

### **Measure accuracy and error of pH and EC:**

The measure accuracy and error of the pH/conductivity analyzer were shown in Table S1. Table S2 showed the pH values and pH differences in all sets of samples. From the tables, 96% of the pH differences were far larger than the measure error, which meant the pH differences in the three stages were not caused by our measurement error. EC was the same situation as pH values (Table S3)

### **Formula for calculating volume-weighted mean concentration:**

$$\bar{C}_i = \frac{\sum_{j=1}^n C_{ji} \times V_j}{\sum_{j=1}^n V_j}$$

Where  $\bar{C}_i$  is the Volume Weighted Mean concentration of the i-th ion;

$C_{ji}$  is the concentration of the i-th ion in the j-th sample;

$V_j$  is the volume of the j-th sample;

n is the number of all samples.

**Table S1:** Measure accuracy and error of pH and EC

Item	Application Range	Accuracy	Error
pH	-2.00 to 16.00 pH	0.01 pH	$\pm 0.02$ pH
EC	0 to 99.99 $\mu\text{S cm}^{-1}$	0.01 $\mu\text{S cm}^{-1}$	$\pm 0.5$ $\mu\text{S cm}^{-1}$
	0 to 999.9 $\mu\text{S cm}^{-1}$	0.1 $\mu\text{S cm}^{-1}$	$\pm 5$ $\mu\text{S cm}^{-1}$

**Table S2:** pH values and pH differences in each set of samples

pH	Stage 1	Stage 2	Stage 3	DB12	DB23	DB31
E-1.1	5.76	5.50	5.64	0.26	-0.14	-0.12
E-1.2	5.94	5.61	5.07	0.33	0.54	-0.87
E-2.1	5.63	5.42	5.58	0.21	-0.16	-0.05
E-2.2	5.67	5.43	5.36	0.24	0.07	-0.31
E-2.3	5.67	6.01	6.64	-0.34	-0.63	0.97
E-2.4	6.27	5.92	6.08	0.35	-0.16	-0.19
E-3.1	5.69	5.70	5.80	<b>-0.01</b>	-0.10	0.11
E-3.2	6.07	5.80	5.69	0.27	0.11	-0.38
E-3.3	6.23	5.80	5.75	0.43	0.05	-0.48
E-3.4	6.16	6.09	5.96	0.07	0.13	-0.20
E-4	5.37	5.51	5.48	-0.14	0.03	0.11
E-5	3.75	3.70	3.60	0.05	0.10	-0.15
E-6	5.68	5.48	5.23	0.20	0.25	-0.45
E-7	5.86	5.78	6.08	0.08	-0.30	0.22
E-8	5.39	5.38	5.02	<b>0.01</b>	0.36	-0.37
E-9	5.98	5.78	5.70	0.20	0.08	-0.28
E-10	4.62	4.46	4.22	0.16	0.24	-0.40

DB12: difference between stage1 and stage 2; DB23: difference between stage2 and stage 3;

DB31: difference between stage3 and stage 1; **Red** means the difference smaller than the error.

**Table S3:** EC values and EC differences in each set of samples

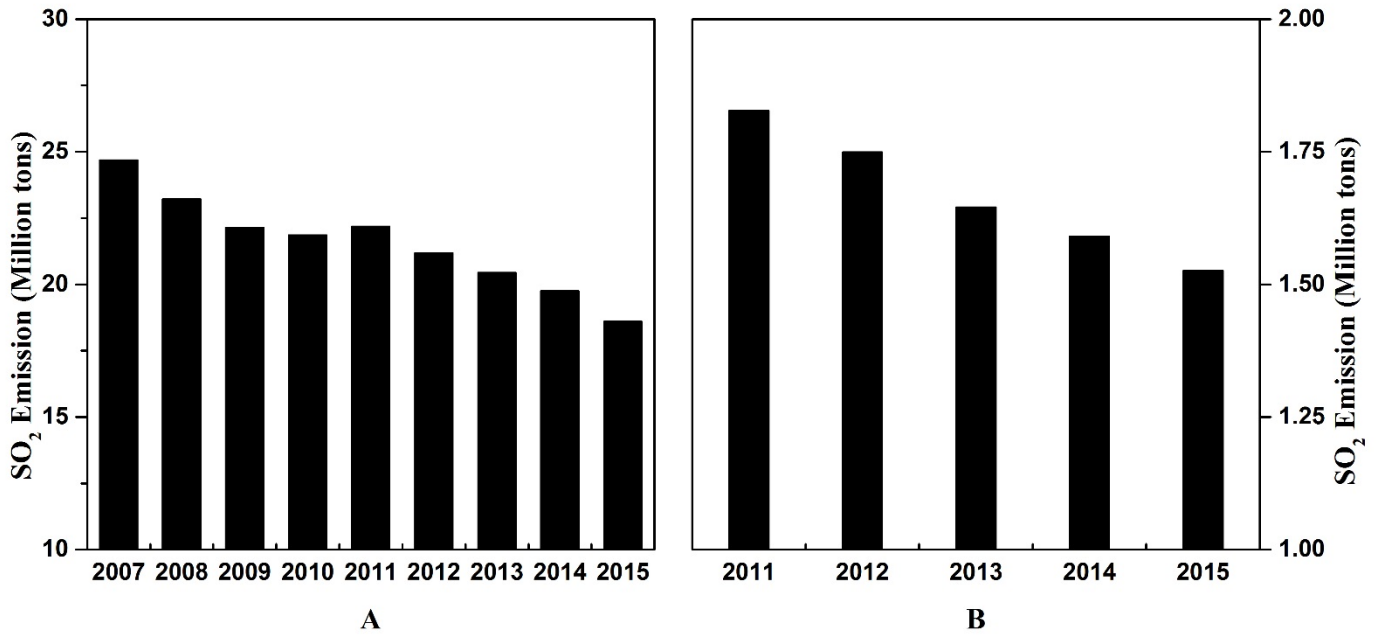
EC/ ( $\mu\text{S cm}^{-1}$ )	Stage 1	Stage 2	Stage 3	DB12	DB23	DB31
E-1.1	95.00	121.9	42.00	-26.9	79.9	-53.00
E-1.2	152.6	144.4	89.90	8.2	54.5	-62.7
E-2.1	91.76	99.78	70.38	-8.02	29.40	-21.38
E-2.2	94.64	94.83	141.3	<b>-0.19</b>	-46.5	46.7
E-2.3	74.11	142.0	195.0	-67.9	-53.0	120.9
E-2.4	71.95	45.11	52.91	26.84	-7.80	-19.04
E-3.1	106.1	94.58	80.28	11.5	14.30	-25.8
E-3.2	96.85	73.65	105.0	23.20	-31.4	8.2
E-3.3	109.9	70.33	139.4	39.6	-69.1	29.5
E-3.4	180.5	160.8	143.8	19.7	17.0	-36.7
E-4	190.5	346.8	252.4	-156.3	94.4	61.9
E-5	162.4	238.8	155.9	-76.4	82.9	-6.5
E-6	363.8	367.9	378.8	<b>-4.1</b>	-10.9	15.0
E-7	249.5	264.3	130.7	-14.8	133.6	-118.8
E-8	501.3	496.2	546.5	5.1	-50.3	45.2
E-9	119.1	99.19	77.18	19.9	22.01	-41.9
E-10	30.21	28.70	25.85	1.51	2.85	-4.36

DB12: difference between stage1 and stage 2; DB23: difference between stage2 and stage 3;

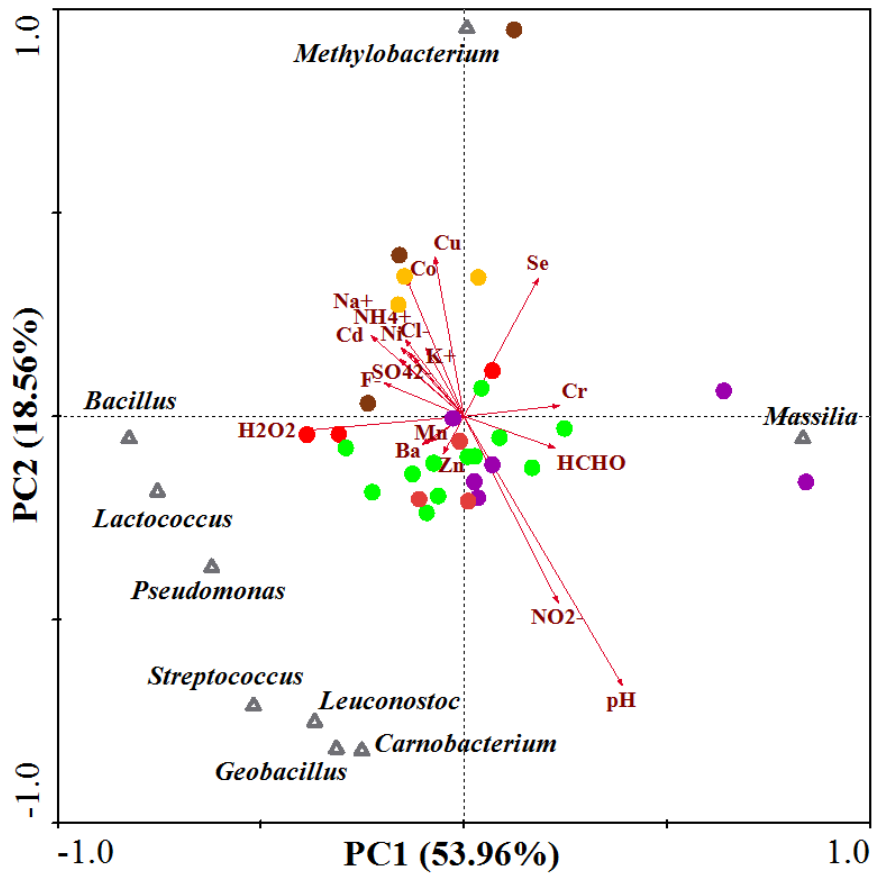
DB31: difference between stage3 and stage 1; **Red** means the difference smaller than the error.

**Table S4:** Bacterial phyla comparison between different kinds of samples.

Sample	Location	Bacterial Phyla						Reference	
		Firmicutes	Proteobacteria	Bacteroidetes	Actinobacteria	Fusobacteria	Cyanobacteria		Verrucomicrobia
Cloud water	Mt. Tai, Chian	80.50%	17.40%	1.20%	0.46%	0.23%			This study
Lake water	Chaiwopu Lake, China		58.1%	15%	3.2%		17.2%	4.3%	(Zeng et al., 2014)
Rain water	Seoul National University, Republic of Korea	23.48%	22.35%	41.29%			0.19%		(Cho and Jang, 2014)
River water	Ohio River, North America	57%		14%	20%				(Kovatch and Schultz, 2013)
Aerosol particle	Denver and Greeley, America	28.2%	34.6%	9.7%	22%				(Bowers et al., 2016)



**Figure S1:** A: SO<sub>2</sub> Emission in China from 2007 to 2015 (data from the Ministry of Environment Protection of the People's Republic of China); B: SO<sub>2</sub> Emission in Shandong Province from 2011 to 2015 (data from Ministry of Environment Protection of Shandong Province)



**Figure S2:** RDA analysis between chemical composition and bacterial community. The same color circles represented samples in the same cloud event. Red, samples in E-1.1; purple, samples in E-2.1 and E-2.3; green, samples in E-3; yellow, samples in E-6; dark red, samples in E-7; brown, samples in E-10.

## Reference

- Bowers, R.M., Clements, N., Emerson, J.B., Wiedinmyer, C., Hannigan, M.P., Fierer, N. (2016) Seasonal variability in bacterial and fungal diversity of the near-surface atmosphere. *Environ. Sci. Technol* 47, 12097-12106.
- Cho, B.C., Jang, G.I. (2014) Active and diverse rainwater bacteria collected at an inland site in spring and summer 2011. *Atmos. Environ.* 94, 409-416.
- Kovatch, J., Schultz, G.E. (2013) Bacterial diversity in a large, temperate, heavily modified river, as determined by pyrosequencing. *Aquat. Microb. Ecol.* 70, 196-179.
- Zeng, J., Deng, L.-J., Lou, K., Zhang, T., Yang, H.M., Shi, Y.W., Lin, Q. (2014) Molecular characterization of the planktonic microorganisms in water of two mountain brackish lakes. *J. Basic Microbiol.* 54, 509-520.