

Responds to the reviewer's comments:

We sincerely thank the reviewer for the valuable comments and suggestions concerning our manuscript entitled "Chemical composition and droplet size distribution of cloud at the summit of Mount Tai, China". These comments are all valuable and helpful for revising and improving our paper. The responses to reviewers are in blue. The changes are marked in red in the revised manuscript.

## Reviewer 1

### Comment 1:

1. Page 3, line 29: Any reference on the measurement of these organic acids using IC? Please provide it.

**Response:** We sincerely thank you for your pertinent comments and valuable suggestions. We cited the relevant references as in Page 4 Line 4.

### Comment 2:

2. Page 3, line 30-31: it's not clear to readers how OC and EC in cloud samples were measured using a sunset OC/EC analyzer. Detailed information is needed here.

**Response:** We added the detailed information about the measurement of OC and EC as in Page 4 Line 5- Line 9:

*"The concentrations of organic carbon (OC) and elemental carbon (EC) in cloud water were determined using a thermal-optical transmittance (TOT) carbon analyzer (Sunset Laboratory, Tigard, OR, USA). For each cloud sample, certain microliters were dropped on the surface of a small standard size punch (~1.5 cm<sup>2</sup>) from a pre-combusted quartz filter and analyzed based on the NIOSH protocol 870 TOT program (Khan et al., 2009; Xu et al., 2017)."*

### Comment 3:

3. The authors report organic acids in cloud samples from the summit of Mt. Tai. What's the main sources of the measured organic acids such as lactic and oxalic acids in cloud waters? Detailed discussion on such a point is needed.

**Response:** We have added the discussion on the sources of organic acids in cloud samples as in Page 6 Line 5- Line 16:

*"The VWM concentrations of acetate, lactate, formate and oxalate were 4.1, 3.0, 1.75 and 0.81 mg L<sup>-1</sup>, respectively, accounting for 7.01% of TDIC. Based on the sources or source strengths of formic acid and acetic acid, the formic-to-acetic acid ratio (F/A) cloud be used as an indicator to determine the sources of organic acids (Sun et al., 2016; Tan et al., 2010). Low ratio indicated the important role of direct emissions (such as biomass emission, combustion activities and automobile exhaust) whereas high ratio indicated the in situ photochemical generation of formic acid (Talbot et al., 1988; Tanner and Law, 2003). In the collected cloud samples, formic acid and acetic acid were highly correlated ( $r=0.758$ ,  $p \leq 0.01$ ). F/A was about 0.78 (lower than 1), figuring out direct emissions were important sources of organic acids*

(Kieber et al., 2002; Li et al., 2011). Oxalic acid was significantly correlated with formic acid ( $r=0.667$ ,  $p \leq 0.01$ ) and acetic acid ( $r=0.638$ ,  $p \leq 0.01$ ). This implied that formic acid, acetic acid and oxalic acid were probably emitted from the same sources and/or accumulated under similar physical conditions (Tanner and Law, 2003). No significant correlations were found between lactic acid and the other three carboxylic acids. No significant correlations were found between lactic acid and other water-soluble ions in the cloud samples. It implied that the emission source of lactic acid was different from formic, acetic and oxalic acids.”

#### Comment 4:

4. Are there any correlations between organic acids and water-soluble cations in the cloud samples?

**Response:** The correlation coefficients between organic acids and water-soluble ions were calculated as shown in Table R1. No significant correlations were found between lactate and water-soluble cations. From acetate to oxalate, the correlations between carboxylic acids and water-soluble cations gradually increased. Especially oxalate, it was strongly correlated with all measured cations. This indicated that although formate, acetate and oxalate mainly derived from direct emissions, no characteristic sources were found for these three carboxylic acids

Table R1. The correlation coefficients between four carboxylic acids and water-soluble ions in the cloud samples.

	lactate	acetate	formate	oxalate
lactate	1.000			
acetate	-0.292	1.000		
formate	-0.215	0.758**	1.000	
oxalate	-0.015	0.638**	0.667**	1.000
Cl <sup>-</sup>	-0.106	0.524*	0.617**	0.860**
NO <sub>3</sub> <sup>-</sup>	-0.027	0.533*	0.484*	0.901**
SO <sub>4</sub> <sup>2-</sup>	-0.066	0.554*	0.688**	0.959**
nss-SO <sub>4</sub> <sup>2-</sup>	-0.065	0.554*	0.687**	0.959**
Na <sup>+</sup>	-0.140	0.438	0.558*	0.673**
NH <sub>4</sub> <sup>+</sup>	-0.060	0.456	0.574*	0.898**
K <sup>+</sup>	-0.039	0.289	0.339	0.793**
Mg <sup>2+</sup>	-0.009	0.459	0.554*	0.896**
Ca <sup>2+</sup>	0.041	0.374	0.478	0.912**

\*\*  $p \leq 0.01$  \*  $p \leq 0.05$

#### Reference

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