Response to comments

Comment from Referee #1:

General Comments: 1. The introduction (starting around line 77) is improved, but the reader would still benefit from a brief, referenced paragraph that discusses the link between particle size and cloud droplet size for a given chemical composition. I think this will really help lead into the discussion of enrichment. It is not abundantly clear to the reader is the Twohy 1989 result is that small particle grow to small droplets and large particles grow to large droplets, independent of composition, or if the connection to small droplets is via $(NH_4)_2SO_4$ and to large droplets through NaCl. This is the foundation of the manuscript and could be made much more clear. Essentially, this should be a concise introduction to the physics behind how chemical enrichment in large (or small) cloud droplets could come about.

Reply: We would like to thank the reviewer for his/her useful comments and recommendations to improve the manuscript. We had made careful revision according to the suggestions, please refer to the following responses for details.

Twohy et al. (1989) observed that small ammonium sulfate particles grew to small droplets and large sea salt particles grew to large droplets. Previous observations also considered that the supermicron or giant sea salt-containing particles readily became large cloud droplets, and their CCN behavior was less affected by chemical composition (Noone et al., 1988; Monger et al., 1989; Andreae and Rosenfeld, 2008; Tao et al., 2012). So far, the study on the submicron sea salt-containing particles in cloud droplets is scarce in the literature. Additionally, the existence of secondary species (e.g., sulfate, nitrate, or organic species) onto the submicron sea salt-containing particles might significantly impact on their cloud activation (O'Dowd et al., 1999; Gibson et al., 2006; Nguyen et al., 2017). Please refer to Lines 79-88 in the modified manuscript.

2. I think it would be tremendously helpful to have a figure (comparable to figure 3A, but for particles) that shows the number fraction of sea-spray (or sea-salt as termed here) as a function of particle size for the ambient aerosol (not cloud residuals). This

distribution could then be compared to the cloud residual distribution to identify enrichment. The paper currently argues that compositional differences in submicron SSA drive the observed changes in the cloud residual number fractions. This may be true, but more detail on the ambient aerosol distribution would help support (or refute) this argument.

Reply: We had added the size distribution of sea salt particles during cloud-free periods (Figure S4). During cloud-free periods, sea salt particles accounted for 2% of the total ambient aerosol particles, and higher number fraction was observed in the supermicron particles (20%) than in the submicron particles (1%). Meanwhile, the submicron sea salt particles contained higher proportions of organic nitrogen (73-94% versus 58%, by number), hydrocarbon organic species (51-81% versus 2%) and organic acids (35-72% versus 4%) in the ambient aerosol particles than in the cloud residues at cut size of 7.5 μm. This further supports that the enhancement of organic species in the submicron particles likely reduced CCN activity of sea salt particles. Please refer to Lines 210-211, 292-293 and 363-368 in the modified manuscript and Figure S4 in the modified Supplement.

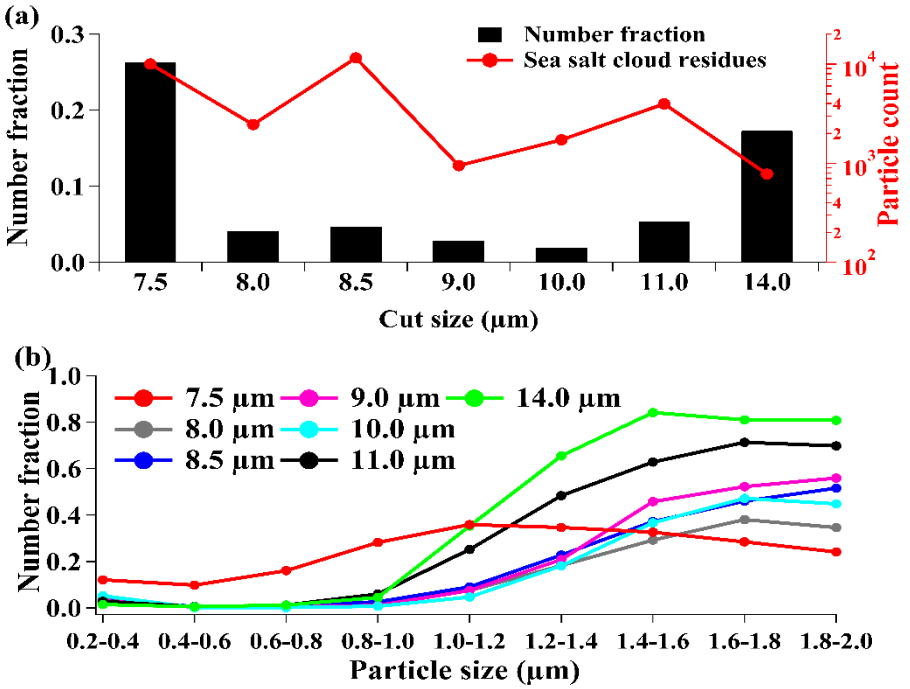

Figure 3. Number fraction or counts of sea salt-containing cloud residues as a function of the cut size (a) and the relative contribution of sea salt-containing cloud residues to the total cloud residues in the given size range (b).

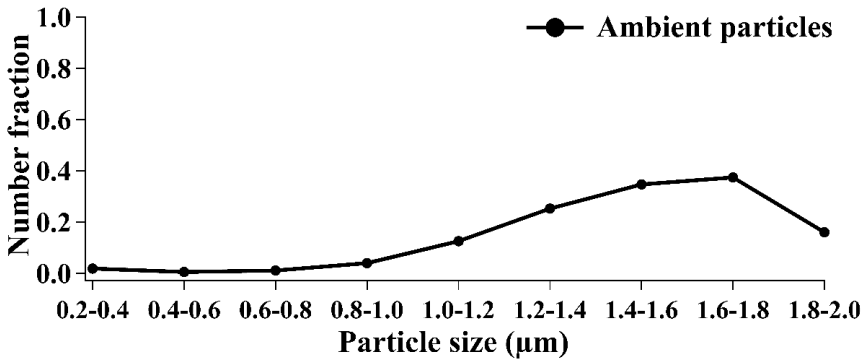

Figure S4. The relative contribution of sea salt-containing particles to the total particles in the given size range during cloud-free periods.

Specific Comments

Line 43: I don't think that Cochran et al is an appropriate primary reference for this statement.

Reply: The reference of Cochran et al. had been replaced by Boucher et al. (2013),

which summarized assessment report of the Intergovernmental Panel on Climate Change (IPCC). Please refer to Line 46 in the modified manuscript.

Line 52: I'm not sure how well founded this sentence is. The uncertainty in the CCN activity of sea-spray aerosol is much more a function of variance in the size distribution of SSA than its composition outside of the smallest SSA. There remain open questions as to why the hygroscopicity of SSA is so high given the organic content. See Collins et al., GRL 2016 (https://doi.org/10.1002/2016GL069922)

Reply: We agreed with the comment. Collins et al. (2016) found that large increases in the organic matter fraction of freshly emitted sea-spray aerosols only induce less than 3% change in CCN concentrations, and they also suggested that secondary aerosol chemistry may be the more important factor linking ocean biogeochemistry to marine clouds. In this study, we found that atmospheric ageing processes would alter chemical composition of sea salt particles, might thus influence CCN behavior. The sentence had been changed to "Therefore, it is important to evaluate the impact of chemical composition and particle size on the CCN behavior of sea salt particles." Please refer to Lines 53-55 in the modified manuscript.

Line 58: Given that SSA is largely organic, consider changing from sea salt aerosol to sea spray aerosol.

Reply: We had changed accordingly. Please refer to Line 59 in the modified manuscript.

Line 64: I don't think the conclusion of Wang et al 2015 is that the enrichment does not always occur. The conclusion is that the fraction of aliphatic organic material in SSA has some level of variability given surface ocean composition.

Reply: We had changed to "the fraction of organics (i.e., aliphatic organic material) in small sea spray aerosols exhibited some levels of variability in the similar simulation of ocean seawater conditions". Please refer to Lines 64-67 in the modified manuscript.

Figure 4 is extremely complicated. I would recommend a more detailed figure caption

that defines what “mixing fraction means”. Is one to interpret that a value of 1 (or red) for nitrate is that all of the particles detected at this size have nitrate?

Reply: The mixing fraction is defined as the number particles of a given compound divided by the total number particles. Yes, value of 1 (red color) for nitrate at the given cut size represented 100% of the total detected particles internally mixed with nitrate. Please refer to the caption of Figure 4 in the modified manuscript.

References:

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