

Response to Referee

This study investigates how the aerosol indirect effect varies with spatial scale and liquid water path. It analyzes the sensitivity of cloud microphysics during two distinct periods with different aerosol loading (high and decreased AOD) over eastern China. This is a very relevant topic and helps us understand how aerosol loading modulates the scale dependence of aerosol indirect effects.

The manuscript is quite clear and scientifically relevant. However, there are some issues with the presentation and discussion that, in my opinion, should be addressed before publication.

The manuscript aims to:

- Reveal patterns of aerosol indirect effects across spatial scales
- Assess the regulatory role of LWP in aerosol-cloud interactions over land in eastern China

The authors are grateful to the Referee for spending valuable time thoroughly reading our manuscript. We also appreciate the expert views provided to guide us in improving the work. All main and specific points have been carefully considered. The comments are listed below in black, and corresponding changes have been made to the manuscript. For your convenience, all modifications in the revised manuscript have been highlighted. Below, we respond to each comment individually, referring to the revised manuscript with line numbers from the clean version, and provide quoted text from the manuscript.

Main points

In the abstract, two distinct regimes of effective radius variation with liquid water path are presented. Later in the manuscript, however, three regimes are discussed and presented in the conclusions.

Answer: Thank you for pointing out this inconsistency. We have revised the abstract to align with the main text and conclusions, now clearly presenting three distinct regimes of effective radius variation with liquid water path, consistent with the later discussion in the manuscript. The specific revisions are as follows:

“The results show three distinct regimes of CER variation with LWP: a rapid growth regime ($LWP < 55/50 \text{ g/m}^2$), a decreasing regime ($LWP = 55-135/50-100 \text{ g/m}^2$) and a slow growth regime ($LWP > 135/100 \text{ g/m}^2$) (thresholds vary by period). The slow growth regime is not analyzed further due to limited data.” in the Abstract (lines 27-30).

The authors present interesting relationships that they discuss could be used as a basis for improving aerosol-cloud parameterization schemes in climate models, but do not demonstrate how these findings translate into model improvements.

As it stands, the connection between the observational analysis and its application in climate models are largely qualitative. If the authors aim to link their main results to aerosol-cloud modeling, it would be helpful to clarify how these findings could be incorporated into parameterization schemes, for example in activation, autoconversion, or in representing the transition from regional-scale effects to model grid scales.

The authors mention similar statements several times throughout the manuscript:

Answer: The reviewer correctly points out that the link between our observational analysis and climate model applications was largely qualitative. We have now added explicit explanations of how our scale-dependent and LWP-modulated ACI sensitivities can be used to constrain and improve aerosol–cloud parameterization schemes, including:

Incorporating scale-dependent sensitivity coefficients (S_{CER} , S_{Nd}) into aerosol activation parameterizations to reflect scale-dependent CCN activation efficiency.

Using LWP regime-dependent sensitivities to improve autoconversion parameterizations by distinguishing rapid-growth and decreasing LWP regimes.

Providing observationally constrained optimal buffer/grid scales to support scale transition from regional processes to climate model grid scales.

Revising all repetitive general statements into specific, process-oriented claims. These revisions make the application of our findings to model development concrete and operational.

Lines 36-38 - “This study reveals the scale-dependence of aerosol-cloud interactions, providing critical observational support for optimizing climate model parameterization schemes.”

Answer: Thank you for the suggestion. The original text has been replaced with the following in the Abstract (lines 38-41): “This study reveals the scale-dependence of aerosol-cloud interactions, providing quantitative observational constraints for optimizing scale-aware aerosol-cloud parameterization schemes, particularly for constraining scale-dependent aerosol activation and cloud droplet autoconversion processes”.

Lines 155-156 - “how the aerosol indirect effect depends on observational spatial scales in eastern China is of great significance for developing parameterization schemes that align with the regional characteristics of aci.”

Answer: Thank you for the suggestion. The original text has been replaced with the following in the Introduction (lines 154-157): “identifying the spatial scale dependence of the aerosol indirect effect over eastern China provides observationally based coefficients. These coefficients can be used to develop regionally adapted, scale-aware parameterization schemes that better represent regional ACI characteristics.”

Lines 173-174 - “The study aims to reveal the patterns of the sensitivity of aerosol indirect effects to spatial scales, provides support for optimizing parameterization schemes and accurate assessment of regional aerosol effects.”

Answer: Thank you for the suggestion. The original text has been replaced with the following in the Introduction (lines 173-176): “This study reveals the spatial scale-dependent sensitivity of aerosol indirect effects, supporting the development of scale-adaptable parameterization modules and improved regional aerosol effect assessment under different model grid resolutions.”

Lines 384-385: “This approach enabled the determination of the optimal buffer size for aerosol indirect effects as function of the size of the study area, ultimately leading to the development of a parameterization scheme for aerosol indirect effects for observations with different spatial resolution and different sizes of the study area over eastern China.”

Answer: Thank you for the suggestion. The original text has been replaced with the following in Section 2.4 (lines 382-385): “This approach determined the optimal buffer size for aerosol indirect effect estimations as a function of study domain size, providing key observational

constraints for constructing scale-adaptive parameterization frameworks suitable for different domain sizes over eastern China.”.

Line 690-692: “Such advancements provide actionable insights for refining parameterization schemes in climate models, thereby enhancing their predictive reliability.”

Answer: Thank you for the suggestion. The original text has been replaced with the following in Section 4.2 (lines 683-686): “These results provide quantitative observational constraints for refining scale-dependent parameterization modules in climate models, particularly for representing the modulation of ACI by spatial scale and LWP, thereby improving model predictive reliability.”

Lines 730-733: “The scale-dependent attenuation of both sensitivities and their modulation by LWP indicate that quantifying aerosol indirect effects requires full consideration of spatial scales and the key role of liquid water, providing observational basis for optimizing climate model parameterization schemes.”

Answer: Following Minor comment 5, we have integrated the main points from Section 4.3.3 into the conclusions as recommended, rather than keeping them as a separate section, thus the text “The scale-dependent attenuation of both sensitivities and their modulation by LWP indicate that quantifying aerosol indirect effects requires full consideration of spatial scales and the key role of liquid water, providing observational basis for optimizing climate model parameterization schemes.” has been removed from the revised manuscript.

Lines 774-777: These findings not only deepen our understanding of aerosol indirect effects but also provide an important observational basis for improving aerosol-cloud parameterization schemes in climate models. The results emphasize that both the phased characteristics of LWP and spatial scale effects must be considered when assessing aerosol indirect effects.

Answer: Thank you for the suggestion. The original text has been replaced with the following in the Conclusions (lines 764-767): “These findings deepen our understanding of aerosol indirect effects and provide quantitative observational constraints for improving aerosol–cloud parameterization in climate models. Our results highlight that ACI parameterization should explicitly incorporate LWP regime-dependent behavior and spatial scale-dependent interaction strengths to improve realistic representation.”

Minor/technical comments

Line 187-188 – The term aerosol-cloud interactions (aci) is defined earlier (Line 45), but the manuscript later alternates with “cloud-aerosol interactions” and “cloud-aerosol coupling”. For clarity and conceptual consistency, it would be preferable to use consistent terminology (aci) throughout.

Answer: Thank you for the suggestion. We have reviewed the manuscript and replaced the instances of “cloud-aerosol interactions” and “cloud-aerosol coupling” with the consistent term “aerosol-cloud interactions (aci)” as defined in Line 45. This change has also been made throughout the manuscript to improve clarity and conceptual consistency.

Line 346 – You may consider rewriting the list as:(10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 120, 140, 150, 160, 180, 200, 250, and 300 km)

Answer: Corrected.

Line 366 – consider simplifying to: from 10 to 300 km

Answer: Corrected.

Line 402 – I would suggest being cautious with the use of the term “pristine”. It may be more appropriate to use a term such as “relatively cleaner”.

Answer: Thank you for your suggestion. We have replaced “pristine” with “relatively cleaner” in line 401 in the revised manuscript.

Lines 727-733 You may consider briefly incorporating the main points from Section 4.3.3 into the conclusions, rather than introducing them as a separate section

Answer: Thank you for your suggestion. We followed your suggestion and integrated the main points from Section 4.3.3 into the conclusions.

The revised text in the Conclusions (line 760-763): “These observed patterns correspond to the fundamental microphysical pathway of aci, whereby increased aerosol loading enhances Nd and reduces CER under constant LWP conditions. The results emphasize that accurate quantification of ACI must explicitly consider both the phase-dependent characteristics of LWP and spatial scale effects when assessing aerosol indirect effects.

These findings deepen our understanding of aerosol indirect effects and provide quantitative observational constraints for improving aerosol–cloud parameterization in climate models. Our results highlight that ACI parameterization should explicitly incorporate LWP regime-dependent behavior and spatial scale-dependent interaction strengths to improve realistic representation.”.

Line 554 & 565 - aerosol- cloud interactions -> aci

Answer: Corrected.

Lines 602-624 - During revision, the authors may consider splitting longer sentences into two or three simpler ones to improve readability.

Answer: Thank you for the suggestion. During revision, we have split the longer sentences in Lines 602–624 into two or three simpler ones to enhance readability. Please see the revised version.

The revised text in the Section 4.1 (line 596-618): “Comparative analysis of scale-conditioned $SCER$ across LWP regimes in periods 1 and 2 revealed markedly enhanced sensitivity of $SCER$ to AOD in the second LWP regime. A trade-off exists between AOD and LWP under conditions of insufficient water vapor. This trade-off leads to smaller CER values. As suggested by Costantino et al. (2013), the LWP response to aerosol invigoration is influenced by two competing mechanisms: a drying effect caused by enhanced entrainment of dry air at cloud top (dominant in optically thin clouds) and a moistening effect from precipitation suppression (dominant in optically thick clouds). For larger LWP, the supply of cloud water is sufficient. The increase in aerosol number concentrations significantly affects the distribution of cloud droplet number concentrations and sizes. This further enhances the sensitivity of CER to AOD. For small aerosol concentrations, the values of $|SCER|$ (Figure 5b, 7b) decreased overall with expanding buffer size within the same study area. For fixed buffer size, $|SCER|$ decreased as the study area increased. The ranges of $|SCER|$ values across different study areas showed a convergent pattern. These values typically remained small and close to zero. During the high AOD period (2008–2014), anthropogenic emissions and dust transport provided abundant CCN. This laid the material foundation for strong aci. It enhanced the synergistic effect of “sufficient liquid water + abundant CCN” in the second LWP regime. This synergistic effect amplified the difference in $SCER$ between the two LWP regimes. In the period of decreasing

AOD (2015–2022), CCN concentration decreased following the implementation of clean air policies (de Leeuw et al., 2021; 2023). This reduction weakened the direct impact of aerosols on CER. However, the LWP-driven microphysical differences persisted. Thus, S_{CER} in the second regime remained significantly smaller than that in the first regime, albeit with a smaller difference. Additionally, the complexity of aerosol types during the high AOD period (e.g., mixing of anthropogenic pollutants and natural dust) may have adjusted the value of S_{CER} . However, it did not alter the dominant role of LWP. This aligns with the theory that “aerosol indirect effects are jointly regulated by concentration and type” (Liu et al., 2017).”

Line 648 – Please define what is meant by “LWP-stratified”. It appears to refer to the binning of LWP described in Lines 424–425, but this is not clearly stated.

Answer: Thank you for pointing this out. We have revised Line 648 to explicitly define “LWP-stratified” as the binning of LWP described in Lines 424–425, making the connection clear.

The revised text in the Section 4.1 (line 642–643): “Additionally, LWP-stratified analysis (i.e., binning LWP into 5 g m^{-2} intervals) further isolates interference. The validation of the core hypothesis provides a reliable premise for accurately quantifying the impact of aerosol concentration changes on the sensitivity of cloud parameters and their spatial scale dependence.”.

Section 3.3 – This section could be slightly shortened to improve readability. It would also be helpful to clarify how increasing the spatial domain may mix cleaner and more polluted conditions, potentially reducing regime contrast and affecting the derived S_{Nd} sensitivities.

Answer: Thank you for your valuable suggestions. We have shortened Section 3.3 to improve readability. Additionally, we have added a clarification on how expanding the spatial domain may mix cleaner and more polluted conditions, thereby reducing regime contrast and potentially influencing the derived S_{Nd} sensitivities. Please see the revised section.

The revised text in Section 3.3 (line 536–569): “The sensitivity S_{Nd} is defined as the slope of a linear fit to a log-log plot of S_{Nd} versus AOD (Eq. 2). For each period, we binned the data into AOD intervals of 0.02 and averaged N_d within each bin. Figure 8 shows the variation of S_{Nd} with buffer size for different study areas. In contrast to S_{CER} , S_{Nd} is predominantly positive ($p < 0.01$) in both periods and decreases with increasing buffer size. During period 1 (2008–2014, high AOD), S_{Nd} for the $6^\circ \times 6^\circ$ study area decreases rapidly to a minimum at buffer sizes of 40–50 km, then increases to a maximum at a buffer size of 120 km. For buffer size ≥ 120 km, the two smallest study areas ($4^\circ \times 4^\circ$ and $6^\circ \times 6^\circ$) yield similar S_{Nd} values, which are substantially larger than those for the two larger study areas ($8^\circ \times 8^\circ$ and $10^\circ \times 10^\circ$). During period 2 (2015–2022, decreasing AOD), we can see an initial increase of S_{Nd} for the study area of $6^\circ \times 6^\circ$ and $8^\circ \times 8^\circ$, and variation of S_{Nd} for the study area of $10^\circ \times 10^\circ$ and $4^\circ \times 4^\circ$. After that, S_{Nd} for the study area of $4^\circ \times 4^\circ$ and $6^\circ \times 6^\circ$ decreases rapidly to a minimum for a buffer size of 80 km, followed by an increase to a maximum for a buffer size of 140 km.

The optimal buffer sizes (maximizing R) are larger for the $8^\circ \times 8^\circ$ and $10^\circ \times 10^\circ$ study areas than for the $8^\circ \times 8^\circ$ and $10^\circ \times 10^\circ$ areas (Fig. 9), reflecting different aci characteristics under varying AOD conditions. Estimates of S_{Nd} and R, stratified by optimal buffer size, for study areas ranging from $4^\circ \times 4^\circ$ to $10^\circ \times 10^\circ$ during the two periods are presented in Appendix A2.

Expanding the study area inevitably increases spatial heterogeneity in aerosol loading, meteorology, and cloud type, while also introducing larger satellite retrieval uncertainties. This spatial mixing blends clean and polluted air masses, thereby introducing a statistical dilution effect on the estimated S_{Nd} . Therefore, the decrease in S_{Nd} with increasing study area size

(Fig. 8) reflects not only a genuine weakening of aerosol-cloud interactions but also the combined effects of meteorological confounding, cloud regime transitions, and retrieval limitations. It is noted that S_{CER} and S_{Nd} exhibit distinct anomalies in large buffer zones, which may be associated with all these factors.

Changes in aerosol chemical composition between the two periods may also modulate S_{Nd} . During 2008–2014, aerosols over eastern China were dominated by sulfate (30%–40% of $PM_{2.5}$ mass; Huang et al., 2014; Zheng et al., 2018). Given the strong hygroscopicity of sulfate-dominated aerosols (Zhang et al., 2012; Liu et al., 2023), their CCN activation efficiency was likely high, providing a favorable physical basis for aci (Lee et al., 2009). In contrast, during 2015–2022, policy interventions (e.g., the Air Pollution Prevention and Control Action Plan; Zheng et al., 2018) drove a structural transition: the sulfate mass fraction dropped sharply to 15%–25% (an absolute reduction of >50%), while less hygroscopic components (nitrate, carbonaceous aerosols, and secondary organic aerosols) increased in relative proportion (Huang et al., 2014; Zheng et al., 2018). As these components generally exhibit weaker hygroscopicity compared with sulfate (Zhang et al., 2012; Liu et al., 2023), this compositional shift may have reduced CCN activation efficiency under the same AOD, thereby weakening the sensitivity of N_d to AOD and altering aci intensity (Lee et al., 2009).”.