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Gaseous and Particle Products Formation from α -Pinene Ozonolysis under Conditions of Different Relative Humidities and Particle Seeds

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2
3 **Gaseous and Particle Products Formation from α -Pinene Ozonolysis under**
4 **Conditions of Different Relative Humidities and Particle Seeds**

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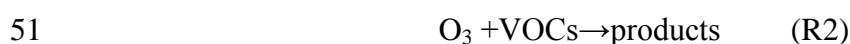
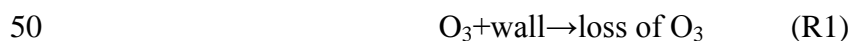
42 Figure S4. Average yields of (a) HCHO, (b) acetone, (c) α -pinene oxide, and (d)
43 cyclohexanone in the experiment

44

45 **Supplementary text**

46 **S1. Rate coefficients determination**

47 In this study, the absolute method was used to calculate the rate coefficient of
48 α -pinene ozonolysis by detecting changes in ozone and α -pinene concentrations, and
49 the loss path of O_3 in the reaction mainly has the following two reactions (R1) (R2),

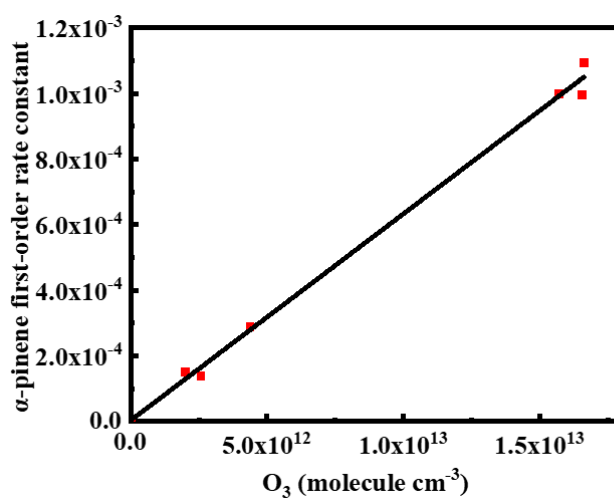


52 According to reactions R1 and R2, the following equations can be given by,

$$\frac{-\ln[\text{VOCs}]}{dt} = k_1 + k_2[O_3]$$

53 where k_1 and k_2 are the reaction rate coefficients of O_3 wall loss (R1) and the
54 reaction of organic matter with O_3 (R2), respectively.

55 Figure S1 shows the relationship between the pseudo-first-order decay rate and
56 the initial O_3 concentration. The slope of the plot is used to calculate the reaction rate
57 coefficient. Table S2 summarizes the initial experimental conditions and final reaction
58 rate coefficients for the reaction of O_3 with α -pinene in this experiment and compares
59 them with the published results.



60

61 **Figure S1.** Relationship between quasi-primary decay rate coefficient and initial O_3 .

62

63 S2 Detection of gaseous products

64 The reactant α -pinene and its gaseous oxidation products (acetone, α -pinene
65 oxide, and macromolecular oxidation products) were monitored by PTR-TOF-MS
66 (Proton Transfer Reaction-Time of Flight-Mass Spectrometer, IONICON 8000). It has
67 a mass resolution of > 8000 (FWHM) and a VOC detection limit of $0.1 \sim 0.2$ ppbv.
68 The TofDaq (Tofwerk AG, Switzerland). was used to analyze the mass spectra of
69 PTR-TOF-MS. The precisions were 7%. A formaldehyde analyzer (AL4021,
70 Aero-Laser, Germany) within the detection limit of 100 pptv based on the Hantzsch
71 reaction (acetylacetone) was used to detect formaldehyde (HCHO) formation in the
72 gas phase continuously.

73 Due to the extremely short lifetime of the OH radical, it is difficult to measure
74 directly. In this study, cyclohexane was used as a scavenger for the OH radical. The
75 reaction between OH radicals and cyclohexane produced cyclohexanone, which was
76 subsequently detected using PTR-TOF-MS. The OH yields can be calculated from the
77 following equation¹

$$Y_{\text{OH}} = \frac{[\text{cyclohexanone}]/\Delta[\text{organic}]}{[\text{cyclohexanone}]/\Delta[\text{cHX}]} = \frac{[\text{cyclohexanone}]/\Delta[\alpha - \text{pinene}]}{0.5}$$

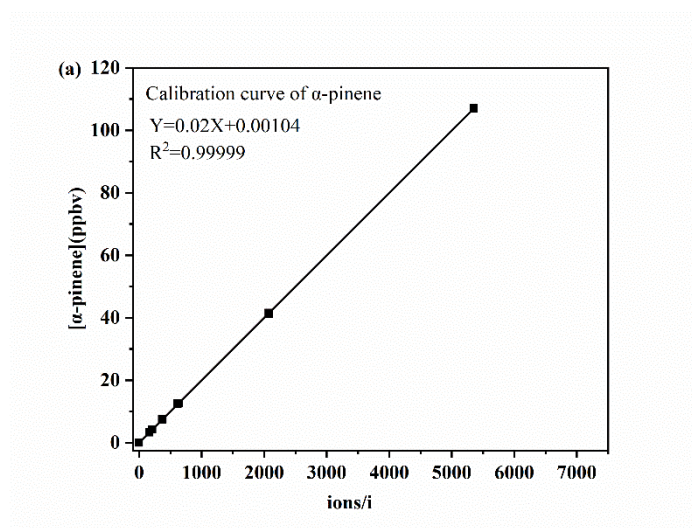
78 [cyclohexanone] and [α -pinene] were their concentration at reaction time t ,
79 which was calculated from their PTR-TOF-MS signal and a calibration curve. The
80 calibration curve was obtained by adding different amounts of cyclohexanone and
81 α -pinene to the chamber as shown in Figure S2.

82

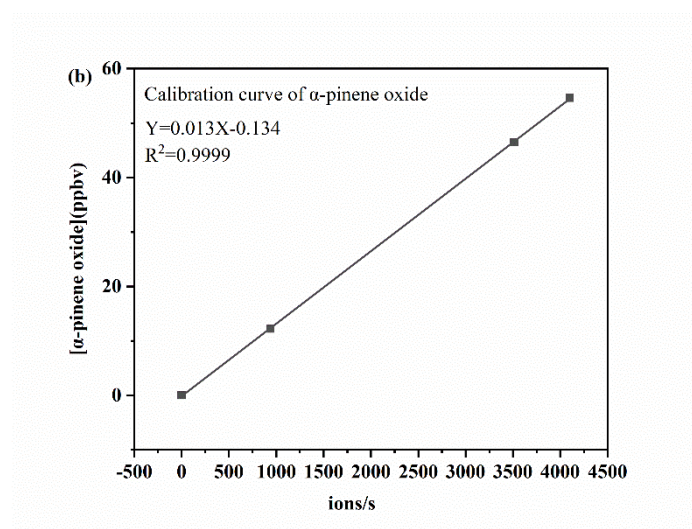
83 **Table S1.** Summary of reaction rate coefficients for α -pinene ozonolysis in this study
 84 and comparison with other studies.

Temp. (K)	$K(\text{cm}^3 \text{molecule}^{-1} \text{s}^{-1})$	Ref.
Absolute Rate Coefficients		
295.7-302.7	6.29×10^{-17}	This study
294	1.6×10^{-16}	2
298	$(3.3 \pm 0.3) \times 10^{-16}$	3
295 ± 1	1.45×10^{-16}	4
298	8.0×10^{-17}	5
276-324	$9.4 \times 10^{-17} \exp[-(731 \pm 173)/T]$	6
301	$(8.6 \pm 1.3) \times 10^{-17}$	7
296 ± 2	$(9.71 \pm 1.06) \times 10^{-17}$	8
243.1-303.2	$1.4 \times 10^{-15} \exp[-(833 \pm 86)/T]$	9
303.2	$(9.0 \pm 0.6) \times 10^{-17}$	
296 ± 1	$(1.1 \pm 0.1) \times 10^{-16}$	10
296	$(1.1 \pm 0.2) \times 10^{-16}$	
Relative Rate Coefficients		
297 ± 2	$(8.54 \pm 1.29) \times 10^{-17}$	7
288-363	$6.0 \times 10^{-16} \exp[-(560 \pm 87)/T]$	11
298	$(9.39 \pm 0.46) \times 10^{-17}$	
288-343	$3.8 \times 10^{-16} \exp[-(447 \pm 303)/T]$	11
298	$(8.26 \pm 0.34) \times 10^{-17}$	
295 ± 0.5	$(1.06 \pm 0.09) \times 10^{-16}$	12
302 ± 1	$(9.26 \pm 0.84) \times 10^{-17}$	13
302 ± 1	$(1.03 \pm 0.04) \times 10^{-16}$	
298 ± 2	$(1.13 \pm 0.14) \times 10^{-16}$	14
298	9.6×10^{-17}	
240-370	$8.22 \times 10^{-16} \exp(-640/T)$	Preferred Values

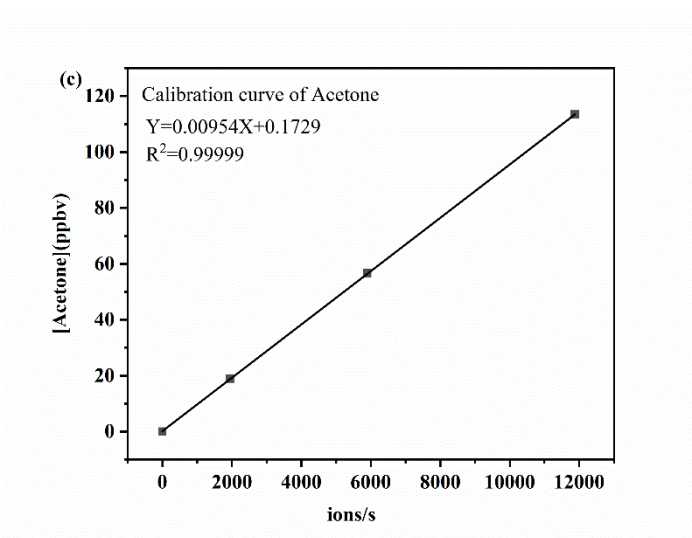
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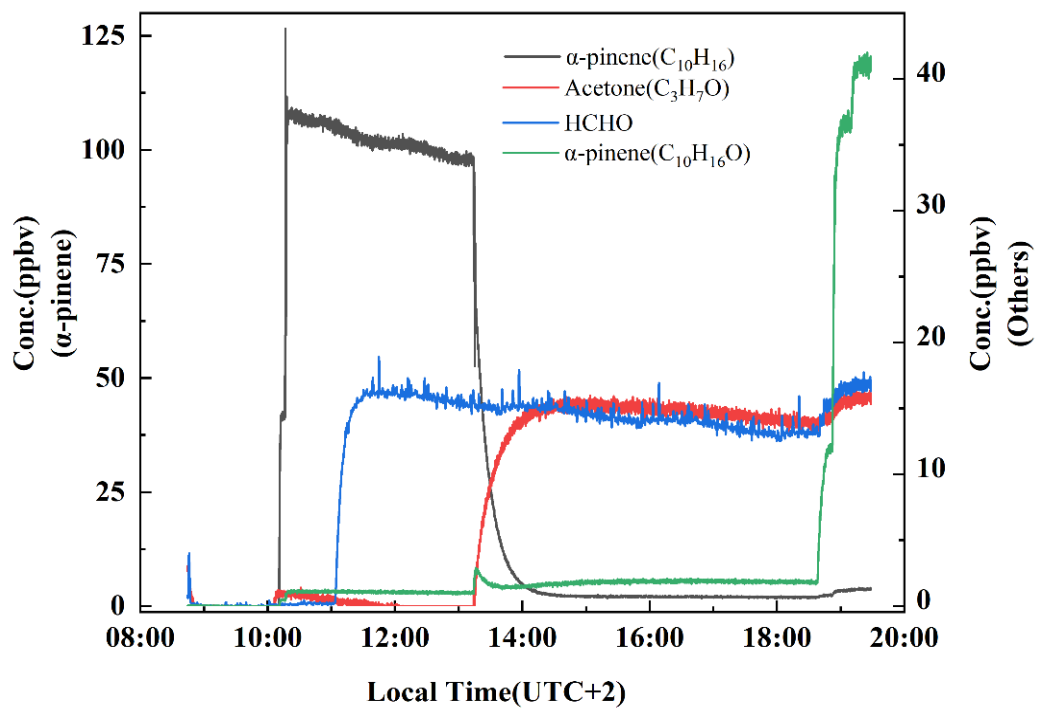
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89 **Figure S2.** Calibration curve for (a) α -pinene concentration, (b) α -pinene oxide and (c)
90 acetone in the experiment.

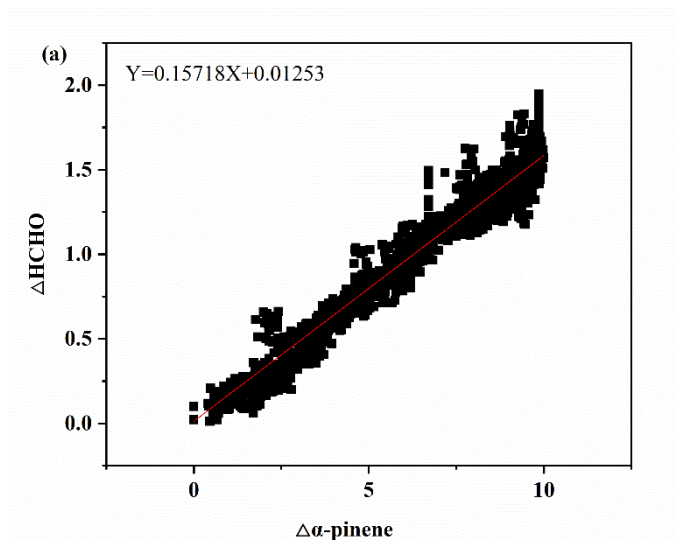


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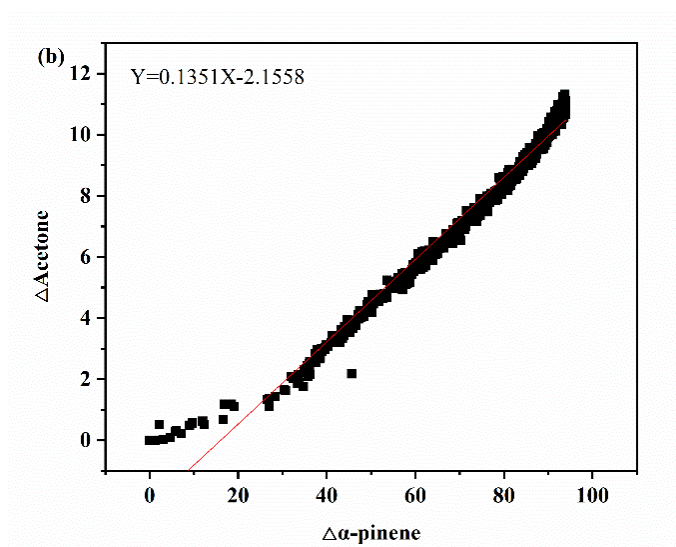
92 **Figure S3** Time series diagram of α -pinene, acetone, and HCHO concentrations.

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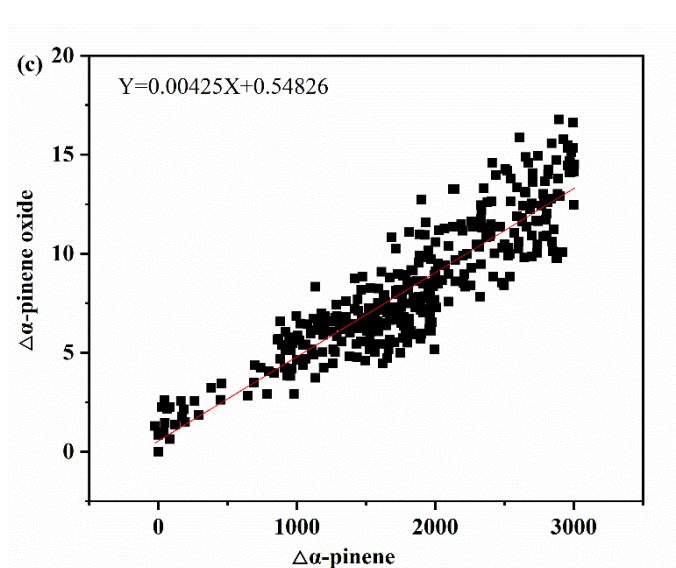
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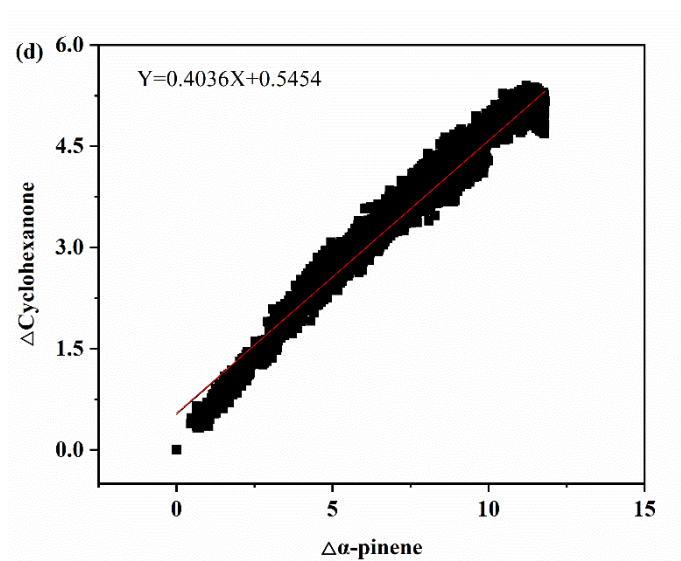


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98 **Figure S4.** Average yields of (a) HCHO, (b) acetone, (c) α -pinene oxide and

99 (d) cyclohexanone in the experiment.

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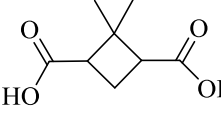
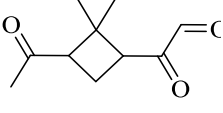
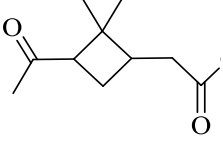
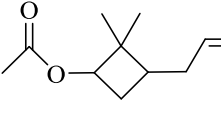
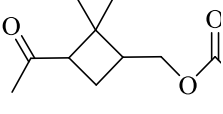
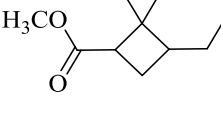
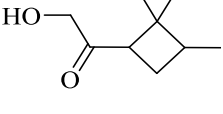
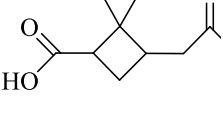
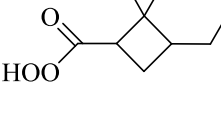
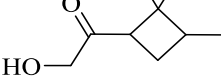
101 **Table S2.** Yield of HCHO and acetone produced during α -pinene ozonolysis in this
102 and previous studies.

Products	Product yield (%)	Reference
HCHO	20.0 \pm 3.3	This work
	28 \pm 3	15
Acetone	12.6 \pm 2.7	This work
	8 \pm 2	16
	3 \pm 1	17
	5.9 \pm 0.5	15

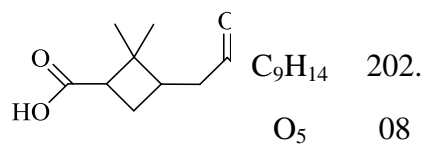
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104 **Table S3.** Structures and IUPAC names of a selection of products typically observed
 105 in the oxidation of α -pinene.

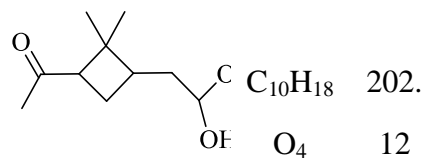
Name	Structure	Molecular Formula	m/z
Formaldehyde		HCHO	30.0 1
Acetone		C ₃ H ₆ O	58.0 4
2-(2,2-dimethyl-3-oxocyclobutyl) acetaldehyde		C ₈ H ₁₂ O ₂	140. 08
α -pinene oxide		C ₁₀ H ₁₆ O	152. 12
Norpinonaldehyde		C ₉ H ₁₄ O ₂	154. 10
Norpinalic acid		C ₈ H ₁₂ O ₃	156. 08
Pinonaldehyde		C ₁₀ H ₁₆ O ₂	168. 12
Pinalic acid		C ₉ H ₁₄ O ₃	170. 09
Norpinonic acid		C ₉ H ₁₄ O ₃	170. 09

Norpinic acid		C_8H_{12} O_4	172. 07
Keto-pinonaldehyde		$C_{10}H_{14}$ O_3	182. 09
Pinonic acid		$C_{10}H_{16}$ O_3	184. 11
(1S,3R)-2,2-dimethyl-3-(2-oxoethyl) cyclobutyl acetate		$C_{10}H_{16}$ O_3	184. 11
(3-acetyl-2,2-dimethylcyclobutyl)methyl formate		$C_{10}H_{16}$ O_3	184. 11
Pinalic acid methyl ester		$C_{10}H_{16}$ O_3	184. 11
OH-pinonaldehyde		$C_{10}H_{16}$ O_3	184. 11
Pinic acid		C_9H_{14} O_4	186. 09
Pinalic-peroxo acid		C_9H_{14} O_4	186. 09
Hydroxy-pinonic acid		$C_{10}H_{16}$ O_4	200. 10

Peroxo-pinic acid



1-(3-(2-hydroperoxy-2-hydroxyethyl)-2,2-dimethyl
cyclobutyl)ethan-1-one



107 **Table S4** The SOA yield under different conditions in the present work and compared
 108 with other studies.

$[\alpha\text{-pinene}]_0$	$[\text{Ozone}]_0$	Temp. (K)	Seed	RH(%)	OH scavenger	$Y_{\text{SOA}}(\%)$	Ref.
107	670	295	N/A	0	Cyclohexane	27.8	This work
			$(\text{NH}_4)_2\text{SO}_4$	0		15.5	
				57		4.2	
12.7	176			0		4.8	
1000	2000	298	$(\text{NH}_4)_2\text{SO}_4$	0	Cyclohexane	78	18-claflin et al.
				50		41	
				85		48	
10	300			0		23	
				65		24	
186		293	N/A	4.1	Cyclohexane	41	¹⁵ -Lee et al.
50	100	298	$(\text{NH}_4)_2\text{SO}_4$	<5	Cyclohexane	23.3	¹⁹ -Nah et al.
52			N/A	<5		22.6	
50	500		$(\text{NH}_4)_2\text{SO}_4$	<5		28.6	
48			N/A	<5		31.8	
92	110	300.8	N/A	1	2-Butanol	21.5	²⁰ -Cocke r et al.
122	380	301.7		57.7		26.2	
177	324	302.7		37.3		28.5	
123	258	302.5		46		25.5	
162	302	300.6	$(\text{NH}_4)_2\text{SO}_4$	52.4		16.4	
114	130	302.1		1		22	
136	310	302.2		48.5		16.6	
70	256	302.4		39		19	
101	580	302.3	NH_4HSO_4	62		14.8	
75	255	300.3		45		12.3	

117	352	302.5		14.4		22.4	
73	250	301.5	CaCl ₂	41.5		14.8	
113	300	302.5		50		19.2	
113	300	301.5		57.5		20.7	
252	753	298	(NH ₄) ₂ SO ₄	26	Diethylether	31	²¹ -Deng
205	>652	298		27		28	et al.
244	749	288		32		39	
220	742	288		34		38	
252	710	278		48		42	
290	>593	278		52		52	
243	>740	298	H ₂ SO ₄ +(NH ₄)	26		28	
314	>600	298	₂ SO ₄	27		27	
228	>622	288		32		35	
291	>596	288		34		38	
200	>612	278		48		47	
252	>595	278		52		56	
242		303	(NH ₄) ₂ SO ₄	40	Methanol	13.8	²² -Zhao
258			N/A	40		14.7	et al.
246			(NH ₄) ₂ SO ₄	40		16.5	
125	Excess O ₃		N/A	10		17	
131			(NH ₄) ₂ SO ₄	10		19.9	
90			N/A	10		14.1	
156	100-500	293.7	H ₂ SO ₄	12.9	N/A	31	²³ -
151		298.2		23		22	Czoschke
138		296.8		45.7		17	et al.
158		295.4	NH ₄ HSO ₄	14.3		9	
158		300.6		23.5		12	
147		297.1		40.3		7	
152		299.7	(NH ₄) ₂ SO ₄	15.3		10	

151		298.2		26		9	
131		298		40.9		8	
394	500	277.4- 281.8	N/A	<10	N/A	51.83	²⁴ -Li et al.
175	307	279.2- 279.8				39.36	
95	370	277.7- 281.6				32.31	
250	390	279.6- 284.7				44.72	
15.16	85	291	N/A	1-4	N/A	21	²⁵ -Ren et al.
49.88	242	287.8		1-2		30	
70	358	297		1-3		39	
81.8	400	283		1-6		46	
146	43	285.5		8-14		48	
48	Excess O ₃	243	N/A	8	Cyclohexane	125	²⁶ -Saath off et al.
		243		70		125	
		252		0		98	
		252		66		116	
		252		70		107	
		273		70		77	
		273		30		44	
		283		30		35	
		293		35		32	
		303		0		26	
		303		40		22	
		313		16		17	

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