

We would like to thank the referee for their positive and insightful comments on the manuscript. Below is our point-by-point response to the comments. Our responses are in black, manuscript text is in italic with new/modified text is marked in blue. The line numbers cited here refer to those in the clean manuscript version.

Referee comment: Section 2.1: The model description is too concise and could be improved a bit. Which 26 species are transferred between the gas and aqueous phases? It would be worth showing the coupled mass transfer ODEs with Schwartz's treatment.

Author response: We agree with the referee that the model description in the section 2.1 was very short. Since also the other referee asked for more detail on the model, we added three tables into the supplement with details on our aqueous phase chemical mechanism (Table S1), phase transfer parameters (Table S2) and initial mixing ratios and concentrations (Table S3). Please find all tables at the end of this response.

We will add the following text at the beginning of Section 2.1:

*We use a multiphase box model with detailed gas and aqueous phase chemistry (75 species, 44 gas phase reactions, 31 aqueous reactions). The chemical aqueous phase mechanism with rate constants is listed in **Table S1**. The chemical gas phase mechanism is based on the NCAR Master mechanism (Aumont et al., 2000; Madronich and Calvert, 1989). The two phases are coupled by 26 phase transfer processes which is described kinetically based on the resistance model by Schwartz (1986). The parameters describing the phase transfer of the soluble species are presented in **Table S2**. In addition, the initial gas phase mixing ratios are listed in **Table S3**. The equations for the mass transfer coefficient k_{mt} and the differential equations for the aqueous and gas phase concentrations can be found in the supplement (**Eq-R1-R3**; Section 'Description of phase transfer') (Seinfeld and Pandis, 1998).*

In addition, we add the following set of equations for the description of the phase transfer to the supplement:

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

Whereas

r_d = cloud droplet radius [cm]

D_g = gas phase diffusion coefficient [cm s⁻¹]

α = mass accommodation coefficient (dimensionless)

M_g = molecular weight of gas [g mol⁻¹]

R = constant for ideal gases (8.314 · 10⁷ erg mol⁻¹ 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:

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

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

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.

Referee comment: Is the size class same as the size bin? On line 101, it is stated that the 5 μm < D_{droplet} < 20 μm, but then one droplet size class has D_{droplet} = 20 μm. Should the range be changed to 5 μm < D_{droplet} ≤ 20 μm? Also, why is only the last size class allowed to have bacteria cells?

Author response: The referee is correct that the size range of the droplets should be written as 5 μm ≤ D_{droplet} ≤ 30 μm .

In our box model, we chose a drop size spectrum with diameters of 5 to 30 μm. The bacteria are only in one of the drop classes (diameter 20 μm). Note that other box model studies usually only consider a single drop size (Deguillaume et al., 2004; Ervens et al., 2003; Tilgner et al., 2013). However, as we have shown previously that the drop size may impact the OH(aq) concentration and distribution (Ervens et al., 2014), we used a polydisperse drop size distribution. While also the uptake of organic compounds may be drop-size-dependent, we did not further explore this effect as it would not add significantly to our conclusions.

Referee comment: Line 105-106: Which organic compound? Is the model run separately for each individual compound? What other inorganic species were considered in the model? What was the cloud water pH in the model simulations discussed in section 3?

Author response: We removed the sentence in line 105/106 (now line: 116) and clarified now which species are initialized in our model. We provide more detail on our model in Table S1 (detailed aqueous phase chemical mechanism) ,Table S2 (phase transfer parameters) and Table S3 (initial concentration of different species).

Referee comment: While the degradation rate constant remains constant between pH values 5 and 8, could it change at lower pH?

Author response:

The referee is correct to point out the possible effects of acidity on bacterial activity. Several studies investigated the acid tolerance mechanism of different microorganisms, e.g., (Casal et al., 2016; Patel et al., 2006) by decarboxylation, deamination (Noh et al., 2018), or cell membrane modification (Zhang et al., 2011). In addition, some bacteria can develop an acid resistance system to survive under acidic conditions (pH=2.5) (Lu et al., 2013). However, these strategies do not necessarily imply that the bacteria maintain the same biodegradation activities at high acidity but they allow survival of the cells in the atmosphere.

When exposed to very broad ranges of external pHs, bacteria can control their intracellular pH (~6.5 -7) by internal buffering (Delort et al., 2017). As biodegradation occurs inside the cell, it takes place at these (nearly) neutral conditions. The efficiency of buffering decreases at extreme conditions, e.g., pH < 2 or pH > 10 (Guan and Liu, 2020). However, such pH range is not representative for cloud water where more moderate pH values (~ 3 – 6) are typically found (Deguillaume et al., 2014).

We will modify the text in Section 2.2.2. as follows:

*Experiments with 17 different cloud bacteria in artificial cloud water with pH = 5.0 and pH = 6.5 showed also nearly identical results (Vařtilingom et al., 2011). ~~so it can be concluded that biodegradation rates are largely independent of pH for values typical in cloud water.~~ Similar results were shown by Razika et al. (2010) who demonstrated that biodegradation rates of phenol by *Pseudomonas aeruginosa* were very similar when incubated at pH = 5.8, 7.0 and 8.0, respectively. When exposed to very broad ranges of external pHs, bacteria can control their intracellular pH (~6.5-7) by internal buffering (Delort et al., 2017). As biodegradation occurs inside the cell, it takes place at these (nearly) neutral conditions. The efficiency of buffering decreases at extreme conditions, e.g., pH < 2 or pH > 10 (Guan and Liu, 2020). However, such pH range is not representative for cloud water where more moderate pH values (~ 3 – 6)(Deguillaume et al., 2014) are typically found.*

In addition, we add in the conclusion section (1.596):

In addition, the biodegradation rates may be affected under highly acidic conditions. Some studies demonstrated that some bacteria can develop an acid resistance to survive under acidic conditions ((Lu et al., 2013). However, these strategies do not necessarily imply that the bacteria maintain the same biodegradation activities at high acidity but they allow survival of the cells in the atmosphere. It can be expected that internal buffering of bacteria cells allows them to maintain their metabolic activity over wide pH ranges (~ 3 to 6) as found in cloud water. Therefore, we do not consider a potential pH dependency of biodegradation rates in our model studies.

Referee comment: Figures 2, 3, 6, 7, 8, 10 are presently displayed as three-dimensional plots, which I found a little difficult to read as parts of the plots are obscured by curved surfaces. Since the Z-axis and colors represent the same dimension, I suggest replotting these figures as two-dimensional color contour plots. This would greatly improve the quality and readability of the figures.

Author response: Thank you for this suggestion, we agree with this suggestion and show in the revised version the figures 2,3,6,7,8 and 10 and Figures S1 and S2 as two-dimensional color contour plots to improve the quality and readability of the figures as suggested.

Referee comment: Table 2: There's an extra multiplication symbol in the 5 C Experimental rate column on 12th row.

Author response: Corrected

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

Reactions	Reactants	Products	k [$M^{-1} s^{-1}$]	E_a/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 + COOHCH ₂ OH	1.2×10 ⁹	
R ₁₆	OH + COOHCH ₂ OH	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 + HOCCOOH	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	Reactants	products	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) and Henry's law constants K_H . (M_g : molecular weight, α : mass accommodation coefficient (*dimensionless*), D_g : gas phase diffusion coefficient)

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	$10^2 - 10^9$
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[1]: (Ervens et al., 2003a), [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}

