

## **SUPPLEMENTARY MATERIAL**

### **H<sub>2</sub>O<sub>2</sub> modulates the energetic metabolism of the cloud microbiome**

N. Wirgot<sup>1</sup>, V. Vinatier<sup>1</sup>, L. Deguillaume<sup>2</sup>, M. Sancelme<sup>1</sup>, and A.-M. Delort<sup>1</sup>

<sup>1</sup>Université Clermont Auvergne, CNRS, Sigma-Clermont, Institut de Chimie de Clermont-Ferrand, 63000 Clermont-Ferrand, France

<sup>2</sup>Université Clermont Auvergne, CNRS, Laboratoire de Météorologie Physique, 63000 Clermont-Ferrand, France

Correspondence to: A.-M. Delort (A-marie.delort@uca.fr)

Table SM1: Summary of values extracted from 37 cloud samples collected at the puy de Dôme station used for principal component analysis (PCA). The values in grey were used to perform the Spearman's rank correlation test.

Date	Cloud N°	Origin	Composition	ATP	Bacteria	Fungi	H <sub>2</sub> O <sub>2</sub>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>	Acetate	Formate	Oxalate	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	Mg <sup>2+</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	T	pH	
				pmol ml <sup>-2</sup>	cell ml <sup>-1</sup>	cell ml <sup>-1</sup>	µmol L <sup>-1</sup>													°C	
17/11/04	31	North/West	Marine	0.36	1.58 10 <sup>5</sup>	4.84 10 <sup>5</sup>	9.90	17.51	48.31	24.70	6.14	9.53	1.33	52.95	46.92	8.80	3.63	15.07	-0.20	5.10	
16/12/04	32	South/West	Continental	0.27	6.84 10 <sup>4</sup>	3.52 10 <sup>5</sup>	1.20	11.86	6.56	73.03	0.31	1.88	0.31	107.64	24.36	7.94	3.30	53.12	-0.10	6.70	
19/01/05	33	North/West	Highly marine	0.35	7.61 10 <sup>4</sup>	1.33 10 <sup>5</sup>	2.00	31.79	24.22	393.98	5.11	4.44	0.81	329.00	49.24	47.92	12.52	54.70	-3.40	6.70	
18/04/05	34	South/West	Marine	0.04	7.68 10 <sup>4</sup>	2.99 10 <sup>5</sup>	5.80	7.98	9.91	10.07	8.42	7.16	1.24	8.29	28.71	0.40	3.78	11.99	3.10	6.10	
06/06/05	36	West	Marine	0.29	9.99 10 <sup>4</sup>	1.55 10 <sup>5</sup>	17.03	25.20	32.34	9.50	7.37	9.33	3.76	21.13	75.54	1.43	1.19	4.45	8.20	5.70	
28/09/05	37	North/West	Highly marine	0.96	1.04 10 <sup>5</sup>	1.89 10 <sup>5</sup>	16.64	37.57	102.30	113.61	20.94	28.71	2.66	214.94	125.76	15.47	5.23	35.93	5.30	5.90	
03/10/05	38	North/East	Continental	0.80	5.42 10 <sup>4</sup>	8.34 10 <sup>5</sup>	10.35	64.81	144.99	30.36	6.71	8.88	2.50	49.89	138.62	3.70	3.01	14.14	2.40	4.30	
13/12/07	42	North/East	Continental	0.06	NA	NA	1.20	39.72	198.40	20.24	10.24	5.79	2.93	19.14	148.17	3.54	11.86	58.00	-6.10	4.70	
29/02/08	44	West	Continental	0.30	NA	NA	18.00	24.55	65.92	17.22	26.80	17.99	1.28	15.16	148.54	1.80	5.75	22.29	1.50	5.20	
21/04/08	45	South/West	Continental	0.26	4.50 10 <sup>4</sup>	1.10 10 <sup>5</sup>	11.40	44.69	65.52	31.68	17.73	17.92	2.78	33.32	122.45	4.89	9.66	53.62	2.80	4.60	
24/02/09	52	North	Continental	0.02	5.80 10 <sup>4</sup>	8.40 10 <sup>5</sup>	1.80	28.55	148.59	37.78	4.33	6.45	1.13	35.73	145.14	17.12	16.33	12.98	-0.10	4.10	
30/03/09	53	North/East	Polluted	0.01	6.80 10 <sup>4</sup>	1.60 10 <sup>5</sup>	2.70	73.80	516.51	193.83	41.22	13.67	3.45	171.86	363.23	13.51	71.57	52.28	1.00	3.98	
03/11/09	54	West	Marine	0.19	2.20 10 <sup>4</sup>	9.20 10 <sup>5</sup>	0.90	2.34	13.33	30.47	4.40	14.95	1.68	36.99	6.64	12.09	20.51	NA	7.90	5.16	
23/11/09	55	West	Marine	0.15	3.80 10 <sup>4</sup>	2.80 10 <sup>5</sup>	3.90	9.29	34.81	97.51	6.66	10.14	3.25	95.12	31.07	12.60	17.22	NA	5.60	5.84	
01/06/10	62	West	Marine	0.50	2.80 10 <sup>4</sup>	9.02 10 <sup>5</sup>	3.60	3.49	4.48	2.25	3.44	6.19	1.44	1.82	5.99	NA	NA	NA	NA	6.11	
08/06/10	63	South	Continental	2.37	8.07 10 <sup>4</sup>	2.55 10 <sup>5</sup>	33.40	51.42	74.53	7.25	22.89	42.04	9.50	8.35	210.10	NA	NA	NA	13.60	5.38	
16/06/10	64	North/East	Continental	0.99	4.42 10 <sup>4</sup>	2.24 10 <sup>5</sup>	22.30	46.10	70.60	7.70	25.40	42.70	9.70	10.10	100.30	2.10	1.50	3.80	9.60	6.17	
18/06/10	65	North/East	Continental	2.13	8.94 10 <sup>4</sup>	3.00 10 <sup>5</sup>	57.70	64.00	228.70	11.30	23.20	33.20	9.30	8.80	122.30	2.70	2.20	3.80	NA	3.90	
16/09/10	66	West	Marine	0.42	2.71 10 <sup>4</sup>	4.53 10 <sup>5</sup>	16.90	3.97	17.80	1.49	7.10	8.60	2.60	2.30	32.42	0.24	0.65	0.50	11.60	5.71	
19/01/11	68	North	Continental	0.24	9.15 10 <sup>5</sup>	2.01 10 <sup>5</sup>	1.40	42.01	288.09	80.07	30.26	6.10	1.08	180.86	134.44	3.16	65.27	223.08	-3.00	7.06	
02/02/11	69	West	Marine	0.05	5.41 10 <sup>5</sup>	1.12 10 <sup>5</sup>	4.10	4.47	48.80	11.08	0.24	2.65	0.22	34.32	4.24	0.48	7.64	146.72	NA	5.79	
24/02/11	70	West	Marine	0.23	8.37 10 <sup>5</sup>	4.83 10 <sup>5</sup>	0.80	1.28	3.31	8.18	8.47	2.17	12.93	12.46	6.34	0.12	4.80	28.20	5.20	5.30	
28/03/11	71	South/West	Marine	1.15	8.02 10 <sup>5</sup>	4.52 10 <sup>5</sup>	5.00	3.48	6.57	6.22	7.05	4.16	2.23	6.92	42.51	0.07	3.60	5.05	5.50	5.93	
31/03/11	72	West	Marine	1.11	7.95 10 <sup>5</sup>	4.52 10 <sup>5</sup>	20.80	12.67	25.97	26.39	16.55	12.74	1.82	40.21	75.12	0.36	12.51	15.24	9.00	6.96	

Table SM1(end): Summary of values extracted from 37 cloud samples collected at the puy de Dôme station used for principal component analysis (PCA). The values in grey were used to perform the Spearman's rank correlation test.

Date	Cloud N°	Origin	Composition	ATP	Bacteria	Fungi	H <sub>2</sub> O <sub>2</sub>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>	Acetate	Formate	Oxalate	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	Mg <sup>2+</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	T	pH	
				pmol ml <sup>-2</sup>	cell ml <sup>-1</sup>	cell ml <sup>-1</sup>	μmol L <sup>-1</sup>													°C	
20/07/11	78	North/West	Marine	1.28	1.24 10 <sup>4</sup>	6.59 10 <sup>2</sup>	7.43	7.08	10.43	24.09	3.31	11.29	3.05	22.72	32.68	12.29	9.15	7.10	8.30	5.50	
07/11/11	79	South/East	Marine	0.62	1.08 10 <sup>4</sup>	6.00 10 <sup>2</sup>	0.91	1.31	5.01	0.30	4.29	5.60	1.03	0.64	52.65	0.82	1.55	3.23	7.00	4.57	
20/01/12	80	North/West	Marine	1.03	1.77 10 <sup>4</sup>	8.01 10 <sup>2</sup>	5.86	1.78	2.15	13.35	7.44	5.30	1.11	80.56	39.25	12.02	6.13	5.22	-0.40	4.90	
23/01/12	81	North/West	Marine	2.29	6.00 10 <sup>4</sup>	8.20 10 <sup>2</sup>	3.38	4.84	11.18	23.38	15.26	15.61	6.69	145.72	188.64	20.26	6.20	9.34	-1.20	5.82	
19/03/12	82	North	Marine	0.49	1.65 10 <sup>3</sup>	1.71 10 <sup>2</sup>	3.66	1.63	1.96	0.16	4.23	6.16	2.97	1.02	75.44	0.95	0.06	6.11	-1.50	5.25	
04/04/12	83	North/West	Continental	5.62	1.49 10 <sup>4</sup>	1.32 10 <sup>2</sup>	10.42	10.68	49.93	12.16	NA	38.98	6.47	93.54	531.13	14.26	10.71	34.63	-0.40	5.60	
18/04/12	84	West	Marine	2.06	3.90 10 <sup>3</sup>	1.14 10 <sup>2</sup>	4.65	1.64	1.87	5.95	3.91	7.66	1.23	36.03	38.10	6.00	1.44	5.20	0.20	5.47	
25/06/12	85	West	Marine	2.07	4.47 10 <sup>3</sup>	1.09 10 <sup>2</sup>	31.76	1.15	3.02	0.93	NA	18.23	3.32	5.99	77.85	2.22	4.02	4.33	NA	5.52	
13/09/12	86	West	Marine	0.64	5.20 10 <sup>3</sup>	1.96 10 <sup>2</sup>	5.99	0.49	1.01	1.03	3.23	3.24	1.04	8.81	16.82	1.35	5.53	4.51	7.20	5.89	
10/10/12	87	South/West	Marine	1.55	1.97 10 <sup>4</sup>	1.97 10 <sup>2</sup>	1.88	5.33	10.95	26.73	4.00	13.58	0.74	21.22	39.31	8.33	7.57	2.34	NA	6.22	
28/11/12	88	North	Polluted	1.71	6.50 10 <sup>3</sup>	6.50 10 <sup>2</sup>	1.24	111.09	346.50	47.55	18.46	13.08	6.45	17.64	59.26	15.84	53.58	6.55	-2.70	4.55	
22/05/13	90	North	Marine	1.52	1.16 10 <sup>4</sup>	8.30 10 <sup>2</sup>	30.82	43.39	43.16	9.58	11.94	14.86	3.19	6.03	90.83	5.16	2.40	14.06	2.40	4.90	
29/05/13	91	North/West	Marine	2.06	6.64 10 <sup>3</sup>	1.57 10 <sup>2</sup>	5.25	28.51	9.33	16.25	21.83	9.76	3.33	13.27	57.44	6.22	8.01	22.21	0.00	5.48	

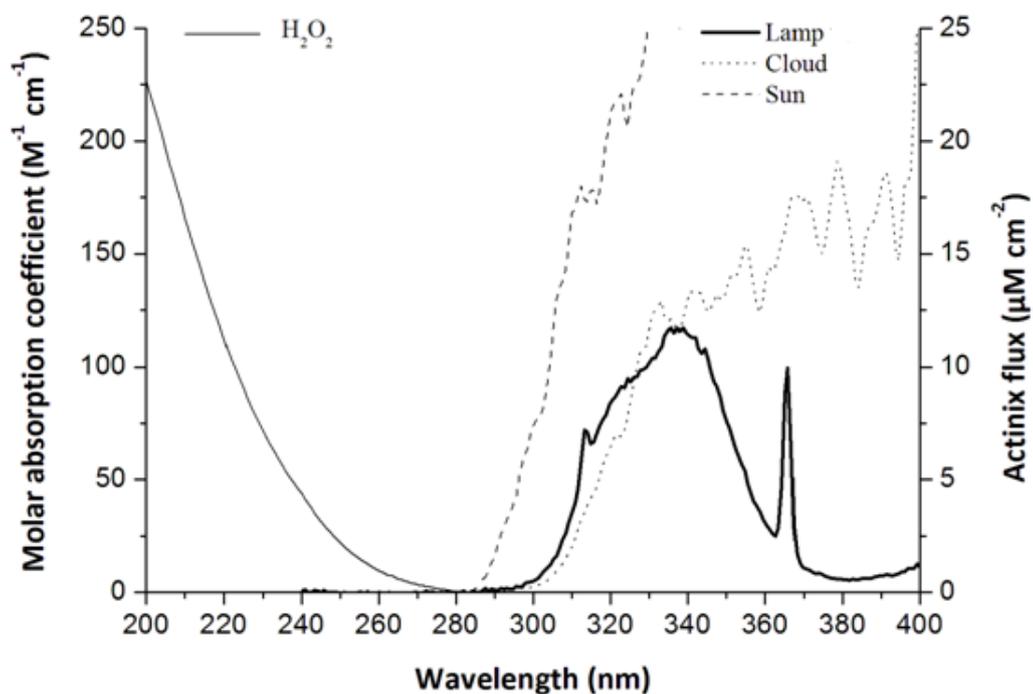


Figure SM1: Light spectrum of the lamp (Sylvania Reptistar) used for experiments compared to spectra of solar light measured on a sunny day and inside a cloud (measured on October 16, 2013 at 3:57-3:58 PM). Solar light and solar light inside a cloud were measured on the top of the puy de Dôme. H<sub>2</sub>O<sub>2</sub> absorbance spectrum.

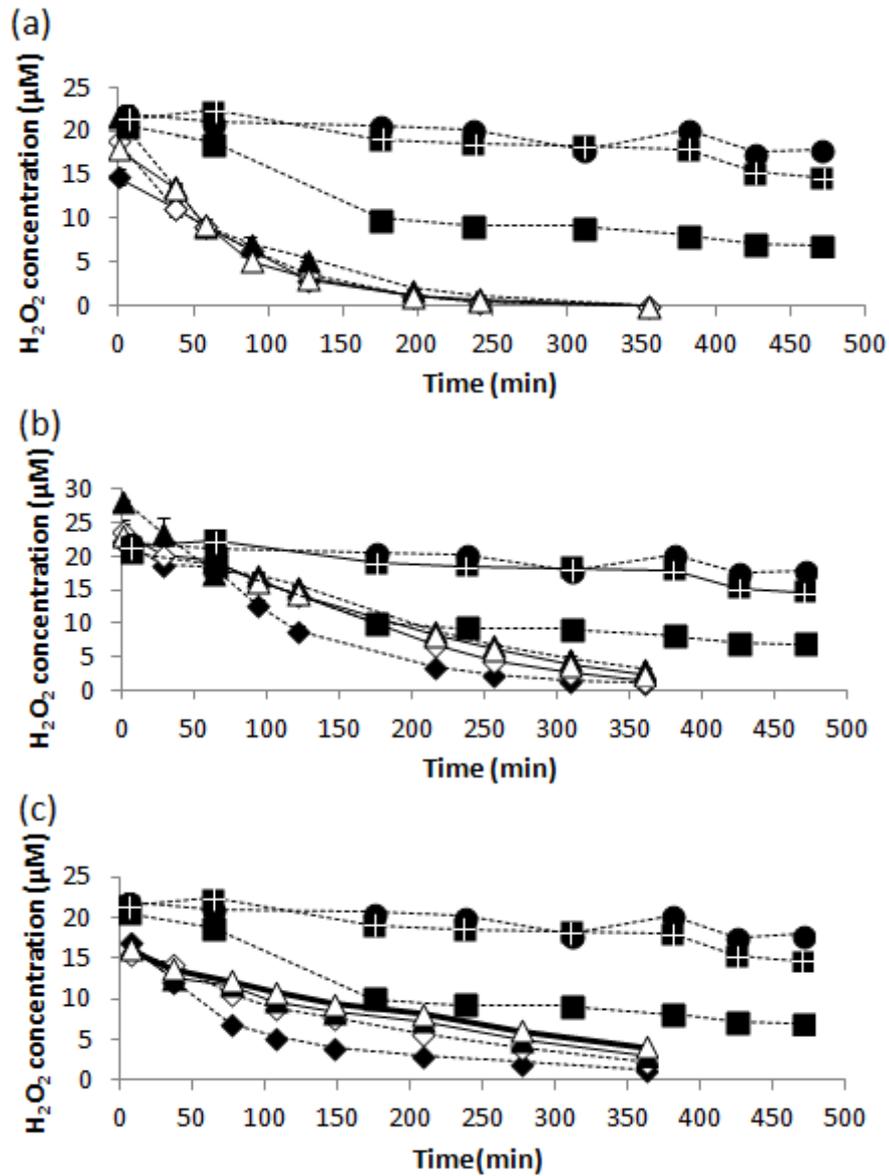


Figure SM2: Initial degradation of H<sub>2</sub>O<sub>2</sub> (µM) as a function of time (min) summarizing abiotic conditions: Light + Fe-[EDDS] (black square), Light (black circle), Fe-[EDDS] (black square with white cross) and biotic conditions: Light + Fe-[EDDS] + Bacteria<sup>(a,b,c)</sup> (black diamond), Fe-[EDDS] + Bacteria<sup>(a,b,c)</sup> (white diamond), Light + Bacteria<sup>(a,b,c)</sup> (black triangle), Bacteria<sup>(a,b,c)</sup> (white triangle). (a) *Pseudomonas graminis* 13b-3 (b) *Pseudomonas syringae* 13b-2 (c) *Sphingomonas* sp. 14b-5. Three replicates were done. Error bars (very low values) represent the standard errors (SEs) of the enzymatic assay (5%).

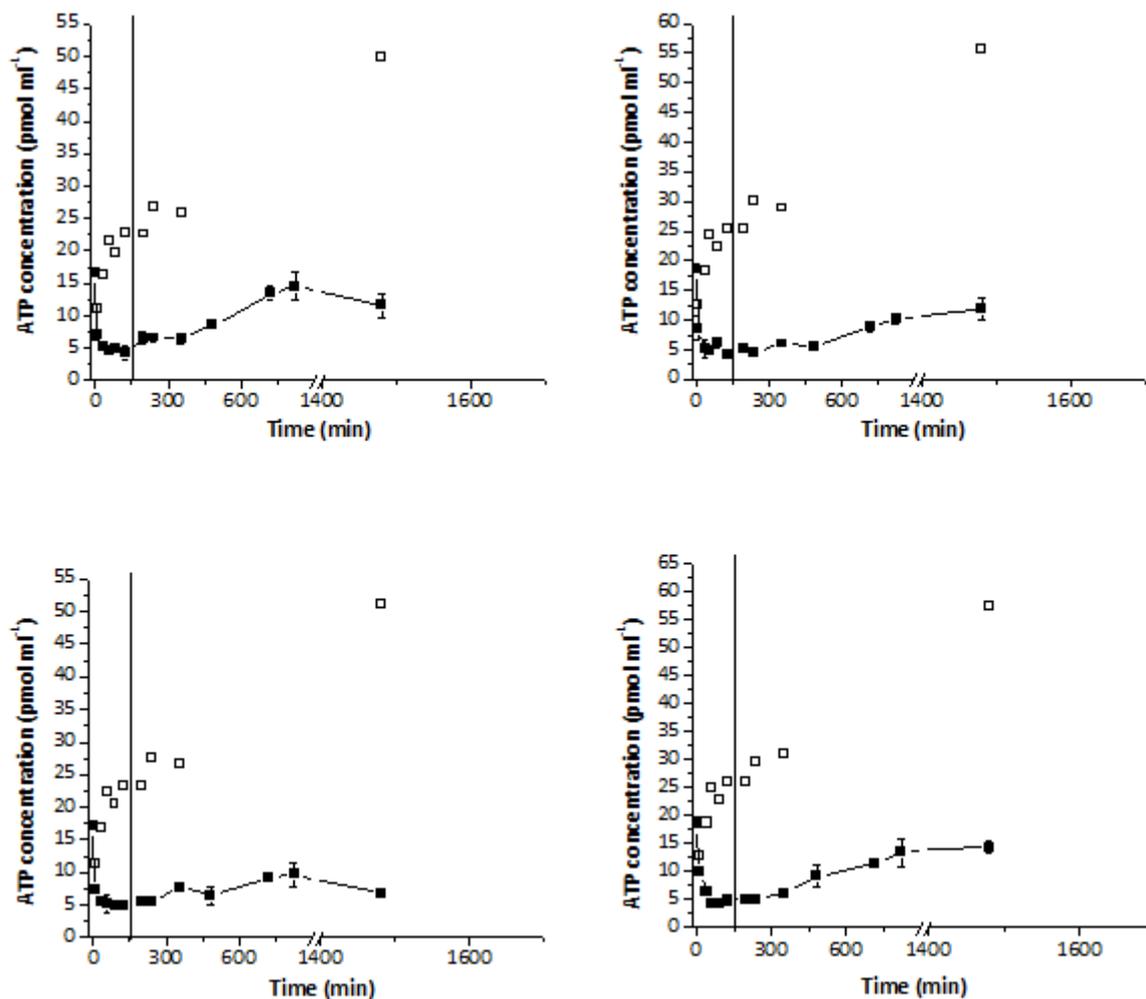


Figure SM3: ATP concentration ( $\mu\text{M}$ ) for the *Pseudomonas graminis* (13b-3) strain as a function of time (min) in the absence (white square) and the presence (black square) of  $\text{H}_2\text{O}_2$  at  $20 \mu\text{M}$ : (a) Cloud + Bacteria +  $\text{H}_2\text{O}_2$ ; (b) Cloud + Bacteria +  $\text{H}_2\text{O}_2$  + Light; (c) Cloud + Bacteria +  $\text{H}_2\text{O}_2$  + Fe-[EDDS]; (d) Cloud + Bacteria +  $\text{H}_2\text{O}_2$  + Fe-[EDDS] + Light.

The vertical bar illustrates the time corresponding to the total degradation of  $\text{H}_2\text{O}_2$ .

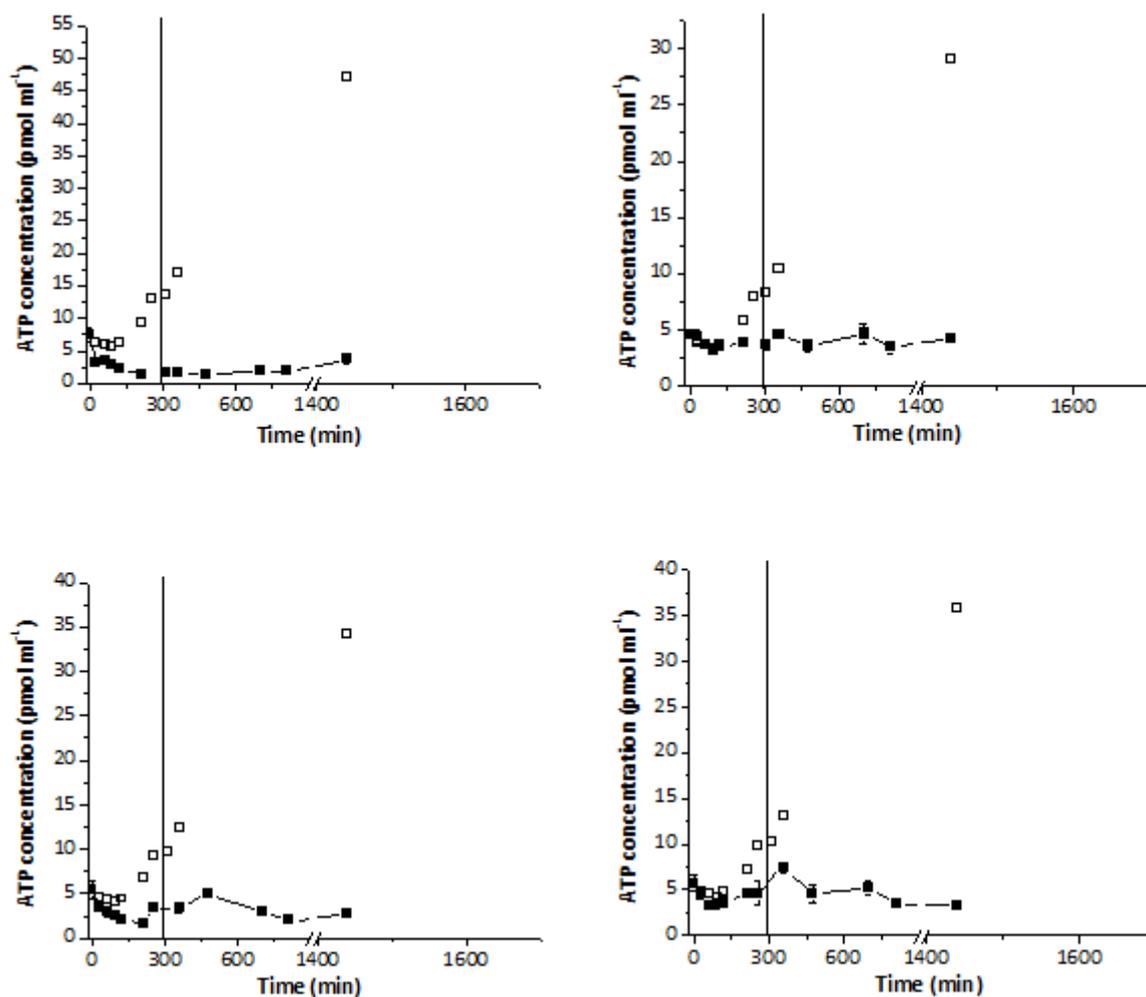


Figure SM4: ATP concentration ( $\mu\text{M}$ ) for the *Pseudomonas syringae* (13b-2) strain as a function of time (min) in the absence (white square) and the presence (black square) of  $\text{H}_2\text{O}_2$  at  $20 \mu\text{M}$ : (a) Cloud + Bacteria +  $\text{H}_2\text{O}_2$ ; (b) Cloud + Bacteria +  $\text{H}_2\text{O}_2$  + Light; (c) Cloud + Bacteria +  $\text{H}_2\text{O}_2$  + Fe-[EDDS]; (d) Cloud + Bacteria +  $\text{H}_2\text{O}_2$  + Fe-[EDDS] + Light. The vertical bar illustrates the time corresponding to the total degradation of  $\text{H}_2\text{O}_2$ .

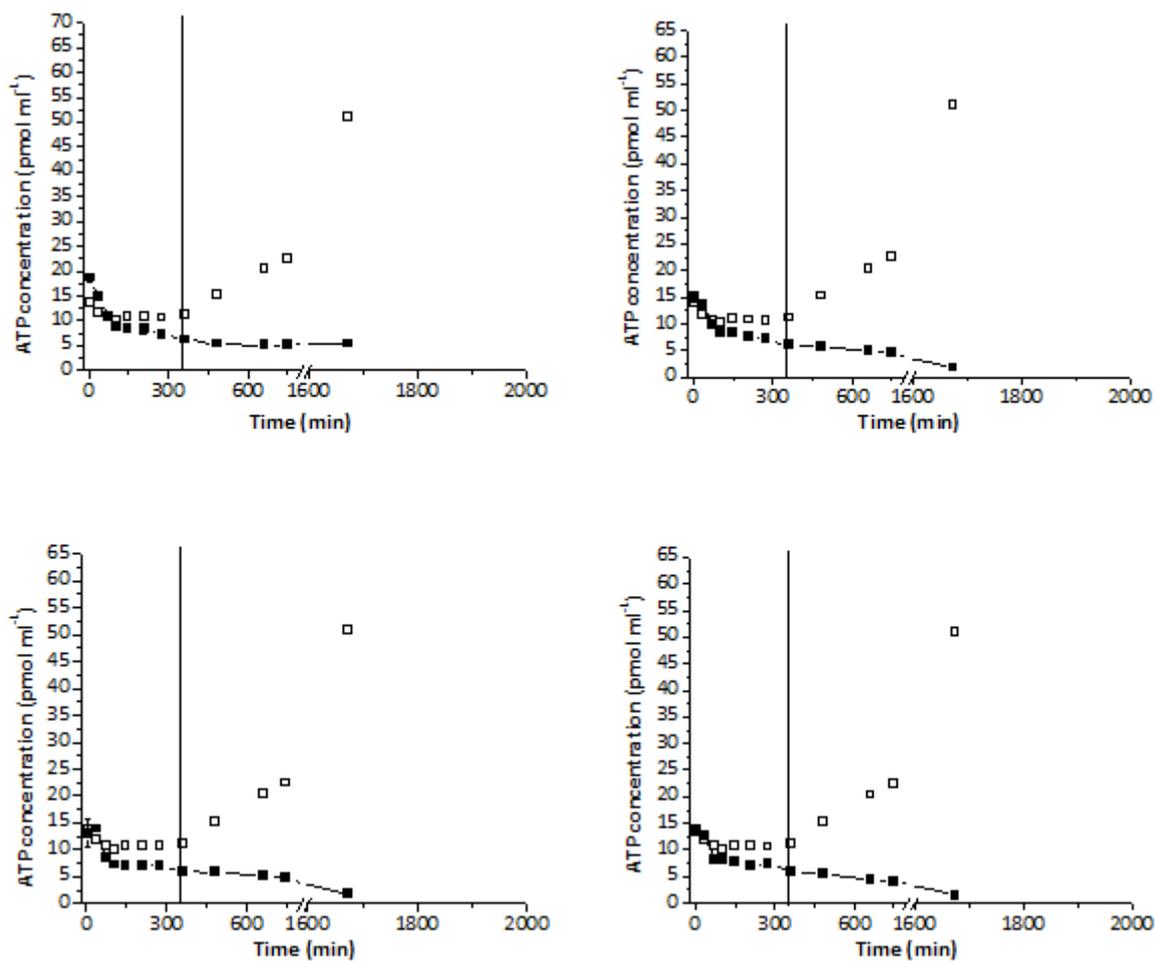


Figure SM5: ATP concentration ( $\mu\text{M}$ ) for the *Sphingomonas sp.* (14b-5) strain as a function of time (min) in the absence (white square) and the presence (black square) of  $\text{H}_2\text{O}_2$  at  $20 \mu\text{M}$ : (a) Cloud + Bacteria +  $\text{H}_2\text{O}_2$ ; (b) Cloud + Bacteria +  $\text{H}_2\text{O}_2$  + Light; (c) Cloud + Bacteria +  $\text{H}_2\text{O}_2$  + Fe-[EDDS]; (d) Cloud + Bacteria +  $\text{H}_2\text{O}_2$  + Fe-[EDDS] + Light. The vertical bar illustrates the time corresponding to the total degradation of  $\text{H}_2\text{O}_2$ .