



# Supplement of

# Bacteria in clouds biodegrade atmospheric formic and acetic acids

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Chemical reactions					
	Reactants		Products	$k [M^{-1} s^{-1}]$	$E_a/R[K]$
R1	$SO_2 + O_3$	$\rightarrow$	$S(VI) + O_2$	$2.4 \cdot 10^4$	
R2	$HSO_3^- + O_3$	$\rightarrow$	$S(VI) + O_2$	$3.7 \cdot 10^5$	5530
R3	$SO_3^{2-} + O_3$	$\rightarrow$	$S(VI) + O_2$	$1.5 \cdot 10^9$	5280
R4	$H_2O_2 + HSO_3^- + H^+$	$\rightarrow$	$S(VI) + H_2O$	$7.2 \cdot 10^7 \text{ M}^{-2} \text{ s}^{-1}$	4000
R5	$HO_2 + HO_2$	$\rightarrow$	$H_2O_2 + O_2$	$8.3 \cdot 10^5$	2720
R6	$O_2^-$ + HO <sub>2</sub>	$\rightarrow$	$H_2O_2 + O_2$	$9.7 \cdot 10^7$	1060
R7	$OH + CH_2(OH)_2$	$\rightarrow$	$HO_2 + HCOOH$	$1 \cdot 10^{9}$	1000
R8	$OH + CH_3OOH$	$\rightarrow$	$CH_3O_2 + H_2O$	$2.4 \cdot 10^7$	1680
R9	$OH + CH_3OOH$	$\rightarrow$	$HO_2 + HCOOH$	$61 \cdot 10^{6}$	1680
R10	$O_3 + O_2^- (+ H^+)$	$\rightarrow$	$OH + 2 O_2$	$1.5 \cdot 10^9$	2200
R11	OH + CHOCHO	$\rightarrow$	$HO_2$ + CHOCOOH	$1.1 \cdot 10^9$	1516
R12	OH + CHOCOOH	$\rightarrow$	$HO_2 + H_2C_2O_4$	$3.6 \cdot 10^8$	1000
R13	$OH + CHOCOO^{-}$	$\rightarrow$	$HO_2 + H_2C_2O_4$	$2.9 \cdot 10^9$	4300
R14	$OH + C_2 O_4^{2-}$	$\rightarrow$	$O_2^- + 2 CO_2 + OH^-$	$1.6 \cdot 10^8$	4300
R15	$OH + HC_2O_4^-$	$\rightarrow$	$HO_2 + 2 CO_2 + OH^-$	$1.9 \cdot 10^8$	2800
R16	$OH + H_2C_2O_4$	$\rightarrow$	$HO_2 + 2 CO_2 + H_2O$	$1.4 \cdot 10^{6}$	
R17	$OH + CH_3C(O)COO^-$	$\rightarrow$	$HO_2 + CO_2 + CH_3COO^-$	$7.2 \cdot 10^8$	
R18	$OH + CH_3C(O)COOH$	$\rightarrow$	$HO_2 + H_2O + CH_3COOH$	$1.2 \cdot 10^8$	
R19	$OH + CH_3CHO$	$\rightarrow$	$HO_2 + CO_2 + CH_3COOH$	$3.6 \cdot 10^9$	
R20	$OH + CH_3C(O)CHO$	$\rightarrow$	$HO_2 + CHC(O)COOH$	$1.1 \cdot 10^{9}$	1516
R21	$OH + HCOO^{-}$	$\rightarrow$	$HO_2 + CO_2 + H_2O$	$3.2 \cdot 10^9$	1000
R22	OH + HCOOH	$\rightarrow$	$HO_2 + CO_2 + H_2O$	$1.3 \cdot 10^8$	1000
R23	$CH_3O_2 + CH_3O_2$	$\rightarrow$	$CH_2O + CH_3OH + HO_2$	$1.7 \cdot 10^8$	2200
R24	$H_2O_2 + OH$	$\rightarrow$	$HO_2 + H_2O$	$3 \cdot 10^{7}$	1680
R25	OH + WSOC	$\rightarrow$	WSOC + $HO_2$	$3.8 \cdot 10^8$	
R26	$OH + CH_2OHCHO$	$\rightarrow$	$CH_2OHCOOH + HO_2$	$1.2 \cdot 10^9$	
R27	$OH + CH_2OHCOOH$	$\rightarrow$	$CHOCOOH + HO_2$	$5.4 \cdot 10^8$	
R28	$OH + CH_2OHCOO^-$	$\rightarrow$	$CHOCOOH + HO_2$	$1.2 \cdot 10^9$	
R29	$OH + CH_3COOH$	$\rightarrow$	0.85 CHOCOOH + $0.15$ HCHO + HO <sub>2</sub>	$1.5 \cdot 10^{7}$	1330
R30	$OH + CH_3COO^-$	$\rightarrow$	0.85 CHOCOOH + $0.15$ HCHO + HO <sub>2</sub>	$1 \cdot 10^{8}$	1800
R31	$CH_3(O)O_2 + O_2^-$	$\rightarrow$	CH <sub>3</sub> COOH	$1 \cdot 10^{9}$	
R32	$CH_3(O)O_2 + CH_3(O)O_2$	$\rightarrow$	2 CH <sub>3</sub> O <sub>2</sub>	$1.5 \cdot 10^8$	
Biodegradation				$k_{bact}$ [L cell <sup>-1</sup> s <sup>-1</sup> ]	
R33	HCOOH/HCOO <sup>-</sup> + Bacteria	$\rightarrow$	Products	$1.5 \cdot 10^{-13}$	
R34	CH <sub>3</sub> COOH/CH <sub>3</sub> COO <sup>-</sup> + Bacteria	$\rightarrow$	Products	$8.7 \cdot 10^{-14}$	

**Table S1.** Irreversible processes in the aqueous phase. The kinetic data are the same as used in the model study by Khaled et al. (2021). The biodegradation rate constants are calculated based on the lab data by Vaïtilingom et al. (2010).

				$K_a$ [M]
E1	$H_2O$	$\rightleftharpoons$	$OH^-+H^+$	$1.0 \cdot 10^{-14}$
E2	$HO_2$	$\rightleftharpoons$	${\rm O_2}^-$ + H <sup>+</sup>	$1.60 \cdot 10^{-5}$
E3	CHOCOOH	$\rightleftharpoons$	$CHOCOO^{-} + H^{+}$	$6.60 \cdot 10^{-4}$
E4	HCOOH	$\rightleftharpoons$	$HCOO^- + H^+$	$1.77 \cdot 10^{-4}$
E5	$H_2C_2O_4$	$\rightleftharpoons$	$\mathrm{HC_2O_4^-}$ + $\mathrm{H^+}$	$6.40 \cdot 10^{-2}$
E6	$\mathrm{HC_2O_4}^-$	$\rightleftharpoons$	$C_2O_4^{2-} + H^+$	$5.25 \cdot 10^{-5}$
E7	$HNO_3$	$\rightleftharpoons$	$NO_3^- + H^+$	22
E8	$SO_2 \cdot H_2O$	$\rightleftharpoons$	$HSO_3^- + H^+$	0.013
E9	$HSO_3^-$	$\rightleftharpoons$	$SO_3^{2-} + H^+$	6.60·10 <sup>-8</sup>
E10	$H_2SO_4$	$\rightleftharpoons$	$\mathrm{HSO_4}^-$ + $\mathrm{H^+}$	1000
E11	$\mathrm{HSO_4}^-$	$\rightleftharpoons$	$SO_4^{2-} + H^+$	0.102
E12	$NH_3$	$\rightleftharpoons$	$\mathrm{NH_4^+}$ + $\mathrm{OH^-}$	$1.76 \cdot 10^{-5}$
E13	$CH_3COOH$	$\rightleftharpoons$	$CH_3COO^- + H^+$	$1.77 \cdot 10^{-5}$

Table S2. Aqueous phase equilibria. The data are the same as used in the model study by Khaled et al. (2021).

**Table S3.** Phase transfer parameters. The parameters are the same as used in the model study by Khaled et al. (2021). The Henry's law constant for acetic acid is the same as used by Brimblecombe and Clegg (1988), its  $\alpha$  and  $D_g$  values are estimated to be same as for similar compounds.

Species	$M_g$	α	$D_g$	K <sub>H</sub>
	$[g mol^{-1}]$		$[\text{cm}^2 \text{ s}^{-1}]$	$[M atm^{-1}]$
O <sub>3</sub>	48	0.05	0.148	$1.14 \cdot 10^{-2}$
$H_2O_2$	34	0.1	0.118	$1.02 \cdot 10^5$
OH	17	0.05	0.153	25
$HO_2$	33	0.05	0.104	$9.10^{3}$
НСНО	30	0.02	0.164	$4.99 \cdot 10^3$
$CH_3O_2$	47	0.0038	0.135	310
$CH_3OOH$	48	0.0038	0.135	310
$HNO_3$	63	0.054	0.132	$2.1 \cdot 10^5$
$N_2O_5$	108	0.0037	0.110	1.4
$SO_2$	64	0.035	0.128	1.23
HCOOH	46	0.012	0.153	5530
$(CHO)_2$	58	0.023	0.115	$4.19 \cdot 10^5$
CH <sub>3</sub> COCHO	72	0.1	0.115	$3.2 \cdot 10^4$
$NH_3$	17	0.1	0.1	60.7
CH <sub>3</sub> COOH	60	0.1	0.1	5500

Table S4. Gas phase formation and loss processes of formic and acetic acids. The full gas phase mechanism is the same as used in Barth et al. (2021).

	Reactants		Products	$k [cm^3 s^{-1}]$	$E_a/R[K]$
Rg1	Isoprene + $O_3$	$\rightarrow$	0.2 CH <sub>3</sub> COOH + 0.1 OH + 0.27 HO <sub>2</sub> +	9.6·10 <sup>-18</sup>	
			$0.06 \text{ HO}_2 + 0.6 \text{ HCHO} + \text{CH}_3\text{COOH} +$		
			0.4 MACR + 0.3 MVK + 0.07 C <sub>3</sub> H <sub>6</sub> +		
			0.2 CH <sub>3</sub> (O)O <sub>2</sub>		
Rg2	$CH_3(O)O_2 + HO_2$	$\rightarrow$	$0.1 O_3 + 0.7 CH_3C(O)OOH + 0.3$	$1.8 \cdot 10^{-11}$	360
			CH <sub>3</sub> COOH		
Rg3	HCOOH + OH	$\rightarrow$	$CO_2 + H_2O + HO_2$	$1.5 \cdot 10^{-12}$	
Rg4	$CH_3COOH + OH$	$\rightarrow$	$0.7 \text{ OH} + 0.7 \text{ CH}_3 \text{CHO} + 0.3 \text{ C}_2 \text{H}_5 \text{O}_2$	$1 \cdot 10^{-11}$	200

**Table S5.** Initial mixing ratios of gas phase species [ppb] and concentrations of aqueous phase species [ $\mu g m_{air}^{-3}$ ]; all other species are not initialized

Gas phase species	Mixing ratio [ppb]
O <sub>3</sub>	60
$H_2O_2$	1
НСНО	0.1
HCOOH	0.5
CH <sub>3</sub> COOH	0.2
$HNO_3$	1
$SO_2$	2
$NH_3$	1
NO	3
$NO_2$	0.1
CO	150
Isoprene	1
Toluene	2
Ethylene	0.5
$CH_4$	1850 (const.)
Aqueous phase species	Concentration
WSOC	$20 \ \mu M$
Bacteria cells	$0.1 \ \mathrm{cm}_{air}^{-3}$

**Table S6.** Rates for all processes shown in Figure 6, at the end of 1-hour simulation time and for  $D_d = 20 \ \mu$ m. The upper part of the table lists the rates in units of  $10^{-16} \ \text{mol g}_{air}^{-1} \ \text{s}^{-1}$ . These numbers allow the comparison of rates related scaled by the aqueous phase volumes. The bottom part of the table reports the same rates (only aqueous phase) in units of  $10^{-9} \ \text{mol L}_{aq}^{-1} \ \text{s}^{-1}$ . These units allow comparing the two droplet classes. If the chemical composition were identical in both classes, the rates should be identical, too. Any deviation is caused by the biodegradation in drop class 2.

Acid	pН	$PT_1$	$S_{aq1}$	$L_{aq1}$	$PT_2$	$S_{aq2}$	$L_{aq2}$	$L_{Bact}$	$S_g$	$L_g$
		$10^{16} \text{ mol } g_{air}^{-1} \text{ s}^{-1}$								
Formic acid	3	-2.5	2.9	-0.45	0.87	0.003	-4.10-4	-0.87	0	-0.77
	4.6	3.7	5.3	14	2.0	0.006	-0.011	-2.0	0	-0.22
	5.6	1.2	11	27	0.49	0.013	-0.008	-0.5	0	-0.015
Acetic acid	3	-3.0	3.1	-0.004	0.15	0.003	-3.5.10-6	-0.15	0.38	-6.4
	4.6	-8.2	8.9	-0.05	0.31	0.009	-5·10 <sup>-5</sup>	-0.32	0.85	-5.4
	5.6	-10	16	-0.74	0.98	0.02	-7.10-4	-1.0	1.1	-3.4
· · · ·		$10^{-9}$ mol $L_{aq}^{-1}$ s <sup>-1</sup>								
Formic acid	3	-0.61	0.72	-0.11	210	0.72	-0.11	-210		
	4.6	0.91	1.3	-3.5	500	1.4	-2.8	-500		
	5.6	0.3	2.8	-6.6	120	3.1	-2.0	-120		
Acetic acid	3	-0.74	0.76	-8.7·10 <sup>-4</sup>	37	0.76	-8.5·10 <sup>-4</sup>	-38		
	4.6	-2.0	2.2	-0.012	77	2.2	-0.012	-79		
	5.6	-2.5	4.0	-0.18	240	4.6	-0.17	-250		



**Figure S1.** Results from 900 1-hour simulations (30 pH values, 30 drop diameters, Total formic acid concentrations (gas + aqueous) and absolute concentration difference ( $(c_0 - c_t)$  in the absence of bacteria (a, c) and in the presence of bacteria (b, d). e) Relative difference in concentration due to bacteria according to Equation 6. The threshold of  $c_0 - c_{t,nocell}$  in panel e) is chosen to avoid displaying very high values by dividing by an irrelevant small concentration difference



**Figure S2.** Results from 900 1-hour simulations (30 pH values, 30 drop diameters, Total acetic acid concentrations (gas + aqueous) and absolute concentration difference ( $(c_0 - c_t)$  in the absence of bacteria (a, c) and in the presence of bacteria (b, d). e) Relative difference in concentration due to bacteria according to Euqation 6

#### S1 Derivation of the factor q based on the model studies by Fankhauser et al. (2019) and Pailler et al. (2023)

The study by Fankhauser et al. (2019) provides predicted aqueous and gas phase concentrations in the absence and presence of bacteria cells. Pailler et al. (2023) reports aqueous phase concentrations and total concentrations (gas + aqueous). Based on their figures, we are able to derive (approximate) q values to explore the extent to which uptake limitations occurred. The q value is defined as the ratio of predicted aqueous and gas phase concentrations divided by teh Henry's law constant (Equation 14)

Figure 2 by Fankhauser et al. (2019) shows aqueous phase concentrations of  $\sim 0.3 \cdot 10^{-5}$  M and  $\sim 0$  in the absence and presence of bacteria cells, respectively. For both simulations, their Figure S3 shows a gas phase concentration of  $\sim 2 \cdot 10^{p}$  molec cm<sup>-3</sup>. In the absence of bacteria cells, their concentration ratio is, thus,

$$\frac{c_{aq}[M]}{p_g[atm]} = \frac{0.3 \cdot 10^{-5} M}{2 \cdot 10^9 molec\,cm^{-3}/2.5 \cdot 10^{19} molec\,cm^{-3} atm^{-1}} \sim 37800\,Matm^{-1} \tag{S1}$$

This concentration ratio corresponds to

$$q = \frac{C_{aq}}{p_g K_{H,eff}} = \frac{37800 \,Matm^{-1}}{36500 \,Matm^{-1}} \sim 1 \tag{S2}$$

indicating thermodynamic equilibrium between the gas and aqueous phases in absence of bacteria and biodegradation.

In the presence of bacteria cells, the aqueous phase concentration was basically completely depleted ( $\sim 0$ , Fankhauser et al. (2019)'s Figure 2B) implying a q value much lower than unity by several orders of magnitude, and also lower than our value of  $q \sim 0.8$  predicted for bacteria-containing droplets (our Figure 5c). We cannot reconcile the reasons for this difference by several orders of magnitude. However, the significantly lower q based on the results by Fankhauser et al. (2019) suggests that there was no thermodynamic equilibrium between the gas and aqueous phases in their model and that despite a relatively high gas phase concentration, the phase transfer was not sufficiently fast to replenish the formic acid concentration in the aqueous phase. Therefore, we conclude that the biodegradation of formic acid in Fankhauser's model was *significantly limited* due to the relatively inefficient uptake of formic acid from the gas phase (and chemical production within the droplet) as compared to the efficient loss by biodegradation.

Since Fankhauser et al. (2019) do not report gas phase concentrations of acetic acid, we cannot perform the corresponding analysis for this acid. However, given that the aqueous phase concentration was only  $\sim 10\%$  lower in the presence of bacteria may imply that the biodegradation did not lead to such strong subsaturation of acetic acid in the droplets, in agreement with our results (*q*(*aceticacid*)  $\sim 1$  at pH = 4.5, Figure 5b).

Pailler et al. (2023) report that net phase transfer rates in their simulations are zero in the presence and absence of bacteria, indicating thermodynamic equilibrium. This claim can be corroborated by an estimate of the corresponding q value based on their Figures 3 and 6 (very approximate since the exact data were not given in table form): E.g. for day conditions during summer in the presence of bacteria cells:  $c_{aq}$ (HCOOH) 20  $\mu$ M,  $c_{total}$ (HCOOH) ~3.6e9 molec cm<sup>-3</sup> results in

$$\frac{c_{aq}[M]}{p_g[atm]} = \frac{20 \cdot 10^{-6} M}{(4.3 \cdot 10^9 - 3.6 \cdot 10^9) molec \, cm^{-3}/2.5 \cdot 10^{19} molec \, cm^{-3} atm^{-1}} = 7 \cdot 10^5 M atm^{-1} \tag{S3}$$

leading to

$$q = \frac{7 \cdot 10^5 Matm^{-1}}{5.2 \cdot 10^5 Matm^{-1}} \sim 1 \tag{S4}$$

whereas the denominator is the effective Henry's law constant for pH = 5.5. using the  $K_H$  and  $K_a$  as applied in the CLEPS mechanism that is used by Pailler et al. (2023). This result shows that in their model, the aqueous phase concentration was not limited by uptake and the acids were in thermodynamic equilibrium.

### S2 Temperature dependence of biodegradation

Figure S3 shows  $k_{bact}$  vs T<sup>-1</sup> [K<sup>-1</sup>] (Table S7) compared to the corresponding trends for the OH reactions of formic acid/formate and acetic acid/acetate. This comparison suggests that the temperature dependencies for acetic acid are similar whereas the biodegradation of formic acid/formate seems more strongly T-dependent than the corresponding OH reactions.



**Figure S3.** Temperature dependencies for chemical rate constants for the OH reactions  $(k_{OH})$  and biodegradation  $(k_{bact})$  of formic and acetic acids. The temperature dependencies for the OH reactions are taken from Chin and Wine (1994), while bacterial species data were derived by Khaled et al. (2021) based on the measurements by Vaïtilingom et al. (2010) at 278.15 K and 290.15 K. The vertical lines denotes the temperature at which the model simulation were performed.

**Table S7.**  $k_{bact}$  values for formic and acetic acid, (Khaled et al., 2021) based on the measured biodegradation rates by (Vaïtilingom et al., 2010)

Species	$k_{bact}(17^{\circ}\mathrm{C})$	$k_{bact}(5^{\circ}C)$
	$/10^{-13} \mathrm{Lc}$	$ell^{-1} s^{-1}$
F	ormic acid	
Sphingomonas sp.	0.3	1.0
P. graminis	4.3	3.2
Pseudomonas sp.	1.5	0.28
P. viridiflava	5.4	1.5
A	Acetic acid	
Sphingomonas sp.	1.1	0.0054
P. graminis	10	1.0
Pseudomonas sp.	0.87	0.53
P. viridiflava	1.8	0.4

Using the Arrhenius equation

$$k_2 = k_1 \cdot exp\left[-\frac{E_a}{R}\left(\frac{1}{T_2} - \frac{1}{T_1}\right)\right] \tag{S5}$$

we can derive the activation energies  $E_a$  of the biodegradation processes as 90 kJ mol<sup>-1</sup> and 27 kJ mol<sup>-1</sup> for *Pseudomonas* sp., but with large ranges of (-65 - +90 kJ mol<sup>-1</sup> and 273 - 285 kJ mol<sup>-1</sup>) for all species investigated by (Vaïtilingom et al., 2010). Using the  $E_a$  values, the biodegradation rates at our model temperature would have been ~ 40% and ~ 13% lower than the measurements at 17 °C.

However, given the uncertainties associated with the measured biodegradation rates and their derived temperature dependencies (based on the two data points only!), we do not put much confidence into the derived temperature dependencies. This uncertainty is even further supported by the large scatter of values and slopes for the temperature dependent values of the other bacteria species measured by Vaïtilingom et al. (2010) (Figure S3). Considering these large uncertainties, the use of the directly measured data at 17°C seems justified.

Activation energies for biodegradation processes are rarely measured directly. A study on biodegradation of aromatic compounds showed an average value of 65 kJ mol<sup>-1</sup> (Knudsmark Sjøholm et al., 2021) which is within the range of the values determined (for quite different organic species).

The comparison of the  $E_a$  values for the OH reactions of the acids show similar trend but much lower values as compared to those for biodegradation with 8 kJ mol<sup>-1</sup> and 9 kJ mol<sup>-1</sup> for formic acid and formate and 11 kJ mol<sup>-1</sup> and 15 kJ mol<sup>-1</sup> for acetic acid and acetate (Chin and Wine, 1994).

The temperature dependence for biological processes is often expressed by means of the  $Q_{10}$  factor according to

$$R_{cell}(T_2) = R_{cell}(T_1) \cdot Q_{10}^{(T_2 - T_1)/10}$$
(S6)

i.e., the  $Q_{10}$  factor quantifies the change in the rate constant in a temperature interval of 10 K

$$Q_{10} = \frac{k_2}{k_1}$$
(S7)

Based on Equation S7, we derived  $Q_{10}$  values for the temperature interval 285 - 295 K (Table S8). They are in general agreement with those for cell generation rates ( $\sim 2 \le Q_{10} \le \sim 3$ ). By combining Equations S5 and S6, one can derive a simple relationship of  $Q_{10}$  and  $E_a$ , which is valid if  $T_1$  and  $T_2$  differ by 10 K (i.e.  $(T_2-T_1)/10 = 1$ ):

$$Q_{10} = exp\left[-\frac{E_a}{R}\left(\frac{1}{T_2} - \frac{1}{T_1}\right)\right]$$
(S8)

**Table S8.**  $Q_{10}$  factors ( $12^{\circ}C \le T \le 22^{\circ}C$ ) and activation energies derived from the biodegradation rates measured at 5°C and 17°C by Vaitilingom (2010).  $Q_{10}$  is temperature dependent and only valid for the indicated specific temperature interval.

	F	Formic acid	Acetic acid		
	$Q_{10}$	$E_a$ /kJ mol <sup>-1</sup>	$Q_{10}$	$E_a$ /kJ mol <sup>-1</sup>	
Pseudomonas sp.	3.9	90	1.5	27	
Sphingomonas sp.	0.4	-65	72	286	
Pseudomonas graminis	1.3	16	6.4	124	
Pseudomonas viridiflava	2.8	69	3.4	81	

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