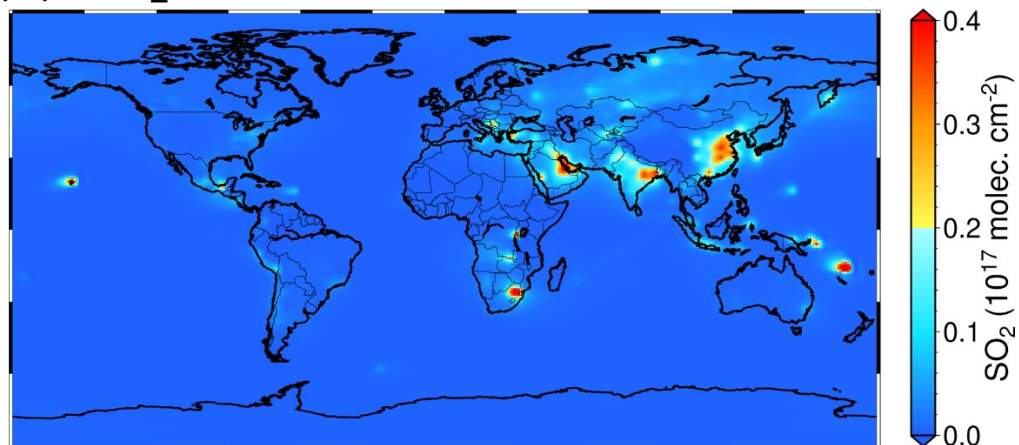


Text S2 □ *Additional information of “Results and Discussion” section*

(a) O<sub>3</sub>



(b) SO<sub>2</sub>



(c) Total alkenes

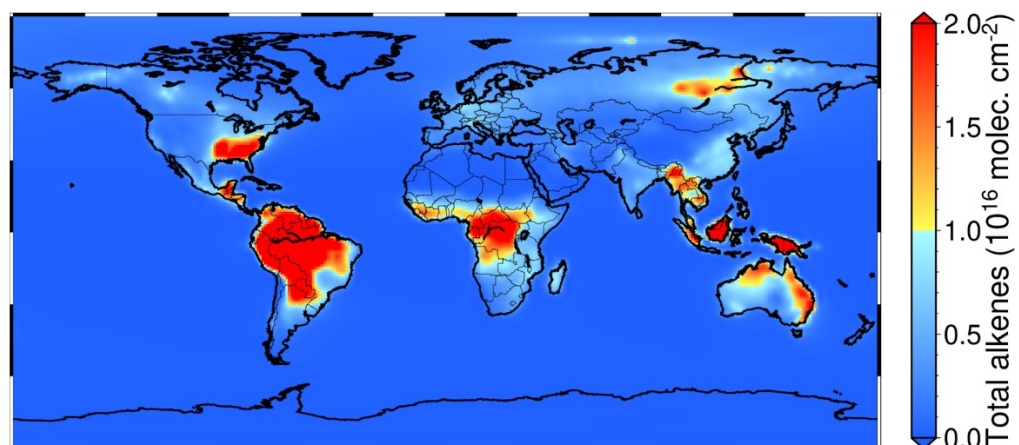


Figure S2: Annual mean column densities of (a) O<sub>3</sub>, (b) SO<sub>2</sub>, and (c) total alkenes. O<sub>3</sub> and alkenes are the precursors of Stabilised Criegee intermediates (sCIs), whereas SO<sub>2</sub> is the precursor of SO<sub>4</sub><sup>2-(p)</sup>.

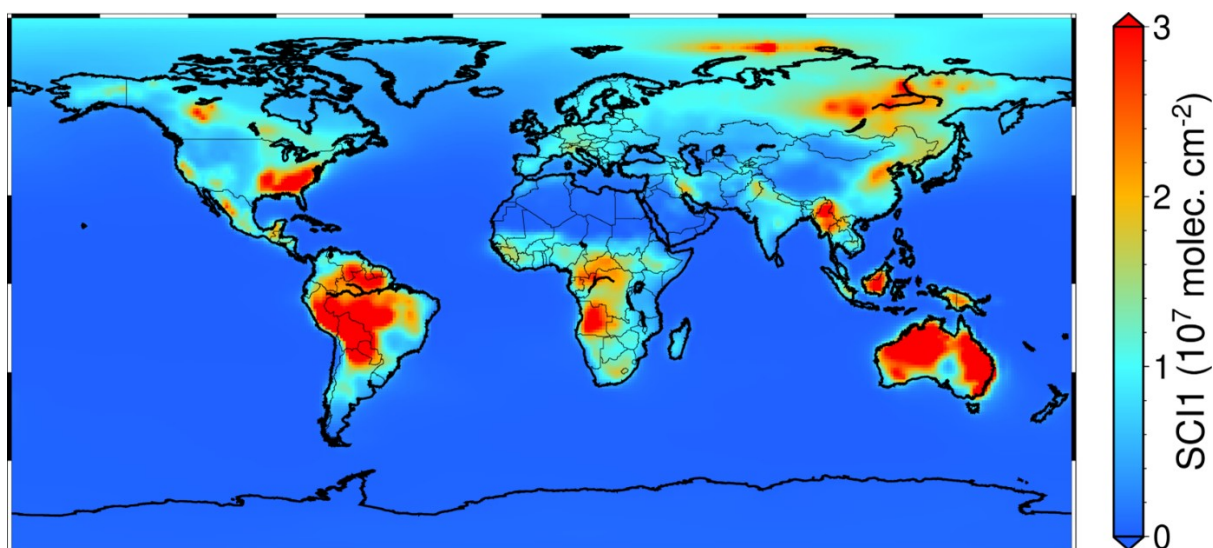


Figure S3: Annual mean column density of SCI1 derived from ozonolysis of alkenes emitted by both anthropogenic and natural sources.

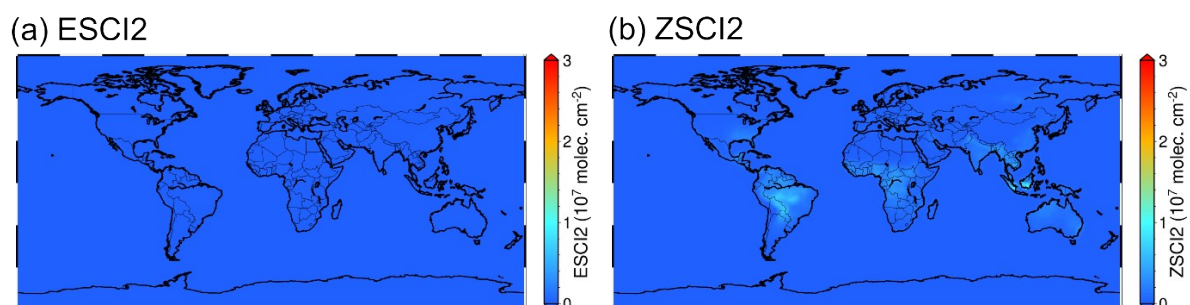


Figure S4: Annual mean column densities of ESCI2 and ZSCI2 derived from the ozonolysis of alkenes emitted by both anthropogenic and natural sources, respectively.

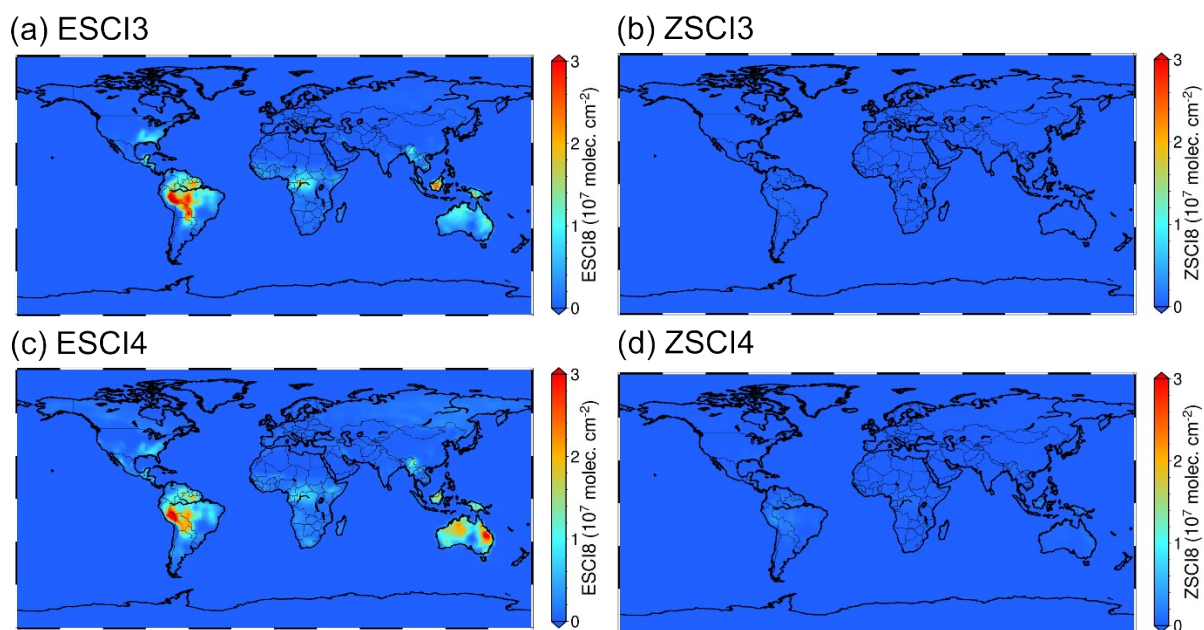


Figure S5: Annual mean column densities of *E*- and *Z*-isomers of Stabilised Criegee intermediates (sCIs) derived from isoprene ozonolysis.

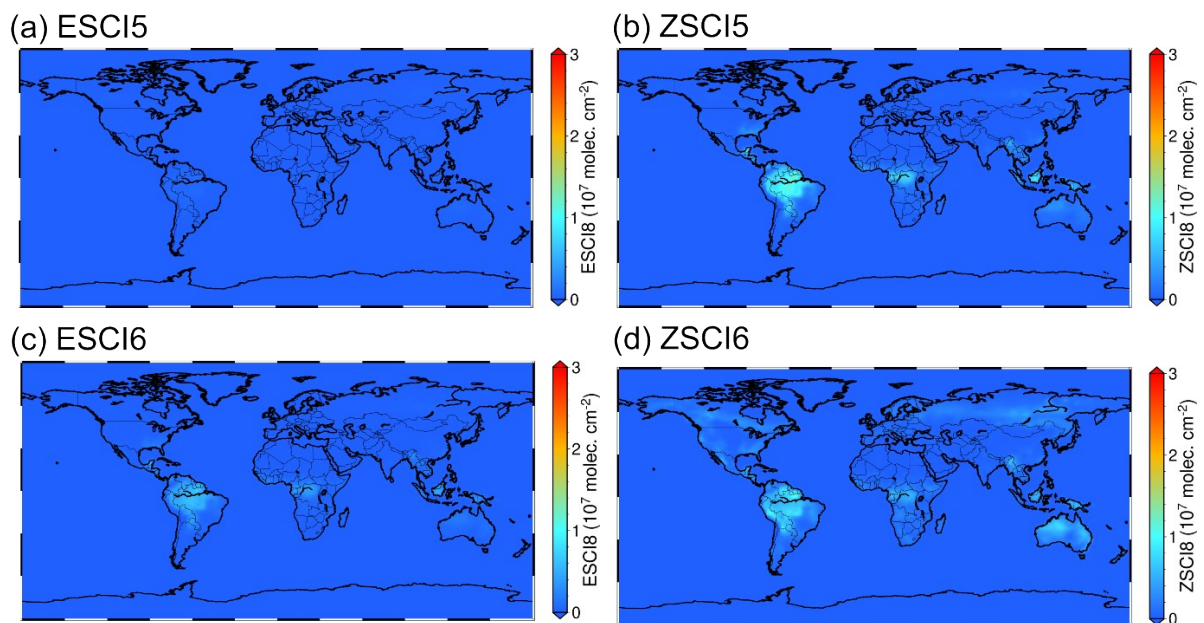
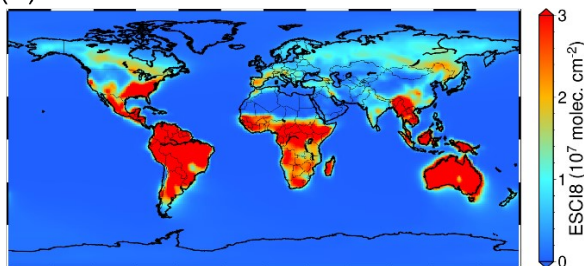


Figure S6: Annual mean column densities of *E*- and *Z*-isomers of Stabilised Criegee intermediates (sCIs) derived from the ozonolysis of monoterpenes.



(a) ESCI7



(b) ZSCI7

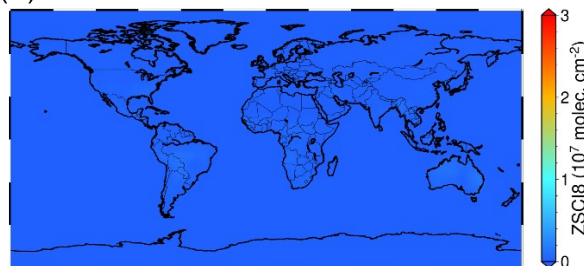
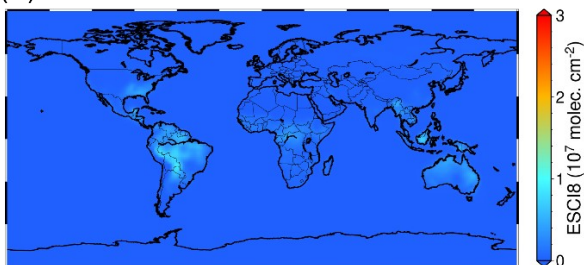


Figure S7: Annual mean column densities of *E*- and *Z*-isomers of Stabilised Criegee intermediates (sCIs) derived from the ozonolysis of methyl vinyl ketone.

(a) ESCI8



(b) ZSCI8

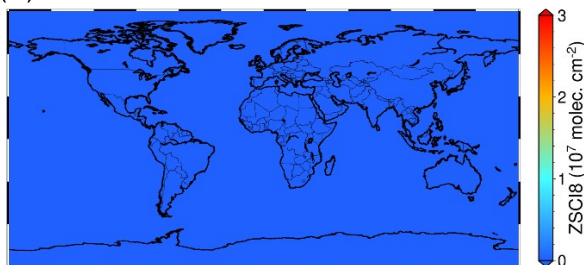


Figure S8: Annual mean column densities of *E*- and *Z*-isomers of Stabilised Criegee intermediates (sCIs) derived from the ozonolysis of methacrolein.

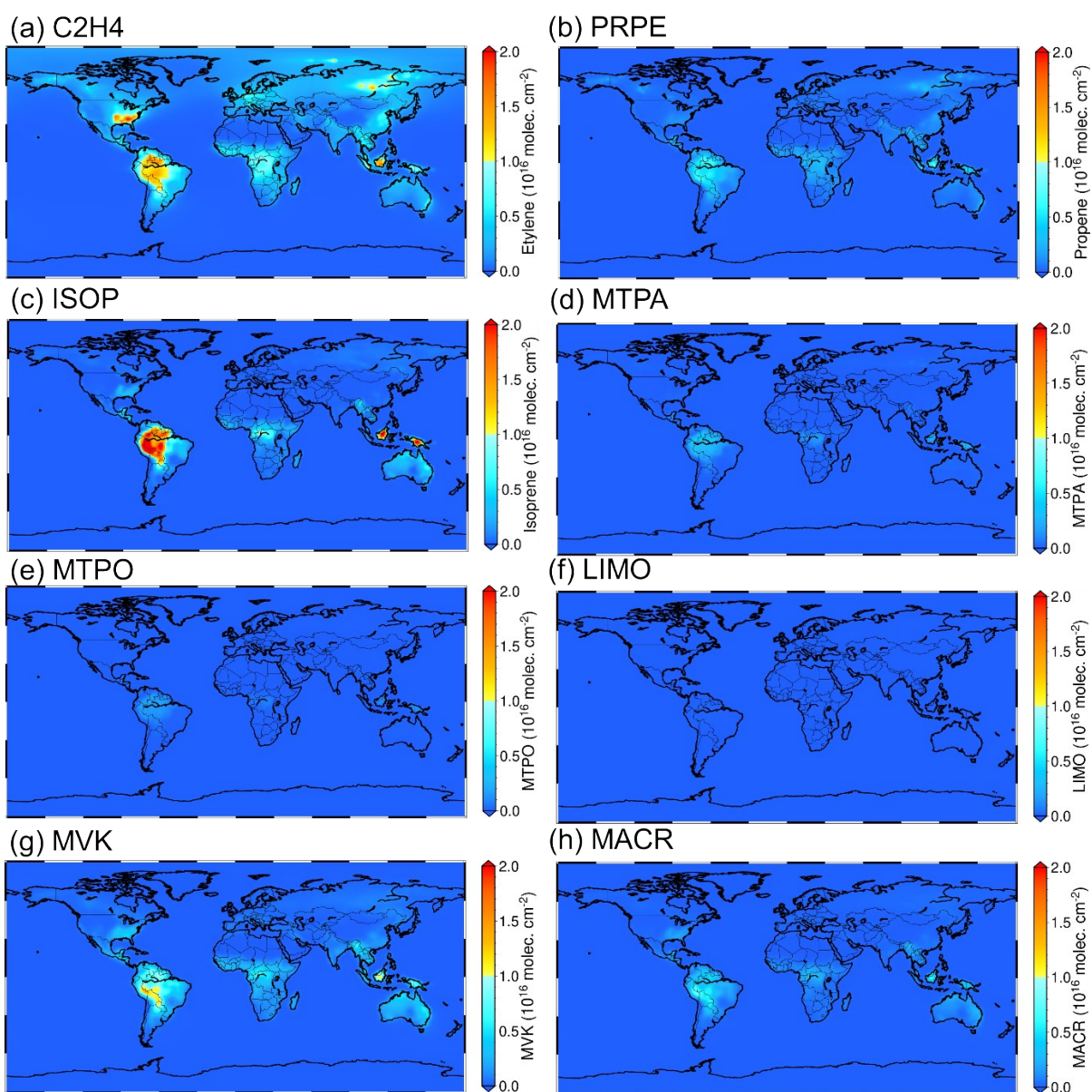


Figure S9: Annual mean column density of alkenes: (a) ethylene, (b) propene, (c) isoprene, (d)  $\alpha$ -pinene, (e) monoterpenes aside from  $\alpha$ -pinene, (f) limonene, (g) methyl vinyl ketone, and (h) methacrolein.

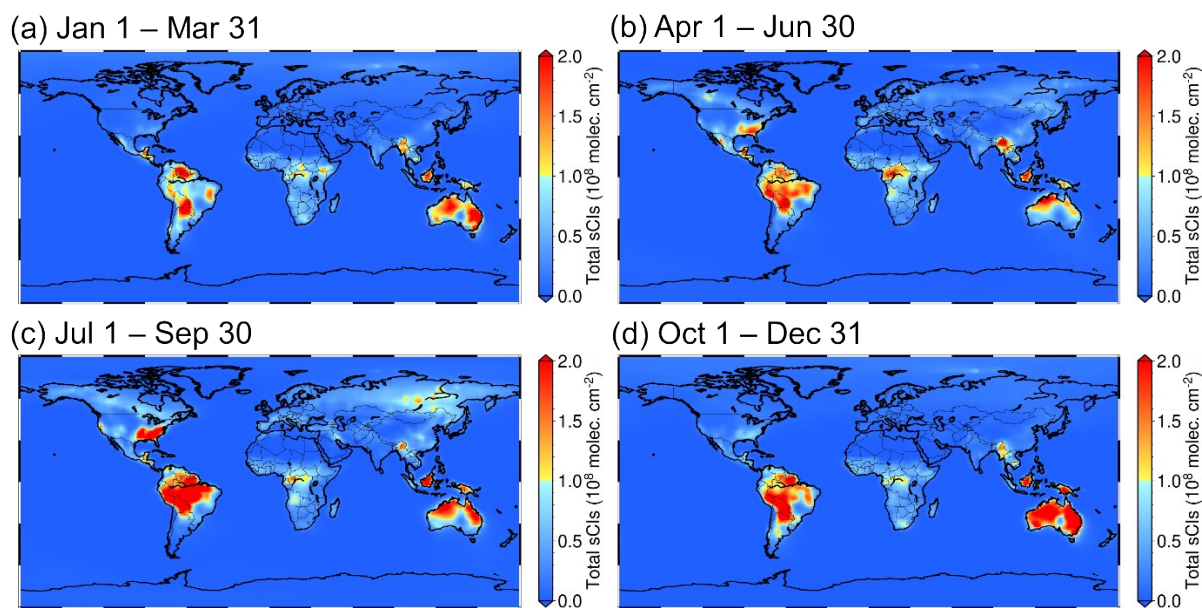
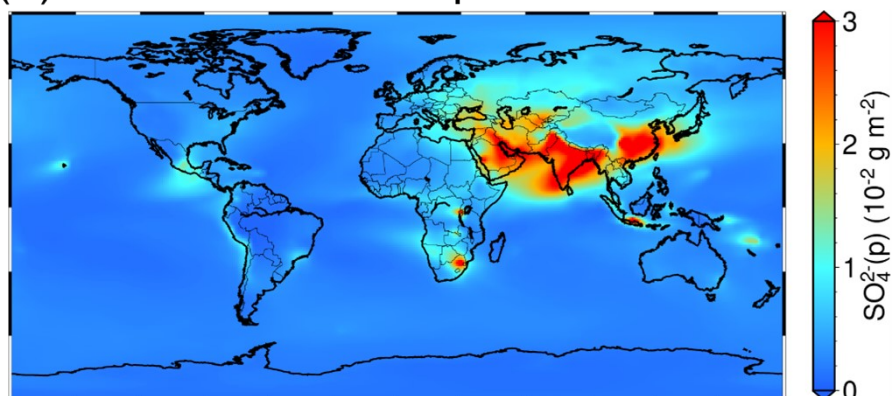
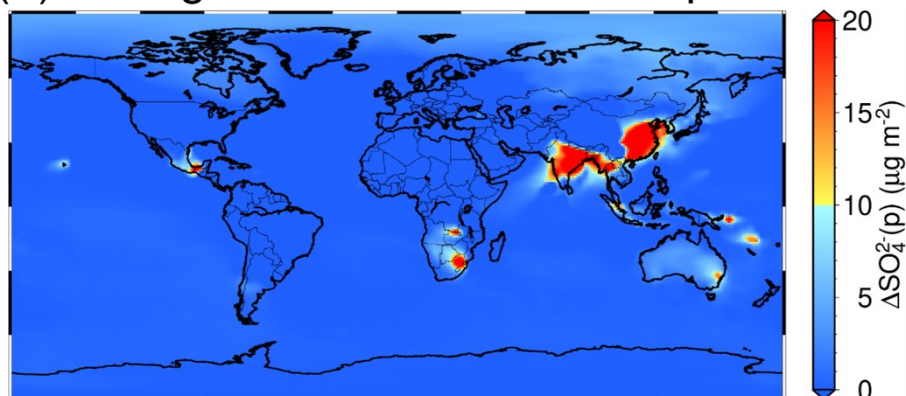


Figure S10: Seasonal trend of the total column density of sCIs in 2019: (a) January, February, and March; (b) April, May, and June; (c) July, August, and September; and (d) October, November, and December.

(a) Jan-Mar-mean sulphate



(b) Change of Jan-Mar-mean sulphate



(c) Ratio of the change

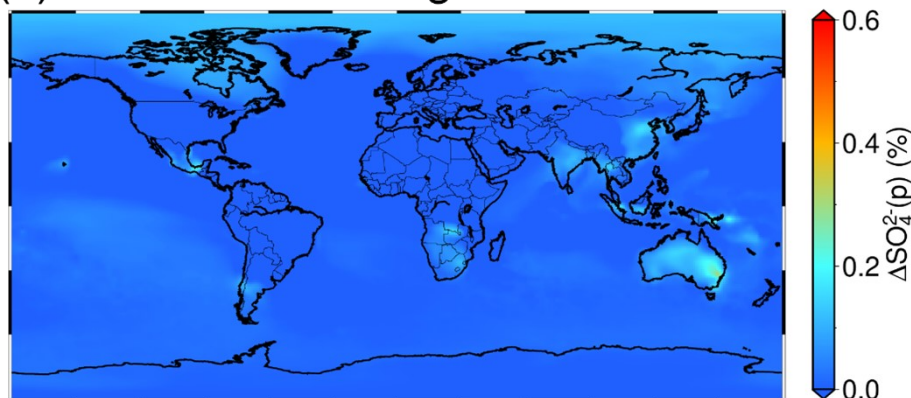
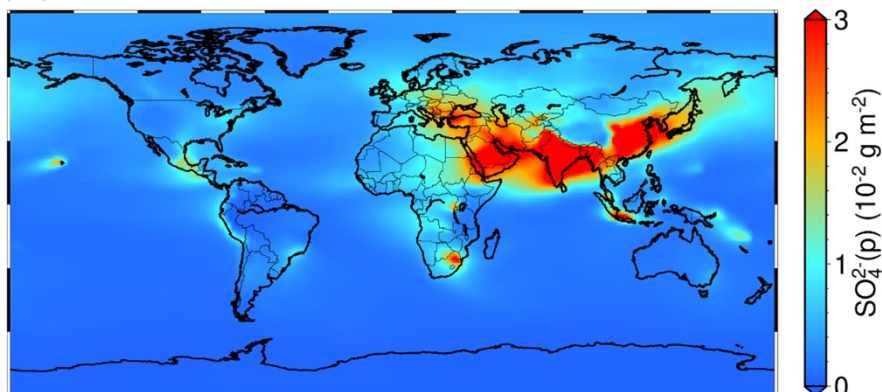


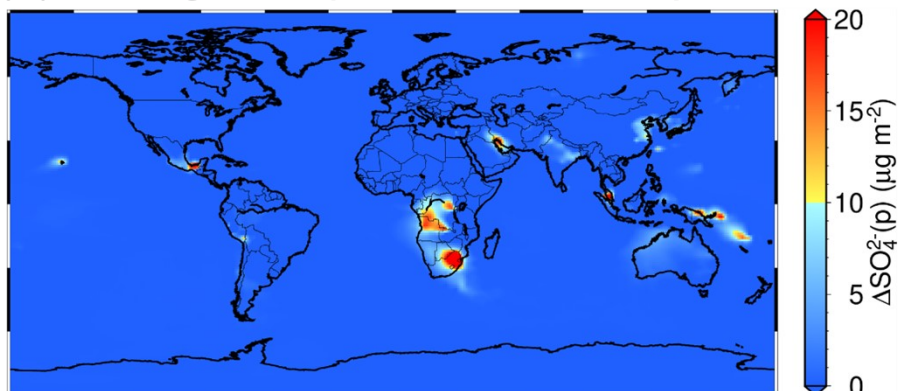
Figure S11: Mean column density from January to March for (a) sulphate ( $\text{g m}^{-2}$ ), (b) the change in column density of sulphate before and after the introduction of Criegee chemistry ( $\text{mg m}^{-2}$ ), and (c) the rate of change of sulphate before and after the introduction of Criegee chemistry (%). Note that the colour range for (c) is set to 0.0–0.6 %, which differs from the colour range of Fig. 4(c) in the main manuscript (0.0–0.4%) for improved readability.



(a) Apr-Jun-mean sulphate



(b) Change of Apr-Jun-mean sulphate



(c) Ratio of the change

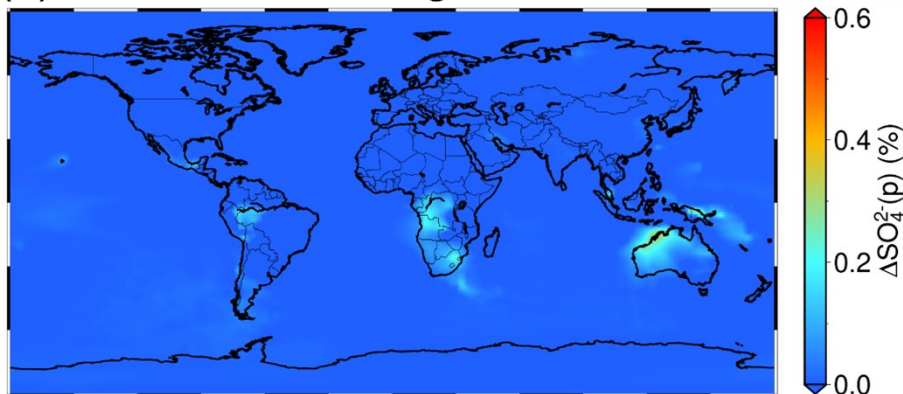
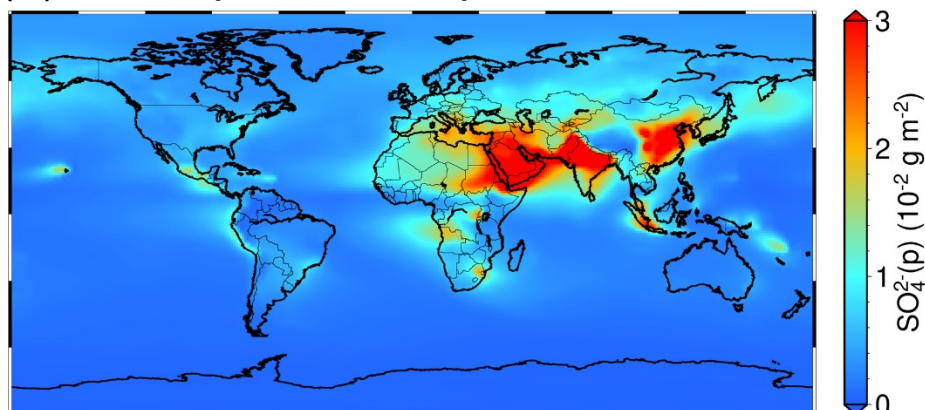


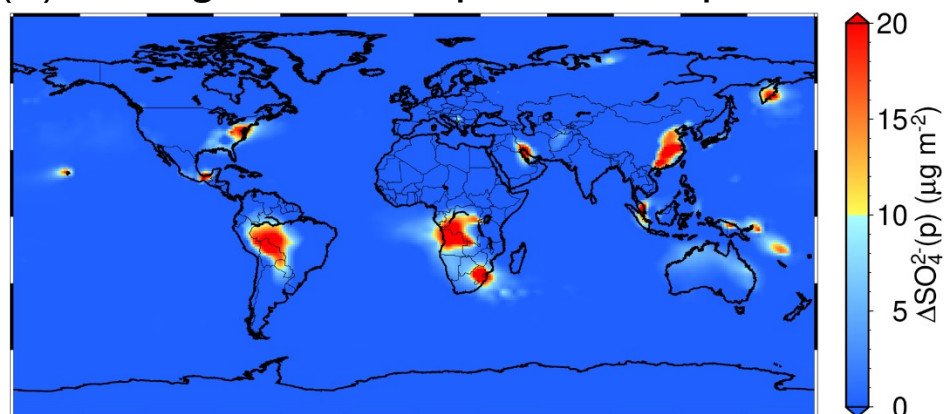
Figure S12: Mean column density from April to June for (a) sulphate ( $\text{g m}^{-2}$ ), (b) the change in column density of sulphate before and after the introduction of Criegee chemistry ( $\text{mg m}^{-2}$ ), and (c) the rate of change of sulphate before and after the introduction of Criegee chemistry (%). Note that the colour range for (c) is set to 0.0–0.6 % which differs from the colour range of Fig. 4(c) in the main manuscript (0.0–0.4%) for improved readability.



(a) Jul-Sep-mean sulphate



(b) Change of Jul-Sep-mean sulphate



(c) Ratio of the change

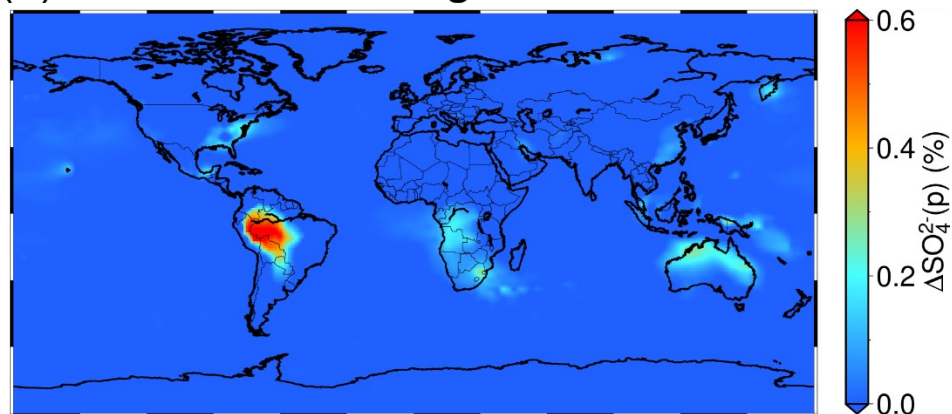
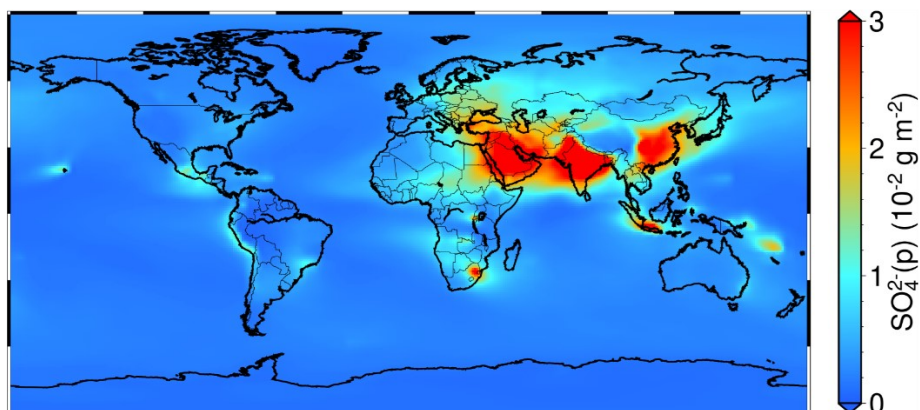
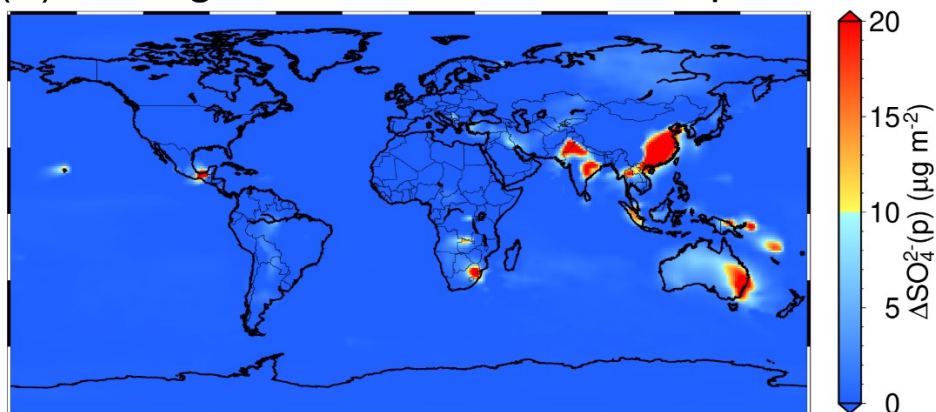


Figure S13: Mean column density from July to September for (a) sulphate ( $\text{g m}^{-2}$ ), (b) the change in column density of sulphate before and after the introduction of Criegee chemistry ( $\text{mg m}^{-2}$ ), and (c) the rate of change of sulphate before and after the introduction of Criegee chemistry (%). Note that the colour range for (c) is set to 0.0–0.6%, which differs from the colour range of Fig. 4(c) in the main manuscript (0.0–0.4%) for improved readability.

(a) Oct-Dec-mean sulphate



(b) Change of Oct-Dec-mean sulphate



(c) Ratio of the change

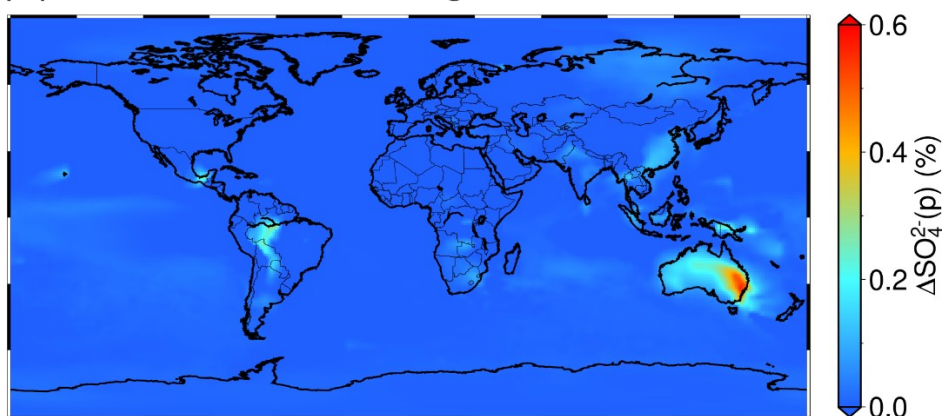


Figure S14: Mean column density from October to December for (a) sulphate ( $\text{g m}^{-2}$ ), (b) the change in column density of sulphate before and after the introduction of Criegee chemistry ( $\text{mg m}^{-2}$ ), and (c) the rate of change of sulphate before and after the introduction of Criegee chemistry (%). Note that the colour range for (c) is set to 0.0–0.6%, which differs from the colour range of Fig. 4(c) in the main manuscript (0.0–0.4%) for improved readability.