

We sincerely appreciate the reviewers for taking the time to review our manuscript and for providing valuable comments. Based on the reviewers' suggestions, we have made revisions to the paper. Below are our responses to each of the reviewers' comments, with the reviewers' comments in black, our responses in red, and the revised manuscript content in italicized orange font.

This manuscript investigates the aerosol and CCN properties over the South China Sea area. The manuscript generally provides some interesting results regarding the seasonal variation and the anthropogenic influences in this area. However, the writing of the manuscript needs a lot of improvements. The focus of the manuscript may also need some changes. Below are some suggestions:

1. I find it very distracting when results from this study are mixed with a lot of results from previous studies in Section 3. It is very difficult to get the point of this study when it is mixed with other studies. For example, lines 227-234, some measurements of particle number concentration are listed. It actually doesn't mean much if only to compare which one is higher and which one is lower. I would suggest adding a table to summarize these measurements so that it is a lot easier for the readers to see which one is higher and which one is lower. Alternatively, results from previous studies can be organized into the Introduction section so that the readers can have a better background from the beginning. The comparison between the previous results and this study can also be organized into a new Discussion section. Basically, I would prefer that Section 3 focuses on the results only from this study. Similarly, lines 236-238, lines 249-251, lines 256-265, lines 298-308, lines 321-323, lines 344-345, lines 362-367, lines 374-377, lines 380-388, and lines 392-396 are all results from previous studies. Please put these previous results in a table, or put them into Introduction or a new Discussion section. This would help the readers to integrate the results from this study.

**Reply:** We have rewritten the first section of the results to focus specifically on the content of our study (lines 269-299):

*Figure 2 presented the timeseries of PNSD (a1 and a2), NR-PM<sub>1</sub> mass concentrations and fractions (b1 and b2, c1 and c2), number concentrations of CCN (d1 and d2), and hygroscopicity  $\kappa$ -values (e1 and e2) during two campaigns in summer and winter. During the summer cruise, we observed two distinct periods around the onset of the summer monsoon. The South China Sea (SCS) summer monsoon began in the sixth pentad of May (Chao et al., 2022). In winter, the influence of the winter monsoon persisted throughout the entire observation period (Fig. 1c). Despite our measurements being limited to the northern SCS in winter, the impact of the Northeast Monsoon on the SCS was evident.*

*The average particle number concentration in summer ( $6966\text{ cm}^{-3}$ ) was higher than in winter ( $4988\text{ cm}^{-3}$ ), primarily due to the higher number concentration of Aitken-mode particles in summer (Fig. 3a-b). In summer, particles were concentrated in smaller sizes, whereas in winter, particle size distribution was relatively balanced between the Aitken mode ( $2185\text{ cm}^{-3}$ ) and the accumulation mode ( $2176\text{ cm}^{-3}$ ) (Fig. 3a-b).*

*The average mass concentration of NR-PM<sub>1</sub> was  $3.76\text{ }\mu\text{g m}^{-3}$  in summer and increased to  $9.39\text{ }\mu\text{g m}^{-3}$  in winter (Fig. 3c-d). In summer, the dominant aerosol component was*

sulfate (45.5%), followed by organics (35.8%), ammonium (12.9%), nitrate (4.0%), and chloride (1.9%) (Fig. 3c), similar to the pattern observed in the northern SCS during the summer of 2018 (Fig. 3e) (Liang et al., 2021). However, in winter, organics became the predominant aerosol component (37%), with nitrate (22.2%) replacing sulfate (18.9%) as the highest proportion of inorganic components (Fig. 3d).

The average number concentration of cloud condensation nuclei ( $N_{CCN}$ ) in summer was higher than in winter at all supersaturation (SS) levels (Table 1). The absolute difference in the  $N_{CCN}$  between summer and winter was greater at high SS ( $\Delta N_{CCN}=2099\text{ cm}^{-3}$  and  $1865\text{ cm}^{-3}$  at 0.4% SS and 0.7% SS, respectively) compared to low SS ( $\Delta N_{CCN}=341\text{ cm}^{-3}$  at 0.2% SS), likely due to the significant difference in Aitken-mode particles between the two seasons (Fig. 3a-b).

Aerosol hygroscopicity ( $\kappa$ ) was similar at low SS but differed significantly at high SS between summer and winter (Table 1). The hygroscopicity pattern varied between seasons: in summer,  $\kappa$  increased with SS (from 0.49 to 0.72 between 0.2% SS and 0.4% SS), while in winter,  $\kappa$  decreased with SS (from 0.50 to 0.15 between 0.1% SS and 0.7% SS) (Fig. 3a-b). The winter  $\kappa$  pattern was similar to observations in the Western North Pacific (Table 1) (Kawana et al., 2020). Additionally, the winter  $\kappa$  values were comparable to those in Guangzhou, adjacent to the SCS, indicating that the northern SCS is influenced by air masses from Mainland China under the significant influence of the Northeast Monsoon during winter.

Additionally, we have included relevant results from other studies in Figure 3 and Table 1 to facilitate easier comparison for readers.

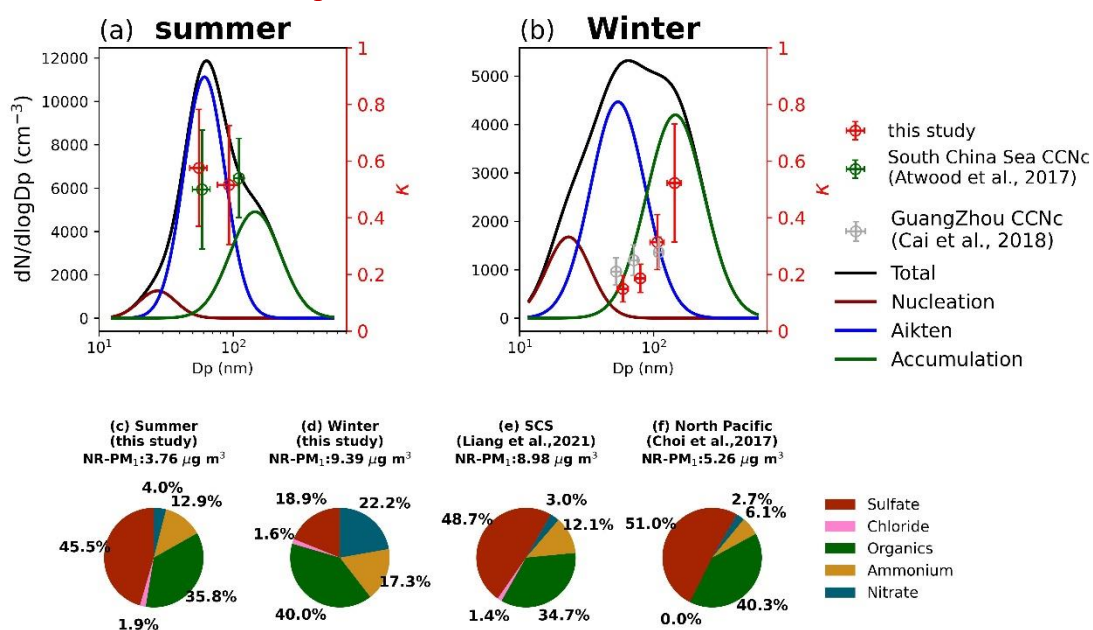


Figure 3. Particle number size distribution in summer (a) and winter (b); The red markers represent the activation diameters and hygroscopicity parameters corresponding to 0.1%, 0.2%, 0.4%, and 0.7% supersaturations in this study (without 0.1% in summer). The green markers represent the hygroscopicity parameters reported in Atwood et al. (2017) for the southern South China Sea during summer. The gray

markers represent the hygroscopicity parameters documented in Cai et al. (2018) for the Pearl River Delta region during winter. The fraction of NR-PM<sub>1</sub> in summer (c) and winter (d) in this study, in northern SCS reported by Liang et al. (2021) (e), and in North Pacific reported by Choi et al. (2017) (f).

Table 1. The number concentration of particle and cloud condensation nuclei at different supersaturation (SS), the hygroscopicity and activation ratio (AR) at different SS in different studies.

Location	period	$N_{CN} (cm^{-3})$	$N_{CCN} (cm^{-3})$	Hygroscopicity ( $\kappa$ )	AR	Reference
South China Sea	2021.05.05-2021.06.09	6966±9249	2019±2993 (0.20% SS)	0.49±0.42 (0.20% SS)	0.43±0.17 (0.20% SS)	This study
			4445±7018 (0.40% SS)	0.74±0.51 (0.40% SS)	0.68±0.19 (0.40% SS)	
			4786±6402 (0.70% SS)		0.89±0.12 (0.70% SS)	
Northern South China Sea	2021.12.19-2021.12.29	4988±3474	1100±1287 (0.10% SS)	0.50±0.21 (0.10% SS)	0.23±0.10 (0.10% SS)	This study
			1678±1046 (0.20% SS)	0.31±0.10 (0.20% SS)	0.35±0.12 (0.20% SS)	
			2346±1767 (0.40% SS)	0.19±0.05 (0.40% SS)	0.48±0.14 (0.40% SS)	
			2921±1917 (0.70% SS)	0.15±0.05 (0.70% SS)	0.60±0.16 (0.70% SS)	
Northern South China Sea	2018.8.6-2018.8.27	3463	1544 (0.34% SS)	0.38±0.09 (0.18% SS) 0.40±0.08 (0.34% SS) 0.38±0.08 (0.59% SS)	/	Cai et al., 2020
Remote South China Sea	2012.9.14-2012.9.26	503±455	450±388 (0.14% SS)		0.47±0.16 (0.14% SS)	Atwood et al., 2017
			675±516 (0.38% SS)	0.54±0.14 (0.14% SS)	0.72±0.17 (0.38% SS)	
			698±555 (0.53% SS)	0.50±0.21 (0.38% SS)	0.79±0.15 (0.53% SS)	
			724±512 (0.71% SS)		0.85±0.13 (0.71% SS)	
Western North Pacific	2015.3.4-2015.3.26	/	/	0.75±0.21 (0.11% SS)	0.40±0.22 (0.11% SS)	Kawana et al., 2020
				0.51±0.16 (0.24% SS)	0.50±0.22 (0.24% SS)	
				0.45±0.16 (0.60% SS)	0.70±0.23 (0.60% SS)	
Guangzhou	2014.11-2014.12	/	3103±1913 (0.10% SS)	0.37±0.11 (0.10% SS)	0.26±0.10 (0.10% SS)	Cai et al., 2018
			5095±2972 (0.20% SS)	0.29±0.09 (0.20% SS)	0.41±0.14 (0.20% SS)	
			6524±3783 (0.40% SS)	0.18±0.07 (0.40% SS)	0.53±0.15 (0.40% SS)	
			7913±4234 (0.70% SS)	0.15±0.06 (0.70% SS)	0.64±0.13 (0.70% SS)	

2. It is very interesting to see that aerosol and CCN properties are quite different in summer and winter. This indicates that the seasonal variations of aerosol and CCN properties should be considered in regional or climate models when studying aerosol-cloud interaction for this area. I especially agree that particle composition and kappa are quite different in summer and winter. However, it is not very clear if particle number concentration is significantly different in summer and winter. The manuscript emphasizes that summer has much higher number concentration when the marine atmosphere is influenced by terrestrial air masses (lines 226-227). I agree. But how about the average number concentration in the whole observed period in summer? Is it still much higher than that in winter? For many days when the atmosphere is influenced by mixed air masses, particle number concentration seems to be similar in summer and winter. So please check if the number concentration in the whole summer period is on average much higher than that in winter. The panel d in Figure 2 cannot provide such information because summer and winter are not plotted with the same scale. It would help if the properties are plotted in the same scale for summer and winter in Figure 2. In addition, regarding the dominant mode in PNSD, it is said in line 30 that PNSD has a dominance of Aitken mode in summer and a dominance of accumulation mode in winter. But based on Figure 2 and Figure 3, I would say that both Aitken mode and accumulation mode are important in winter. It is not appropriate to conclude that the dominant mode in winter is accumulation mode. So I would see more evidence regarding the dominant mode. Maybe you could calculate the total number concentration in Aitken mode and in accumulation mode and compare to see which one is dominant.

Reply: Thanks for reviewer's comment. The total particle number concentration was higher in summer ( $6966 \text{ cm}^{-3}$ ) than in winter ( $4988 \text{ cm}^{-3}$ ) (Table 1). We have replotted the Figure 2 to make sure the particle number concentration value is in the same scale. Besides, the Aitken mode particle concentration was similar to accumulation mode particle in winter, so we revised our description in lines 278-280:

*In summer, particles were concentrated in smaller sizes, whereas in winter, particle size distribution was relatively balanced between the Aitken mode ( $2185 \text{ cm}^{-3}$ ) and the accumulation mode ( $2176 \text{ cm}^{-3}$ ) (Fig. 3a-b).*

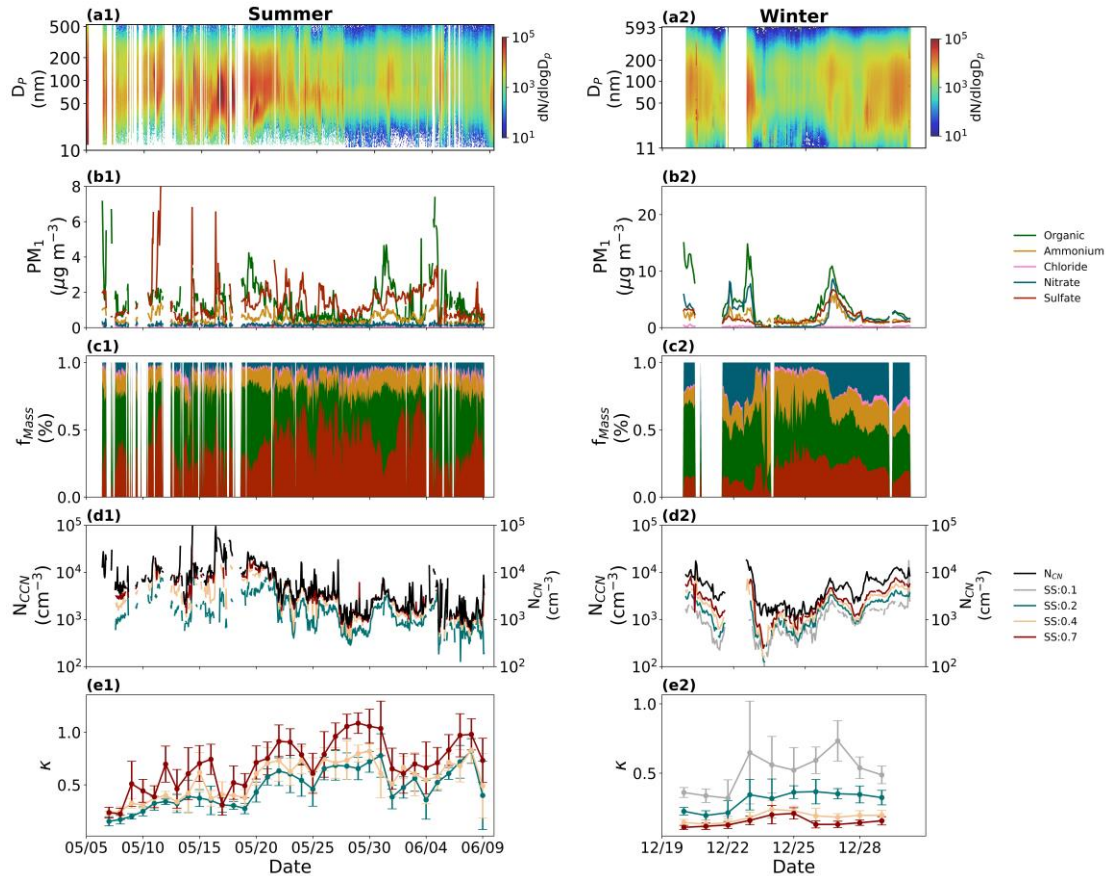


Figure 2. Timeseries of (a) particle number size distribution, (b) mass concentration of NR-PM<sub>1</sub>, and (c) its fraction, (d) mass concentration of organic carbon and elemental carbon, (e) number concentration of total particle and cloud condensation nuclei under the supersaturation of 0.1%, 0.2%, 0.4%, and 0.7%, and (f) aerosol hygroscopicity. The number 1 in figure number means timeseries in summer and number 2 means it in winter.

3. I think the size-resolved AR should be shown, especially because D50 is determined by fitting the AR and dry diameter at each supersaturation.

Reply: We have added the figure of size-resolved AR fitting result as Fig. S3.



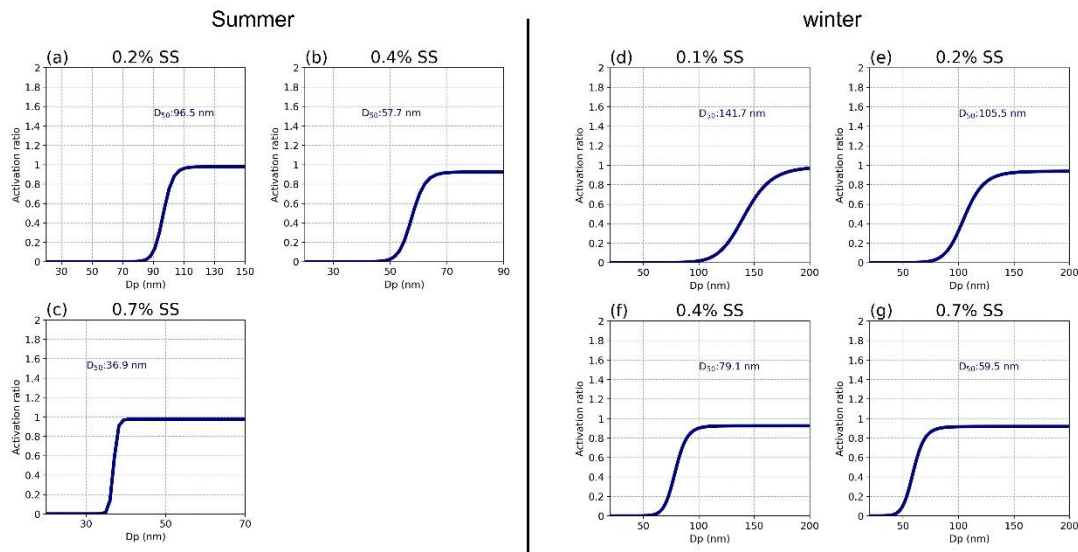


Figure S3. The average size-resolved activation ratio (AR) fitting result at 0.2% SS (a), 0.4% SS (b), and 0.7% SS (c) in summer; The average size-resolved activation ratio (AR) fitting result at 0.1% SS (d), 0.2% SS (e), 0.4% SS (f), and 0.7% SS (g) in winter. 4. The method in 2.2.1 (equation 1) and 2.2.2 (equation 4) seem to be in conflict. In equation 1, I assume that the activation ratio is size-resolved? However, in Equation 4, the activation ratio is the bulk activation ratio? So please clarify whether AR represents the size-resolved or the bulk activation ratio.

Reply: We obtained the activation diameter ( $D_{50}$ ) from size-resolved AR and diameter according to SMCA method. The CCN concentration ( $N_{CCN}$ ) was calculated based on the  $D_{50}$  and observed PNSD from SMPS. Then the AR represent bulk AR from ratio of CCN concentration to total particle concentration. We have added the following sentence in lines 188 and 220 for clarification:

*where AR is the size-resolved AR (line 192)*

*It is noting that the AR here is bulk AR. (line 224)*

5. Regarding the size-dependent kappa value in lines 282-283, please clarify the reasons why kappa value is size-dependent.

Reply: To clearly express our observational results, we have revised our statement to indicate that hygroscopicity varies with changes in supersaturation (SS) (lines 293-295):

*The hygroscopicity pattern varied between seasons: in summer,  $\kappa$  increased with SS (from 0.49 to 0.72 between 0.2% SS and 0.4% SS), while in winter,  $\kappa$  decreased with SS (from 0.50 to 0.15 between 0.1% SS and 0.7% SS) (Fig. 3a-b).*

And we discuss the reason why kappa value is size-dependent in the following section. The possible reason for higher hygroscopicity at high SS in summer than winter is the MSA oxidized from DMS produced by phytoplankton (lines 341-345):

*However, aerosol hygroscopicity at small sizes was much lower in the “Mainland China” period than in the “Luzon” period (Fig. 8), contributing to the low AR in the “Mainland China” period (Fig. 7). This lower hygroscopicity could be due to lower sulfate concentration, oxidized by DMS, in winter than in summer, as higher sea surface*

*temperatures in summer (29.3°C) compared to winter (18.0°C) promote DMS production by phytoplankton (Bates et al., 1987).*

Besides, Additionally, the low hygroscopicity of small particles in winter may be due to externally mixed BC and an increased proportion of organics. (lines 373-382):

*In winter, the “External-mixed” scheme always showed a better result than “Internal-mixed” scheme at high SS (0.4% SS and 0.7% SS), indicating that particles in small size were mainly externally mixed. Considering the low hygroscopicity of small-sized particles in winter, it is likely that a significant fraction of these particles consists of externally mixed BC, which probably originated from fresh anthropogenic emissions and remains unmixed with other inorganic salts and organics. As BC ages, inorganic and organic components adhere to it, which would lead to the increase of diameter and particles tended to be internally mixed (Sarangi et al., 2019). This transition resulted in higher hygroscopicity in large-sized particle compared to the smaller-sized particles. Besides, overestimation of aerosol hygroscopicity at high SS could be owing to a higher fraction of non- or less- hygroscopic component (such as organic and BC) at small particle sizes.*

6. Regarding the AR ratio in lines 390-391 in Section 3.3, what does “beyond D50” mean? The meaning of this sentence is not clear. Please revise. This is very important for understanding the sensitivity tests afterwards. I think it’s quite interesting that PNSD is the most important factor influencing Nccn in summer, whereas hygroscopicity is the most important factor in winter (lines 416-417). This is based on the sensitivity tests performed in this study. I wonder if the authors can provide any underlying physical reasons for this.

Reply: Thanks for reviewer’s question. In reference to the two reviewer’s comments and other literatures, we consider that the original method, based on sensitivity experiments, may not accurately explain the effects of PNSD and hygroscopicity on CCN concentrations. Therefore, we removed the original method in revised version and apply CCN closure method, which has been widely used in other researches and can provides more accurately result, to analyze the impact of aerosol composition and mixing state on CCN activities. The CCN closure analysis was shown in 3.3 (lines 358-388).

*CCN closure study was widely applied to investigate the impacts of different factors on the CCN activity (Patel et al., 2021; Cai et al., 2018; Meng et al., 2014; Deng et al., 2013). In this study, two schemes considering aerosol composition and mixing state based on CCN closure method mentioned in 2.2.3 were applied. The fitting parameter and coefficient of determination ( $R^2$ ) was shown in Table 3 and the fitting plots from two schemes were shown in Fig. S8 and Fig. S9. Besides, the NMB from these two schemes was presented in Fig. 8.*

*In summer, the NMB always lower than 0, which indicated that simulated aerosol hygroscopicity was lower than observed value (Fig. 8). Sea salt which cannot be detected by the ToF-ACSM may account for higher fraction in summer due to low aerosol concentration in summer (Fig. 3c), resulting in the underestimation of aerosol hygroscopicity. The NMB exhibits different trends with changes in SS in “Luzon” and “Indochinese Peninsula” period. Better fitting result appeared in high SS in “Indochinese Peninsula” period, while it appeared in low SS in “Luzon” period (Fig. 8),*



which indicated that aerosol fraction had different trend as particle size increased in these two periods. Besides, "Internal-mixed" scheme had more precious result than it in "External-mixed" scheme in summer (Fig. 8), suggesting the aerosol was primary internally mixed in summer.

In winter, the "External-mixed" scheme always showed a better result than "Internal-mixed" scheme at high SS (0.4% SS and 0.7% SS), indicating that particles in small size were mainly externally mixed. Considering the low hygroscopicity of small-sized particles in winter, it is likely that a significant fraction of these particles consists of externally mixed BC, which probably originated from fresh anthropogenic emissions and remains unmixed with other inorganic salts and organics. As BC ages, inorganic and organic components adhere to it, which would lead to the increase of diameter and particles tended to be internally mixed (Sarangi et al., 2019). This transition resulted in higher hygroscopicity in large-sized particle compared to the smaller-sized particles. Besides, overestimation of aerosol hygroscopicity at high SS could be owing to a higher fraction of non- or less- hygroscopic component (such as organic and BC) at small particle sizes. The predicted  $N_{CCN}$  at 0.1% SS are 10%-20% lower than the observed concentrations, whereas the predicted value at 0.2% SS more closely aligns with the observed concentrations (Fig. 8). It could be owing to the higher fraction of sea salt at larger particle size. However, due to instrument limitations, black carbon and sea salt cannot be detected by the ToF-ACSM. More observations containing sea salt and black carbon are needed in the future to better assess their effects on aerosol hygroscopicity in SCS. In addition, further study size-resolved aerosol composition can also enhance the understanding on CCN activity in the SCS.

7. Section 3.4 seems to have conflicting results with Section 3.3. It is shown in Section 3.3 that PNSD determines  $N_{ccn}$  in summer and hygroscopicity determines  $N_{ccn}$  in winter. However, in Section 3.4, it is shown that PNSD is important for  $N_{ccn}$  in both summer and winter. I think Section 3.3 and Section 3.4 should be better integrated. The writing should be more concise and focused.

Reply: Thanks for reviewer's question. Firstly, we consider that this method, based on sensitivity experiments, may not provide accurate explanation on the effects of PNSD and hygroscopicity on CCN concentrations. Therefore, we adopted the more widely used CCN closure method as a replacement. Additionally, we reconsidered the influence of terrestrial air masses in summer and winter. Cluster analysis revealed two distinct continental air masses in summer: one from the direction of Luzon Island and the other from the Indochinese Peninsula. To provide clearer and more comprehensible results, we integrated Section 3.4 into Sections 3.2 and 3.3.

Minor points:

1. Please provide a little discussion on cloud climatology for the studied area. It would be good to see that there is a relatively high cloud fraction, especially warm cloud in the studied area.

Reply: To our current knowledge, there is still a lack of research on the warm cloud on the SCS. Thus, we introduced the seasonal variation of high cloud fraction in SCS in lines 118-120:

*Additionally, the high cloud fraction over the SCS varies from approximately 0.3 to 0.7 across different months, indicating that aerosol-cloud interactions in the region may differ between seasons (Lu et al., 2022).*

2. Lines 431-433 are very similar to lines 448-450. The writing should be improved.

Reply: We employed a new method, CCN closure analysis, to revise Section 3.3, replacing the original Sections 3.3 and 3.4.

3. The manuscript title and the titles in Section 3 are kind of confusing. A lot of “impact of ... on...” Or “influence of ... on ...” are seen in the titles. For example,

Title of the manuscript: “anthropogenic influence on CCN activation”

Title of 3.2: Impact of chemical composition on hygroscopicity

Title of 3.2.1: impact of inorganic components

Title of 3.2.2: impact of organic components

Title of 3.4: influence of spatial distribution of particle properties on NCCN

In addition, the title of 3.1 is too simple. This title does not provide much information.

There are also some inconsistency in the titles. For example, I can see that seasonal variation is a focus of this study based on the title of the manuscript. However, only the title of 3.3 has “seasons”. Based on the current titles, it is hard to figure out which part in Section 3 actually discusses “seasonal variations”.

Reply: We have changed our title:

3.1 CCN concentration and aerosol characteristics over SCS in summer and winter

3.2 Anthropogenic influence on CCN concentration in different season

3.3 CCN closure analysis

In Section 3.1, we briefly introduce the observation result in summer and winter. The detail discussion about the impact of different types of terrestrial air masses on CCN activities in summer and winter. In the last section, we further discuss the influence of aerosol composition and mixing state on CCN activities in SCS.

4. In section 3, there are some sentences that are repetitive. For example, lines 266-268, and lines 280-282. Section 3 should be integrated in a better way. After the results from previous studies are moved to other places, Section 3 can be more focused and better integrated.

Reply: We have added Table 1 and replotted the Figure 3 to present the differences between this study and other researches. Besides, we have rewritten the 3.1 to focus on presenting the result on this study.

5. It seems the organic carbon and elemental carbon are missing in Figure 2. See the figure caption, (d).

Reply: We have deleted the data of organic carbon and elemental carbon in our discussion and changed the figure caption in Figure 2.

6. Equation 1:  $N_{ccn}/N_{cn}$  should be replaced with AR, to be consistent with the use of AR in Equation 4.

Reply: We have changed  $N_{ccn}/N_{cn}$  to AR.

7. Equation 2: the two formula should be put in separate lines.

Reply: We have put them in separate lines.

8. Line 448: “in winter, the increasing trend...” should be changed to “in winter, the decreasing trend”.

Reply: We have deleted this sentence.

9. Line 39: “impact of PNSD on AR was greater than on aerosol hygroscopicity in summer” should be changed to “impact of PNSD on AR was greater than aerosol hygroscopicity in summer”. In addition, “vice versa” is not a good expression in this

sentence.

Reply: We have deleted this sentence.