Comments by Referees are in blue.

Our replies are in black.

Changes to the manuscript are highlighted in red both in here and in the revised manuscript.

Reply to Ref #2

This paper uses two complementary techniques to explore the water uptake properties of commonly found Mg and Ca salts in mineral dust and sea salts that are relevant to atmospherically aged particles. The manuscript is thorough and very well written. I recommend this work for publication after minor revisions.

Reply: We would like to thank Ref #2 for his/her very positive review of our manuscript. These comments, which largely helped us improve our manuscript, have been adequately addressed in our revised manuscript, as detailed below.

Major Comments:

I have several major comments: 1. More discussion of salts found in freshly emitted and heterogeneously processed sea salt aerosols would have balanced out the intro and discussion in this paper.

Reply: In the revised manuscript we have added a few sentence (<u>line 78-83</u>) in the introduction section to further explain the relevance of our work for sea salt aerosol and saline mineral dust, as suggested.

In addition, we have added a new section (Section 3.3.2, line 666-700), entitled "Atmospheric implications", to further discuss atmospheric implications of our work.

2. The conclusions and implications section would have benefited from discussion of the implications for water uptake and CCN activation of freshly emitted and processed dusts and sea salts.

Reply: We fully agree with the referee. In the revised manuscript we have changed the title of Section 4 to "Summary and Conclusion", since this section is more like a summary of our work.

Furthermore, we have added a new section (Section 3.3.2, line 666-700), entitled "Atmospheric implication", to discuss the implications of our work for mineral dust, saline mineral dust and sea salt aerosols. Please refer to Section 3.3 in the revised manuscript for details.

3. The authors should create another table or a κ plot with their κ (GF) values and to report these values in the abstract. This will attract more attention to their work.

Reply: The following changes have been implemented in the revised manuscript:

- 1) We have created a new table (Table 5, Page 26) to compare κ_{gf} measured in this work with κ_{ccn} measured by previous studies, and relevant text in Section 3 has been updated accordingly.
- 2) We have also mentioned κ_{gf} results in the abstract (line 40-43): "The hygroscopic growth factors at 90% RH were found to range from 1.26±0.04 for Ca(HCOO)₂ and 1.79±0.03 for Ca(NO₃)₂, and the single hygroscopicity parameter ranged from 0.09-0.13 for Ca(CH₃COO)₂ to 0.49-0.56 for Ca(NO₃)₂."

Specific Comments:

Abstract

1. I recommend pointing out that your results also fit theoretical predictions from the Clausius-Clapeyron equation and to report $\kappa(GF)$ values obtained from this work. This is important for incorporating your results into models.

Reply: The following changes have been made in the revised manuscript:

- 1) In the abstract (line 34-35), we have included the following sentence to point out that our results fit theoretical predictions: "We further found that the dependence of DRH on temperature can be approximated by the Clausius-Clapeyron equation."
 - 2) We have also mentioned κ_{gf} results in the abstract (line 40-43), as suggested.

Introduction

1. Please also reference [Gaston et al., 2017] which explored the CCN activity of playa dusts.

Reply: In the revised manuscript (line 80-83), the work by Gaston et al. was cited to mention the role of CaCl₂ in saline dust particles: "Furthermore, the CCN activity of saline mineral dust was explored (Gaston et al., 2017), and good correlations were found between the CCN activities of saline mineral dust particles and the abundance of the soluble components (e.g., CaCl₂) they contained."

2. Lines 75-78: CaCl₂ would also be important for sea spray aerosol.

Reply: The following change has been made in the revised manuscript (line 76-80) to mention the importance of CaCl₂ in sea salt aerosol: "In addition, MgCl₂ and CaCl₂ are important components in sea salt (as known as sea spray) aerosol. The presence of MgCl₂ and CaCl₂, in addition to NaCl, can alter the hygroscopicity of sea salt aerosol (Gupta et al., 2015; Zieger et al., 2017); to be more specific..."

3. Lines 80-83: were these previous studies incomplete that the work warrants further investigation? How so?

Reply: As we pointed out in the original manuscript (80-83), hygroscopic growth of these aerosol particles was only investigated by one or two studies, and therefore further studies are warranted.

Methods

1. Were diameters corrected for shape factors particularly for dry particle diameters?

Reply: No shape factors were used in our work to correct the dry particle diameters. In Section 2.1 of the revised manuscript (line 141-150), we have added one paragraph to further explain the D-TMDA method. The reviewer is kindly referred to the revised manuscript for further details.

Results

1. Line 414: Gaston et al., 2017 also measured $\kappa(CCN)$ for CaCl₂ and for a MgCl₂ hydrate.

Reply: We would like to thank the ref #2 for bringing our attention to the results reported by Gaston et al. (2017). We were aware of this paper, but did not pay attention to the supplementary information in which κ_{ccn} was reported for CaCl₂ and MgCl₂. The following changes have been made in the revised manuscript:

- 1) We have created a table (Table 5, Page 26) to compare κ_{gf} measured in this work with κ_{ccn} measured by previous studies, and in this table the results reported by Gaston et al. (2017) were included.
- 2) We have rephrased our discussion on comparison between our and previous work, and further details can be found in Section 3.1.4 of the revised manuscript (line 482-497).
- 2. The authors are encouraged to create another table or a κ plot with their κ (GF) values and to report these values in the abstract. This will attract more attention to their work.

Reply: The following changes have been made in the revised manuscript, as suggested:

- 1) A table (Table 5, Page 26) has been created to compare κ_{gf} measured in this work with κ_{ccn} measured by previous studies;
 - 2) We have also mentioned κ_{gf} values in the abstract (line 40-43).

Conclusions:

1. The authors are encouraged to point out the broader implications of their work for the water uptake and cloud nucleating properties of fresh and processed dusts and sea salts.

Reply: This is a very good point. In the revised manuscript we have changed the title of Section 4 to "Summary and Conclusion", since this section is more like a summary of our work.

Furthermore, we have added a new section (Section 3.3.2, line 666-700), entitled "Atmospheric implication", to discuss atmospheric implications of our work for mineral dust, saline mineral dust and sea salt aerosol. Please refer to Section 3.3 in the revised manuscript for details.

2. The authors are encouraged to also point out the ability of the Clausius-Clapeyron equation to predict the temperature-dependent behavior of the water uptake properties of some of the salts.

Reply: As suggested, the following change has been made in the revised manuscript (<u>line 708-709</u>): "...both showing negative dependence on temperature, and the dependence of their DRH on temperature can be approximated by the Clausius-Clapeyron equation."