Summary:

Using quantum chemical calculations combined with high level ab initio method; the authors studied the catalytic ability of the most common dicarboxylic acid in the atmosphere - oxalic acid - for the hydration reaction of SO_3. Further, taking the real atmosphere into consideration, they found that oxalic acid-catalyzed hydration reaction can compete with the water-catalyzed reaction in the upper troposphere, which has certain significance for the formation of H_2SO_4 in the atmosphere.

The work is performed with care and I believe it can be published after the following concerns are fully addressed.

Major comments:

1. In previous paper (Hazra et al, *J. Am. Chem. Soc*. **2011**, *133*, 17444), hydrolysis of SO_3 catalyzed by formic acid in the gas phase has been studied and the result shows a near barrierless mechanism for sulfuric acid formation. Moreover, we note that formic acid is considered to be the most abundant carboxylic acid, ubiquitous in the atmosphere (Millet et al. *Atmos. Chem. Phys.*, **2015**, *15*, 6283; Bannan et al. *J. Geophys. Res. - Atmos*, **2017**, *122*, 488). Thus, to make this story more interesting, I think the authors should highlight the specific characteristic of oxalic acid compared with formic acid, and add more discussion about the advantages of oxalic acid acting as a catalyst.

2. For method and basis set of the single point energy, the CCSD(T) method with the 6-311++G(3df,3pd) is not a good match, I think it is available using at least cc-pVTZ level. If the author prefer the 6-311++G(3df,3pd) basis set when using CCSD(T) to perform the single point energy, please give the corresponding reasons.

4.In Section 3.1: it is better to compare the results with previous studies to enrich the text.

Minor comments:

1. Page1, line 12 **(Abstract),** "can involve in" should be "can be involved in".

2. Page1, line 16 **(Abstract),** "the rate of SO_3 hydration" should be "the rates of SO_3 hydration".

3. Page 2, line 5: is it better to change the second "can" to "thus"?

4. Page 2, line 11: “firstly is” should be “is firstly”.

5. Page 2, line 11: “rearrange” should be “rearranges”.

6. Page 2, line 14: “takes” should be “take”

7. Page 2, line 17: “reduce” should be “reduces”

8. Page 3, line 15: “for” should be “of”

9. **Page 3, line 17**: “... the hydration of SO_3 in the second water ...” should be “... the hydration of SO_3 with the second water ...”.

10. Page 3, line 20: “reactant, complex, transition state” should be “reactants, complexes, transition states”

11. **Page 4, lines 9-10**: “..., the electronic energies based on the ..., while the partition functions obtained from ...” should be “..., the electronic energies were based on the ..., while the partition functions were obtained from ...”

12. **Page 4, line 14**: “..., it can conclude that the ...” should be “..., it can be concluded that the ...”

13. **Page 5**: “The corresponding rate constants are that” should be “The corresponding reaction rates are that”.

14. Page 6, line 10: “react” should be “reacts”

15. Page 6, line 11: “rearrange” should be “rearranges” and “begin” should be “begins”

16. Page 7, line 10: “Table S2-S5” should be “Tables S2-S5”

17. Page 10, line 23: is it better to change “real” to “potential”

18. Page 11, line 3: is it better to change “The energy barrier of hydration reaction of SO3 is about or below 1 kcal mol^{-1}” to “Other conformers can catalyze the hydration reaction and the corresponding energy barrier is a little higher or less than 1 kcal mol^{-1}”

19. Page 11, line 4: add “that” after “signify”

20. Page 11, line 5: add “from the view of barrier” after “H_2SO_4”

21. Page 22, the caption of Figure 4. Please add the unit of hydrogen bond length.