

We provide detailed responses to all comments below; the responses are shown in blue, and all revisions in the manuscript are highlighted in red.

Comment to “Vertical structure and controlling factors of cloud condensation nuclei activation over eastern China: Insights from aircraft measurements and interpretable machine learning”

This study examines the factors associated with variations in cloud condensation nuclei (CCN) number concentrations across different altitudes, aerosol vertical structures, and supersaturation conditions. A key strength of the work is the application of nonlinear multivariate analysis using Generalized Additive Models (GAMs), together with SHapley Additive exPlanations (SHAP), an approach used to quantify the contribution of individual variables to model predictions, to aircraft observations. These methods allow the authors to evaluate the relative importance of multiple aerosol and meteorological variables while accounting for nonlinear relationships. The analysis provides useful insight into the factors associated with vertical CCN variability and contributes to the broader understanding of aerosol-cloud interactions. However, I have several comments regarding the methodology and interpretation of the results that should be addressed before publication.

We sincerely thank the reviewer for the positive evaluation of our study and for recognizing the value of combining aircraft observations with GAM and SHAP analyses to investigate factors associated with vertical CCN variability. We also appreciate the reviewer’s constructive comments regarding the methodology and interpretation of the results. We have carefully addressed all comments and revised the manuscript accordingly, with particular attention to clarifying variable definitions, improving the description of vertical CCN variability, justifying the aerosol-structure classification criteria, refining the interpretation of SHAP results, and more clearly discussing the potential covariability between temperature and altitude. We believe these revisions have substantially improved the clarity and robustness of the manuscript.

Abstract.

Several terms in the abstract should be more clearly defined. Specifically, the “Decrease,” “Increase,” and “Stable” aerosol vertical structures are introduced without explaining what these classifications represent or how they are defined. In addition, “small-size aerosol concentrations (SA)” is too vague; please specify the particle diameter range included in this definition here.

We thank the reviewer for this helpful comment. We agree that these terms should be defined when they first appear in the Abstract. We have therefore clarified that the “Increase,” “Decrease,” and “Stable” aerosol vertical structures refer to aerosol number concentration profiles that increase, decrease, or remain relatively stable with altitude, respectively. The detailed quantitative classification criteria are provided in the Methods section. We have also specified that small-size aerosol number concentration (SA) represents particles with diameters of 0.1–1 μm .

Line 25: higher small-size aerosol concentrations (SA, 0.1–1 μm)

Line 16: Cloud condensation nuclei (CCN) play a critical role in cloud droplet formation and microphysical processes. Based on aircraft observations, this study investigated the factors controlling CCN number concentrations (NCCN) under different aerosol vertical structures (Decrease, Increase, and Stable, representing decreasing, increasing, and relatively stable aerosol concentrations with altitude, respectively) and supersaturation (SS) conditions using generalized additive models (GAM) combined with SHapley Additive exPlanations (SHAP).

Line 24-26. “GAM results identified temperature (T), SA, relative humidity (RH), and horizontal wind speed (WS) as important explanatory variables for NCCN variations, with contributions of 20%–56%, 9%–45%, 9%–40%, and 3%–19%, respectively.”

The use of “respectively” here is difficult to follow given the number of variables and contribution ranges. Consider restructuring the sentence to pair each variable directly with its corresponding range for clarity.

We thank the reviewer for this suggestion. We have revised the sentence by directly pairing each explanatory variable with its corresponding relative SHAP contribution range, which improves clarity and avoids the use of “respectively.”

Line 25: GAM results identified several important explanatory variables for NCCN variations, with relative SHAP contributions of 19%–57% for temperature (T), 9%–45% for SA, 9%–40% for relative humidity (RH), and 3%–32% for horizontal wind speed (WS).

Line 42.

Please revise the citation formatting when references are incorporated into the sentence. For example, “(Prabhakaran et al., 2025) found that...” should be written as “Prabhakaran et al. (2025) found that...”. Please check for and correct this citation formatting throughout the manuscript.

We thank the reviewer for pointing this out. We have corrected the citation format where references are incorporated grammatically into the sentence (e.g., “Prabhakaran et al. (2025) found that...”), and have carefully checked the manuscript to ensure consistent citation formatting throughout.

Line 68: For example, Prabhakaran et al. (2025) found that under a warming climate, aerosol perturbations influence the stratocumulus-to-cumulus transition (SCT) over the northeastern Pacific;

Line 47. “...(2003)observed...”

Change to “...(2003) observed...”. Please check that there is a space between citations and the surrounding text throughout the manuscript (also seen in Line 68).

We thank the reviewer for pointing this out. The missing spaces around citations have been corrected, including “(2003) observed” and the similar issue in Line 68. We have also checked the entire manuscript to ensure consistent spacing between citations and the surrounding text.

Line 73: Borys et al. (2003) observed that high anthropogenic aerosol concentrations substantially modified the microphysical structure of lower “feeder” layers in orographic clouds

Line 65. “... T structure...”

Please define temperature (T) at its first occurrence in the main text. Although T is defined in the abstract, abbreviations should also be defined when first introduced in the main text.

We thank the reviewer for pointing this out. We have defined temperature (T) at its first occurrence in the main text and checked the manuscript to ensure that abbreviations are defined when first introduced.

Line 102: For example, Zhang et al. (2022) found that over the North China Plain, the vertical distribution of NCCN was jointly influenced by temperature(T) structure, air-mass origin, anthropogenic emissions, and topography.

Line 279: Relative humidity (RH)

Line 281: Horizontal wind speed (WS)

Line 81.

Ren et al. (2025) is missing from the reference list.

We thank the reviewer for pointing out this omission. The reference for Ren et al. (2025) has now been added to the reference list, and we have also checked the manuscript to ensure that all citations in the text are included in the reference list.

Line 756: Ren, J., Zou, S., Xu, H., Liu, G., Wang, Z., Zhang, A., Zhao, C., Hu, M., Shang, D., Tang, L., Huang, R.-J., Sun, Y., Zhang, F., 2025. Machine learning significantly improves the simulation of hourly-to-yearly scale cloud nuclei concentration and radiative forcing in polluted atmosphere. EGU sphere 1–32. <https://doi.org/10.5194/egusphere-2025-1483>

Line 184. “...while SA, MA, and LA were...”

Please define all these abbreviations here instead of Line 306. SA is only defined in abstract.

We thank the reviewer for pointing this out. We have now defined the aerosol size categories at their first occurrence in the main text as small aerosols (SA, 0.1–1 μm), medium aerosols (MA, 1–2 μm), and large aerosols (LA, 2–3 μm). The corresponding definitions later in the manuscript have been adjusted to avoid repetition.

Line 246: To reduce multicollinearity among aerosol variables, total aerosol number concentration (Na) was excluded, while small-size aerosol number concentration (SA, 0.1–1 μm), medium-size aerosol number concentration (MA, 1–2 μm), and large-size aerosol number concentration (LA, 2–3 μm) were retained to represent size-resolved aerosol populations, improving interpretability and avoiding redundancy.

Lines 210-220.

The description of the CCN activation ratio as exhibiting an “increase-decrease-increase” pattern is overly qualitative. Please specify the relevant altitude ranges and quantify the corresponding range and magnitude of changes in the activation ratio across these altitudes. This would provide a clearer assessment of the vertical variability than simply describing the direction of change.

We thank the reviewer for this helpful comment. We agree that the original description of the vertical variation in the CCN activation ratio was too qualitative. We have therefore revised this section to provide the approximate altitude ranges over which the activation ratio generally increased, decreased, and increased again for each supersaturation level. We also retained the quantitative activation-ratio ranges around ~4000 m (3%–8% for $\text{SS} \leq 0.6\%$ and 21%–35% for $\text{SS} > 0.6\%$). Because local deviations occur within individual altitude intervals, these altitude ranges are described as approximate transitions in the overall composite profiles rather than strict boundaries. The revised text now provides a more quantitative description of the observed vertical variability.

Line 301: When observations from all flight missions were grouped by altitude and analyzed separately for each SS level (Fig. S2), CCN activation ratios generally exhibited an increase–decrease–increase pattern with altitude, although local deviations from this overall tendency were present. At $\text{SS} = 0.2\%$ and 0.6% , activation ratios generally increased below approximately 2000 m, decreased toward ~4000 m, and then increased again at higher altitudes. A similar pattern was observed at $\text{SS} = 0.4\%$, with the transition from increasing to decreasing occurring near ~1500 m. At higher SS, the initial increasing tendency extended to higher altitudes, reaching approximately 2500 m at $\text{SS} = 0.8\%$ and ~3000 m at $\text{SS} = 1.0\%$, followed by a decrease toward ~4000 m and a subsequent increase above this level. Relatively low activation ratios were observed around 4000 m in the composite profiles (3%–8% for $\text{SS} \leq 0.6\%$ and 21%–35% for $\text{SS} > 0.6\%$). Overall, activation ratios under $\text{SS} \geq 0.8\%$ were higher than those under lower supersaturation conditions. It should be noted that the CCNC operated sequentially across different supersaturation levels during each flight, resulting in partial coupling between altitude and supersaturation. Therefore, the vertical variations presented here represent composite statistics based on the observations available at each SS level rather than strictly altitude-controlled comparisons among supersaturation conditions. Accordingly, differences among SS levels may partly reflect variations in sampling altitude and flight trajectory and should be interpreted with appropriate caution.

Lines 224-226.

The criteria used to classify adjacent bins as Stable ($<100 \text{ cm}^{-3}$ absolute difference and $<20\%$ relative change) appear somewhat arbitrary. Please provide justification for these threshold values, including whether they are based on instrument uncertainty, prior literature, or the distribution of the observations.

We thank the reviewer for this important comment. We agree that the basis for these thresholds should be clarified. The thresholds were not directly taken from instrument uncertainty or previous literature; rather, they were developed empirically from the present observational dataset.

Specifically, we first examined the distribution and vertical profiles of aerosol number concentration and identified the magnitude of changes associated with visually apparent increasing, decreasing, and relatively stable segments. Based on these observed characteristics, an absolute change of $<100 \text{ cm}^{-3}$ together with a relative change of $<20\%$ was adopted as a data-driven criterion for identifying Stable segments. This criterion was intended to distinguish small fluctuations from more pronounced vertical trends and to ensure that the objective classification was broadly consistent with the structures evident in the observed profiles. We have clarified this rationale in the revised manuscript.

Line 329: To investigate the impact of aerosol vertical structure on CCN activation characteristics, aerosol number concentration profiles were divided into consecutive vertical trend segments according to changes between adjacent altitude bins (Fig. S7). The classification thresholds were empirically determined from the present observational dataset based on the distribution of aerosol number concentration and visual inspection of the vertical profiles. Specifically, adjacent bins were classified as Stable when the absolute difference in aerosol number concentration was $<100 \text{ cm}^{-3}$ and the relative change was $<20\%$, which was intended to distinguish relatively small fluctuations from more pronounced vertical trends. Changes exceeding these thresholds were classified as Increase or Decrease according to the direction of the change. The resulting segments were subsequently grouped into the three aerosol vertical structure categories.

Lines 256-268. “Fig. 4 shows that the size distributions of CCN-activated droplets measured by the CCNC are generally unimodal. The modal diameter is approximately $2 \mu\text{m}$ under low SS conditions (0.2 and 0.4), whereas it shifts to approximately $5 \mu\text{m}$ under higher SS conditions (0.8 and 1.0).”

The droplet size distributions do not appear strictly unimodal in all cases. Particularly at lower SS, there appears to be a smaller mode or shoulder at $<2 \mu\text{m}$ in addition to a broader population at $2\text{--}3 \mu\text{m}$. Could this mode becoming less apparent at higher SS also be related to the enhanced condensational growth described later in this section?

We thank the reviewer for this careful observation. We agree that the CCNC droplet size distributions are not strictly unimodal under all supersaturation conditions. At lower SS, a shoulder or weak secondary population can indeed be observed below approximately $2 \mu\text{m}$ in addition to the dominant population at larger diameters. We have therefore revised the description from “unimodal distributions” to “distributions dominated by a main mode, with a

small-particle shoulder under some low-SS conditions.”

The reduced prominence of this small-particle shoulder at higher SS is consistent with stronger condensational growth within the CCNC. As SS increases, activated droplets can grow to larger diameters before detection, causing the dominant mode to shift toward larger sizes and making the smaller-diameter population less apparent. We have added this interpretation to the revised manuscript while avoiding a strict causal attribution.

Line 365: Fig. 4 shows that the size distributions of CCN-activated droplets measured by the CCNC are generally dominated by a main mode, although a small-particle shoulder below $\sim 2\ \mu\text{m}$ is apparent under some low-SS conditions. The dominant modal diameter is approximately $2\text{--}3\ \mu\text{m}$ at low SS (0.2% and 0.4%) and shifts toward $\sim 5\ \mu\text{m}$ at higher SS (0.8% and 1.0%). The reduced prominence of the small-particle shoulder and the shift toward larger diameters at higher SS are consistent with enhanced condensational growth of activated droplets within the CCNC, rather than changes in the critical activation diameter of aerosol particles. Under higher SS conditions, smaller aerosol particles can also be activated, while stronger water vapor supersaturation promotes faster condensational growth after activation. For instance, in F1 (Fig. 4a–e), the modal diameter at SS = 0.2% is approximately $2\ \mu\text{m}$ for all three aerosol vertical structures, whereas at SS = 1.0% it increases to 5.5, 5.5, and $4.5\ \mu\text{m}$ for the Increase, Stable, and Decrease structures, respectively.

Line 305-306. “...medium-size aerosol numberconcentration (MA), and large-size aerosol number concentration (LA)...”

Please define SA when it is first introduced in the main text and provide the particle size ranges corresponding to SA, MA, and LA. Currently, SA is only written out in the abstract, and the size ranges for these categories are only provided in Fig. 5.

We thank the reviewer for this comment. We have now defined the aerosol size categories at their first occurrence in the main text as small aerosols (SA, $0.1\text{--}1\ \mu\text{m}$), medium aerosols (MA, $1\text{--}2\ \mu\text{m}$), and large aerosols (LA, $2\text{--}3\ \mu\text{m}$). The corresponding size ranges are therefore explicitly provided in the text rather than only in Fig. 5.

Line 246: To reduce multicollinearity among aerosol variables, total aerosol number concentration (Na) was excluded, while small-size aerosol number concentration (SA, $0.1\text{--}1\ \mu\text{m}$), medium-size aerosol number concentration (MA, $1\text{--}2\ \mu\text{m}$), and large-size aerosol number concentration (LA, $2\text{--}3\ \mu\text{m}$) were retained to represent size-resolved aerosol populations, improving interpretability and avoiding redundancy.

Line 346-349. “Specifically, in the Decrease and Stable scenarios, T exhibits the highest contribution, exceeding 25% in both cases. In the Increase scenario, T’s contribution ranking decreases under certain SS conditions but still remains above 15%, suggesting that T maintains high importance across different scenarios and is closely associated with variations in NCCN”

Figure S1 shows that CCN number concentration generally decreases with altitude, as also

discussed previously in the manuscript. Because temperature is also expected to vary systematically with altitude, please address the potential covariability between altitude and temperature and how this may affect the inferred importance of temperature for CCN number concentration. In particular, the high contribution attributed to temperature may partly reflect the vertical dependence of CCN number concentration rather than a direct temperature effect. This is only discussed in the conclusion. If sufficient data are available, consider including altitude as an additional model variable to help separate these effects.

We thank the reviewer for this important comment. We agree that the strong covariability between temperature and altitude needs to be explicitly considered when interpreting the relatively high SHAP importance of temperature. In the full cloud-free dataset, temperature was strongly negatively correlated with altitude (Pearson $r = -0.950$ and Spearman $\rho = -0.954$), confirming that temperature largely reflects the vertical thermodynamic structure sampled by the aircraft.

Because including altitude together with temperature in the same GAM would introduce substantial collinearity and would not necessarily allow their individual effects to be cleanly separated, we instead conducted an additional sensitivity analysis using a near-constant-altitude segment at approximately 5.8 km. Under $SS = 0.6\%$, this subset contained 506 observations spanning only 17 m in altitude but 2.27 °C in temperature. Under these near-constant-altitude conditions, the relative SHAP importance of temperature decreased compared with that obtained from the full dataset, while the SHAP dependence on temperature still showed an overall positive tendency.

These results indicate that the high apparent importance of temperature in the full dataset is partly associated with its strong covariance with altitude and the corresponding vertical variation in NCCN. However, the persistence of a temperature-related signal under near-constant-altitude conditions suggests that this association is not explained solely by the large-scale vertical temperature gradient. We have added these analyses to the revised manuscript and now emphasize that the SHAP results represent statistical associations rather than direct causal effects of temperature.

Line 250: To assess the influence of temperature–altitude coupling inherent in aircraft observations, an additional sensitivity analysis was conducted. First, the temperature–altitude relationship was quantified for the full cloud-free dataset using Pearson and Spearman correlation coefficients, both for the combined dataset and for the three aerosol vertical-structure categories. Second, a quasi-level flight segment near 5.8 km altitude was selected, in which altitude remained nearly constant while temperature still varied measurably. Summary statistics of altitude, temperature, and CCN concentration were calculated separately for each supersaturation level. The temperature–altitude correlations within this subset were also evaluated for each supersaturation level. Among these subsets, the $SS = 0.6\%$ data were selected for a GAM/SHAP sensitivity analysis because they provided the largest sample size and the widest temperature range while maintaining very small altitude variability. The results were compared with those obtained from the full dataset.

Line 504: Because temperature and altitude are physically linked in aircraft observations, we further evaluated the robustness of the temperature–NCCN relationship through a sensitivity

analysis. In the full cloud-free dataset, temperature was strongly negatively correlated with altitude, with Pearson $r = -0.950$ and Spearman $\rho = -0.954$ (Fig.S30). Similar negative correlations were found within the Increase, Stable, and Decrease structure categories, although the correlation strength varied among them. These results confirm that the high importance of temperature in the full-dataset GAM/SHAP analysis must be interpreted with caution.

Line 511: To reduce the influence of the large-scale vertical temperature gradient, a quasi-level flight segment near 5.8 km altitude was selected. In this segment, the altitude range was only 10–17 m depending on supersaturation level, whereas temperature still varied by about 0.9–2.3 °C. The temperature–altitude correlations within this subset were much weaker and no longer systematically negative, indicating that the original large-scale coupling was substantially mitigated. Among the five supersaturation levels, the SS = 0.6% subset was used for the GAM/SHAP sensitivity analysis because it had the largest sample size ($N = 506$) and the widest temperature range (2.27 °C) while maintaining very small altitude variability. In this near-constant-altitude subset, the relative SHAP importance of temperature decreased compared with that in the full dataset (Fig. S32), but the SHAP dependence still showed an overall increase with temperature (Fig.S33). This suggests that altitude-related covariation explains part, but not all, of the temperature importance identified in the full dataset. Nevertheless, this result should still be interpreted as a statistical association rather than a direct causal temperature effect.

Line 576: A sensitivity analysis based on a quasi-level flight segment further showed that the importance of temperature decreased after substantially reducing altitude variability, although a positive temperature–NCCN association remained. This indicates that part of the temperature importance in the full dataset is related to altitude-associated covariation, while the remaining relationship should still be interpreted as a statistical association rather than a direct causal effect.

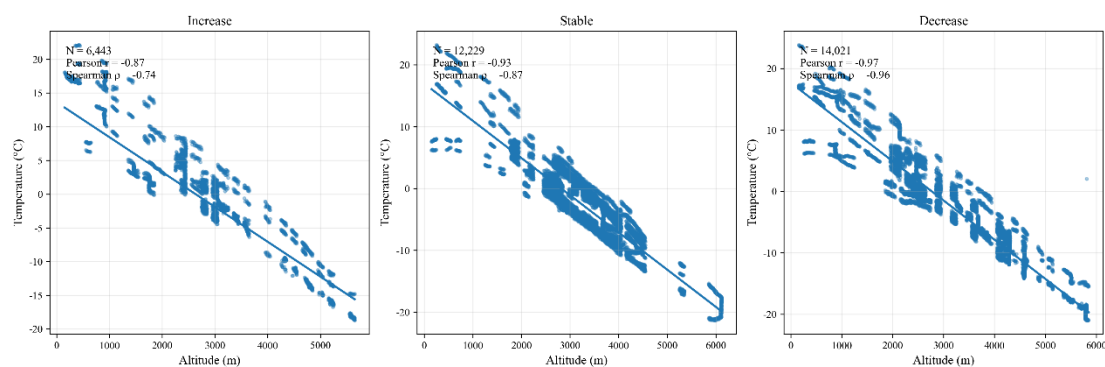


Fig.S30. Scatter plots of temperature versus altitude under different aerosol vertical structures, with the corresponding Pearson and Spearman correlation coefficients.

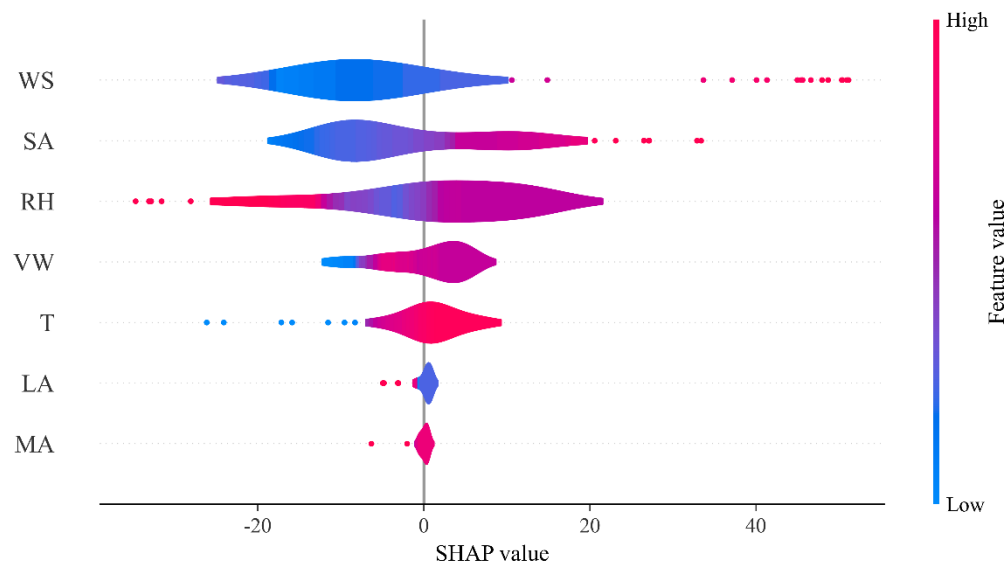


Fig.S31. SHAP violin summary plots from the near-constant-altitude sensitivity analysis.

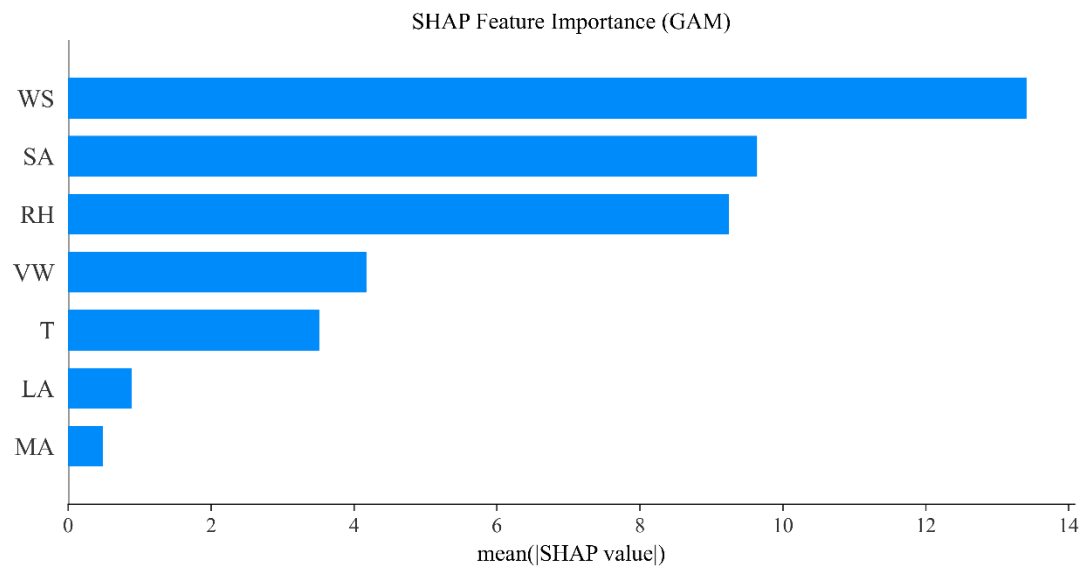


Fig.S32. Relative SHAP contributions and ranking of different predictors in the near-constant-altitude sensitivity analysis.

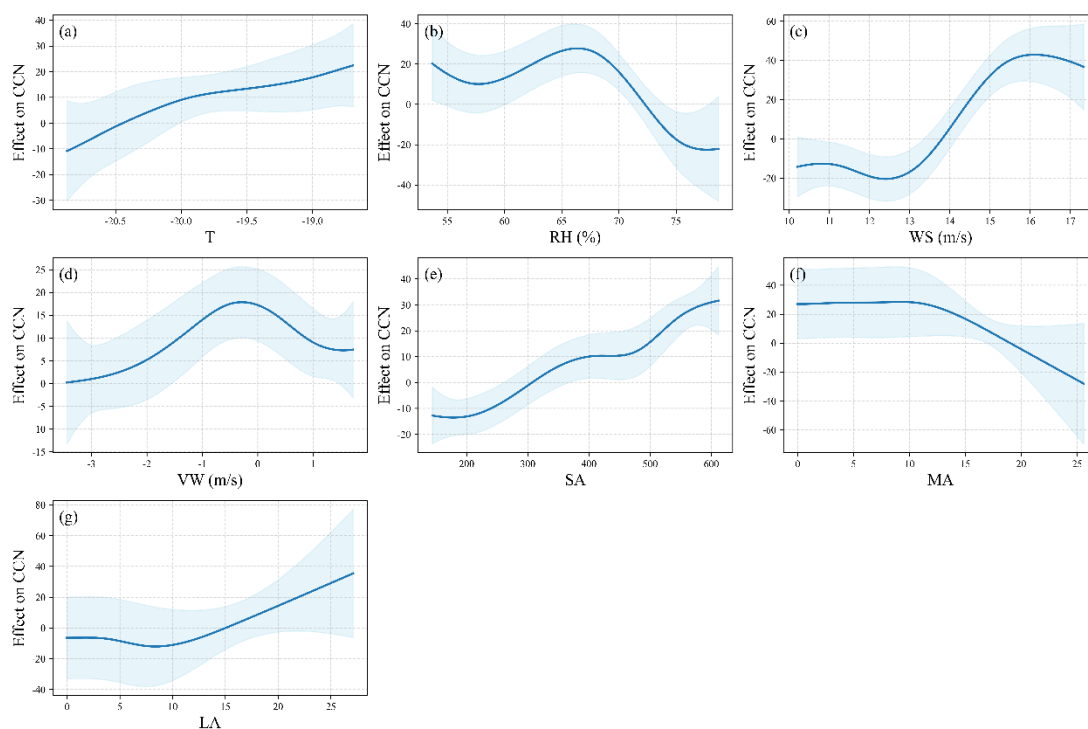


Fig.S33. SHAP dependence plots for different predictors in the near-constant-altitude sensitivity analysis.

Figure S9.

It is difficult to evaluate the shape of the particle number size distributions in Fig. S9 because particle diameter is plotted on a linear rather than logarithmic scale. I recommend plotting diameter on a logarithmic scale to better assess the reported unimodal distribution.

We thank the reviewer for this helpful suggestion. To facilitate evaluation of the particle number size distribution, we have added an additional figure (Fig. S34) in the Supplementary Material showing the same particle size distributions with particle diameter plotted on a logarithmic scale. The original figure is retained for comparison, while the logarithmic representation provides a clearer view of the spectral shape, particularly at smaller particle sizes.

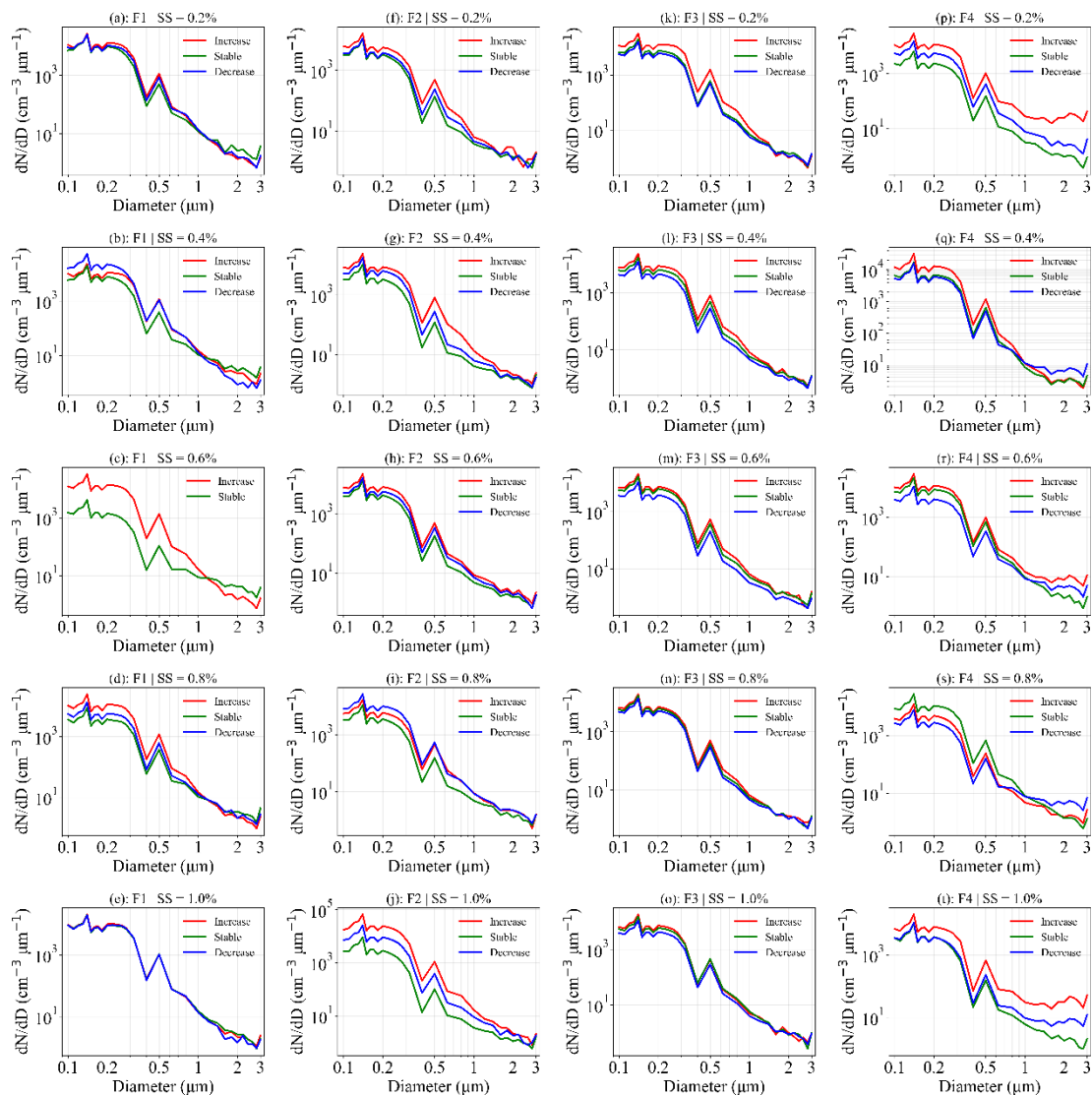


Fig.S34. Aerosol size distributions under different supersaturations and aerosol vertical structures for flights F1–F4, with particle diameter plotted on a logarithmic scale. Each column corresponds to the same flight, and each row corresponds to the same supersaturation. Red, green, and blue curves represent the Increase, Stable, and Decrease structures, respectively.

Figure 6

I recommend using conventional SHAP summary (essentially a beeswarm) plots, which would show both the magnitude and direction of each predictor's contribution to the modeled CCN concentration, rather than only the aggregated contribution percentages shown in Figure 6. Percentages can just be added as text on the plot.

We thank the reviewer for this helpful suggestion. We agree that sample-level SHAP summary plots are useful for showing both the magnitude and direction of each predictor's contribution. In the present study, Figure 6 was designed to provide a concise comparison of the relative SHAP contributions across different aerosol vertical structures and supersaturation conditions. More detailed sample-level SHAP distributions are already provided in the Supplementary Material as SHAP violin summary plots (e.g., Fig. S28). These plots display the

distribution and sign of SHAP values for each predictor, while the color scale represents the corresponding feature values.

To avoid making the main figure overly complex, we therefore retained the relative contribution percentages in Figure 6 and revised the manuscript to more clearly direct readers to the supplementary SHAP violin summary plots for information on the magnitude, direction, and distribution of individual SHAP effects.

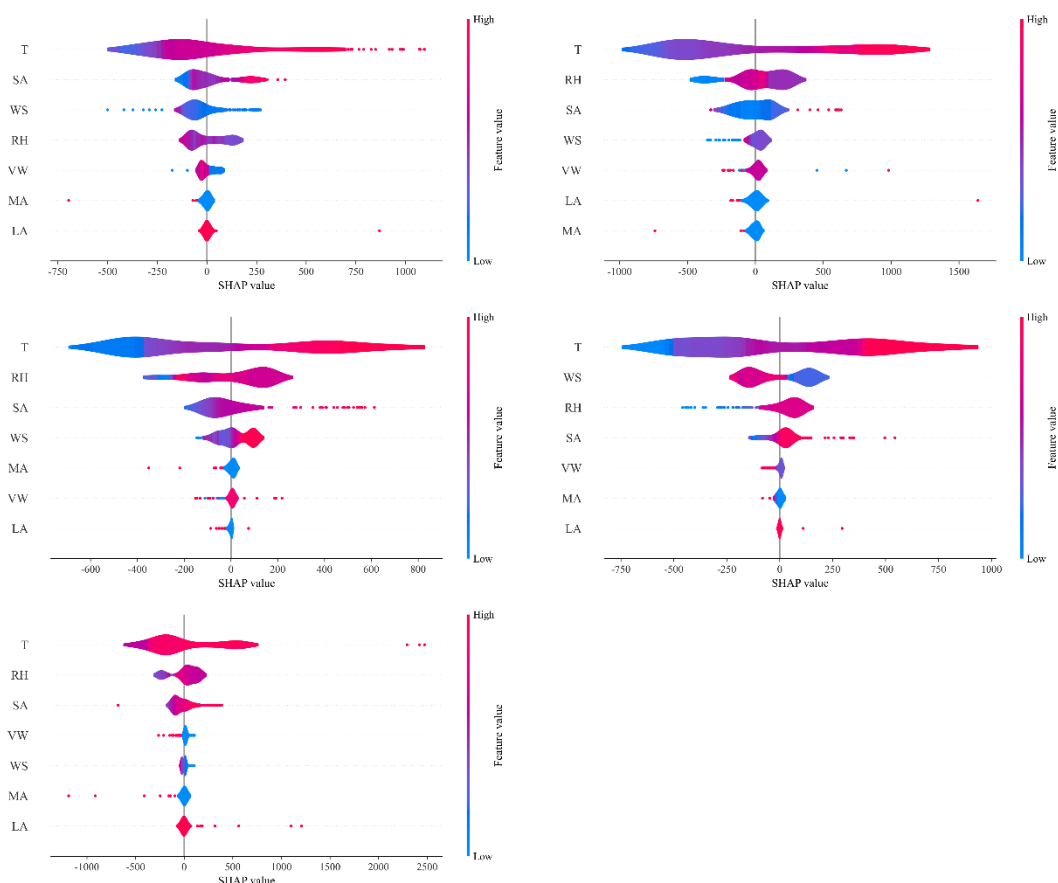


Fig.S27. Stacked SHAP values of different factors under the decrease aerosol vertical profile

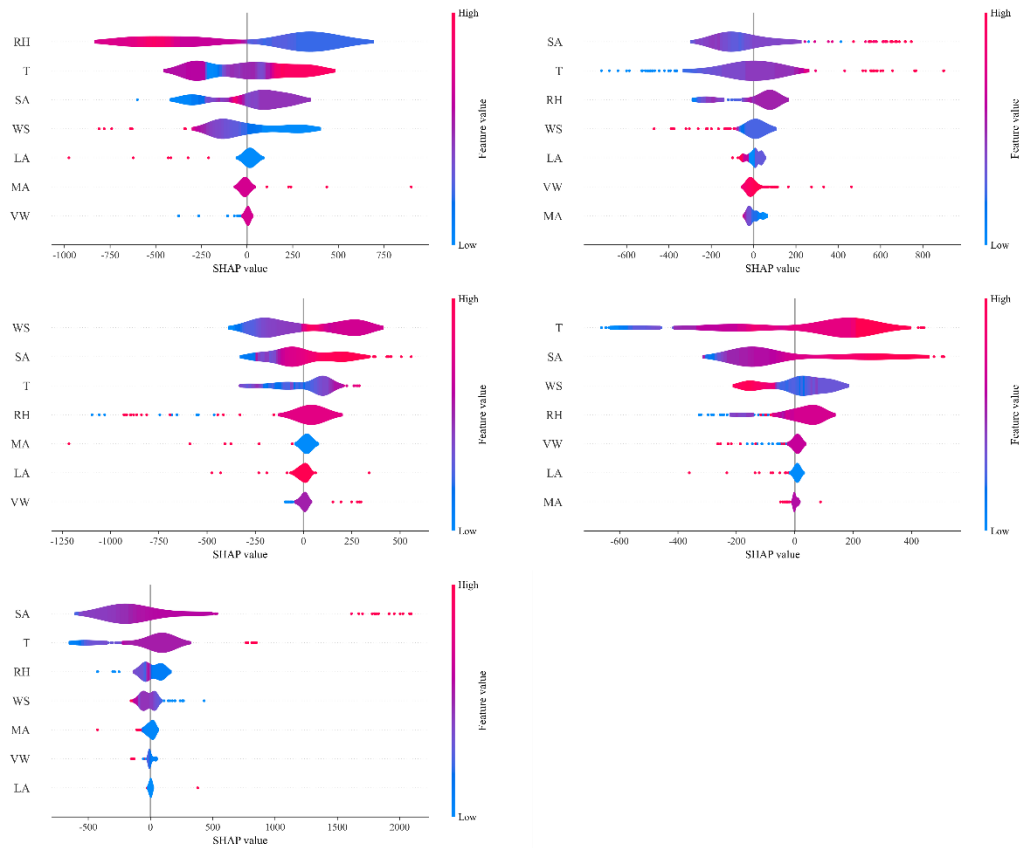


Fig.S28. Stacked SHAP values of different factors under the increase aerosol vertical profile

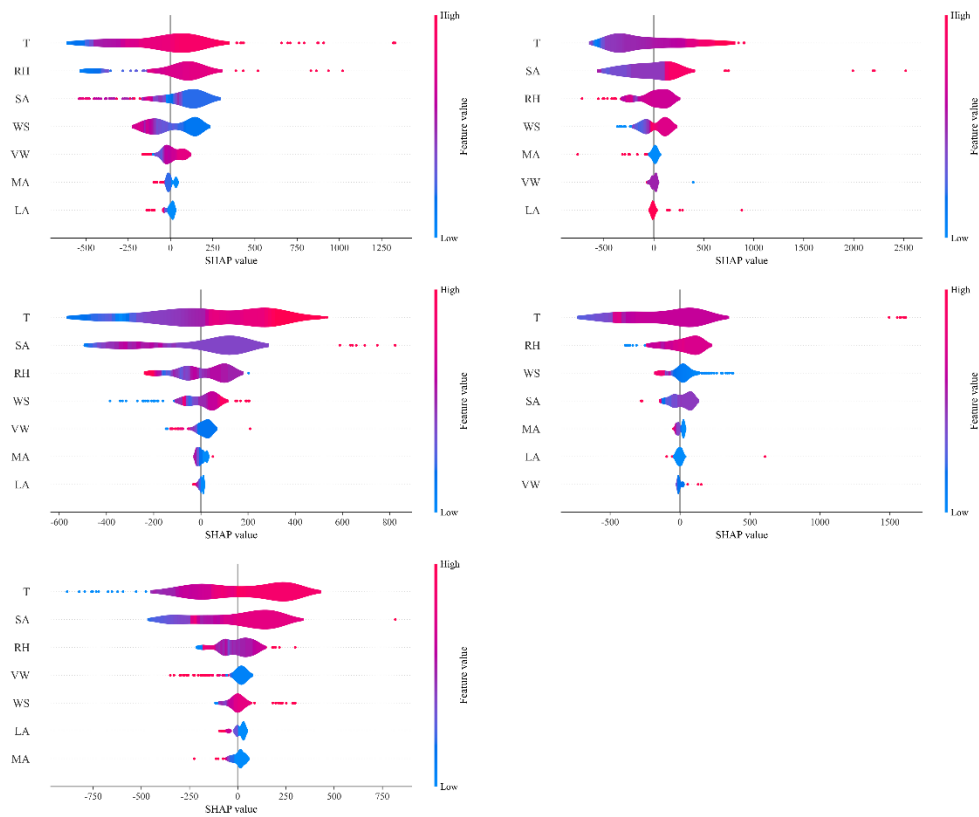


Fig.S29. Stacked SHAP values of different factors under the stable aerosol vertical profile