Reply to RC1

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.

This manuscript presents the airborne cloud condensation nuclei (CCN) measurements taken during the ARIAs (Air chemistry Research In Asia) campaign. The authors use HYSPLIT trajectories to identify the source regions of different air masses measured during the campaign, and present the results separately for air masses coming from two main directions (northwest and southeast). They show the impact of atmospheric stability on the vertical distribution of CCN. Furthermore, they parametrize the number concentration of CCN (Nccn) in terms of aerosol optical properties. The manuscript presents a novel height resolved in-situ Nccn data and has good potential for publication in ACP only after implementing and addressing the following comments.

Lines 90-92, 'Tao et al. (2018) proposed … system'. I don't understand how this is related to the idea of this paragraph. Did they give any empirical relationship between Nccn and optical properties? If yes, then it should be stated.

RE: Yes, they gave the empirical relationship. This sentence is revised as: "Tao et al. (2018) established a lookup table that includes σ, hygroscopicity parameter (κ), and Ångström exponent ($\mathring{A}$) for estimating N_{CCN} based on the measurement of a three-wavelength humidified nephelometer system.".

Lines 92-93, 'Most of these… in situ Nccn profiles'. In atmospheric remote sensing, the word "profile" usually refers to a vertical representation. The parametrization schemes are mostly focused on estimating Nccn at ground. So there's no way one can compare/validate them with Nccn "profiles". I suggest replacing the word. Overall, I found the fourth paragraph of introduction to be confusing and suggest to modify it. It starts with the in situ Nccn "profile" measurements and the challenges involved in it. Thereafter how researchers have come up with empirical relations to estimate Nccn at "ground" using column integrated aerosol optical properties (AOD, AI, SAE). The ending sentence again discuss the how there's no validation with in situ Nccn "profile".

RE: That's a good suggestion. The fourth paragraph is revised as shown in the below.

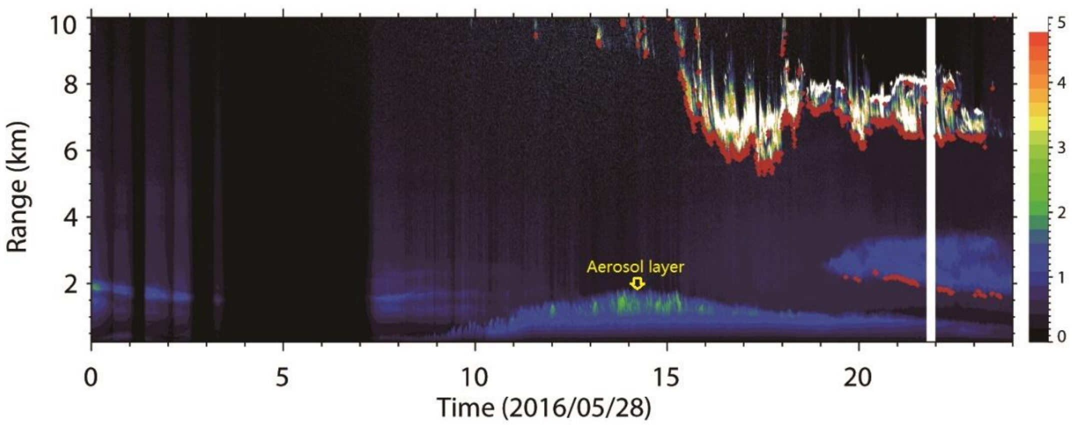
"A commonly used platform to observe vertical distributions of N_{CCN} and CCN activation ability is an aircraft (e.g., J. Li et al., 2015b; Jayachandran et al., 2020a; Manoj et al., 2021; Z. Cai et al., 2022). However, limited by high costs and technological complexity, current aircraft measurements are insufficient to quantify ACI. Some studies have thus attempted to estimate N_{CCN} using aerosol optical data that are much more plentiful (e.g., Andreae, 2009; Liu and Li, 2014; Tao et al., 2018). For example, Andreae (2009) built an exponential function between N_{CCN} and aerosol optical depth (AOD). Liu and Li (2014) found that the relationship between N_{CCN} and AOD becomes invalid when the relative humidity (RH) exceeds 75% and they developed new parameterized relationships to estimate N_{CCN} accounting for RH, particle size, and single scattering albedo (SSA). Tao et al. (2018) established a lookup table that includes σ, hygroscopicity parameter (κ), and Ångström exponent ($\mathring{A}$) for estimating N_{CCN} based on the measurement of a three-wavelength humidified nephelometer system. The vertical distributions of N_{CCN} were also estimated using lidar data. Lv et al. (2018) developed an algorithm for profiling N_{CCN} using backscatter coefficients at 355, 532, and 1,064 nm and extinction coefficients at 355 and 532 nm from multiwavelength lidar systems. Satellite lidar data of the Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) have also been employed to retrieve the profiles of lidar of CCN (Mamouri and Ansmann, 2016; Choudhury and Tesche 2022). Most of the retrieved N_{CCN} profiles are yet to be validated against in situ N_{CCN} profile measurements."

Lines 100-104. The manuscript presents vertical distribution of Nccn for different regions within the NCP. Currently, we have satellite-based Nccn retrieval algorithms, for instance, Mamouri and Ansmann (2016) and Choudhury and Tesche (2022), to estimate profiles of Nccn from CALIPSO measurements. The in-situ measurements presented here will also be beneficial in validating such algorithms. This information is missing in the motivation.

RE: The sentence: "The in-situ measurements presented here are beneficial in validating lidar- or satellite-based N_{CCN} retrieval algorithms (e.g., Choudhury and Tesche, 2022)." is added.

Lines 242-243: The Nccn values first increases till the base of the first temperature inversion layer (TIL). It is quite strange as the Nccn in the previous case with one TIL were more or less uniform below the layer, perhaps due to vertical mixing, which is not seen for this case with two inversion layers. Is there a possible reason behind this pattern?

RE: This is because the vertical mixing and the terrain effect make aerosol accumulate below the planetary boundary layer (PBL) on some days, which can be seen on the lidar image of a micro-pulse lidar (MPL) deployed during our field campaign at the Xingtai (XT) supersite. Unfortunately, the MPL data are missing during the RF1_1 on May 8, 2016. Figure S3h in the supplement suggests that N_{CCN} also increases with height below the planetary boundary layer (PBL) during the RF8_1 on May 28, 2016. The MPL image shown below indicates that aerosol accumulates obviously in the upper PBL in the daytime. According to our measurements, this phenomenon has no relationship with TIL amount.



MPL image on May 28, 2016 at Xingtai (XT) supersite.

Table 1. As the flights measurements are taken in a spiral path, please mention the maximum horizontal distance covered by individual flight segments chosen in this study. This is important as you consider them as individual profiles later in the paper.

RE: The maximum spiral radius during every vertical spiral flight is added in Table 1. The updated table is 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, 20160521	12:04–14:41	RF6_1	QZ	0.3–3.1	36	~ 5.0
		RF6_a	track from QZ to XT	~2.5	18	–
		RF6_2	XT	0.3–2.6	43	~ 5.0
		RF6_b	track from XT to LC	~1.1	13	–
RF7, 20160528	10:21–13:25	RF7_a	track around XT	~3.1	20	–
		RF7_1	XT	0.5–3.1	49	~ 5.0
		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, 20160528	16:30–18:24	RF8_a	track around XT	~0.6	15	–
		RF8_1	XT	0.5–3.1	36	~ 5.0
RF11, 20160611	11:07–12:28	RF11_a	track around XT	~0.6	16	–
		RF11_1	XT	0.3–3.2	50	~ 4.0

Some important technical information are missing. Did you smooth the flight measurements before the analysis? The pre-processing done to the measurements should be discussed in Section 2. Please also provide the uncertainty or retrieval errors associated with the in-situ measurements.

RE: The CCNc data with instable sample or sheath flow are excluded. Considering the time reaching equilibrium at different *SS* levels, data acquired in the final 30 s at any *SS* level are used. The measurements of temperature (T) and potential temperature (θ) are averaged in the intervals of 50 m in altitude. No other smoothing is applied. The uncertainty in nephelometer data is less than 10% (Anderson et al., 1996; Anderson and Ogren, 1998). The uncertain of effective water vapor supersaturation in CCNc is less than 5% (Rose et al., 2008).

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 in the supplement.

Reference:

Anderson, T. L., Covert, D. S., Marshall, S. F., Laucks, M. L., Charlson, R. J., Waggoner, A. P., Ogren, J. A., Caldow, R., Holm, R. L., Quant, F. R., Sem, G. J., Wiedensohler, A., Ahlquist, N. A., and Bates, T. S.: Performance Characteristics of a High-Sensitivity, Three-Wavelength, Total Scatter/Backscatter Nephelometer, Journal of Atmospheric and Oceanic Technology, 13, 967-986, https://doi.org/10.1175/1520-0426(1996)013<0967:PCOAHS>2.0.CO;2, 1996.

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.

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.

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.

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.

Lines 346-351: The definition and expression of scattering Ångström exponent should not be included in the "Results and Discussion" section. Please place it either in Section 2 or create a separate section.

RE: Agree. It is moved to Section 2.

Line 371: The section title is misleading. It is not the estimation of NCCN. It is where you parametrize NCCN in terms of aerosol optical properties. Please modify it.

RE: The section title is revised as: "Parametrizing N_{CCN} in terms of aerosol optical properties".

Lines 379-383: Please refer Shinozuka et al. (2015) and correct the statements. Shinozuka et al. (2015) parameterize NCCN in terms of "extinction coefficient (at 500 nm)" and "Angstrom exponent" (calculated from extinction coefficients at 450 and 550 nm) for dry particles. They did not use scattering coefficient or scattering Angstrom exponent for the same. Also for equation 3, it should be stated that in Shinozuka et al. (2015), only the parameter β depends on the Angstrom exponent (computed from extinction coefficients).

RE: Shinozuka et al. (2015) identified N_{CCN} at 0.4±0.1% SS with $10^{0.3\alpha}\sigma_{\mathrm{ext}}^{0.75}$ where σ_{ext} is the 500 nm extinction coefficient by dried particles and α is the extinction Angstrom exponent. They determined the slope in $\log_{10}N_{CCN}$ vs. $\log_{10}\sigma_{ext}$ to be constant based on a variety of airborne and ground-based observations. However, we found that the slope varies with the extinction Angstrom exponent at some sites (such as the site of Black Forest, Germany) form Fig. 3a and Table 2 in Shinozuka et al. (2015). Therefore, we used the modified parameterization. The sentence is revised as: "Shinozuka et al. (2015) identified N_{CCN} at 0.4±0.1% *SS* with $10^{0.3\alpha}\sigma_{ext}^{0.75}$ where σ_{ext} is the 500 nm extinction coefficient by dried particles and α is the extinction Angstrom exponent. According to our measurements, a modified parameterization is used in this study:

$$N_{CCN}=10^{\beta}\cdot\sigma^{\gamma} \quad (4)$$ ".

Lines 387-388: Coefficient of determination or R2 and correlation are synonymously used. R2 quantifies the goodness of fit (here linear fit) or performance of the model (here linear model) in simulating the variable of concern (here fitting parameters β and γ). I suggest using either correlation coefficient or slope of the linear fit. Also, is Figure 7 really important to include in the manuscript? I would suggest omitting the figure. If the authors want to retain it, they should justify the significance of the observed relations between SAE and the fitting parameters.

RE: Shinozuka et al. (2015) investigated the relationships of the slope (β) and intercept (γ) with the extinction Angstrom exponent (α) shown in their Fig. 3. Following their methods, we also analyze these relationships in our study. The sentence is revised as: "The correlation coefficients (R^2) are lower in northwesterly air masses than in southeasterly air masses, likely due to more complex aerosol sources in northwesterly air masses." The figure is moved to the supplement.

Figure 7 (if retained) and Figure 8 should include the total number of points used in the comparison. I also suggest including two more lines in the figure representing one order of magnitude more and less than the 1:1 line in Figure 8 for better visualization.

RE: Agree. The updated figure is shown in the below.

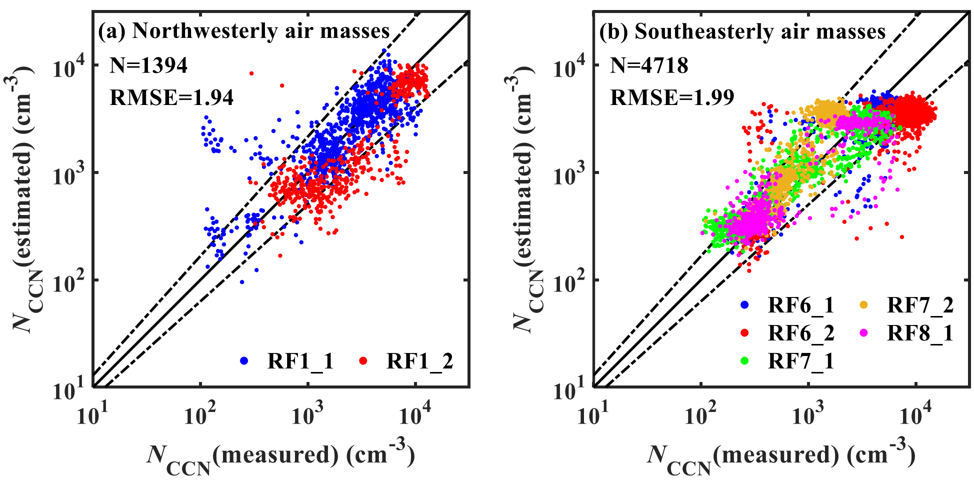


Figure 7. Comparisons between measured N_{CCN} at 0.7% *SS* and estimated N_{CCN} at 0.7% *SS* using Eqs. (4) and (5) for different vertical spiral flights in (a) northwesterly and (b) southeasterly air masses. The black solid lines are 1:1 line and the dash lines indicate the boundaries representing ±10% deviations of N_{CCN} (estimated) from N_{CCN} (measured) in the log-log plot. The 10% deviation means that the deviation of individual data points is typically within a factor of 1.26 of the best

estimates. The point number (*N*) and root mean square error (RMSE) in each panel are given.

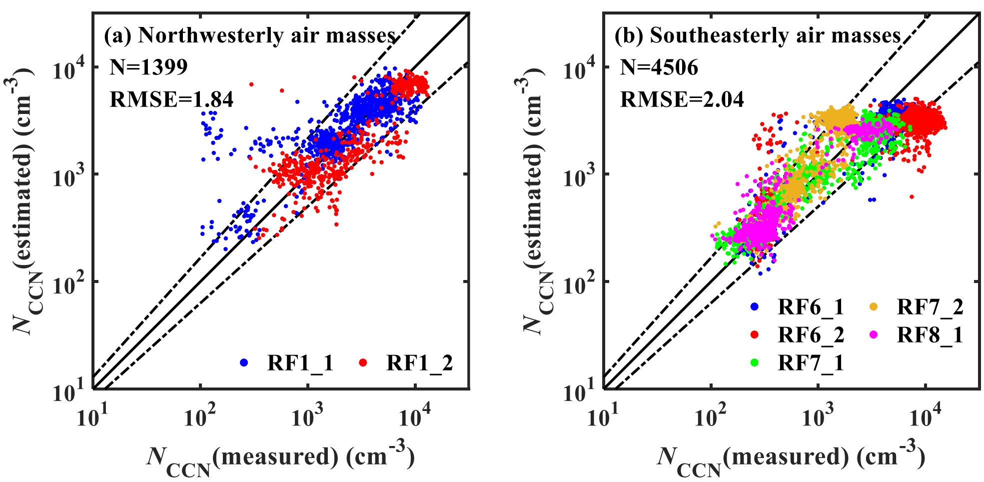
Lines 399-404: Qualitative interpretation from a log-log plot can be misleading. What seems to be different by a few millimeters in the plot can be different by orders of magnitude in reality. I suggest using parameters like normalized mean error or bias and root mean square error (normalized by mean) in percentage to get a better quantitative comparison. Such parameters should then be used to quantify the error associated with the proposed parametrization.

RE: The vertical variation of N_{CCN} is in five orders of magnitude from a few to tens of thousands per cubic centimeter. Therefore, a log-log plot is commonly used. The root mean square error (RMSE) is calculated and shown in the above figure.

Is there any specific reason why the authors use aerosol scattering coefficient instead of extinction coefficient (scattering + absorption). The authors identify anthropogenic emissions as one of the aerosol types in their analysis, which may also include absorbing aerosols. Using scattering coefficient in such scenarios may result in mis-representation of absorbing aerosols in the parametrization, which is perhaps one of the reasons behind the errors in the predicted Nccn.

RE: It is true that absorbing aerosols can also serve as CCN, but their hygroscopicity is generally weak, such as black carbon (BC). In this study, we find that N_{CCN} profile is impacted largely by anthropogenic emissions, especially air masses from the southeast. The impact of anthropogenic emissions does not only refer to primary processes. Our previous studies suggest that high concentration of gaseous precursors from anthropogenic emissions and strong atmospheric oxidization capacity lead to frequent new particle formation (NPF) and rapid particle growth in the NCP (Y. Wang et al., 2018, 2021; Zhang et al., 2018). These processes can produce many hydrophilic secondary aerosols (such as sulfate, nitrate, and so on), leading to the large increase of N_{CCN}. Moreover, the absorbing aerosols are much less than the scattering aerosols, which can be reflected from the value of single scattering albedo (SSA). F. Wang et al. (2018) reported that regional mean value of SSA at 550 nm in this campaign is 0.85±0.02.

The N_{CCN} closure results using the data of extinction coefficients to estimate N_{CCN} are shown below. The performance is similar with those using the data of scattering coefficients shown in the manuscript, indicating unimportant role of absorbing aerosols in the estimation of N_{CCN}.



Reference:

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.

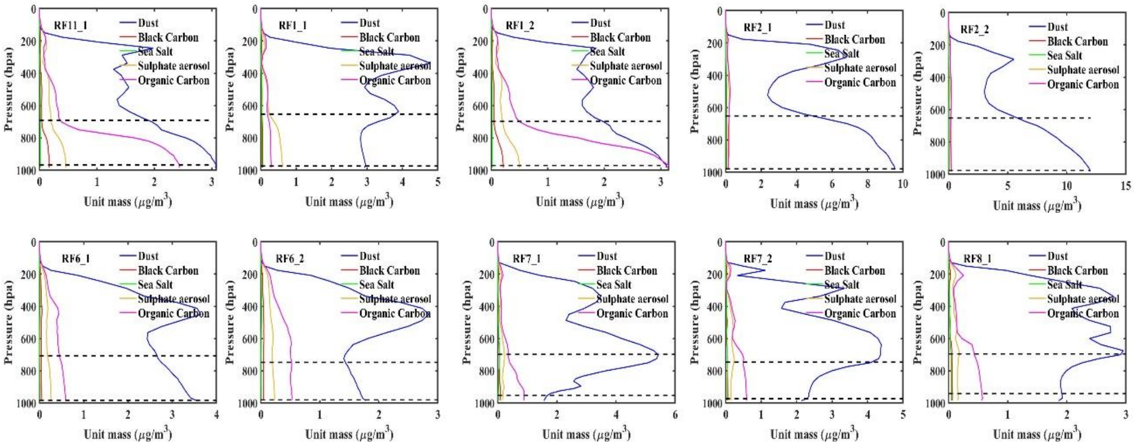
Wang, Y., Li, Z., Zhang, Y., Du, W., Zhang, F., Tan, H., Xu, H., Fan, T., Jin, X., Fan, X., Dong, Z., Wang, Q., and Sun, Y.: Characterization of aerosol hygroscopicity, mixing state, and CCN activity at a suburban site in the central North China Plain, Atmospheric Chemistry and Physics, 18, 11739-11752, https://doi.org/10.5194/acp-18-11739-2018, 2018.

Wang, Y., Wang, J., Li, Z., Jin, X., Sun, Y., Cribb, M., Ren, R., Lv, M., Wang, Q., Gao, Y., Hu, R., Shang, Y., and Gong, W.: Contrasting aerosol growth potential in the northern and central-southern regions of the North China Plain: Implications for combating regional pollution, Atmospheric Environment, 267, 118723, https://doi.org/10.1016/j.atmosenv.2021.118723, 2021.

Zhang, Y., Du, W., Wang, Y., Wang, Q., Wang, H., Zheng, H., Zhang, F., Shi, H., Bian, Y., Han, Y., Fu, P., Canonaco, F., Prévôt, A. S. H., Zhu, T., Wang, P., Li, Z., and Sun, Y.: Aerosol chemistry and particle growth events at an urban downwind site in North China Plain, Atmospheric Chemistry and Physics, 18, 14637-14651, https://doi.org/10.5194/acp-18-14637-2018, 2018.

For identifying the aerosol types in the analyzed samples, HYSPLIT back trajectory analysis is used to track the source regions and the regions through which the air parcels have passes before reaching the target. However, this is based on the assumption that the lifetime of aerosols is long enough to retain its source identity. One of the ways to crosscheck the aerosol types is to use CALIPSO aerosol product (CALIPSO, 2018) for the identified cases. If there is no CALIPSO overpass over the region of interest at the desired time, one can also use re-analysis datasets like CAMS (Inness et al., 2019) and/or MERRA-2 (Molod et al., 2015) to identify the aerosol types that are dominant at different height levels. I suggest using either one of these datasets to check if the assumed aerosol signatures are correct.

RE: HYSPLIT has been used in a variety of simulations describing the atmospheric transport, dispersion, and deposition of pollutants. CALIPSO aerosol product is not suitable for our study chiefly because of lack of overpass over our observation site. We check the data of aerosol chemical composition from MERRA-2 (shown in the below). The results have a big difference with our measurement by an Aerodyne aerosol chemical speciation monitor (ACSM) at XT supersite (Zhang et al., 2018). The patterns of aerosol profiles from MERRA-2 are also not consistent with our measurements. On the contrary, our measurement data can be used to validate MERRA-2.



The vertical profiles of aerosol chemical composition from MERRA-2

Reference:

Zhang, Y., Du, W., Wang, Y., Wang, Q., Wang, H., Zheng, H., Zhang, F., Shi, H., Bian, Y., Han, Y., Fu, P., Canonaco, F., Prévôt, A. S. H., Zhu, T., Wang, P., Li, Z., and Sun, Y.: Aerosol chemistry and particle growth events at an urban downwind site in North China Plain, Atmospheric Chemistry and Physics, 18, 14637-14651, https://doi.org/10.5194/acp-18-14637-2018, 2018.

Minor comments:

Please modify Figure 1 caption to include the meaning of RF1, RF2… RF11.

RE: The sentence "The number after 'RF' indicates the research flight number" is added in the Fig. 1 caption.

Line 128. Please include the word in bracket. … 182 m above [mean] sea level …

RE: The word is added.

Line 295. Remove the word in the bracket. "…profiles [ss] are influenced…"

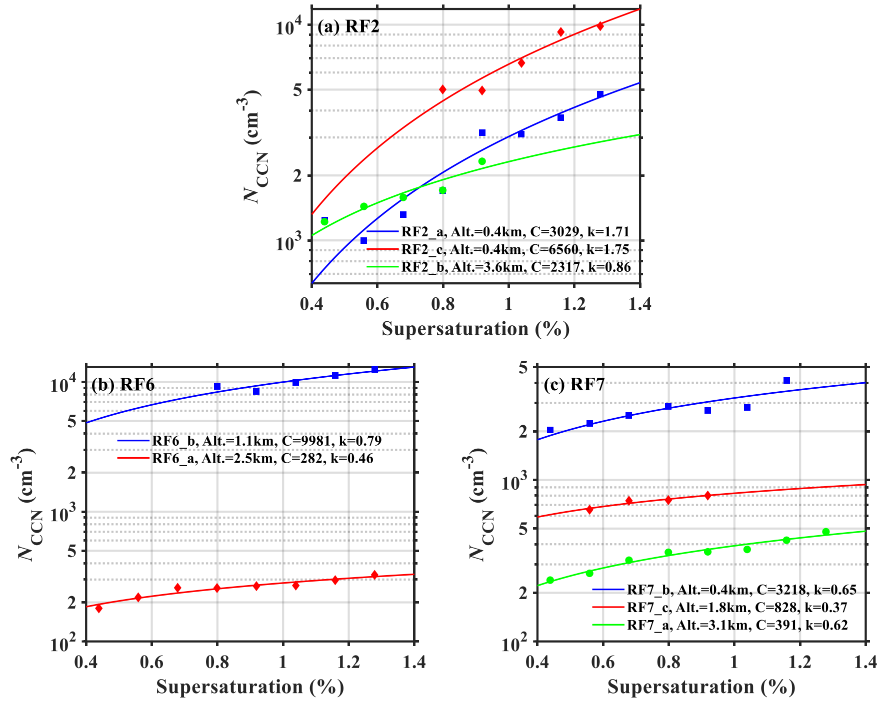
RE: The word is removed.

Lines 301-302. Rephrase the sentence to "Twomey (1959) first reported an exponential relationship between Nccn and ss."

RE: It is revised. Thanks.

In Figure 5, please mention that the y-axis is in log-scale. Please mark at least two (or three, if possible) tick labels in the y-axis of each plot.

RE: The sentence "The y-axis is logarithmic." is added in the Figure caption. The updated figure is shown in the below.



Line 345. The acronym "SAE" is previously defined in the paper. There is no need to define it again here.

RE: It is revised. Thanks.

Lines 349-351. Replace the word "dominated" by "are dominant".

RE: It is revised. Thanks.

References:

CALIPSO: Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation Lidar Level 2 Aerosol Profile, V4-20, NASA Langley Atmospheric Science Data Center DAAC [data set], https://doi.org/10.5067/CALIOP/CALIPSO/LID_L2_05KMAPRO-STANDARD-V4-20, 2018.

Choudhury, G. and Tesche, M.: Estimating cloud condensation nuclei concentrations from CALIPSO lidar measurements, Atmos. Meas. Tech., 15, 639–654, https://doi.org/10.5194/amt-15-639-2022, 2022.

Inness, A., Ades, M., Agustí-Panareda, A., Barré, J., Benedictow, A., Blechschmidt, A.-M., Dominguez, J. J., Engelen, R., Eskes, H., Flemming, J., Huijnen, V., Jones, L., Kipling, Z., Massart, S., Parrington, M., Peuch, V.-H., Razinger, M., Remy, S., Schulz, M., and Suttie, M.: The CAMS reanalysis of atmospheric composition, Atmos. Chem. Phys., 19, 3515–3556, https://doi.org/10.5194/acp-19-3515-2019, 2019.

Mamouri, R.-E. and Ansmann, A.: Potential of polarization lidar to provide profiles of CCNand INP-relevant aerosol parameters, Atmos. Chem. Phys., 16, 5905–5931, https://doi.org/10.5194/acp-16-5905-2016, 2016.

Molod, A., Takacs, L., Suarez, M., and Bacmeister, J.: Development of the GEOS-5 atmospheric general circulation model: evolution from MERRA to MERRA2, Geosci. Model Dev., 8, 1339–1356, https://doi.org/10.5194/gmd-8-1339-2015, 2015.