

Reply to AC2

We thank the reviewer for providing insightful comments and helpful suggestions that have substantially improved the manuscript. Below we have included the review comments in black followed by our responses in blue. In the revision of this manuscript, we have highlighted those changes accordingly.

The manuscript on “Vertical profiles of cloud condensation nuclei number concentration and its empirical estimate from aerosol optical properties over the North China Plain” by R. Zhang and co-authors made airborne measurements of vertical profiles of CCN concentrations and scattering coefficients over the southern plain of Hebei province. Using this data, they have investigated the influence of thermal structure (TIL) and air mass origin on vertical profiles of CCN. The CCN concentration is estimated using the scattering coefficient and its spectral variation.

Considering the limitations and uncertainties associated with the retrieval of vertical profiles of aerosols and CCN using different techniques, the direct measurements of these parameters onboard the aircraft are very important. But I feel disappointed with the way the authors described their experimental details. Details of the sampling inlet are not provided. (i) What is the effect of aircraft propeller on aerosol sampling? (ii) Whether sampling flow was iso-kinetic? (iii) What was the sampling efficiency of the inlet used? (iv) What was the cruising speed of the Y-12 Turboprop? (v) How do authors account for ram heating? (vi) How do authors account for the flow instabilities during ascending and descending phases of spiral flights? (vii) How much is the total sampling time available for each vertical level? (viii) Whether CCN measurements at all the supersaturations were carried out at each altitude? If not, how do authors decouple the change in CCN due to supersaturation change and also due to vertical variation?

RE: Thanks for the suggestion. The details of the flight plans, sampling method, and initial investigations into the impact of air mass on air chemistry have been published (Benish et al., 2020, 2021; F. Wang et al., 2018), and cited in our manuscript. It would be duplication if they were included in the main text, but we summarize them here below for the sake of the reviewer and we have included the description in the supplement.

(i): The sampling device (shown in the figures below) is above the front of the airplane cabin, which is not affected by the propeller after the plane takes off.



(ii) and (iii): The sampling flow was iso-kinetic. As described in F. Wang et al. (2018), the conical double diffuser aerosol inlet, designed for a Twin Otter, was installed on the Y-12. This inlet system was manufactured by Droplet Measurements Technologies (MP-1806-A and MP-1807-A, Boulder, CO, USA) (Hegg et al., 2005). It has been used extensively on the University of Maryland's Cessna 402 (Brent et al., 2015). The passing efficiency is expected to be near 100% for particle diameters up to 2.5 μm and near 50% for particles between 3 and 4 μm (Huebert et al., 2004; McNaughton et al., 2007).

(iv): As described in F. Wang et al. (2018), the typical cruising speed of aircraft is 60-70 m s^{-1} , with ascent/descent rates of 2–5 m s^{-1} .

(v): Ascents and descents were gentle to avoid turbulence taking about 20 min to ascend 3000 m or $\sim 150 \text{ m/min}$. The ram heating was considered by adjusting the measured air temperature and relative humidity:

$$\text{Temp_adj} = (\text{Temp} + 273.15) / (1 + 0.2 * \text{rf} * \text{M}^2) - 273.15;$$

Where,

Temp_adj – adjusted air temperature by taking the ram heating effect into account

Temp – measured air temperature ($^{\circ}\text{C}$)

rf – recovery factor (rf = 0.896445604404384)

M – mach number, which is calculated from the measured true air speed and calculated speed of sound:

$$\text{M} = \text{Airspeed_True} / \text{Speed_sound}$$

$$\text{Speed_sound} = 331.3 * \text{sqrt}((\text{Temp} + 273.15) / 273.15)$$

Relative humidity was also adjusted by multiplying the ratio of saturated water pressures under measured and adjusted air temperature:

$$\text{RH_adj} = \text{RH} * (\text{svpt} / \text{svpat});$$

where,

$$\text{svpt} = 6.1121 * \exp((18.678 - \text{Temp} / 234.5) * (\text{Temp} / (257.14 + \text{Temp})));$$

$$\text{svpat} = 6.1121 * \exp((18.678 - \text{Temp_adj} / 234.5) * (\text{Temp_adj} / (257.14 + \text{Temp_adj})));$$

(vi): When not on a smooth ascent or descent the sampling time at each level varied from ~ 2 to ~ 20 min.

(vii): The sampling duration of every vertical spiral or level flight is added in the updated Table 1 shown in the below.

Flight number, date	Time range (CST)	Flight code	Region covered	Vertical height a.s.l. (km)	Sampling duration (min)	Maximum spiral radius (km)
RF1, 20160508	13:02– 14:29	RF1_1	XT	0.3–3.7	38	~ 10
		RF1_a	track from XT to LC	~ 3.6	20	–
		RF1_2	LC	0.3–3.2	15	~ 10
RF2, 20160515	12:17– 15:04	RF2_a	track from LC to JL	~ 0.4	18	–
		RF2_1	JL	0.3–3.6	40	~ 5.0
		RF2_2	QZ	0.3–3.6	38	~ 5.0
		RF2_b	track from QZ to JL	~ 3.6	7	–
		RF2_c	track from JL to LC	~ 0.4	10	–

		RF6_1	QZ	0.3–3.1	36	~ 5.0
RF6,	12:04–	RF6_a	track from QZ to XT	~2.5	18	–
20160521	14:41	RF6_2	XT	0.3–2.6	43	~ 5.0
		RF6_b	track from XT to LC	~1.1	13	–
		RF7_a	track around XT	~3.1	20	–
RF7,	10:21–	RF7_1	XT	0.5–3.1	49	~ 5.0
20160528	13:25	RF7_b	track from XT to JL	~0.4	10	–
		RF7_2	JL	0.3–2.5	26	~ 4.0
		RF7_c	track from JL to LC	~1.8	7	–
RF8,	16:30–	RF8_a	track around XT	~0.6	15	–
20160528	18:24	RF8_1	XT	0.5–3.1	36	~ 5.0
RF11,	11:07–	RF11_a	track around XT	~0.6	16	–
20160611	12:28	RF11_1	XT	0.3–3.2	50	~ 4.0

(viii): As described in section 2.2, CCNc-200 has two columns that can simultaneously measure N_{CCN} at two different supersaturation (SS) levels without mutual interference. In this campaign, only one SS level (0.7%) was set in the first column during all flights, but eight different SS levels (0.44%, 0.56%, 0.68%, 0.80%, 0.92%, 1.04%, 1.16%, and 1.28%) were set in the second column with a measurement time interval of 90 s for each SS level. We can get continuous N_{CCN} data at 0.7% SS but not at other SS during any flight. However, N_{CCN} data with different SS at a certain altitude can be obtained during the level flights. Therefore, we can analyze the vertical profile of N_{CCN} with 0.7% SS (Fig. 3–4) and CCN spectra using N_{CCN} data with different SS at certain altitudes (Fig. 5).

Reference:

- Benish, S. E., He, H., Ren, X., Roberts, S. J., Salawitch, R. J., Li, Z., Wang, F., Wang, Y., Zhang, F., Shao, M., Lu, S., and Dickerson, R. R.: Measurement report: Aircraft observations of ozone, nitrogen oxides, and volatile organic compounds over Hebei Province, China, *Atmospheric Chemistry and Physics*, 20, 14523–14545, <https://doi.org/10.5194/acp-20-14523-2020>, 2020.
- Benish, S. E., Salawitch, R. J., Ren, X., He, H., and Dickerson, R. R.: Airborne Observations of CFCs Over Hebei Province, China in Spring 2016, *Journal of Geophysical Research: Atmospheres*, 126, e2021J-e35152J, <https://doi.org/10.1029/2021JD035152>, 2021.
- Brent, L., Thorn, W., Gupta, M., Leen, B., Stehr, J., He, H., Arkinson, H., Weinheimer, A., Garland, C., and Pusede, S.: Evaluation of the use of a commercially available cavity ringdown absorption spectrometer for measuring NO₂ in flight, and observations over the Mid-Atlantic States, during DISCOVER-AQ, *Journal of Atmospheric Chemistry*, 72, 503–521, <https://doi.org/10.1007/s10874-013-9265-6>, 2015.
- Hegg, D. A., Covert, D. S., Jonsson, H., and Covert, P. A.: Determination of the transmission efficiency of an aircraft aerosol inlet, *Aerosol Science and Technology*, 39, 966–971, <https://doi.org/10.1080/02786820500377814>, 2005.
- Huebert, B., Bertram, T., Kline, J., Howell, S., Eatough, D., and Blomquist, B.: Measurements of organic and elemental carbon in Asian outflow during ACE-Asia from the NSF/NCAR C-130, *Journal of Geophysical Research: Atmospheres*, 109, D19S11, <https://doi.org/10.1029/2004JD004700>, 2004.
- McNaughton, C. S., Clarke, A. D., Howell, S. G., Pinkerton, M., Anderson, B., Thornhill, L.,

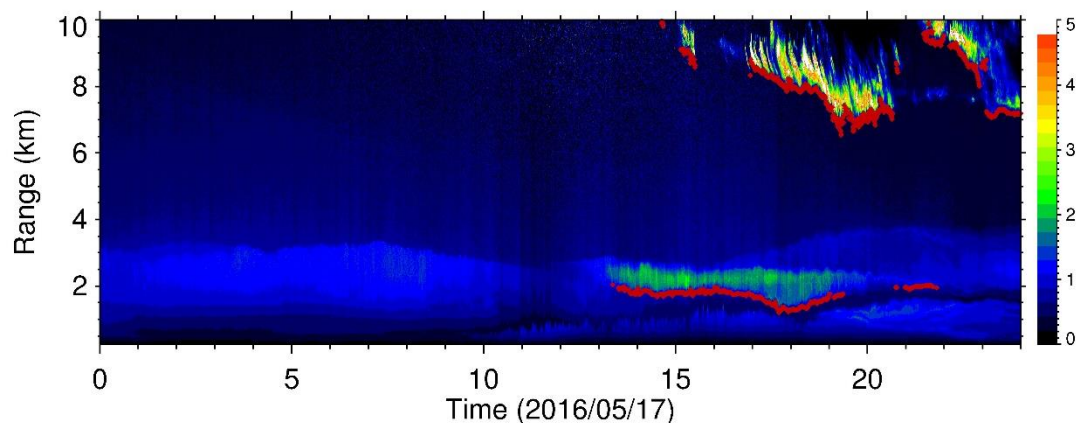
Hudgins, C., Winstead, E., Dibb, J. E., and Scheuer, E.: Results from the DC-8 Inlet Characterization Experiment (DICE): Airborne versus surface sampling of mineral dust and sea salt aerosols, *Aerosol Science and Technology*, 41, 136–159, <https://doi.org/10.1080/02786820601118406>, 2007.

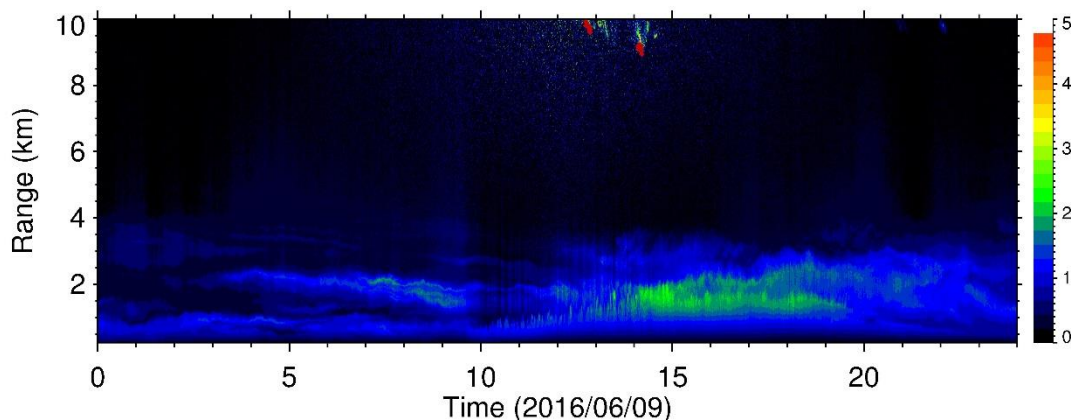
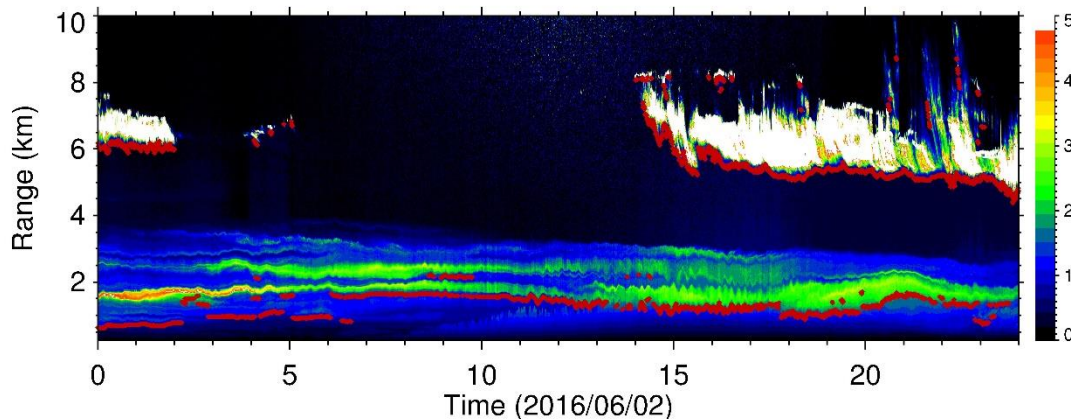
Wang, F., Li, Z., Ren, X., Jiang, Q., He, H., Dickerson, R. R., Dong, X., and Lv, F.: Vertical distributions of aerosol optical properties during the spring 2016 ARIAs airborne campaign in the North China Plain, *Atmospheric Chemistry and Physics*, 18, 8995-9010, <https://doi.org/10.5194/acp-18-8995-2018>, 2018.

The authors mentioned that CCN profiles have a strong dependence on the number and thickness of TIL. This is mostly due to the TIL influence on the vertical transport of aerosols. On the other hand, the influence of air mass trajectory indicates long-range transport. In other words, when long-range transport dominates at higher altitudes, the influence of vertical transport of aerosols from the lower atmosphere is irrelevant. If long range transport is the prominent mechanism, then how could authors associate TIL with CCN concentration?

RE: The N_{CCN} profiles (Fig. 4) differ significantly for air masses from the northwest vs southeast. In air masses from the northwest, air from the western desert or plateau carries a large number of aerosols to the NCP. Aerosols do not accumulate near the surface due to the rapid dispersion. Therefore, the TIL effect on N_{CCN} profiles is weak under the impact of air masses from the northwest. This is why the vertical variation of N_{CCN} is small in northwesterly air masses (Fig. 4b). In southeasterly air masses, aerosols near the surface accumulate easily due to the terrain blocking effect by the Taihang Mountains, leading to the strong vertical transport of aerosols. At the same time, the long-range transport of aerosols from northwest is weak. Thus, much lower N_{CCN} above 2 km than near the surface. The effect of long-range transport is dominant in northwesterly air masses, while the effect of vertical transport (the effect of TIL) is dominant in southeasterly air masses. The two effects are caused by the different meteorological conditions and special terrain distribution.

The long-range transports of aerosols play an important role in the structure of N_{CCN} profiles. However, the role of TIL cannot be ignored. In this campaign, a micro-pulse lidar (MPL) was deployed at the Xingtai (XT) supersite. On some days, there are different aerosol layers, which can be reflected by the MPL images. Some examples are shown below (green colors in the lower atmosphere indicate aerosol layers). Aerosol vertical stratification should be related to the influence of TILs.





MPL images on different days

How do authors link CCN spectra with activation efficiency? In lines 313-314, the authors mentioned that “A lower value means a stronger aerosol activation ability (i.e., more coarse-mode particles or stronger aerosol hygroscopicity), and vice versa.” This is not always true when hygroscopicity changes with the size of the particles.

RE: According to Köhler theory, aerosol hygroscopicity or activation ability is controlled by the Raoult effect and the Kelvin effect. An increase in particle size can enhance aerosol hygroscopicity or activation ability due to the Kelvin effect. Many studies used the k parameter to analyze aerosol activation ability. For example, Jefferson (2010) suggested that the k parameter indicates the steepness of the change in CCN concentration with SS . Low values of k are typical of highly soluble aerosol such as sea salt and high k values of low-solubility aerosols.

Reference:

Jefferson, A.: Empirical estimates of CCN from aerosol optical properties at four remote sites, *Atmospheric Chemistry and Physics*, 10, 6855-6861, <https://doi.org/10.5194/acp-10-6855-2010>, 2010.

How much time CCN counter required for attaining set supersaturation, especially when supersaturation changes from 1.28% to 0.44%? What is the sanctity of 0.7% supersaturation? Why lower supersaturations (<0.4%) are excluded from the sampling? What is the broad range of atmospheric supersaturation observed over the study region?

RE: In this campaign, the supersaturation of CCNc is adjusted by the control of flow rate (Rose et al., 2008). Therefore, the supersaturation adjustment for any change is rapid. Eight different SS levels are set in the second column with an observation interval of 90 s. Considering the equilibrium time of SS levels, data from the final 30 s data at any SS level in the cycle for the second column are used in this study. We focus on the impact of aerosols on the convective clouds. Therefore, the set supersaturation is high. In addition, the low N_{CCN} value at low SS in the free troposphere approaching CCNc measurement limit can make a large uncertainty.

Reference:

Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O., and Pöschl, U.: Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment, *Atmospheric Chemistry and Physics*, 8, 1153-1179, <https://doi.org/10.5194/acp-8-1153-2008>, 2008.

What kind of drier was used to remove the humidity of the air sampled by the nephelometer? Whether this could maintain a constant RH throughout the campaign?

RE: We did not dry the air sampled by the Nephelometer. Instead, we adjust for increased scattering with increased relative humidity with a correction factor, $f(RH)$, which is calculated by:

$$f(RH) = \left[\frac{(100 - RH_{neph})}{(100 - RH_{amb})} \right]^\gamma \quad B_{scat_adj} = B_{scat} \times C \times f(RH)$$

Where,

RH_{neph} – Internal Nephelometer RH

RH_{amb} – Adjusted Ambient RH that takes the ram heating into account (see above for the adjustment)

B_{scat_adj} – Adjusted Scattering Coefficient

B_{scat} – Measured Scattering Coefficient

γ – Measured Dry vs Humid Factor

C – Angular Truncation Factor, which is empirically derived based on the method by Anderson and Ogren (1998):

λ	Angular Truncation Factor	Angstrom Exponent	Detection Limit
450 nm	$C^{450} = 1.165 - 0.046 \times A_{500}$	$A_{500} = -\log(b_{scat}^{450}/b_{scat}^{550})/\log(450/550)$	4.4E-07 m ⁻¹
550 nm	$C^{550} = 1.152 - 0.044 \times A_{575}$	$A_{575} = -\log(b_{scat}^{450}/b_{scat}^{700})/\log(450/700)$	1.7E-07 m ⁻¹
700 nm	$C^{700} = 1.120 - 0.035 \times A_{625}$	$A_{625} = -\log(b_{scat}^{550}/b_{scat}^{700})/\log(550/700)$	2.6E-07 m ⁻¹

Reference:

Anderson, T. L. and Ogren, J. A.: Determining aerosol radiative properties using the TSI 3563 integrating Nephelometer, *Aerosol Science and Technology*, 29, 57–69, <https://doi.org/10.1080/02786829808965551>, 1998.

Line 173: Replace “this” with “integrating”

RE: Revised. Thanks.

There are data gaps in Figure 5. For example (i) panel a RF2_c: no CCN data is shown for ss<0.8%.

Similar is the case with panel b RF6_b and panel C RF7_c. Explain?

RE: The sampling duration for every vertical spiral or level flight is added in the updated Table 1 shown in the reply to first comment. In some level flights of short sampling durations, we couldn't obtain N_{CCN} data at all SS . Even so, there are sufficient data points in Fig. 5 for fitting.

What is the reason for high CCN activation at higher altitudes than lower levels? Normally, fine mode aerosols are transported to higher altitudes and these particles have lower CCN efficiency than coarse mode aerosols.

RE: What is stated by the reviewer may be generally true, but it varies from case to case. Scattering Ångström exponent (SAE) is often used to qualitatively assess the dominant size mode of aerosol activation, reflecting the particle number size distribution (PNSD) pattern (e.g., Hamonou et al., 1999). A large SAE (> 2) generally implies that fine-mode aerosols are dominant (e.g., smoke particles), while a small SAE (< 1) means that coarse-mode aerosols are dominant (e.g., dust particles). The SAE profiles shown in Fig. 6a suggests that the coarse mode particles are dominant above 2 km, while fine-mode aerosols are dominant below 2 km. The origin of the air shifts generally to the northwest with increasing altitude and the origin of the aerosols changes with it. In addition, particles can age and grow during the transport processes due to the atmospheric chemical reactions such as cloud processing. Liu et al., (2019) indicated that the mass fraction of hydrophilic secondary aerosols is higher in the upper atmosphere than near the surface based on the in-situ aircraft measurements in Beijing in the NCP.

Reference:

Hamonou, E., Chazette, P., Balis, D., Dulac, F., Schneider, X., Galani, E., Ancellet, G., and Papayannis, A.: Characterization of the vertical structure of Saharan dust export to the Mediterranean basin, *Journal of Geophysical Research: Atmospheres*, 104, 22,257-22,270, <https://doi.org/10.1029/1999jd900257>, 1999.

Liu, Q., Quan, J., Jia, X., Sun, Z., Li, X., Gao, Y., and Liu, Y.: Vertical Profiles of Aerosol Composition over Beijing, China: Analysis of In Situ Aircraft Measurements, *Journal of the Atmospheric Sciences*, 76, 231-245, <https://doi.org/10.1175/JAS-D-18-0157.1>, 2019.

How does long-range transport increase SAE? Generally, ageing and chemical processing during the long-range transport increases the size of the particles and reduces SAE. Moreover, ultrafine secondary particles have less residence time and they may not get transported to longer distances to increase SAE.

RE: Agree. In section 3.3.1, we indicate that the long-distance transport of coarse-mode aerosols (like dust particles) decreases SAE in the free troposphere.

Better association between CCN at high SS and scattering coefficients are expected because both CCN and scattering coefficients depend on the entire size distribution of the aerosol system. On the other hand, predicting CCN concentration for lower SS is challenging, since a small portion of the aerosol NSD (coarse mode) gets activated. Using the high-resolution data (1 sec), the authors should show the CCN vs scattering coefficients for low and high supersaturations.

RE: Agree. In the campaign, N_{CCN} at 0.7% SS is measured by a separate column of CCNc but not for other SS s. Therefore, the data of N_{CCN} at 0.7% SS is enough to do the closure study. However,

the samples of N_{CCN} at other SS are too few to obtain a meaningful closure result. The figure below depicts N_{CCN} closure test at other SS . Overall, the closure performance is similar to that at 0.7% SS but the number of data points at any SS is low.

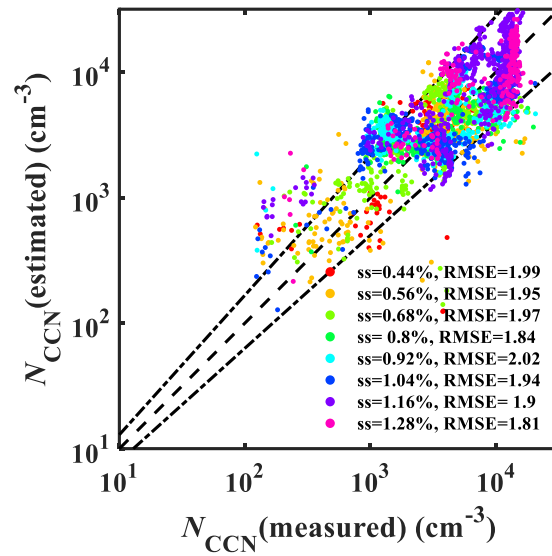
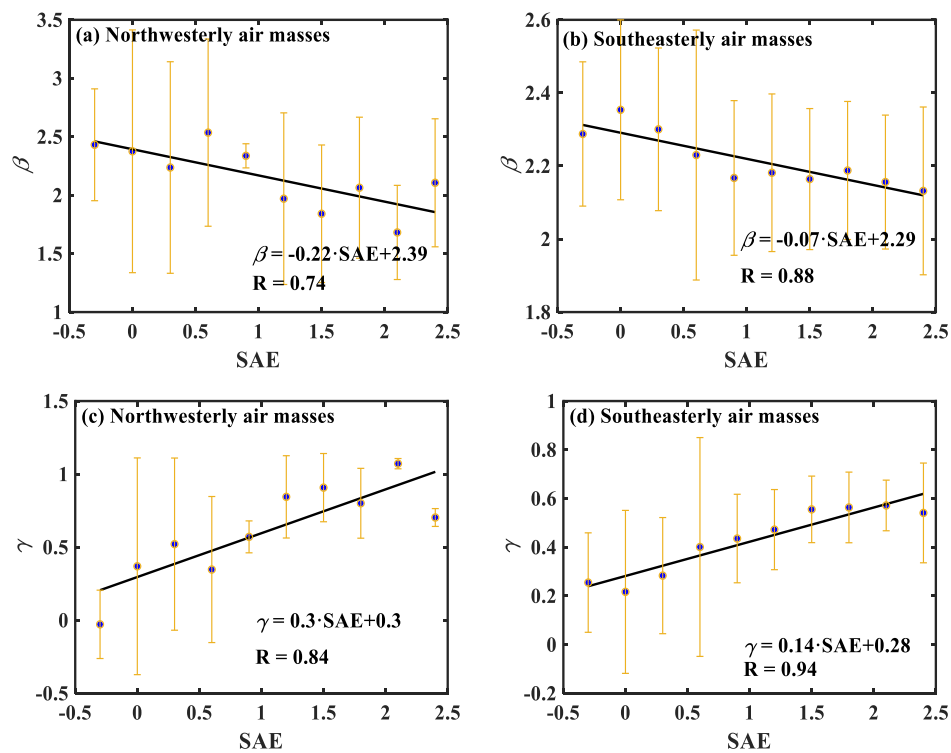


Figure 7: Standard deviation of the β and γ should be included.

RE: Agree. The updated figure is shown in the below. The figure has been removed in the supplement.



The β and γ showed better association with SAE during the southeast airmass period than the

northwest airmass. But the CCN estimated using b and g did not show good association for southeast airmass. Please explain this discrepancy.

RE: The data from two vertical spiral flights (RF1_1 and RF1_2) in northwesterly air masses are used to do the N_{CCN} closure work, while the data from five spiral flights (RF6_1, RF6_2, RF7_1, RF7_2, and RF8_1) in southeasterly air masses are used. More data in southeasterly air masses can be used, leading to better fitting for β and γ . However, the larger difference of N_{CCN} values between five spiral flights in southeasterly air masses worsen the closure performance. More aircraft data will be needed to establish a more reasonable parameterization scheme for N_{CCN} at different SS in the NCP.