

1 **Supplementary Information**

2 **Effects of pH and light exposure on the survival of bacteria and their ability to biodegrade**  
3 **organic compounds in clouds: Implications for microbial activity in acidic cloud water**

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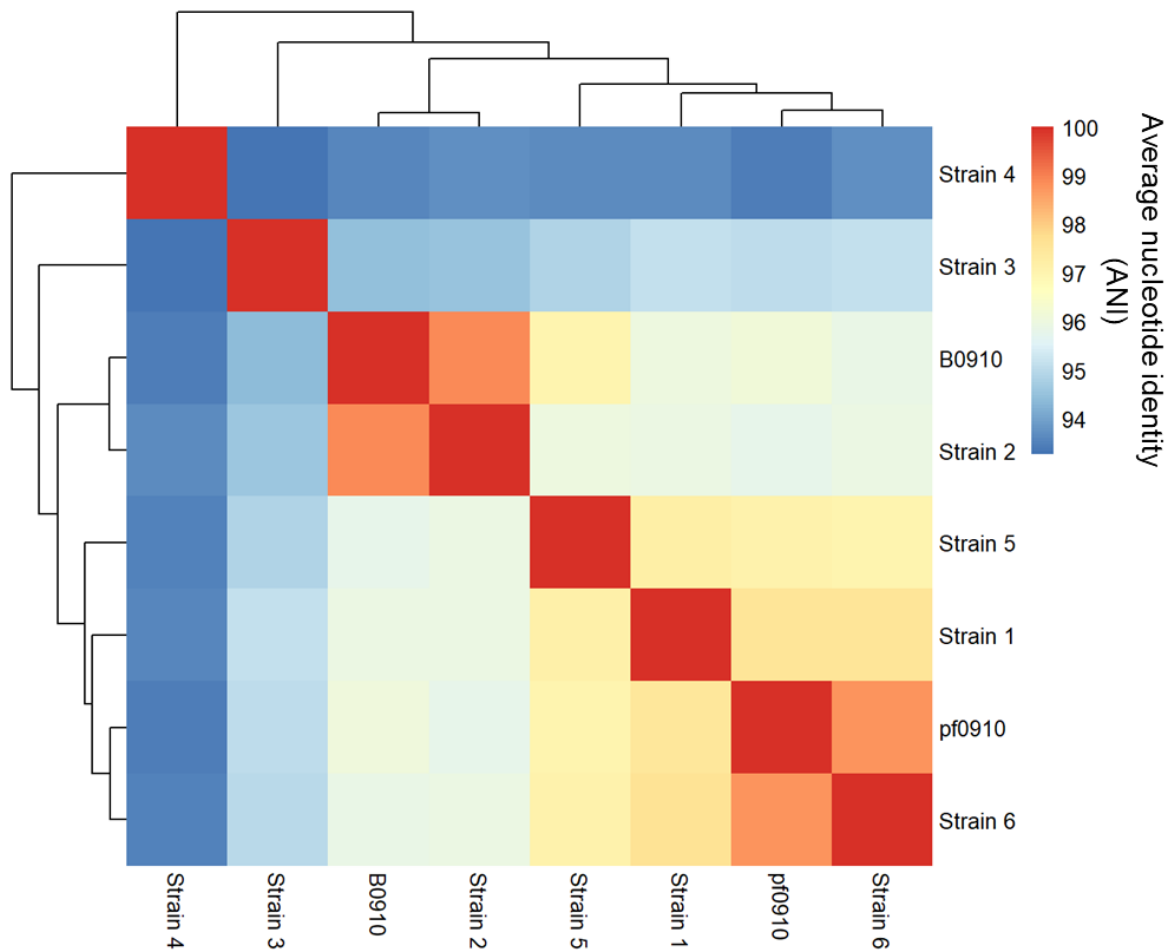
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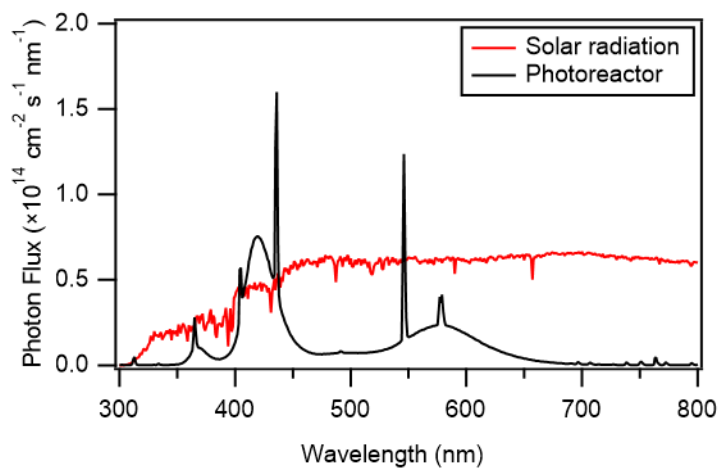
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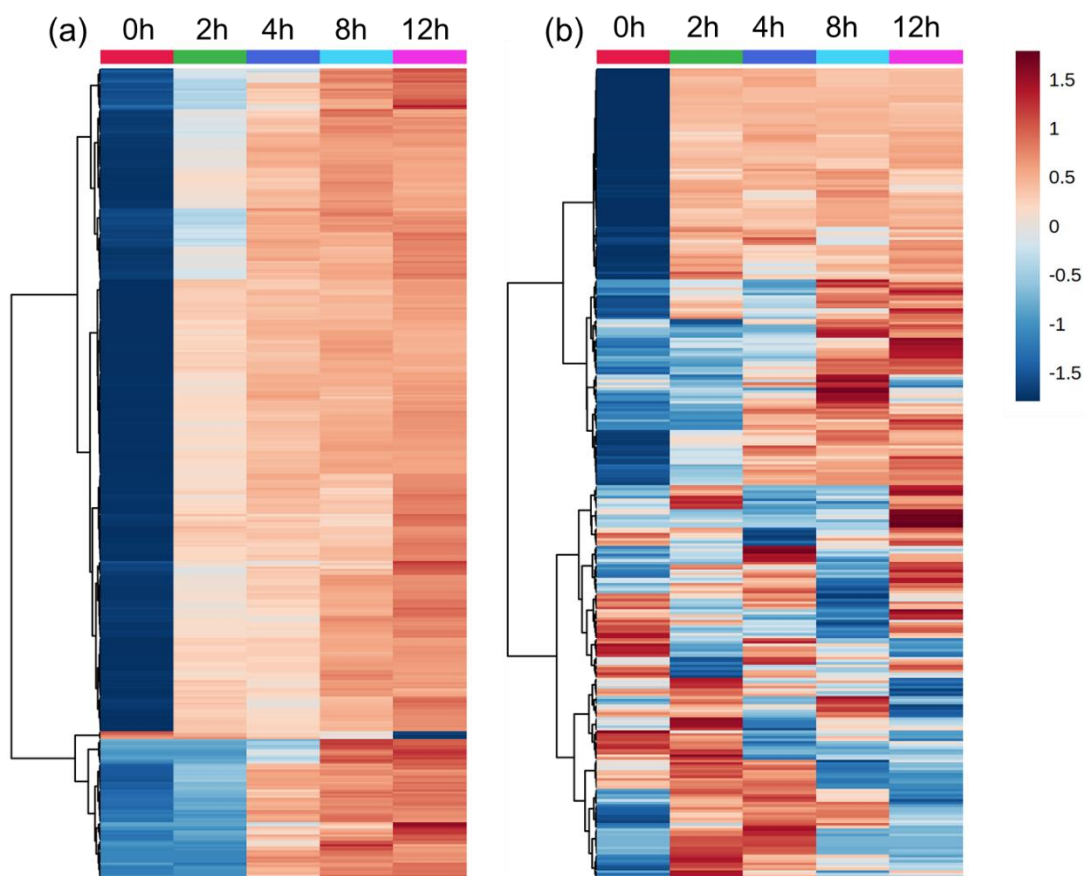
19 **Figure S1.** Average nucleotide identity (ANI) value of *Enterobacter* strains B0910, pf0910,  
 20 and six others. **Strain 1:** *Enterobacter hormaechei* subsp. *oharae* DSM 16687; **Strain 2:**  
 21 *Enterobacter hormaechei* subsp. *hoffmannii* DSM 14563; **Strain 3:** *Enterobacter hormaechei*  
 22 ATCC 49162; **Strain 4:** *Enterobacter quasihormaechei*. GCF 004331385.1; **Strain 5:**  
 23 *Enterobacter xiangfangensis* LMG27195; **Strain 6:** *Enterobacter hormaechei* subsp.  
 24 *steigerwaltii* DSM 16691. Strains 1 to 6 are the closest identified neighbors with strains B0910  
 25 and pf0910.



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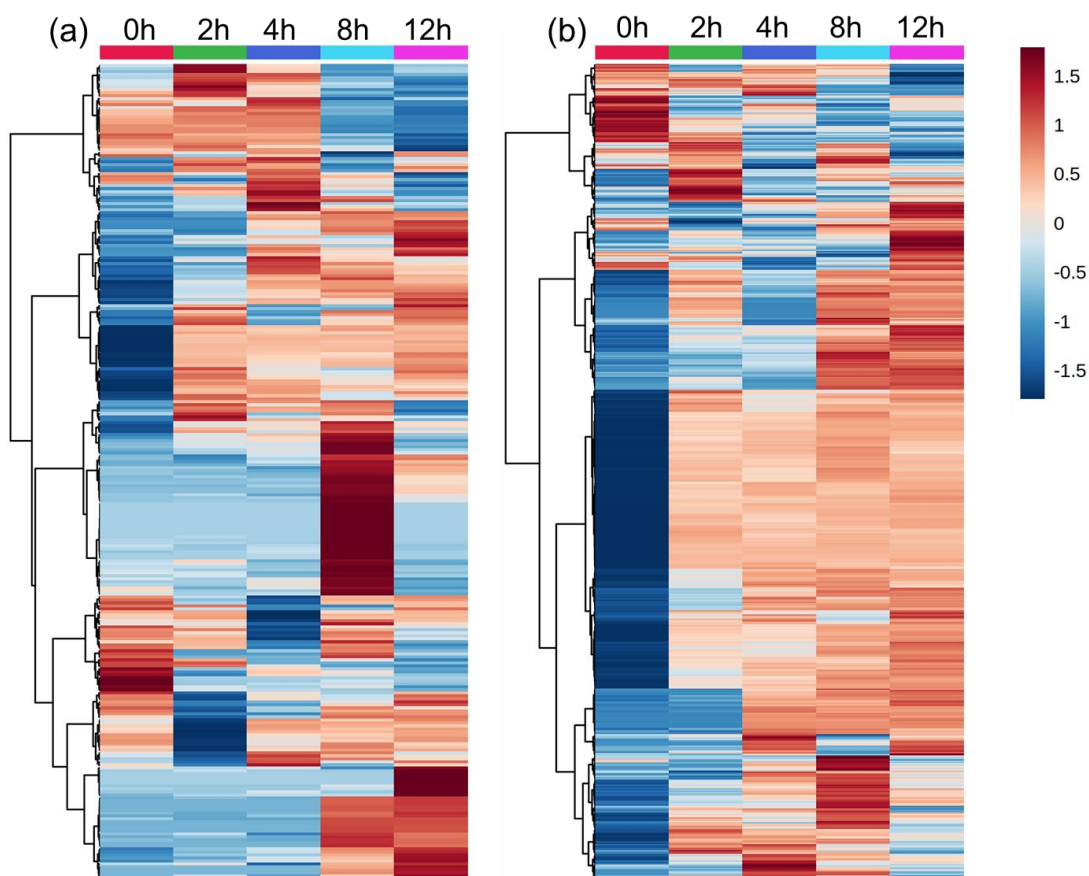
27 **Figure S2.** Photon flux inside of the photoreactor (black) and actinic flux for a fall day in Hong  
28 Kong in the morning (red). One lamp with output centered at  $\sim 365$  nm (RPR-3500A, Southern  
29 New England Ultraviolet Company), four lamps with outputs centered at  $\sim 421$  nm (RPR-  
30 4190A, Southern New England Ultraviolet Company), and three lamps with outputs centered  
31 at  $\sim 580$  nm (RPR-5750A, Southern New England Ultraviolet Company) were used to  
32 illuminate solutions in the photoreactor.

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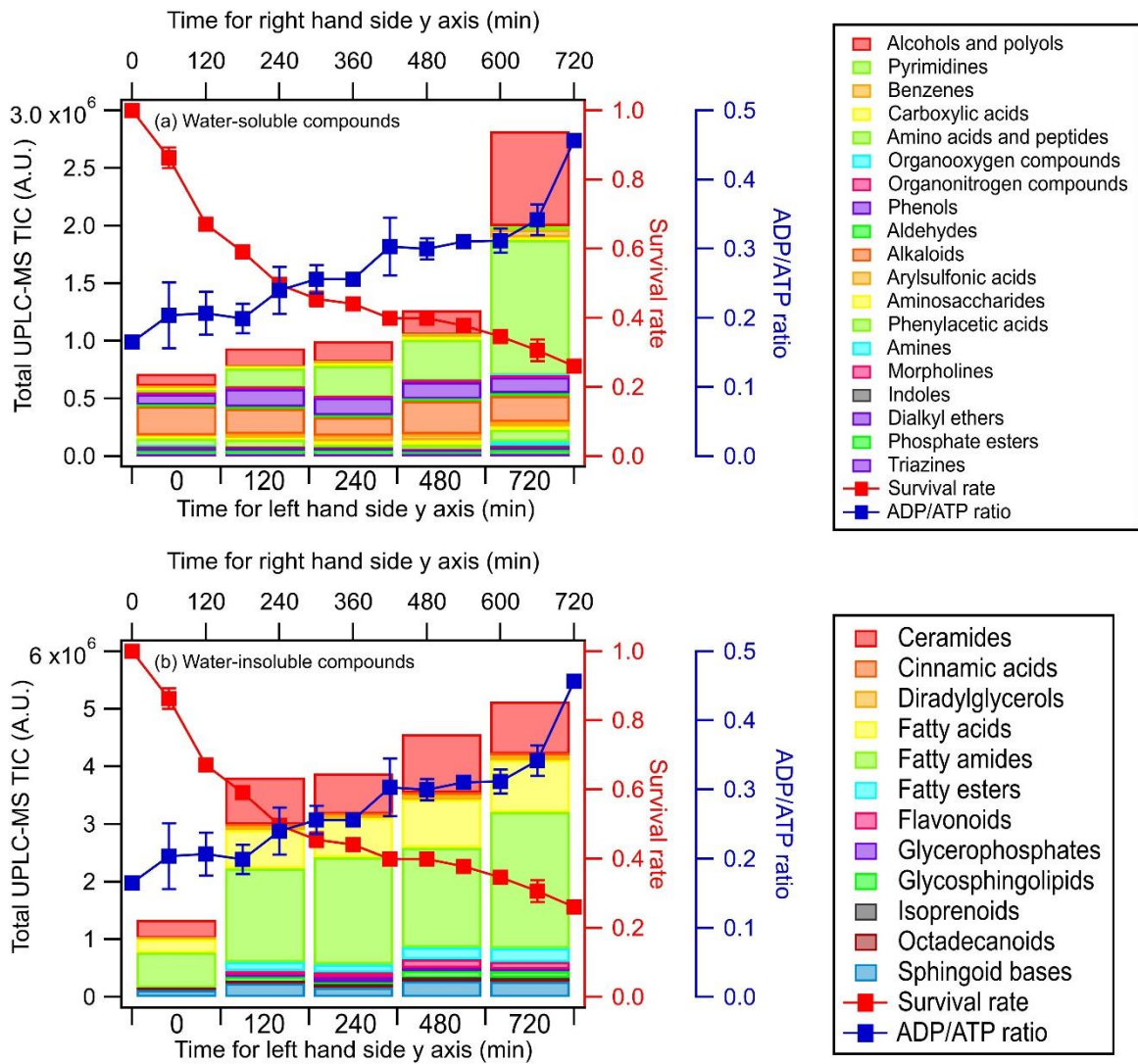
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35 **Figure S3.** Heat maps showing the time evolution of (a) water-soluble compounds and (b)  
 36 water-insoluble compounds from *E. hormaechei* B0910 during exposure to simulated sunlight  
 37 at pH 4.3. The heat maps were generated from non-targeted UPLC-MS analysis of samples  
 38 with different light exposure times. 259 water-soluble compounds and 215 water-insoluble  
 39 compounds were selected based on PLS-DA results (VIP > 1.0 criteria). The average UPLC-  
 40 MS intensity of each compound at each light exposure time was obtained from the nine  
 41 replicates. The average UPLC-MS intensities were subsequently log<sub>10</sub> transformed and auto  
 42 scaled (i.e., mean-centered and divided by the standard deviation of each variable). The color  
 43 scale ranges from red color for high abundance to blue for low abundance.



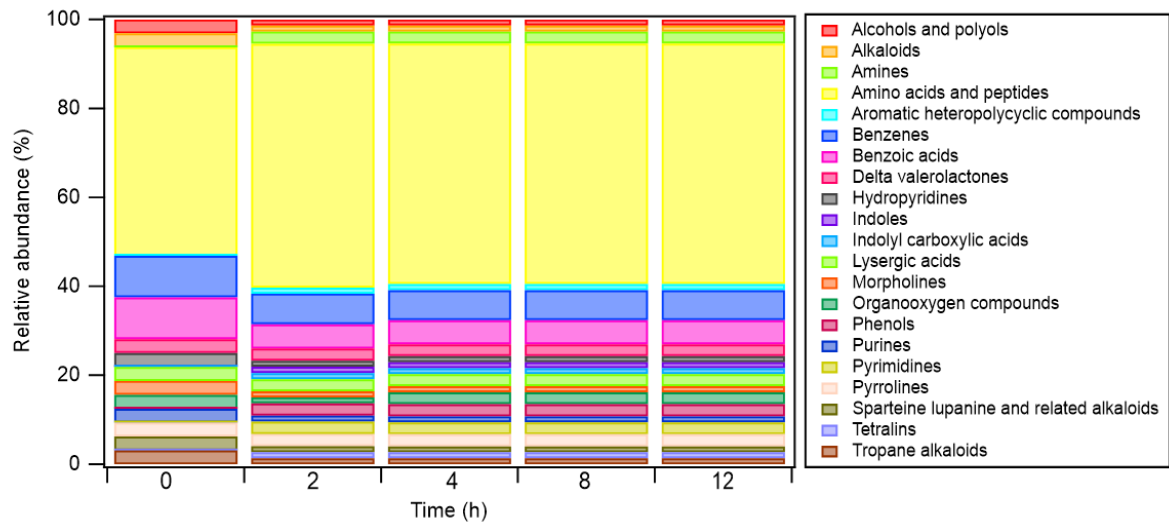
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45 **Figure S4.** Heat maps showing the time evolution of (a) water-soluble compounds and (b)  
 46 water-insoluble compounds from *E. hormaechei* pf0910 during exposure to simulated sunlight  
 47 at pH 4.3. The heat maps were generated from non-targeted UPLC-MS analysis of samples  
 48 with different light exposure times. 209 water-soluble compounds and 251 water-insoluble  
 49 compounds were selected based on PLS-DA results (VIP > 1.0 criteria). The average UPLC-  
 50 MS intensity of each compound at each light exposure time was obtained from the nine  
 51 replicates. The average UPLC-MS intensities were subsequently log<sub>10</sub> transformed and auto  
 52 scaled (i.e., mean-centered and divided by the standard deviation of each variable). The color  
 53 scale ranges from red color for high abundance to blue for low abundance.

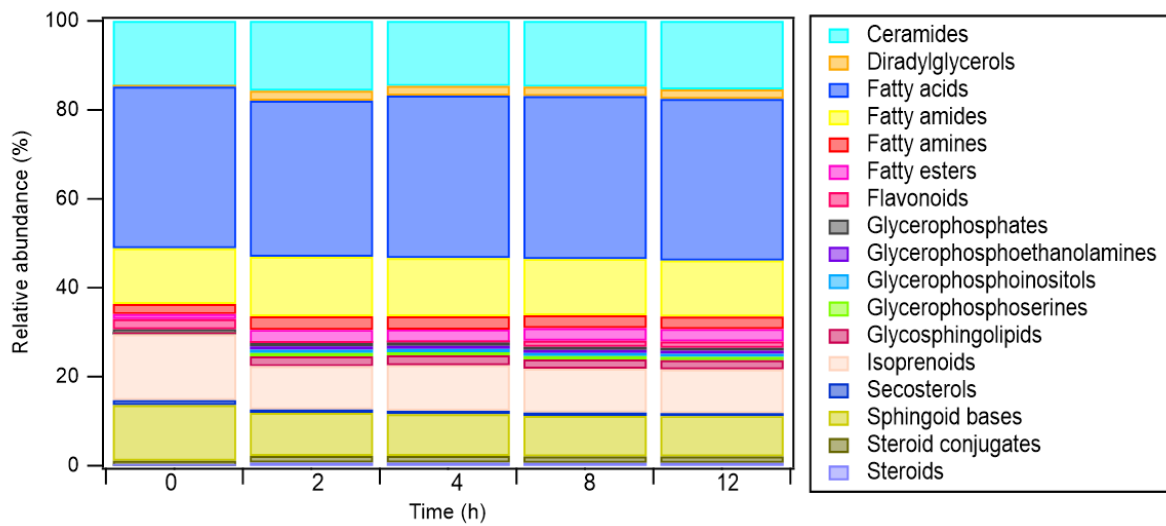


54  
 55 **Figure S5.** Time evolution of the UPLC-MS total ion chromatograph (TIC) signals of (a)  
 56 water-soluble compounds, and (b) water-insoluble compounds from *E. hormaechei* pf0910  
 57 during exposure to simulated sunlight at pH 4.3 over time. These compounds are classified  
 58 based on their chemical functionality. Also shown are the time evolution of the survival rate  
 59 and ADP/ATP ratio of *E. hormaechei* pf0910.

(a) Water-soluble compounds



(b) Water-insoluble compounds



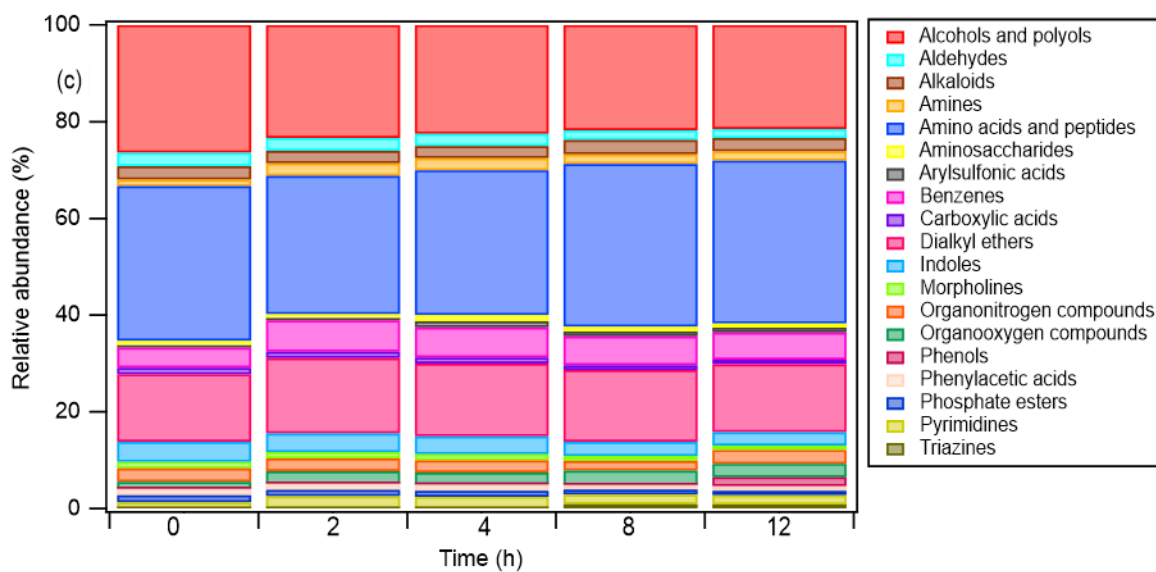
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61 **Figure S6.** Relative abundance of the different classes of (a) water-soluble compounds, and (b)

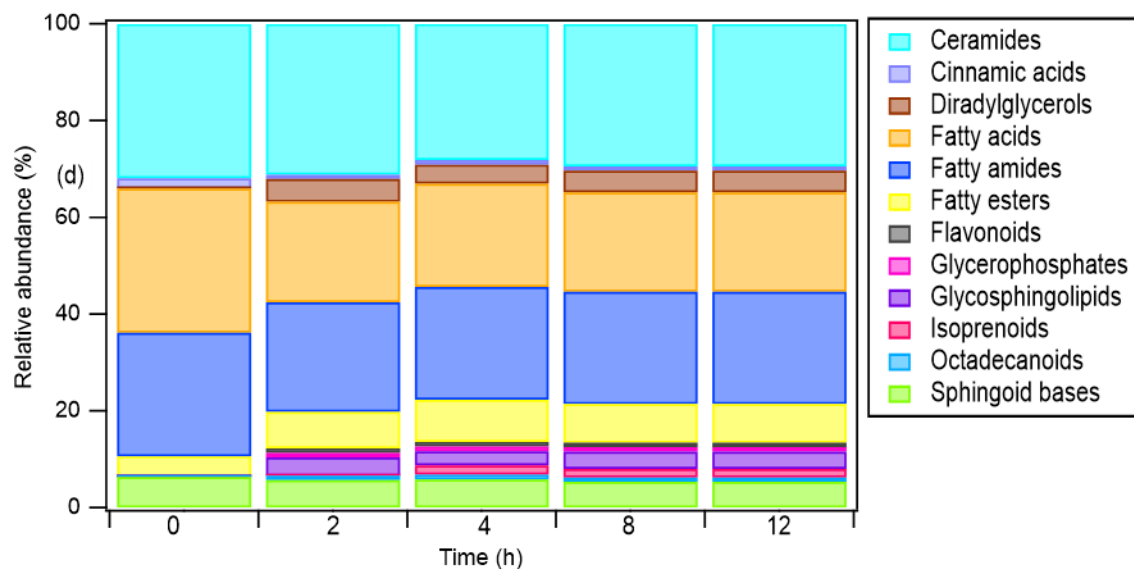
62 water-insoluble compounds from *E. hormaechei* B0910 during exposure to simulated sunlight

63 at pH 4.3.

(a) Water-soluble compounds



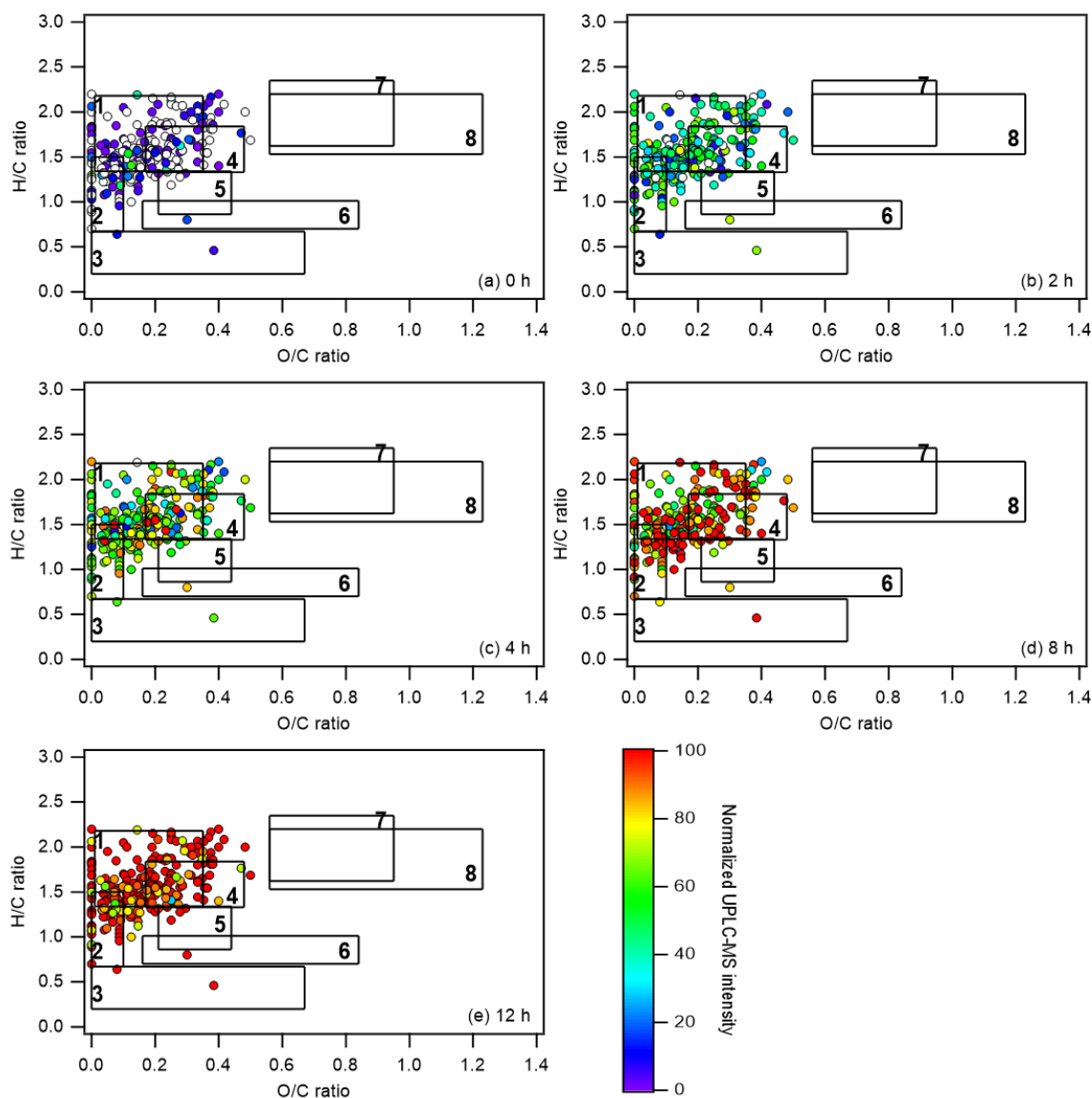
(b) Water-insoluble compounds



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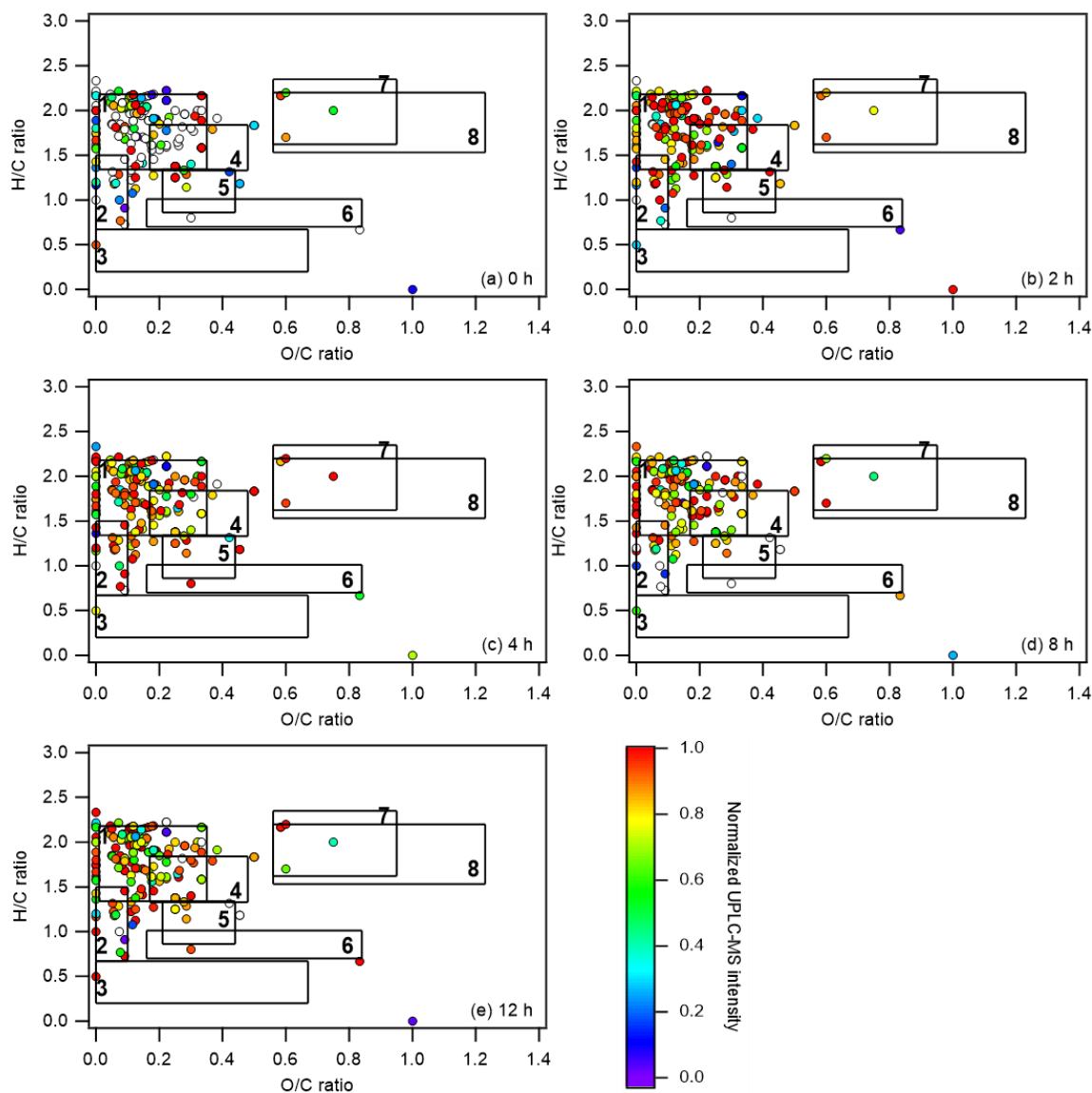
65 **Figure S7.** Relative abundance of the different classes of (c) water-soluble compounds, and (d)  
66 water-insoluble compounds from *E. hormaechei* pf0910 during exposure to simulated sunlight  
67 at pH 4.3.





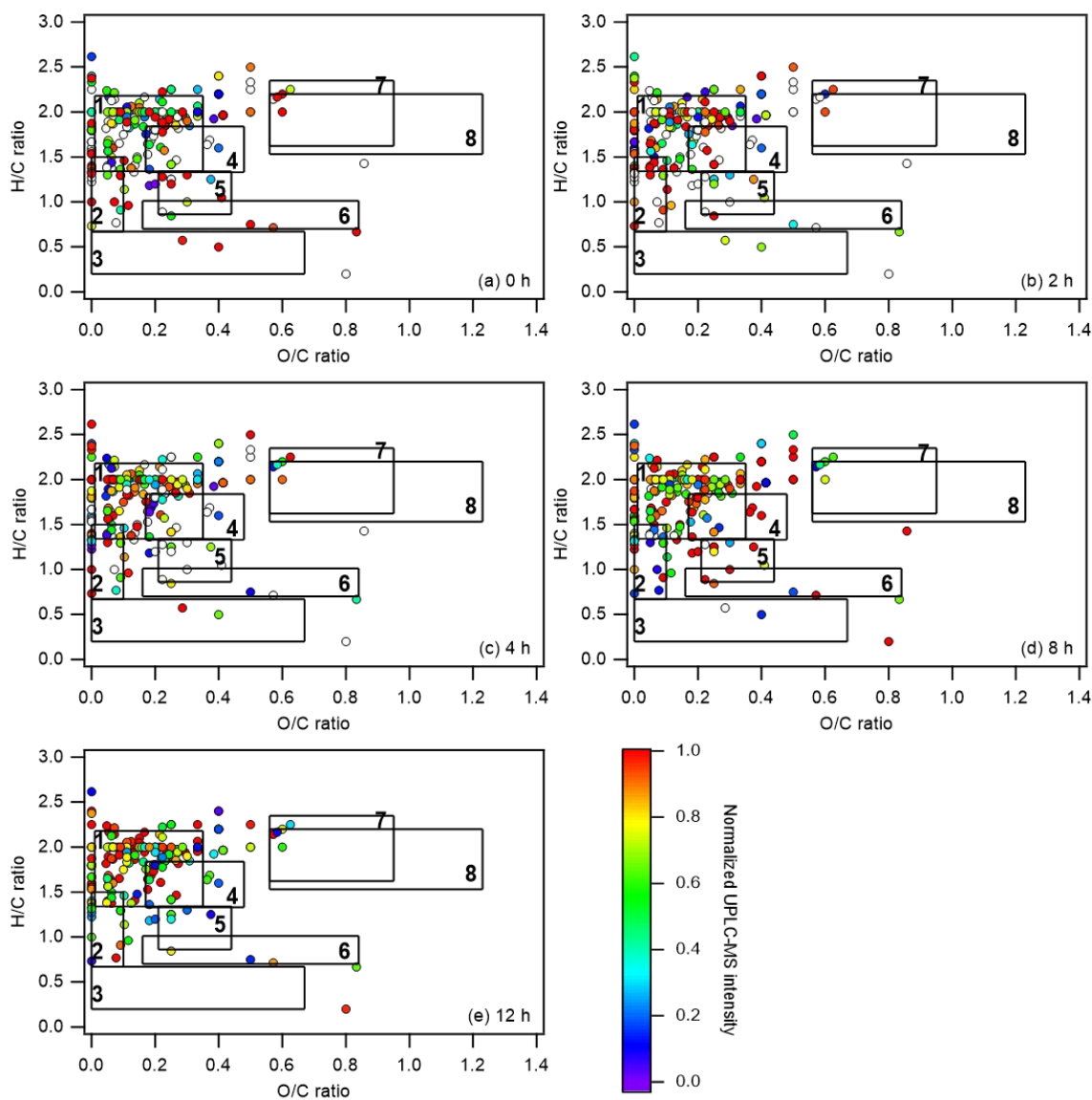
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69 **Figure S8.** Van Krevelen diagrams of water-soluble compounds from *E. hormaechei* B0910  
70 during exposure to simulated sunlight at pH 4.3 taken at different time points of the experiment:  
71 (a) 0 h, (b) 2 h, (c) 4 h, (d) 8 h, and (e) 12 h. The color of each symbol denotes its UPLC-MS  
72 intensity at that specific time point normalized to its maximum UPLC-MS intensity obtained  
73 during the entire experiment. Symbols that are colored white indicates that these compounds  
74 were not detected at that specific time point. The Van Krevelen diagrams are divided into eight  
75 chemical classes based on their O/C and H/C ratios: (1) lipids, (2) unsaturated hydrocarbons,  
76 (3) condensed aromatic structures, (4) peptides, (5) lignin, (6) tannin, (7) amino sugars, and (8)  
77 carbohydrates.



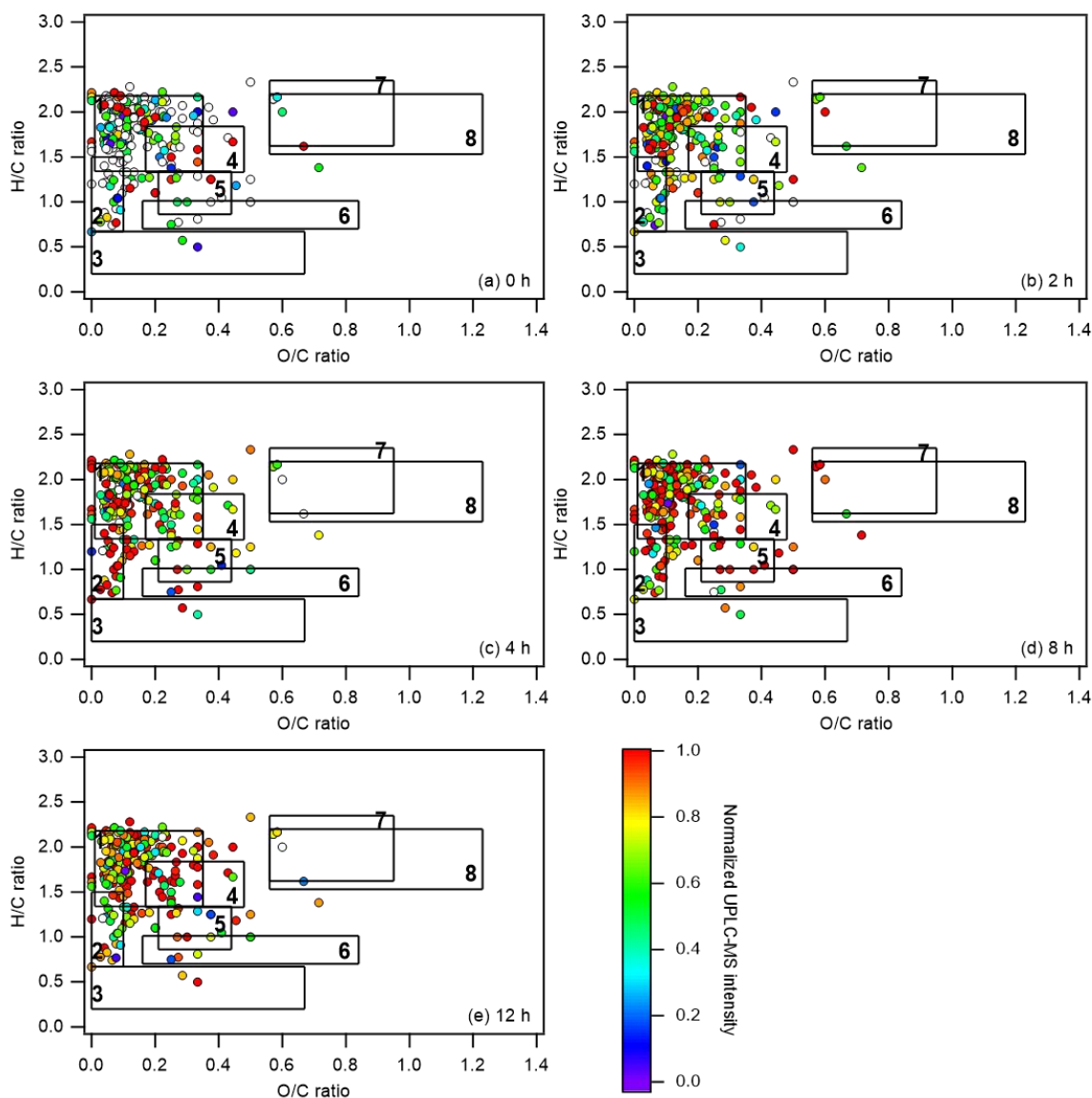
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79 **Figure S9.** Van Krevelen diagrams of water-insoluble compounds from *E. hormaechei* B0910  
 80 during exposure to simulated sunlight at pH 4.3: (a) 0 h, (b) 2 h, (c) 4 h, (d) 8 h, and (e) 12 h.  
 81 The color of each symbol denotes its UPLC-MS intensity at that specific time point normalized  
 82 to its maximum UPLC-MS intensity obtained during the entire experiment. Symbols that are  
 83 colored white indicates that these compounds were not detected at that specific time point. The  
 84 Van Krevelen diagrams are divided into eight chemical classes based on their O/C and H/C  
 85 ratios: (1) lipids, (2) unsaturated hydrocarbons, (3) condensed aromatic structures, (4) peptides,  
 86 (5) lignin, (6) tannin, (7) amino sugars, and (8) carbohydrates.



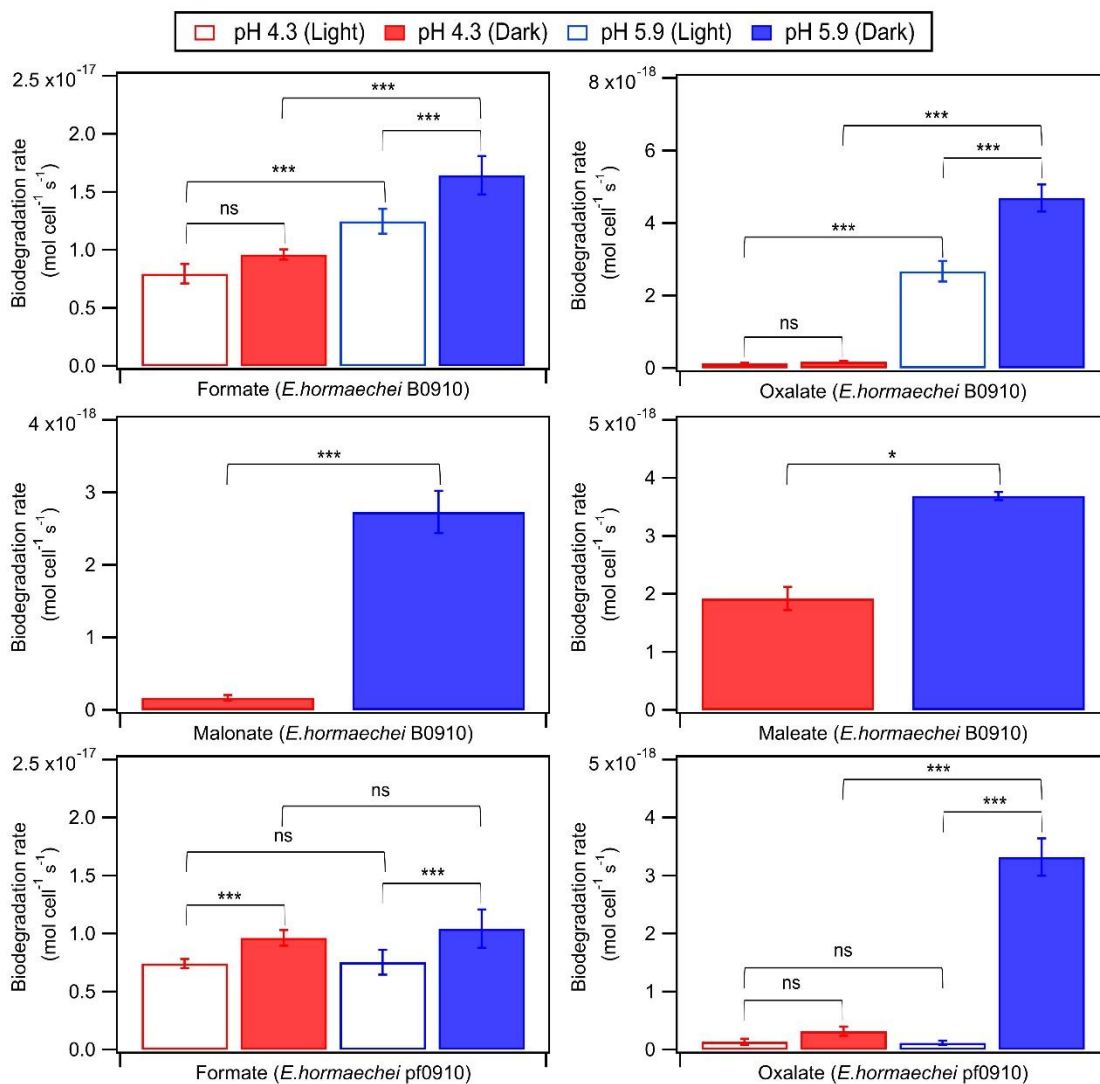
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88 **Figure S10.** Van Krevelen diagrams of water-soluble compounds from *E. hormaechei* pf0910  
 89 during exposure to simulated sunlight at pH 4.3: (a) 0 h, (b) 2 h, (c) 4 h, (d) 8 h, and (e) 12 h.  
 90 The color of each symbol denotes its UPLC-MS intensity at that specific time point normalized  
 91 to its maximum UPLC-MS intensity obtained during the entire experiment. Symbols that are  
 92 colored white indicates that these compounds were not detected at that specific time point. The  
 93 Van Krevelen diagrams are divided into eight chemical classes based on their O/C and H/C  
 94 ratios: (1) lipids, (2) unsaturated hydrocarbons, (3) condensed aromatic structures, (4) peptides,  
 95 (5) lignin, (6) tannin, (7) amino sugars, and (8) carbohydrates.



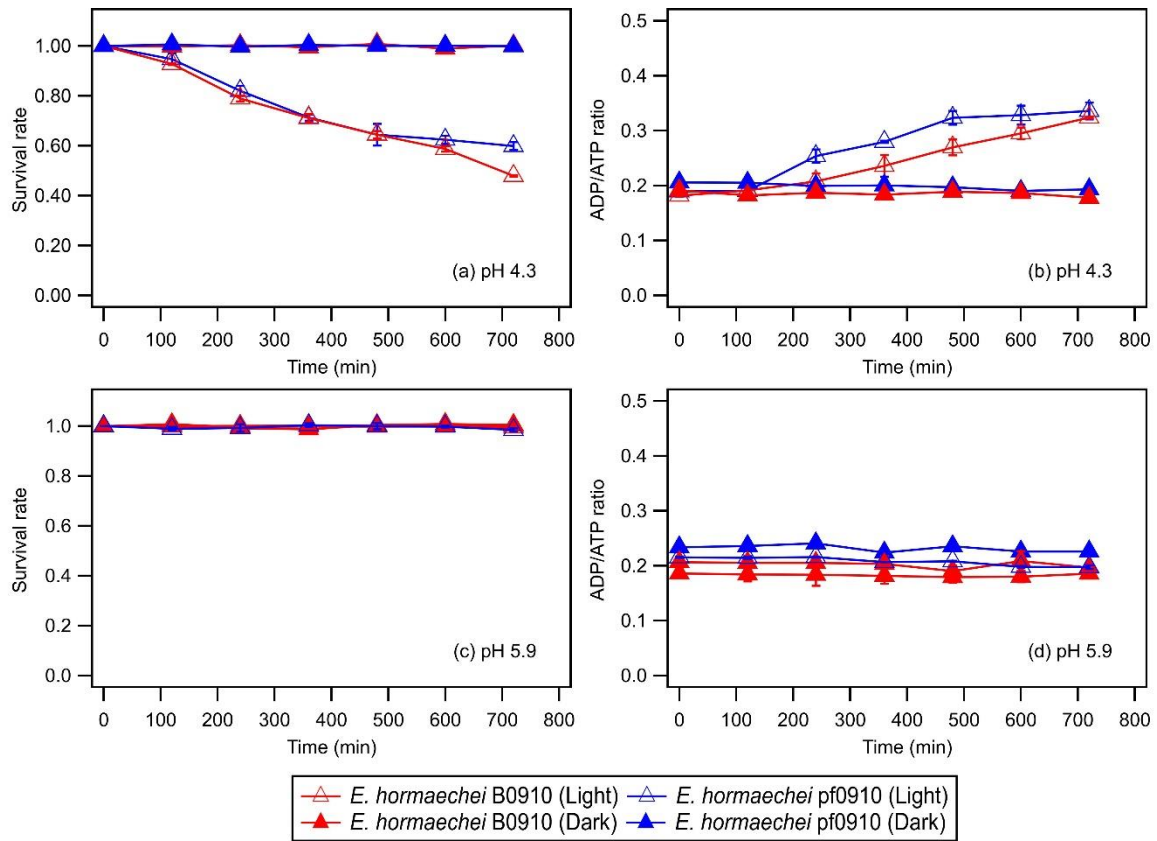
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97 **Figure S11.** Van Krevelen diagrams of water-insoluble compounds from *E. hormaechei*  
 98 pf0910 during exposure to simulated sunlight at pH 4.3: (a) 0 h, (b) 2 h, (c) 4 h, (d) 8 h, and (e)  
 99 12 h. The color of each symbol denotes its UPLC-MS intensity at that specific time point  
 100 normalized to its maximum UPLC-MS intensity obtained during the entire experiment.  
 101 Symbols that are colored white indicates that these compounds were not detected at that  
 102 specific time point. The Van Krevelen diagrams are divided into eight chemical classes based  
 103 on their O/C and H/C ratios: (1) lipids, (2) unsaturated hydrocarbons, (3) condensed aromatic  
 104 structures, (4) peptides, (5) lignin, (6) tannin, (7) amino sugars, and (8) carbohydrates.



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106 **Figure S12.** Biodegradation rates of oxalate, maleate, and malonate by (a) *E. hormaechei*  
 107 B0910 and (b) *E. hormaechei* pf0910 under light and dark conditions at pH 4.3 and pH 5.9.  
 108 Error bars represent one standard deviation from the mean of biological triplicates (ns: not  
 109 significant, \*:  $p$  value < 0.05, \*\*:  $p$  value < 0.01, \*\*\*:  $p$  value < 0.001).



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111 **Figure S13.** Survival and ADP/ATP ratios of *E. hormaechei* B0910 and *E. hormaechei* pf0910  
 112 under illuminated and dark conditions at pH 4.3 and pH 5.9 in the solutions containing the  
 113 seven carboxylic acids. Error bars represent one standard deviation from the mean of biological  
 114 triplicates.

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126 **Table S1.** Chemical composition of the artificial cloud water used to prepare bacterial cells  
 127 and perform experiments that investigated the effects of cloud water pH and light exposure on  
 128 the survival and energetic metabolism of bacteria. In the experiments, the pH of the artificial  
 129 cloud water was adjusted while keeping the final organic and inorganic ion composition the  
 130 same.

Organic ion	$\mu\text{M}$	Inorganic ion	$\mu\text{M}$
Formate	17.1	$\text{Na}^+$	93
Acetate	10.2	$\text{NH}_4^+$	235
Pyruvate	2.7	$\text{K}^+$	8
Oxalate	10.3	$\text{Mg}^{2+}$	23
		$\text{Ca}^{2+}$	49
		$\text{Cl}^-$	138
		$\text{SO}_4^{2-}$	305

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134 **Table S2.** Chemical composition of the artificial cloud water used for carboxylic acid  
 135 biodegradation experiments.

Organic ion	$\mu\text{M}$	Inorganic ion	$\mu\text{M}$
Formate	50	$\text{Na}^+$	930
Acetate	50	$\text{NH}_4^+$	2350
Pyruvate	50	$\text{K}^+$	80
Oxalate	50	$\text{Mg}^{2+}$	230
Succinate	50	$\text{Ca}^{2+}$	490
Maleate	50	$\text{Cl}^-$	1380
Malonate	50	$\text{SO}_4^{2-}$	3050
Glutarate	50		
MSA	50		

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142 **Table S3.** Stoichiometric ranges of the eight chemical classes in VK diagrams (Bianco et al.,  
 143 2018; Laszakovits and Mackay, 2022).

<b>Chemical class</b>	<b>H/C</b>	<b>O/C</b>
Amino sugar (Laszakovits and Mackay, 2022)	$1.62 \leq \text{H/C} \leq 2.35$	$0.56 \leq \text{O/C} \leq 0.95$
Carbohydrate (Laszakovits and Mackay, 2022)	$1.53 \leq \text{H/C} \leq 2.20$	$0.56 \leq \text{O/C} \leq 1.23$
Lignin (Laszakovits and Mackay, 2022)	$0.86 \leq \text{H/C} \leq 1.34$	$0.21 \leq \text{O/C} \leq 0.44$
Lipid (Laszakovits and Mackay, 2022)	$1.34 \leq \text{H/C} \leq 2.18$	$0.01 \leq \text{O/C} \leq 0.35$
Peptide (Laszakovits and Mackay, 2022)	$1.33 \leq \text{H/C} \leq 1.84$	$0.17 \leq \text{O/C} \leq 0.48$
Tannin (Laszakovits and Mackay, 2022)	$0.70 \leq \text{H/C} \leq 1.01$	$0.16 \leq \text{O/C} \leq 0.84$
Unsaturated hydrocarbons (Bianco et al., 2018)	$0.67 \leq \text{H/C} \leq 1.5$	$0 \leq \text{O/C} \leq 0.10$
Condensed aromatic structures (Bianco et al., 2018)	$0.20 \leq \text{H/C} \leq 0.67$	$0 \leq \text{O/C} \leq 0.67$

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145 **Table S4.** Genes involved in the biodegradation of carboxylic acids in the two *E. hormaechei*  
 146 strains.

Carboxylic acid	Genes	<i>E. hormaechei</i> B0910		<i>E. hormaechei</i> pf0910	
		Biodegradation	CDS	Biodegradation	CDS
		Yes/No	Absent/Present	Yes/No	Absent/Present
Formic acid	Formate dehydrogenase	Yes	AECJMNAI_01279; AECJMNAI_01280; AECJMNAI_01281; AECJMNAI_03442	Yes	CFAIMJNC_02488; CFAIMJNC_02489; CFAIMJNC_02490; CFAIMJNC_04681
	Oxalate decarboxylase	Yes	Absent	Yes	Absent
	Oxalate oxidase	Yes	Absent	Yes	Absent
Oxalic acid	Formyl-CoA:oxalate CoA-transferase	Yes	Absent	Yes	Absent
	Succinyl-CoA:oxalate CoA-transferase	Yes	Absent	Yes	Absent
	Hypothetical protein (Cupin 2 protein) <sup>a</sup>	Yes	AECJMNAI_00423	Yes	CFAIMJNC_01624
Malonic acid	Malonate decarboxylase	Yes	AECJMNAI_00917; AECJMNAI_00920; AECJMNAI_00921; AECJMNAI_00922; AECJMNAI_00924	No	CFAIMJNC_02108; CFAIMJNC_02111; CFAIMJNC_02112; CFAIMJNC_02113; CFAIMJNC_02115
	Malonate CoA-transferase	Yes	Absent	No	Absent
	Malonate-semialdehyde dehydrogenase	Yes	Absent	No	Absent
	Malonyl-CoA/methylmalonyl-CoA synthetase	Yes	Absent	No	Absent
	Maleate isomerase	Yes	Absent	No	Absent
	Maleate hydratase	Yes	Absent	No	Absent
	3-isopropylmalate dehydratase <sup>a</sup>	Yes	AECJMNAI_02091; AECJMNAI_02092	No	CFAIMJNC_03425; CFAIMJNC_03426
Acetic acid	Acetyl-CoA synthetase	No	AECJMNAI_01727	No	CFAIMJNC_02972
	Acetate kinase	No	AECJMNAI_04676	No	CFAIMJNC_00936
	Aldehyde dehydrogenase	No	AECJMNAI_01165	No	CFAIMJNC_02371

	ActP <sup>b</sup>	No	AECJMNAI_01725	No	CFAIMJNC_02970
	SatP <sup>b</sup>	No	AECJMNAI_02044	No	CFAIMJNC_03365
Methane sulfonic acid	Alkanesulfonate monooxygenase	No	AECJMNAI_03015; AECJMNAI_03017	No	CFAIMJNC_04351; CFAIMJNC_04353
Glutaric acid	Succinate-semialdehyde dehydrogenase / glutarate-semialdehyde dehydrogenase <sup>c</sup>	No	AECJMNAI_00812; AECJMNAI_01960	No	CFAIMJNC_02004; CFAIMJNC_03281
	Glutaryl-CoA synthetase	No	Absent	No	Absent
	Glutarate dioxygenase	No	Absent	No	Absent

147 <sup>a</sup> Genes are not canonical but may involve in the biodegradation of carboxylic acids.

148 <sup>b</sup> Transporter proteins involved in uptake of acetic acid for biodegradation

149 <sup>c</sup> No reverse catalysis in the direction from glutarate to glutarate-semialdehyde has been  
150 reported in the literature.

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153 **Table S5.** Concentration of radicals and cells used to estimate the loss rates by biodegradation

154 and chemical reactions in Table S6.

Radical concentration/ Cell concentration	Area	Concentration	Reference
·OH (M)	Remote	$2.2 \times 10^{-14}$	(Herrmann et al., 2010)
	Marine	$2.0 \times 10^{-12}$	(Herrmann et al., 2010)
	Urban	$3.5 \times 10^{-15}$	(Herrmann et al., 2010)
NO <sub>3</sub> · (M)	Remote	$5.1 \times 10^{-15}$	(Herrmann et al., 2010)
	Marine	$6.9 \times 10^{-15}$	(Herrmann et al., 2010)
	Urban	$1.4 \times 10^{-13}$	(Herrmann et al., 2010)
Cell (cell L <sup>-1</sup> )		$8.0 \times 10^7$	(Amato et al., 2007)

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156 **Table S6.** Estimations of the loss rates of formate, oxalate, and malonate by biodegradation and chemical reactions (i.e.,  $\cdot\text{OH}$  oxidation (daytime) and  $\text{NO}_3\cdot$  (nighttime)). These loss rates were calculated based on  
 157 concentrations and pH measured at the different sites. Equations used in these calculations can be found in Section S6. References used to obtain the pH of cloud/rainwater and carboxylic acids are indicated in superscripts.  
 158 The biodegradation and chemical reaction loss rates calculated here were used to generate Figure 5.

159 **Daytime**

Location (remote)	Category	pH	Formate ( $\mu\text{M}$ )	Formate loss rate ( $\text{M s}^{-1}$ )			Oxalate ( $\mu\text{M}$ )	Oxalate loss rate ( $\text{M s}^{-1}$ )		
				bio (pH ~4)	bio (pH ~5)	$\cdot\text{OH}$ (remote)		bio (pH ~4)	bio (pH ~5)	$\cdot\text{OH}$ (remote)
Mount Lu(Sun et al., 2016)	Cloud	3.81 (Sun et al., 2016)	10.83 (Sun et al., 2016)	$1.34 \times 10^{-10}$		$5.72 \times 10^{-10}$	4.95 (Sun et al., 2016)	$1.06 \times 10^{-12}$		$1.74 \times 10^{-11}$
Mount Lu(Sun et al., 2016)	Rain	4.44 (Sun et al., 2016)	10.21 (Sun et al., 2016)	$1.26 \times 10^{-10}$		$5.39 \times 10^{-10}$	2.54 (Sun et al., 2016)	$5.44 \times 10^{-13}$		$8.92 \times 10^{-12}$
Mount Heng(Wang et al., 2011)	Cloud	3.8 (Wang et al., 2011)	19.65 (Wang et al., 2011)	$2.43 \times 10^{-10}$		$1.04 \times 10^{-9}$	5.11 (Wang et al., 2011)	$1.10 \times 10^{-12}$		$1.80 \times 10^{-11}$
Mount Heng(Wang et al., 2011)	Rain	4.35 (Wang et al., 2011)	14.30 (Wang et al., 2011)	$1.77 \times 10^{-10}$		$7.55 \times 10^{-10}$	1.66 (Wang et al., 2011)	$3.55 \times 10^{-13}$		$5.83 \times 10^{-12}$
Mangdang Mountain(Cheng et al., 2011)	Rain	4.81 (Cheng et al., 2011)	7.90 (Cheng et al., 2011)	$9.78 \times 10^{-11}$		$4.17 \times 10^{-10}$	1.80 (Cheng et al., 2011)	$3.86 \times 10^{-13}$		$6.34 \times 10^{-12}$
Taiwan(Tsai and Kuo, 2013)	Cloud	3.91 (Tsai and Kuo, 2013)	5.74 (Tsai and Kuo, 2013)	$7.11 \times 10^{-11}$		$3.03 \times 10^{-10}$	6.60 (Tsai and Kuo, 2013)	$1.42 \times 10^{-12}$		$2.32 \times 10^{-11}$
Kleiner Feldberg, Germany(Wobrock et al., 1994)	Cloud	3.9-4.6 (Wobrock et al., 1994)	3.26 (Wobrock et al., 1994)	$4.03 \times 10^{-11}$		$1.72 \times 10^{-10}$	ND			
Whiteface Mountain, USA(Khwaja et al., 1995)	Cloud	3.1-4.4 (Khwaja et al., 1995)	25.20 (Khwaja et al., 1995)	$3.12 \times 10^{-10}$		$1.33 \times 10^{-9}$	9.66 (Khwaja et al., 1995)	$2.07 \times 10^{-12}$		$3.40 \times 10^{-11}$
Rax, Austria(Löflund et al., 2002)	Cloud	3.84 (Löflund et al., 2002)	13.25 (Löflund et al., 2002)	$1.64 \times 10^{-10}$		$7.00 \times 10^{-10}$	5.11 (Löflund et al., 2002)	$1.10 \times 10^{-12}$		$1.80 \times 10^{-11}$
Sonnblick, Austria(Brantner et al., 1994)	Cloud	5.0-6.5 (Brantner et al., 1994)	6.30 (Brantner et al., 1994)		$9.79 \times 10^{-11}$	$3.33 \times 10^{-10}$	1.89 (Limbeck and Puxbaum, 2000)		$3.61 \times 10^{-12}$	$6.65 \times 10^{-12}$
Mount Tai, China(Guo et al., 2012)	Cloud	4.6 (Guo et al., 2012)	31.80 (Guo et al., 2012)	$3.94 \times 10^{-10}$		$1.68 \times 10^{-9}$	11.10 (Guo et al., 2012)	$2.38 \times 10^{-12}$		$3.91 \times 10^{-11}$
Shangzhong(Xu et al., 2009)	Rain	ND	4.95 (Xu et al., 2009)	$6.13 \times 10^{-11}$		$2.61 \times 10^{-10}$	1.16 (Xu et al., 2009)	$2.48 \times 10^{-13}$		$4.07 \times 10^{-12}$
São Paulo State, Brazil(Coelho et al., 2011)	Rain	4.96 (Coelho et al., 2011)	7.80 (Coelho et al., 2011)		$1.21 \times 10^{-10}$	$4.12 \times 10^{-10}$	1.20 (Coelho et al., 2011)		$2.29 \times 10^{-12}$	$4.22 \times 10^{-12}$

Location (Marine)	Category	pH	Formate ( $\mu\text{M}$ )	Formate loss rate ( $\text{M s}^{-1}$ )			Oxalate ( $\mu\text{M}$ )	Oxalate loss rate ( $\text{M s}^{-1}$ )		
				bio (pH ~4)	bio (pH ~5)	$\cdot\text{OH}$ (marine)		bio (pH ~4)	bio (pH ~5)	$\cdot\text{OH}$ (marine)
Puerto Rico	Cloud	5.5 (Gioda et al., 2011)	1.00 (Gioda et al., 2011)		$1.55 \times 10^{-11}$	$4.80 \times 10^{-9}$	0.50 (Gioda et al., 2011)		$9.55 \times 10^{-13}$	$1.60 \times 10^{-10}$
Puerto Rico(Gioda et al., 2011)	Rain	5.3 (Gioda et al., 2011)	0.20 (Gioda et al., 2011)		$3.11 \times 10^{-12}$	$9.60 \times 10^{-10}$	0.00 (Gioda et al., 2011)			

Puy de dome(Vaitilingom et al., 2013)	Cloud	6.1 (Vaitilingom et al., 2013)	4.90 (Vaitilingom et al., 2013)	$7.61 \times 10^{-11}$	$2.35 \times 10^{-8}$	1.00 (Vaitilingom et al., 2013)	$1.91 \times 10^{-12}$	$3.20 \times 10^{-10}$
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Location (Urban)	Category	pH	Formate ( $\mu\text{M}$ )	Formate loss rate ( $\text{M s}^{-1}$ )			Oxalate ( $\mu\text{M}$ )	Oxalate loss rate ( $\text{M s}^{-1}$ )		
				bio (pH ~4)	bio (pH ~5)	$\cdot\text{OH}$ (urban)		bio (pH ~4)	bio (pH ~5)	$\cdot\text{OH}$ (urban)
Shenzhen, South China(Huang et al., 2010)	Rain	4.56 (Huang et al., 2010)	2.26 (Huang et al., 2010)	$2.80 \times 10^{-11}$		$1.90 \times 10^{-11}$	0.58 (Huang et al., 2010)	$1.23 \times 10^{-13}$		$3.22 \times 10^{-13}$
Anshun(Zhang et al., 2011)	Rain	4.67 (Zhang et al., 2011)	8.77 (Zhang et al., 2011)	$1.09 \times 10^{-10}$		$7.37 \times 10^{-11}$	2.84 (Zhang et al., 2011)	$6.09 \times 10^{-13}$		$1.59 \times 10^{-12}$
Newark US East Coast(Song and Gao, 2009)	Rain	4.6 (Song and Gao, 2009)	4.44 (Song and Gao, 2009)	$5.50 \times 10^{-11}$		$3.73 \times 10^{-11}$	0.68 (Song and Gao, 2009)	$1.46 \times 10^{-13}$		$3.81 \times 10^{-13}$
Hong Kong SAR(Li et al., 2020)	Cloud	3.87 (Li et al., 2020)	17.10 (Li et al., 2020)	$2.12 \times 10^{-10}$		$1.44 \times 10^{-10}$	10.30 (Li et al., 2020)	$2.21 \times 10^{-12}$		$5.77 \times 10^{-12}$
Puy de dome(Vaitilingom et al., 2013)	Cloud	3.9 (Vaitilingom et al., 2013)	33.20 (Vaitilingom et al., 2013)	$4.11 \times 10^{-10}$		$2.79 \times 10^{-10}$	9.30 (Vaitilingom et al., 2013)	$1.99 \times 10^{-12}$		$5.21 \times 10^{-12}$

160 ND: No data

161 **Nighttime**

Location (remote)	Category	pH	Formate ( $\mu\text{M}$ )	Formate loss rate ( $\text{M s}^{-1}$ )			Oxalate ( $\mu\text{M}$ )	Oxalate loss rate ( $\text{M s}^{-1}$ )			Malonate ( $\mu\text{M}$ )	Malonate loss rate ( $\text{M s}^{-1}$ )		
				bio (pH ~4)	bio (pH ~5)	$\text{NO}_3\cdot$ (remote)		bio (pH ~4)	bio (pH ~5)	$\text{NO}_3\cdot$ (remote)		bio (pH ~4)	bio (pH ~5)	$\text{NO}_3\cdot$ (remote)
Mount Lu(Sun et al., 2016)	Cloud	3.81 (Sun et al., 2016)	10.83 (Sun et al., 2016)	$1.69 \times 10^{-10}$		$2.32 \times 10^{-12}$	4.95 (Sun et al., 2016)	$2.07 \times 10^{-12}$		$1.11 \times 10^{-12}$	ND			
Mount Lu(Sun et al., 2016)	Rain	4.44 (Sun et al., 2016)	10.21 (Sun et al., 2016)	$1.59 \times 10^{-10}$		$2.19 \times 10^{-12}$	2.54 (Sun et al., 2016)	$1.06 \times 10^{-12}$		$5.69 \times 10^{-13}$	ND			
Mount Heng(Wang et al., 2011)	Cloud	3.8 (Wang et al., 2011)	19.65 (Wang et al., 2011)	$3.06 \times 10^{-10}$		$4.21 \times 10^{-12}$	5.11 (Wang et al., 2011)	$2.14 \times 10^{-12}$		$1.15 \times 10^{-12}$	ND			
Mount Heng(Wang et al., 2011)	Rain	4.35 (Wang et al., 2011)	14.30 (Wang et al., 2011)	$2.23 \times 10^{-10}$		$3.06 \times 10^{-12}$	1.66 (Wang et al., 2011)	$6.94 \times 10^{-13}$		$3.71 \times 10^{-13}$	ND			
Mangdang Mountain(Cheng et al., 2011)	Rain	4.81 (Cheng et al., 2011)	7.90 (Cheng et al., 2011)	$1.23 \times 10^{-10}$		$1.69 \times 10^{-12}$	1.80 (Cheng et al., 2011)	$7.55 \times 10^{-13}$		$4.04 \times 10^{-13}$	1.40 (Cheng et al., 2011)		$5.16 \times 10^{-12}$	$4.00 \times 10^{-14}$
Taiwan(Tsai and Kuo, 2013)	Cloud	3.91 (Tsai and Kuo, 2013)	5.74 (Tsai and Kuo, 2013)	$8.95 \times 10^{-11}$		$1.23 \times 10^{-12}$	6.60 (Tsai and Kuo, 2013)	$2.77 \times 10^{-12}$		$1.48 \times 10^{-12}$	0.16 (Tsai and Kuo, 2013)	$3.65 \times 10^{-14}$		$4.57 \times 10^{-15}$
Kleiner Feldberg, Germany(Wobrock et al., 1994)	Cloud	3.9-4.6 (Wobrock et al., 1994)	3.26 (Wobrock et al., 1994)	$5.08 \times 10^{-11}$		$6.98 \times 10^{-13}$	ND				ND			
Whiteface Mountain, USA(Khwaja et al., 1995)	Cloud	3.1-4.4 (Khwaja et al., 1995)	25.20 (Khwaja et al., 1995)	$3.93 \times 10^{-10}$		$5.40 \times 10^{-12}$	9.66 (Khwaja et al., 1995)	$4.05 \times 10^{-12}$		$2.17 \times 10^{-12}$	7.69 (Khwaja et al., 1995)	$1.75 \times 10^{-12}$		$2.20 \times 10^{-13}$
Rax, Austria(Löflund et al., 2002)	Cloud	3.84 (Löflund et al., 2002)	13.25 (Löflund et al., 2002)	$2.07 \times 10^{-10}$		$2.84 \times 10^{-12}$	5.11 (Löflund et al., 2002)	$2.14 \times 10^{-12}$		$1.15 \times 10^{-12}$	1.92 (Löflund et al., 2002)	$4.38 \times 10^{-13}$		$5.49 \times 10^{-14}$
Sonnblick, Austria(Brantner et al., 1994)	Cloud	5.0-6.5 (Brantner et al., 1994)	6.30 (Brantner et al., 1994)		$1.32 \times 10^{-10}$	$1.35 \times 10^{-12}$	1.89 (Limbeck and Puxbaum, 2000)		$1.19 \times 10^{-11}$	$4.24 \times 10^{-13}$	0.38 (Limbeck and Puxbaum, 2000)		$1.42 \times 10^{-12}$	$1.10 \times 10^{-14}$
Mount Tai, China(Guo et al., 2012)	Cloud	4.6 (Guo et al., 2012)	31.80	$4.96 \times 10^{-10}$		$6.81 \times 10^{-12}$	11.10 (Guo et al., 2012)	$4.65 \times 10^{-12}$		$2.49 \times 10^{-12}$	ND			
Shangzhong(Xu et al.,	Rain		4.95	$7.71 \times 10^{-11}$		$1.06 \times 10^{-12}$	1.16	$4.84 \times 10^{-13}$		$2.59 \times 10^{-13}$	ND			

2009)														
São Paulo State, Brazil(Coelho et al., 2011)	Rain	4.96 (Coelho et al., 2011)	7.80 (Coelho et al., 2011)		$1.63 \times 10^{-10}$	$1.67 \times 10^{-12}$	1.20 (Coelho et al., 2011)		$7.57 \times 10^{-12}$	$2.69 \times 10^{-13}$	ND			
Location (marine)		pH	Formate ( $\mu\text{M}$ )	Formate loss rate ( $\text{M s}^{-1}$ )			Oxalate ( $\mu\text{M}$ )	Oxalate loss rate ( $\text{M s}^{-1}$ )			Malonate ( $\mu\text{M}$ )	Malonate loss rate ( $\text{M s}^{-1}$ )		
				bio (pH ~4)	bio (pH ~5)	$\text{NO}_3^-$ (marine)		bio (pH ~4)	bio (pH ~5)	$\text{NO}_3^-$ (marine)		bio (pH ~4)	bio (pH ~5)	$\text{NO}_3^-$ (marine)
Puerto Rico(Gioda et al., 2011)	Cloud	5.5 (Gioda et al., 2011)	1.00 (Gioda et al., 2011)		$2.09 \times 10^{-11}$	$2.90 \times 10^{-13}$	0.50 (Gioda et al., 2011)		$3.16 \times 10^{-12}$	$1.52 \times 10^{-13}$	ND			
Puerto Rico(Gioda et al., 2011)	Rain	5.3 (Gioda et al., 2011)	0.20 (Gioda et al., 2011)		$4.19 \times 10^{-12}$	$5.80 \times 10^{-14}$	ND				ND			
Puy de dôme(Vaitilingom et al., 2013)	Cloud	6.1 (Vaitilingom et al., 2013)	4.90 (Vaitilingom et al., 2013)		$1.03 \times 10^{-10}$	$1.42 \times 10^{-12}$	1.00 (Vaitilingom et al., 2013)		$6.31 \times 10^{-12}$	$3.04 \times 10^{-13}$	0.40 (Vaitilingom et al., 2012)		$1.47 \times 10^{-12}$ $1.55 \times 10^{-14}$	
Location (urban)		pH	Formate ( $\mu\text{M}$ )	Formate loss rate ( $\text{M s}^{-1}$ )			Oxalate ( $\mu\text{M}$ )	Oxalate loss rate ( $\text{M s}^{-1}$ )			Malonate ( $\mu\text{M}$ )	Malonate loss rate ( $\text{M s}^{-1}$ )		
				bio (pH ~4)	bio (pH ~5)	$\text{NO}_3^-$ (urban)		bio (pH ~4)	bio (pH ~5)	$\text{NO}_3^-$ (urban)		bio (pH ~4)	bio (pH ~5)	$\text{NO}_3^-$ (urban)
Shenzhen, South China(Huang et al., 2010)	Rain	4.56 (Huang et al., 2010)	2.26 (Huang et al., 2010)		$3.52 \times 10^{-11}$		0.58 (Huang et al., 2010)		$2.41 \times 10^{-13}$		ND			
Anshun(Zhang et al., 2011)	Rain	4.67 (Zhang et al., 2011)	8.77 (Zhang et al., 2011)		$1.37 \times 10^{-10}$		2.84 (Zhang et al., 2011)		$1.19 \times 10^{-12}$		ND			
Newark US East Coast(Song and Gao, 2009)	Rain	4.6 (Song and Gao, 2009)	4.44 (Song and Gao, 2009)		$6.92 \times 10^{-11}$		0.68 (Song and Gao, 2009)		$2.85 \times 10^{-13}$		0.29(Song and Gao, 2009)		$6.61 \times 10^{-14}$ $2.27 \times 10^{-13}$	
Hong Kong SAR(Li et al., 2020)	Cloud	3.87 (Li et al., 2020)	17.10 (Li et al., 2020)		$2.66 \times 10^{-10}$		10.30 (Li et al., 2020)		$4.32 \times 10^{-12}$		1.36 (Zhao et al., 2019)		$3.10 \times 10^{-13}$ $1.07 \times 10^{-12}$	
Puy de dome(Vaitilingom et al., 2013)	Cloud	3.9 (Vaitilingom et al., 2013)	33.20 (Vaitilingom et al., 2013)		$5.17 \times 10^{-10}$		9.30 (Vaitilingom et al., 2013)		$3.90 \times 10^{-12}$		3.50 (Vaitilingom et al., 2013)		$7.97 \times 10^{-13}$ $2.74 \times 10^{-12}$	

162 ND: No data

## 163 **Section S1. Genome assembly, annotation, and taxonomic analysis**

164 Genome assembly of the sequencing reads was performed using the NECAT pipeline  
165 (v0.0.1\_update20200803) (Chen et al., 2021) with the default parameters. The reads were first  
166 corrected (PREP\_OUTPUT\_COVERAGE = 40, CNS\_OUTPUT\_COVERAGE = 30,  
167 MIN\_READ\_LENGTH = 3000) and then the corrected reads were assembled  
168 (OVLFP\_FAST\_OPTIONS = -n 500 -z 20 -b 2000 -e 0.5 -j 0 -u 1 -a 1000,  
169 OVLFP\_SENSITIVE\_OPTIONS = -n 500 -z 10 -e 0.5 -j 0 -u 1 -a 1000). Both the correction  
170 and assembly steps were progressive with multiple processing steps to improve the accuracy  
171 and completeness. The quality of the assembled genomes was evaluated using the  
172 Benchmarking Universal Single-copy Orthologs (BUSCO v5.3.1) tool based on the database  
173 of enterobacterales\_odb10 (Manni et al., 2021). For both strains B00910 and pf0910, complete  
174 circular chromosomes and plasmids were obtained.

175 Genome annotation was performed using Prokka (v1.14.6) (Seemann, 2014) with the  
176 default parameters. Whole genome-based taxonomic analysis was conducted using the Type  
177 (Strain) Genome Server (TYGS) (Meier-Kolthoff and Göker, 2019). Average Nucleotide  
178 Identity (ANI) was calculated by fastANI (v1.33) (Jain et al., 2018). Metabolic pathways were  
179 analyzed using the KEGG Mapper (Kanehisa et al., 2022) and the RAST server (Aziz et al.,  
180 2008). The sequences of the two genomes have been deposited in NCBI under the BioProject  
181 number PRJNA812965.

## 182 **Section S2. Extraction of water-insoluble and water-soluble biological material and** 183 **organic compounds for UPLC-MS analysis**

184 A modified Bligh & Dyer (BD) protocol was performed to extract water-insoluble  
185 organic compounds (Sündermann et al., 2016). Briefly, 3 mL of methanol (Duskan, LC-MS  
186 grade)/chloroform (RCI, HPLC grade) (1:2, v/v) was added to a filtered 5 mL sample solution  
187 and vortexed for 5 min, after which the samples were centrifuged at 3000 rpm for 10 min at 10  
188 °C. The bottom layer was collected into a clean 2 mL centrifuge tube and dried in a concentrator  
189 using nitrogen gas. The dried extracts were redissolved in 500 µL of acetonitrile (Duskan, LC-  
190 MS grade) and stored at -20 °C prior to UPLC-MS analysis. Solid-phase extraction (SPE) was

191 performed to remove the inorganic salts and extract the water-soluble organic compounds using  
192 hydrophobic lipophilic balanced (HLB) cartridges (Oasis HLB, 6cc 500 mg). The HLB  
193 cartridges were first preconditioned with 1 mL methanol and 2 mL Milli-Q water. A 10 mL  
194 filtered sample solution was then loaded into the SPE cartridge and washed with 20 mL Milli-  
195 Q water under vacuum at a flow rate of 5 mL/min. The elution was performed by adding 1.5  
196 mL methanol (Duskan, LC-MS grade). The eluent was evaporated to dryness under nitrogen  
197 gas and reconstituted in 500  $\mu$ L acetonitrile (Duskan, LC-MS grade).

### 198 **Section S3. UPLC-MS operation, data processing, and statistical analysis**

199 Chromatographic separation was performed on a Kinetex HILIC LC column (100  $\times$  2.1  
200 mm, 2.6  $\mu$ m, 100  $\text{\AA}$ , Phenomenex). The flow rate was fixed at 0.3 mL/min with ultra-pure  
201 water containing 5 mM ammonium acetate (Fisher, LC-MS grade) as mobile phase A and  
202 acetonitrile (Duskan, LC-MS grade) for mobile phase B. The following gradient program was  
203 used: 0 to 2 min 95% A; 2 to 4 min linear gradient to 80% B; 4 to 11 min linear gradient to  
204 65% B; 11 to 12.5 min 65% B; 12.5 to 13 min linear gradient to 95% B; 13 to 15 min  
205 equilibration wash with 95% B. Injection volume was set at 10  $\mu$ L. The information dependent  
206 analysis (IDA) acquisition was acquired with MS scan (100 to 1200 m/z) followed by MS/MS  
207 scan (50 to 1200 m/z) in positive ion mode. The following MS conditions were used: 30 PSI  
208 curtain gas, 60 PSI ion source gas, 3000 V ESI ion spray voltage, 320  $^{\circ}$ C source temperature,  
209 10 V collision energy for MS, and 80 V declustering potential. MS/MS was acquired with a  
210 collision energy was 20 V with 5 V spread. The raw MS data was processed for peak detection,  
211 retention time correction, alignment, and integration using the XCMS software built into the  
212 web-based Galaxy platform (<https://umsa.cerit-sc.cz/>) (Gowda et al., 2014). The processed data  
213 was then uploaded to MetaboAnalyst 5.0 (<https://www.metaboanalyst.ca/>) (Pang et al., 2021)  
214 to identify cellular compounds that had prominent ion intensities.

215 The raw UPLC-MS data first underwent preprocessing, normalization, and quality  
216 control steps using the XCMS software built into the web-based Galaxy platform (available at:  
217 <https://umsa.cerit-sc.cz/>). The raw data was processed for peak detection, alignment, and  
218 framing. This generated a table that displayed the retention time, mass-to-charge ratio (m/z),

219 and the intensity/peak area for each peak. The quality control step was performed to assess the  
220 stability of the intensities of peaks (“features”) between samples. This was performed using  
221 quality control samples, which were mixtures of equal amounts of experimental samples taken  
222 at each time point of the experiment. The relative standard deviation (RSD) of each feature in  
223 the quality control sample was compared to those in the experimental samples. Features with  
224 higher RSD in the quality control sample than in the experimental samples were excluded,  
225 while features with  $RSD < 30\%$  were retained for further analysis. Multivariable statistical  
226 analysis was performed on the retained features using principal component analysis (PCA) with  
227 95% confidence ellipse and partial least squares discrimination analysis (PLS-DA) to identify  
228 potential discriminations between the experimental samples. Heatmaps were generated to  
229 determine how the retained features changed at different time points during the experiment. A  
230 selection of discriminant ions and buckets was done based on the variable importance in  
231 projection (VIP) values. Features with VIP values greater than 1.0 were used for the  
232 identification step. MS/MS analysis was performed for the structural identification of  
233 compounds. The structure of each compound was deduced based on its adducts, isotopes, and  
234 MS/MS fragments using the SCIEX OS-Q software (AB Sciex). Information about  
235 compounds’ chemical structures, m/z, and retention times were subsequently uploaded to  
236 MetaboAnalyst 5.0 (<https://www.metaboanalyst.ca/>), which used this information to identify  
237 the compounds.

#### 238 **Section S4. IC operation**

239 Carboxylic acid concentrations were measured using a Dionex ICS-1100  
240 (ThermoFisher Scientific) system. Separation was achieved using a Dionex IonPac AS18 (4 ×  
241 250 mm) anion exchange column (Thermo Scientific) equipped with a Dionex IonPac AG18  
242 (4 × 50 mm) guard column (Thermo Scientific). 16 mM potassium hydroxide (Fisher, ≥85%)  
243 was used as the mobile phase at a flow rate of 1.0 mL/min for a 30 min run time. Each aliquot  
244 of solution was passed through a syringe filter before IC analysis.

#### 245 **Section S5. Possible enzymes and mechanisms associated with carboxylic acid** 246 **biodegradation by the two bacterial strains**



247 Table S4 summarized enzymes or metabolic pathways related to the biodegradation of  
248 carboxylic acids. Genes encoding formate dehydrogenases were identified in both genomes,  
249 which is consistent with the observed formate biodegradation. However, no known genes for  
250 oxalic acid biodegradation (Liu et al., 2021) were found in the genomes of both strains, which  
251 suggested the presence of yet to be characterized pathways that catalyzed the biodegradation.  
252 Interestingly, a protein with Cupin 2 domain was found in both genomes. The Cupin  
253 superfamily consists of a diverse range of enzymes including oxalate oxidase and oxalate  
254 decarboxylase that can biodegrade oxalic acid (Burrell et al., 2007).

255 Only the *E. hormaechei* B0910 strain was observed to biodegrade malonic acid.  
256 Interestingly, the malonyl-CoA-acyl carrier transcacylase observed in the *E. hormaechei*  
257 pf0910 strain seems to be a fusion protein, which may render it ineffective in utilizing malonic  
258 acid. Although no gene encoding maleate isomerase was identified in the genomes of both  
259 strains, the maleic acid biodegradation observed can be attributed to the activity of other  
260 enzymes with broad substrates specificity (Hatakeyama et al., 2000). The genes encoding for  
261 the small and large protein subunits that together form the 3-isopropylmalate dehydratase, the  
262 enzyme that isomerizes 2-isopropylmalate to 3-isopropylmalate, were found in both the  
263 *Enterobacter* strains. The small and large protein subunits of this enzyme are homologous to  
264 the small (51% amino acid identity) and large (59% amino acid identity) protein subunit  
265 constituents of maleate hydratase (HbzIJ) from *Pseudomonas alcaligenes* NCIMB 9867 that  
266 converts maleate to D-malate (Liu et al., 2015). Given the high protein homology, we speculate  
267 that the 3-isopropylmalate dehydratase in the *Enterobacter* strains may have a broader substrate  
268 specificity than known and it may be able to biodegrade maleate.

269 The lack of biodegradation of acetic acid, MSA, and glutaric acid in the experiments  
270 could be partly explained by the genomic information. Both strains have genes that encode  
271 enzymes involved in the biodegradation (Table S4) and associated uptake transporters (i.e.,  
272 acetate permease (ActP) and succinate-acetate/proton symporter (SatP)) of acetic acid. The  
273 lack of the corresponding biodegradation in the experiments could be due to the low uptake of  
274 acetic acid by cells as ActP functions to scavenge low concentrations of the compound  
275 (Gimenez et al., 2003) while SatP could be inhibited by formic acid found in the cloud water

276 medium (Sá-Pessoa et al., 2013). Genes encoding the two-component alkanesulfonate  
 277 monooxygenase for MSA biodegradation were found in both strains, but they were likely not  
 278 expressed as sulfur was not deficient in the cloud water medium (Kahnert et al., 2000; Eichhorn  
 279 and Leisinger, 2001), which is consistent with the absence of MSA biodegradation in the  
 280 experiments. While genes encoding succinate-semialdehyde dehydrogenase/glutarate-  
 281 semialdehyde dehydrogenase, which display a reversible conversion between glutarate-  
 282 semialdehyde and glutarate in the KEGG database (Kanehisa et al., 2022), were found in both  
 283 strains, to our knowledge there is no report of experimental results confirming that the reaction  
 284 can go in the reverse direction from glutarate to glutarate-semialdehyde. In addition, a study of  
 285 glutaric semialdehyde dehydrogenase reported the irreversible nature of the catalysis of  
 286 glutarate semialdehyde to glutarate (Ichihara and Ichihara, 1961). Thus, it is not surprising that  
 287 glutarate biodegradation was not observed for the two strains.

## 288 **Section S6. Estimation of biodegradation and chemical reaction rates ( $M s^{-1}$ ) in cloud** 289 **water**

### 290 **S6.1. Biodegradation**

291 The decay in the concentration of a specific carboxylic acid as a function of time during  
 292 a biodegradation experiment can be described by the following equation:

$$293 \frac{d[Acid]}{dt} = k'_{cell} \times [Acid] = k_{cell,acid} \times [cell]_{experiment} \times [Acid]_{experiment}$$

294 where  $k'_{cell}$  ( $s^{-1}$ ) is the pseudo first order rate constant obtained from fitting the decay of the  
 295 carboxylic acid, and  $[Acid]_{experiment}$  ( $mol L^{-1}$ ) is the initial concentration of the carboxylic  
 296 acid used in the biodegradation experiment.  $k'_{cell}$  is the product of the concentration of bacteria  
 297 cells used in the experiment ( $[cell]_{experiment}$ ,  $cell L^{-1}$ ) and the biodegradation rate constant  
 298 ( $k_{cell,acid}$ ,  $L cell^{-1} s^{-1}$ ).

299 The loss rate of the carboxylic acid in cloud water resulting from biodegradation is:

$$300 \frac{d[Acid]_{cloud}}{dt} = k_{cell,acid} \times [cell]_{cloud} \times [Acid]_{cloud}$$

301 where  $[cell]_{cloud}$  ( $cell L^{-1}$ ) is the concentration of bacteria cells present in cloud water  
302 ( $cell L^{-1}$ ), and  $[Acid]_{cloud}$  ( $mol L^{-1}$ ) is the concentration of the carboxylic in cloud water.

## 303 S6.2. Chemical reactions

304 The loss rates of the carboxylic acid in cloud water resulting from reactions with  $\cdot OH$   
305 and  $NO_3\cdot$  are:

$$306 \frac{d[Acid]_{cloud}}{dt} = k_{OH,acid} \times [\cdot OH]_{cloud} \times [Acid]_{cloud}$$

$$307 \frac{d[Acid]_{cloud}}{dt} = k_{NO_3,acid} \times [NO_3\cdot]_{cloud} \times [Acid]_{cloud}$$

308 where  $k_{OH,acid}$  ( $L mol^{-1}s^{-1}$ ) and  $k_{NO_3,acid}$  ( $L mol^{-1}s^{-1}$ ) are the rate constants for the  
309 reactions of the carboxylic acid with  $\cdot OH$  and  $NO_3\cdot$ , respectively, and  $[\cdot OH]_{cloud}$  ( $mol L^{-1}$ )  
310 and  $[NO_3\cdot]_{cloud}$  ( $mol L^{-1}$ ) are the concentrations of  $\cdot OH$  and  $NO_3\cdot$  in cloud water,  
311 respectively.

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