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Supplement of

Biodegradation by bacteria in clouds: an underestimated sink for some organics in the atmospheric multiphase system

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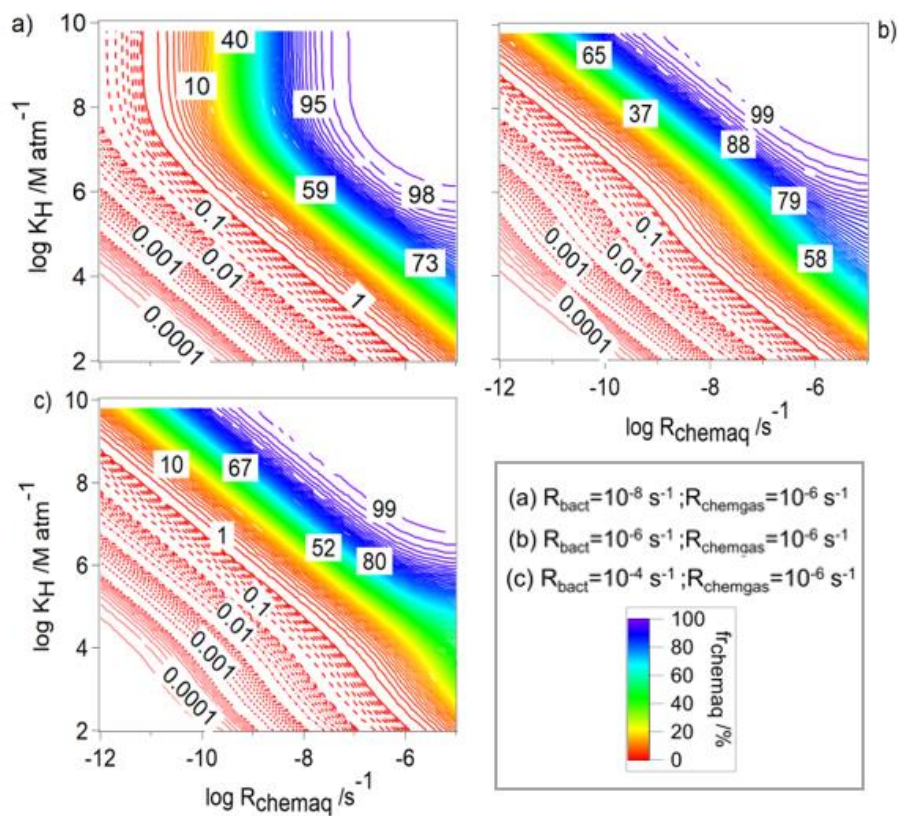


Figure S1. Relative contributions of the aqueous phase chemical processes to the total loss rate of organics (f_{chemaq}) for $R_{\text{chemgas}} = 10^{-6} \text{ s}^{-1}$ and (a) $R_{\text{bact}} = 10^{-8} \text{ s}^{-1}$, (b) $R_{\text{bact}} = 10^{-6} \text{ s}^{-1}$ and (c) $R_{\text{bact}} = 10^{-4} \text{ s}^{-1}$ as a function of R_{chemaq} and K_H . All figure panels have the same scales on the ordinates and abscissas, respectively. These are complementary figures to **Figures 2a-c**.

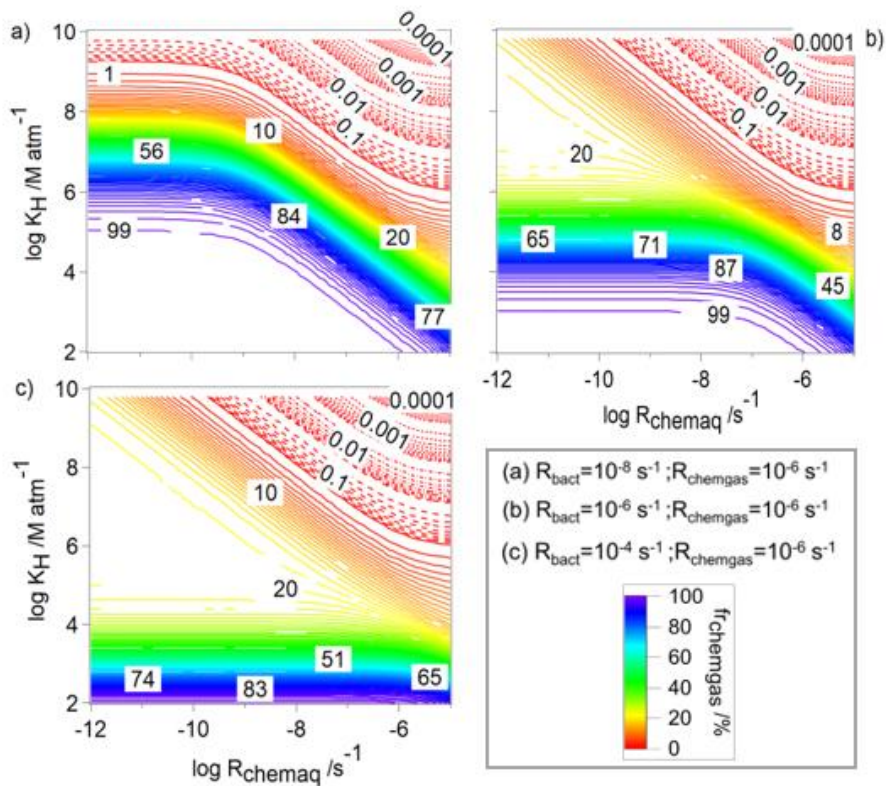


Figure S2: Relative contributions of the gas phase chemical process to the total loss rate of the organics ($f_{r_{chemgas}}$) for $R_{chemgas} = 10^{-6} \text{ s}^{-1}$ and (a) $R_{bact} = 10^{-8} \text{ s}^{-1}$, (b) $R_{bact} = 10^{-6} \text{ s}^{-1}$ and (c) $R_{bact} = 10^{-4} \text{ s}^{-1}$ as a function of R_{chemaq} and K_H . All figure panels have the same scales on the ordinates and abscissas, respectively. These are complementary figures to **Figure 2a-c**.

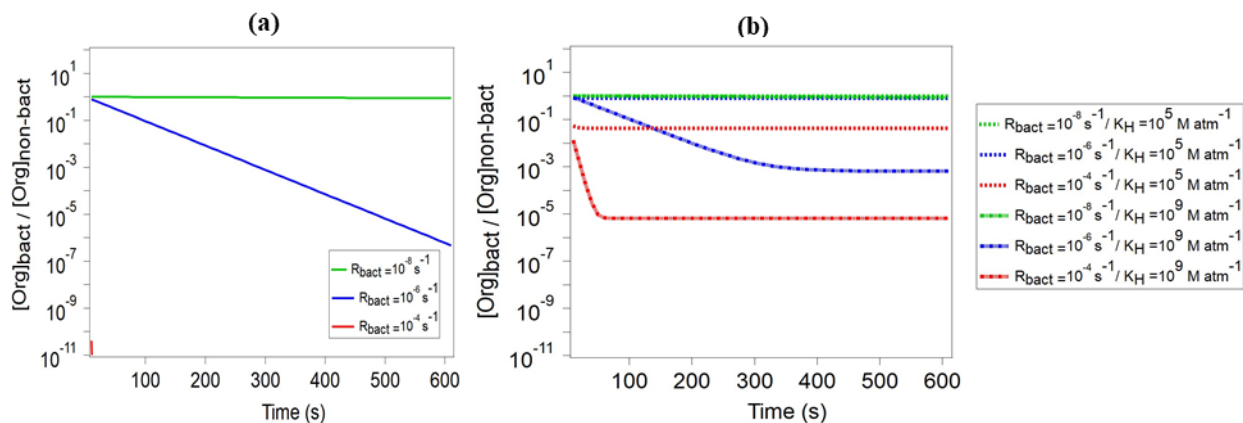


Figure S3: Concentration ratio of the organic compound in bacteria-free ($[\text{Org}]_{\text{non-bact}}$) and bacteria-containing droplets ($[\text{Org}]_{\text{bact}}$) of the same size (20 μm diameter) for (a) CCN-derived compounds and (b) water-soluble organic gases. Results are shown for $R_{bact} = 10^{-8} \text{ s}^{-1}$, 10^{-6} s^{-1} and 10^{-4} s^{-1} , $R_{chemaq} = 10^{-11} \text{ s}^{-1}$, $R_{chemgas} = 10^{-6} \text{ s}^{-1}$. Panel (b) shows in addition results for water-soluble organic gases at $K_H = 10^5$ and 10^9 M atm^{-1} .

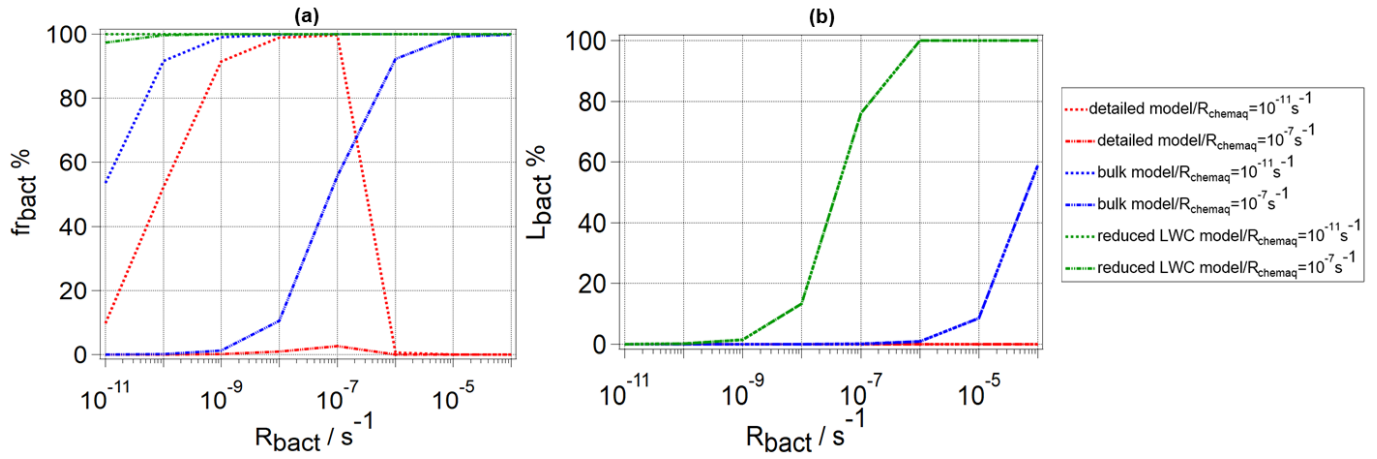


Figure S4: Comparison of (a) fr_{bact} , (b) L_{bact} for CCN-derived compounds for three different approach : detailed model (red line), bulk approach (blue line) and low LWC model (green line) for different R_{bact} , two $R_{chemaq}=10^{-11}$ and $10^{-7} s^{-1}$.

Description of phase transfer in the box model

The mass transfer coefficient $k_{mt} [s^{-1}]$ can be expressed as (Seinfeld and Pandis, 1998)

$$k_{mt} = \left[\frac{r_d^2}{3D_g} + \frac{r_d}{3\alpha} \sqrt{\frac{2\pi M_g}{RT}} \right]^{-1} \quad (\text{Eq-R1})$$

Whereas

r_d = cloud droplet radius [cm]

D_g = gas phase diffusion coefficient [$cm^2 s^{-1}$]

α = mass accommodation coefficient (dimensionless)

M_g = molecular weight of gas [$g mol^{-1}$]

R = constant for ideal gases ($8.314 \cdot 10^7 \text{ erg mol}^{-1} \text{ K}^{-1}$)

T = temperature [K]

The mass transfer coefficient is then applied to determine the sink and source terms due to phase transfer of soluble species:

$$\frac{dc_g}{dt} = k_{mt} \cdot LWC \cdot \left[\frac{c_{aq}}{LWC \cdot K_{H(eff)} \cdot R' \cdot T} - c_g \right] + (P_{gaschem} - L_{gaschem}) \quad (\text{Eq-R2})$$

$$\frac{dc_{aq}}{dt} = k_{mt} \cdot LWC \cdot \left[c_g - \frac{c_{aq}}{LWC \cdot K_{H(eff)} \cdot R' \cdot T} \right] + (P_{aqchem} - L_{aqchem}) \quad (\text{Eq-R3})$$

whereas both the gas and the aqueous phase concentrations have units of mol g(air)⁻¹; LWC is the liquid water content in g/m³, K_{H(eff)} is the effective Henry's law constant in M atm⁻¹, R' is the ideal-gas constant (8.314.10⁷ erg mol⁻¹ K⁻¹), T is the temperature [K] and P and L are the rates of the chemical production and loss reactions in the gas and aqueous phases, respectively.

Description of model parameters

Table S1. Aqueous phase chemical mechanism : aqueous phase irreversible reactions with rate constants k and temperature dependencies Ea/R where available and aqueous phase equilibria reactions (Ervens et al., 2003, 2008) .

Reactions	Reactants	Products	k [M ⁻¹ s ⁻¹]	Ea/R [K]
Aqueous phase irreversible reactions				
R ₁	SO ₂ + O ₃	S(VI) + O ₂	2.4×10 ⁴	
R ₂	HSO ₃ ⁻ + O ₃	S(VI) + O ₂	3.7×10 ⁵	5530
R ₃	SO ₃ ²⁻ + O ₃	S(VI) + O ₂	1.5×10 ⁹	5280
R ₄	H ₂ O ₂ + HSO ₃	S(VI) + H ₂ O	7.2×10 ⁷	14000
R ₅	HO ₂ + HO ₂	H ₂ O ₂ + O ₂	8.3×10 ⁵	2720
R ₆	O ₂ ⁻ + HO ₂	H ₂ O ₂ + O ₂	9.7×10 ⁷	1060
R ₇	OH + CH ₂ O	HO ₂ + HCOOH	1×10 ⁹	1020
R ₈	OH + CH ₃ OOH	CH ₃ O ₂ + H ₂ O	2.4×10 ⁷	1680
R ₉	OH + CH ₃ OOH	HO ₂ + HCOOH	6×10 ⁶	1680
R ₁₀	O ₃ + O ₂ ⁻ (+ H ⁺)	OH + 2 O ₂	1.5×10 ⁹	2200
R ₁₁	OH + CHOCHO	HO ₂ + CHOCOOH	1.1×10 ⁹	1516
R ₁₂	OH + CHOCOOH	HO ₂ + H ₂ C ₂ O ₄	3.6×10 ⁸	1000
R ₁₃	OH + CHOCOO ⁻	H ₂ C ₂ O ₄ ⁻	2.9×10 ⁹	4300
R ₁₄	OH + CH ₃ COCHO	HO ₂ + 0.92 CH ₃ COCOOH + 0.08 CHOCOOH	1.1×10 ⁹	1600
R ₁₅	OH + CH ₂ (OH)CHO	HO ₂ + CH ₂ OHCOOH	1.2×10 ⁹	
R ₁₆	OH + CH ₂ OHCOOH	HO ₂ + CHOCOOH	1.2×10 ⁹	
R ₁₇	OH + C ₂ O ₄ ²⁻	O ₂ ⁻ + 2 CO ₂ + OH ⁻	1.6×10 ⁸	4300
R ₁₈	OH + HC ₂ O ₄ ⁻	HO ₂ + 2 CO ₂ + OH ⁻	1.9×10 ⁸	2800
R ₁₉	OH + H ₂ C ₂ O ₄	HO ₂ + 2 CO ₂ + H ₂ O	1.4×10 ⁶	
R ₂₀	OH + CH ₃ C(O)COOH	HO ₂ + CO ₂ + CH ₃ COO ⁻	7×10 ⁸	
R ₂₁	OH + CH ₃ COCOOH	HO ₂ + H ₂ O + CH ₃ COOH	1.2×10 ⁸	
R ₂₂	OH + HCOO ⁻	HO ₂ + CO ₂ + H ₂ O	3.2×10 ⁹	1000
R ₂₃	OH + HCOOH	HO ₂ + CO ₂ + H ₂ O	1.3×10 ⁸	1000
R ₂₄	OH + CH ₃ COO ⁻	HO ₂ + OH ⁻ + 0.15 CH ₂ O + 0.85 CHOCOOH	1×10 ⁸	1800
R ₂₅	OH + CH ₃ COOH	HO ₂ + H ₂ O + 0.15 CH ₂ O + 0.85 CHOCOOH	1.5×10 ⁷	1330
R ₂₆	CH ₃ O ₂ + CH ₃ O ₂	CH ₂ O + CH ₃ OH + HO ₂	1.7×10 ⁸	2200
R ₂₇	H ₂ O ₂ + OH	HO ₂ + H ₂ O	3×10 ⁷	1680
R ₂₈	OH + CH ₃ CHO	HO ₂ + H ₂ O + CH ₃ COOH	3.6×10 ⁹	580
R ₂₉	O ₂ ⁻ + CH ₃ (CO)OO	CH ₃ COOH	1×10 ⁹	
R ₃₀	CH ₃ C(O)OO + CH ₃ C(O)OO	2 CH ₃ O ₂ + 2 CO ₂	1.5×10 ⁸	
R ₃₁	OH + HO-CH ₂ -CHO	HO ₂ + CH ₂ (OH)COOH	5×10 ⁸	
R ₃₂	OH + WSOC	WSOC + HO ₂	3.8×10 ⁸	
Aqueous phase equilibria				
E			K _a [M]	

E ₁	H ₂ O	OH ⁻ +H ⁺	1.0×10 ⁻¹⁴	
E ₂	HO ₂	O ₂ ⁻ + H ⁺	1.60×10 ⁻⁵	
E ₃	CHOCOOH	CHOCOO ⁻ +H ⁺	6.60×10 ⁻⁴	
E ₄	HCOOH	HCOO ⁻ + H ⁺	1.77×10 ⁻⁴	
E ₅	CH ₃ COCOOH	CH ₃ COCOO ⁻ + H ⁺	4.07×10 ⁻³	
E ₆	CH ₃ COOH	CH ₃ COO ⁻ + H ⁺	1.77×10 ⁻⁵	
E ₇	H ₂ C ₂ O ₄	HC ₂ O ₄ ⁻ +H ⁺	6.40×10 ⁻²	
E ₈	HC ₂ O ₄ ⁻	C ₂ O ₄ ²⁻ + H ⁺	5.25×10 ⁻⁵	
E ₉	HNO ₃	NO ₃ ⁻ + H ⁺	22	
E ₁₀	HOCH ₂ CH ₂ OH	HOCH ₂ CH ₂ O ⁻ + H ⁺	1.54×10 ⁻⁴	
E ₁₁	SO ₂ +H ₂ O	HSO ₃ ⁻ +H ⁺	0.013	
E ₁₂	HSO ₃ ⁻	SO ₃ ²⁻ +H ⁺	6.60×10 ⁻⁸	
E ₁₃	H ₂ SO ₄	HSO ₄ ⁻ + H ⁺	1000	
E ₁₄	HSO ₄ ⁻	SO ₄ ²⁻ + H ⁺	0.102	
E ₁₅	CO ₂ (aq)+H ₂ O	HCO ₃ ⁻ + H ⁺	7.70×10 ⁻⁷	
E ₁₆	HCO ₃ ⁻	CO ₃ ²⁻ + H ⁺	4.84×10 ⁻¹¹	
E ₁₇	NH ₃	NH ₄ ⁺ + OH ⁻	1.76×10 ⁻⁵	
E ₁₈	H ₂ O	H ⁺ + OH ⁻	1×10 ⁻¹⁴	

Table S2. Phase transfer parameters of soluble species to calculate the mass transfer coefficient (Eq-S1). M_g: molecular weight (g/mol), α: mass accommodation coefficient (dimension less), D_g: gas phase diffusion coefficients (cm s⁻¹) and K_H: Henry's law constant [M atm⁻¹]

Species	M _g [g mol ⁻¹]	α ^[1]	D _g [cm s ⁻¹] ^[1]	K _H [M atm ⁻¹]
O ₃	48	0.05	0.148	1.14×10 ⁻² ^[1]
H ₂ O ₂	34	0.1	0.11	1.02×10 ⁵ ^[1]
HO	17	0.05	0.153	25 ^[1]
HO ₂	33	0.01	0.104	9×10 ³ ^[1]
HCHO	30	0.02	0.164	4.99×10 ³ ^[1]
CH ₃ O ₂	47	0.0038	0.135	310 ^[1]
CH ₃ OOH	48	0.0038	0.135	310
HNO ₃	63	0.054	0.132	2.1×10 ⁵ ^[1]
N ₂ O ₅	108	0.0037	0.110	1.4 ^[1]
Hydroxyaldehyde	60	0.03	0.195	4.1×10 ⁴ ^[3]
Glyoxal	58	0.023	0.115	4.19×10 ⁵ ^[1]
Methylglyoxal	72	0.1	0.115	3.2×10 ⁴ ^[3]
HCOOH	74	0.012	0.153	1.77×10 ⁻⁴ ^[1]
Acetic acid	46	0.1	0.1	4×10 ³ ^[4]
Glyoxylic acid	60	0.019	0.124	9×10 ³ ^[2]
Glycolic acid	76	0.1	0.1	9×10 ³ ^[2]
Pyruvate	88	0.1	0.1	3.11×10 ⁵ ^[3]
Oxalate	90	0.1	0.1	9×10 ³ ^[3]
Hydroxyketone	88	0.1	0.1	100 ^[3]
CH ₃ O ₃	75	0.1	0.1	669 ^[3]
Aldehyde	44	0.1	0.1	11.4 ^[3]
SO ₂	64	0.035	0.128	1.23 ^[3]

CO ₂	44	2.e ⁻⁴	0.155	3.11×10 ⁻² [1]
Glycolaldehyde	58	0.1	0.1	4.1×10 ⁴ [3]
C ₂ H ₅ OOH	62	0.1	0.1	310 ^[1]
Organic compound that reacts with OH and is consumed by bacteria	150	0.1	0.1	10 ² - 10 ⁹

[1]: (Ervens et al., 2003), [2]: (Ip et al., 2009), [3]: (Sander, 2015), [4]: (Johnson et al., 1996)

Table S3. Initial gas phase mixing ratios of gas phase species and the concentration of species only in the aqueous phase; all other compounds considered in the mechanism were not initialized

Species formula	Species name	Mixing ratio [ppb]
Gases with phase transfer into the aqueous phase		
O ₃	ozone	39
H ₂ O ₂	hydrogen peroxide	1.202
OH	hydroxyl radical	3.05×10 ⁻¹¹
HO ₂	hydroperoxyl radical	9.07×10 ⁻³
HCHO	formaldehyde	2.519
CH ₃ O ₂	methylperoxy radical	1.38×10 ⁻³
CH ₃ OOH	methyl hydrogen peroxide	0.211
HNO ₃	nitric acid	0.397
N ₂ O ₅	dinitrogen pentoxide	4.806×10 ⁻⁴
CH ₂ (OH)CHO	hydroxy acetaldehyde	0.437
CHOCHO	glyoxal	0.218
CH ₃ COCHO	methyl glyoxal	0.190
HCOOH	formic acid	2.239×10 ⁻³
HAc	Acetic acid	0.198
CH ₃ CO(OO)	acetylperoxy radical	5.5×10 ⁻⁵
SO ₂	sulfur dioxide	0.150
CH ₃ CHO	acetaldehyde	0.409
CO ₂	carbon dioxide	3.96×10 ⁵
CH ₂ (OH)CHO	glycolaldehyde	0.4273
CH ₃ CH ₂ (OOH)	ethyl hydrogen peroxide	2.423×10 ⁻³
Organic compound		1
compounds without phase transfer into the gas phase		
NO	nitric oxide	0.0429
NO ₂	nitrogen dioxide	0.179
NO ₃	nitrate radical	3.5×10 ⁻⁴
HNO ₄	Peroxynitric acid	8.83×10 ⁻³
CO	carbon monoxide	140.3
C ₃ H ₈	isoprene	1.031
MACR	methacrolein	0.282
MVK	methyl vinyl ketone	0.113
C ₂ H ₆	ethane	0.846
C ₂ H ₄	ethene	0.469
C ₃ H ₆	propene	0.118
H ₂	hydrogen	550
CH ₃ CO(OOH)	peracetic acid	0.240
C ₄ H ₁₀	butane	0.142
CH ₃ CH ₂ (OO)	ethylperoxy radical	7.144×10 ⁻⁶
PO ₂	Other peroxy radicals (C ₂ , C ₃)	1.414×10 ⁻⁴

POOH	Hydroperoxides of PO ₂	2.076×10 ⁻³
PAN	peroxy acetyl nitrate	0.586
Isop-OO		4.181×10 ⁻³
CH ₂ =C(CH ₃)CO(OO)	MCO ₃ , peroxy radical from MACR *	1.690×10 ⁻⁴
CH ₂ =CHC(OO)(CH ₃)CH ₂ (ONO ₂)	peroxy radical from NO ₃ +ISOP	2.840×10 ⁻⁵
ONITR	Organic nitrates	0.0921
CH ₃ OH	Methanol	3.403
HOCH ₂ C(OOH)CH ₃ CHCH ₂	ISOPOOH, peroxide from isoprene	0.0318
XO ₂	Additional peroxy radicals	5.318×10 ⁻⁴
XOOH	Hydroperoxides of XO ₂	8.885×10 ⁻³
C ₅ H ₈	terpene	0.0407
terpene-OO	a-pinene peroxy radical	2.020×10 ⁻⁴
terpene-OOH	a-pinene peroxide	2.499×10 ⁻³
MACR-OO	methacrolein peroxy radical	1.678×10 ⁻⁴
MACR-OOH	methacrolein peroxide	3.322×10 ⁻³
MPAN	methacryloyl peroxyxynitrate	0.072
RO ₂	Peroxy radicals from acetone	3.614×10 ⁻⁵
ROOH	Peroxyde from RO ₂	3.220×10 ⁻³
CH ₃ COCH ₃	acetone	1.437
CH ₄	methane	1850 (constant)
Only in the aqueous phase (constant throughout the simulation)		
WSOC (water soluble organic carbon)		0.626
pH		4.5

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