

Spatial-scale dependence of aerosol indirect effects over land in eastern China: A comparative analysis

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Abstract

The aim of this study is to reveal patterns of the sensitivity of aerosol indirect effects to spatial scales and investigate the regulatory role of the liquid water path (LWP) in aerosol-cloud interactions over land in eastern China. Using MODIS and CALIOP satellite observations, we systematically analyzed the relationships between aerosol optical depth (AOD) and cloud properties (cloud droplet effective radius, CER; cloud droplet number concentration, N_d) during two periods: 2008–2014 (period 1, high AOD) and 2015–2022 (period 2, decreasing AOD). 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. The sensitivity of CER to AOD (S_{CER}) exhibited a negative correlation, with stronger sensitivity in the decreasing LWP regime than in the rapid growth regime. The spatial scale (characterized by buffer size and study area) significantly modulated these sensitivities: $|S_{CER}|$ and the positive sensitivity of N_d to AOD (S_{N_d}) both decreased with increasing spatial scale.

34 Optimal buffer sizes range from $6^{\circ}\times 6^{\circ}$ to $10^{\circ}\times 10^{\circ}$: increasing with study area in period 2 but decreasing
35 in period 1 for the decreasing LWP regime. Compared with period 1, $|S_{CER}|$ in period 2 significantly
36 reduced, reflecting the weakened aerosol-cloud interactions due to declining aerosol concentrations.
37 Additionally, the optimal buffer sizes for S_{Nd} were larger in the $8^{\circ}\times 8^{\circ}$ and $10^{\circ}\times 10^{\circ}$ study areas than in
38 $4^{\circ}\times 4^{\circ}$ and $6^{\circ}\times 6^{\circ}$ areas. This study reveals the scale-dependence of aerosol-cloud interactions, providing
39 quantitative observational constraints for optimizing scale-aware aerosol-cloud parameterization
40 schemes, particularly for constraining scale-dependent aerosol activation and cloud droplet
41 autoconversion processes.

42 **Keywords:** Aerosol, Cloud, Liquid water path, Scale effect, Satellite, Eastern China

43 **1 Introduction**

44 Aerosol particles, depending on their chemical composition and size, can serve as cloud condensation
45 nuclei (CCN) in liquid clouds or as ice nucleating particles (INP) in ice clouds. When CCN are activated,
46 they can alter the microphysical properties of clouds and affect precipitation, indirectly impacting the
47 Earth's radiative budget through aerosol-cloud interactions (aci) (Tao et al., 2012; Fan et al., 2016;
48 Rosenfeld et al., 2019; Rao and Dey, 2020; Bellouin et al., 2020; Dagan et al., 2023). An increase in CCN
49 concentrations results in a larger number of cloud droplets (N_d), and if the cloud liquid water path (LWP)
50 remains constant, it leads to a reduction in the cloud droplet effective radius (CER) (Twomey, 1974;
51 Feingold et al., 2003). The reduced CER leads to an increased reflection of solar radiation, i.e. a higher
52 cloud albedo, and enhances radiative forcing due to aci (RFaci). The impact of increasing aerosol particle
53 numbers on cloud properties, while maintaining a constant LWP, is commonly known as the "Twomey"
54 effect (Twomey, 1977; Feingold, et al., 2001; Matheson et al., 2005; Koren et al., 2005; Meskhidze and
55 Nenes, 2010; Costantino et al., 2010; 2013). Another aspect of RFaci involves quick adjustments, which
56 could also cause changes in other cloud characteristics due to the rise in N_d and the decrease of CER. For
57 example, this may lead to a reduction in precipitation efficiency, causing an increase in the LWP and
58 cloud cover. As a result, the reflection of solar radiation is intensified (Albrecht, 1989). These two effects
59 of aci are often categorized as the cloud albedo effect and the cloud lifetime effect (Quaas et al., 2008).
60 Extensive research on the impact of aerosols on the microphysical properties of clouds has been
61 conducted utilizing satellite observations (Liu et al., 2017; Jia et al., 2022), aircraft measurements (Jia et
62 al., 2019; Zheng et al., 2024), ground-based monitoring (Sarna et al., 2016; Zheng et al., 2020), and

63 numerical simulations (Lee et al., 2025; Li et al., 2008). Among these, satellite-based instruments have
64 become a vital observational tool for studying aci due to their wide spatial coverage and high
65 spatiotemporal resolution. However, optical satellite sensors such as the Moderate Resolution Imaging
66 Spectroradiometer (MODIS) cannot effectively penetrate cloud layers (King et al., 2003; Kaufman et al.,
67 2005; Remer et al., 2005), making it difficult to directly retrieve the optical properties of aerosols
68 underneath clouds. Currently, aerosol data are mainly obtained in cloud-free conditions as determined
69 using cloud detection methods. This limitation results in significant spatial mismatches between aerosol
70 and cloud properties, often requiring aggregation of satellite data over large-scale grids for statistical
71 analysis to determine relationships between aerosol and cloud parameters. The discrepancy between this
72 large-scale analysis and the actual process scale frequently leads to biases in quantifying aerosol indirect
73 effects, thereby significantly increasing the uncertainty in radiative forcing estimates (Lebsock et al.,
74 2013; Altaratz et al., 2014; Ma et al., 2015; Possner et al., 2016; Bender et al., 2018).

75 In recent years, studies based on multi-source satellite data or multi-instrument joint observations have
76 demonstrated that aerosol particles significantly influence cloud microphysical properties (Saponaro et
77 al., 2017; Liu et al., 2018; Pandey et al., 2020). Numerous studies have validated the existence of the
78 Twomey effect (Jones et al., 2009; Christensen et al., 2016; Jia et al., 2019). However, also studies have
79 been published with findings that contradict the Twomey effect, particularly over land, where an increase
80 in aerosol optical depth (AOD) results in an increase in CER (Feingold et al., 2001; Yuan et al., 2008;
81 Grandey and Stier, 2010; Tang et al., 2014; Wang et al., 2015; Ma et al., 2018; Jia et al., 2019; Liu et al.,
82 2020). These inconsistent findings highlight the complexity and regional variability of aci mechanisms,
83 and further in-depth research is needed to reveal the underlying processes.

84 Currently, researchers usually use grid methods (such as $1^\circ \times 1^\circ$, $2^\circ \times 2^\circ$, etc.) to study the aerosol indirect
85 effects in large areas (Bréon, 2002; Kaufman et al., 2005; Bulgin et al., 2008; Quaas et al., 2008). For
86 instance, Grandey and Stier (2010) estimated the relationship between aerosols and CER on a global
87 scale ($60^\circ\text{N}\sim 60^\circ\text{S}$) using multiple spatial resolutions ($1^\circ \times 1^\circ$, $4^\circ \times 4^\circ$, $8^\circ \times 8^\circ$, $15^\circ \times 15^\circ$, and $60^\circ \times 60^\circ$). They
88 concluded that the sensitivity of retrieved CER to AOD generally exhibited positive values over land and
89 negative values over oceans, and pointed out that using grids larger than $4^\circ \times 4^\circ$ could introduce significant
90 errors due to the spatial variability of aerosol and cloud parameters. Additionally, the study highlighted
91 that, when using grids larger than $4^\circ \times 4^\circ$ to investigate the relationship between aerosols and CER,

92 significant errors could be introduced in calculating the aerosol indirect effect index due to the spatial
93 variability of aerosol and cloud parameters.

94 For studies focusing on smaller regions, researchers often employ a moving window or a fixed area
95 referred to as a buffer zone, within which the distribution of aerosol concentrations is assumed to be
96 uniform. Spatially matched samples are constructed by averaging AOD and cloud parameters within the
97 window or buffer zone. The choice of the window or buffer size is often arbitrary, and existing studies
98 rarely systematically explore how the detection of aci signals is influenced by the size of the area. For
99 example, Yuan et al. (2008) used a 100 km × 100 km moving window to calculate the mean values of
100 AOD and cloud properties to investigate the relationship between aerosols and CER across seven global
101 regions. Their results indicated that only two of these regions, near the coasts of the Gulf of Mexico and
102 the South China Sea, exhibited a positive correlation between CER and AOD. Similarly, Jones et al.
103 (2009) utilized multi-source remote sensing data and applied a point spread function to derive the mean
104 AOD within a 20 km range, which was designed to match the native 20 km resolution of the
105 corresponding cloud properties (cloud optical thickness, COT; LWP; CER; cloud top pressure, CTP).
106 Their study examined the influence of aerosol types, cloud conditions, and atmospheric factors on aerosol
107 indirect effects across six different oceanic regions globally, finding that the sensitivity of cloud
108 properties to AOD varied substantially with regional characteristics. In addition, significant progress has
109 been made in research utilizing observations from the Cloud-Aerosol Lidar with Orthogonal Polarization
110 (CALIOP) data (Winker et al., 2007). For instance, Costantino et al. (2010) used CALIOP data to
111 investigate the aerosol influence on CER in stratocumulus clouds over the coastal regions of Namibia
112 and Angola. They performed the analysis by co-locating an aerosol index (based on AOD and the
113 Ångström exponent) with CER within a 150 km buffer zone around CALIOP observations. They found
114 that there was no correlation between aerosol load and CER when aerosol and cloud layers were clearly
115 separated, but a strong correlation when lidar profiles indicated mixing. Costantino et al. (2013) further
116 analyzed the statistical relationship between aerosol concentrations and cloud physical parameters by
117 examining aerosol and cloud properties within a 20 km buffer zone around CALIOP samples, integrating
118 vertical profiles of aerosol and cloud data. Their statistics also clearly showed that cloud micro-physical
119 properties were affected by aerosols when aerosol and cloud layers were mixed, decreasing the CER. It
120 is noted that these two studies by Costantino et al. (2010, 2013) reached consistent conclusions about aci

121 (i.e., aerosols modulate CER when layers interact), by adopting different buffer sizes (150 km vs. 20 km)
122 to target distinct study areas. This demonstrates that the buffer size is tailored to the research objectives
123 rather than through a systematic sensitivity analysis. Wang et al. (2015) revealed an inverse “Twomey”
124 effect between aerosols and CER in eastern China by analyzing aerosol concentrations and CER within
125 a 50 km buffer zone around CALIOP samples. Their results showed that larger CER was associated with
126 high AOD, which was attributed to the feedback of microphysical processes from intense competition
127 for vapor in the presence of high aerosol concentrations and the evaporation of smaller, less hygroscopic,
128 droplets. Similarly, Liu et al. (2017) systematically examined the response mechanisms of warm cloud
129 macro- and microphysical parameters to increasing AOD in the Yangtze River Delta region, also using
130 CALIOP samples within a 50 km buffer zone. They found that the relation between cloud properties and
131 AOD depended on the aerosol abundance, with a different behavior for low and high AOD (i.e. $AOD <$
132 0.35 and $AOD > 0.35$). However, both Wang et al. (2015) and Liu et al. (2017) used a fixed 50 km buffer
133 zone without justifying the choice or exploring how varying buffer sizes might alter the strength or
134 robustness of their findings—a common limitation in regional aci studies. More recently, Liu et al. (2024)
135 quantified the relative importance of aerosols, meteorological parameters and their interactions for cloud
136 properties in the eastern coastal and inland regions of China, utilizing MODIS $1^\circ \times 1^\circ$ aerosol and cloud
137 product data. Their study confirmed that CER decreased with the increase in AOD in the moderately
138 polluted atmosphere ($0.1 < AOD < 0.3$) over the East China Sea, whereas, in contrast, CER increased
139 with increasing AOD in the polluted atmosphere ($AOD > 0.3$) over the Yangtze River Delta. These studies
140 have provided critical scientific insights into aci at regional scales, but the lack of systematic scale
141 sensitivity analysis—especially for varying window/buffer sizes within the same regional domain—
142 leaves uncertainties about the generalizability of their conclusions.

143 However, the properties and interaction processes of aerosols and clouds are spatially significantly
144 heterogeneous and scale dependent (McComiskey et al., 2009; McComiskey and Feingold, 2012; Chen
145 et al., 2015; Glotfelty et al., 2020). McComiskey and Feingold (2012) explicitly pointed out that the
146 “scale problem” is a major challenge in quantifying aerosol indirect effects, as the spatial scale of
147 observation can mask or exaggerate the true interaction signals. In previous studies, the definitions of
148 window size and buffer size have often been subjective, inadvertently introducing uncertainties into the
149 research on aci. Although studies have explored the relationship between aerosols and CER across

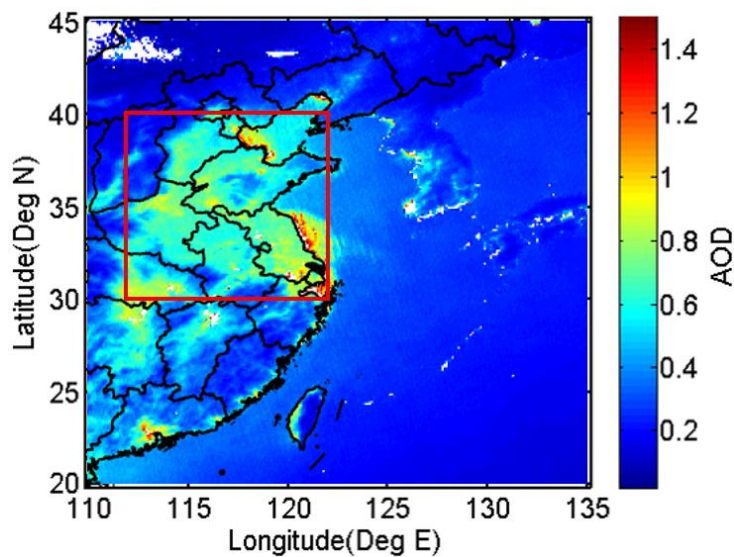
150 different observational scales, these investigations have primarily focused on larger spatial scales, leaving
151 a gap in sensitivity analysis of aerosol indirect effects at smaller regional scales. For example, Grandey
152 and Stier (2010) focused on global-scale grid resolutions but did not explore the scale dependence within
153 regional domains; Wang et al. (2015) and Liu et al. (2017) used fixed buffer sizes (50 km) without
154 investigating how varying buffer sizes affect the results. Therefore, identifying the spatial scale
155 dependence of the aerosol indirect effect over eastern China provides observationally based coefficients.
156 These coefficients can be used to develop regionally adapted, scale-aware parameterization schemes that
157 better represent regional ACI characteristics.

158 Aerosol properties in China have significantly changed between 2008 and 2022 due to economic
159 development and the implementation of emission reduction policies. The AOD over China increased
160 until 2007 to become among the highest worldwide and remained high between 2008 and 2014 with large
161 interannual variations. The enforcement of emission reduction measures, in particular the implementation
162 of the 2013-2017 Clean Air Action Plan, resulted in the decline of the AOD between 2014 and 2018 (de
163 Leeuw et al., 2021; 2022; 2023) and a further decrease resulted in an AOD lower than that in 2000 (de
164 Leeuw et al, 2023). This distinct two-period variation (high AOD vs. decreasing AOD) provides a unique
165 opportunity to investigate how changes in aerosol loading modulate the scale dependence of aerosol
166 indirect effects, a topic that has not been systematically addressed in previous studies. Based on these
167 observations, in this study we conduct a comparative analysis of the sensitivity of cloud parameters (CER
168 and N_d) to AOD variation using data from two distinct periods: 2008-2014 (period 1, high AOD) and
169 2015-2022 (period 2, decreasing AOD). The main objectives of this study are to: (1) reveal patterns of
170 the sensitivity of aerosol indirect effects to spatial scales by investigating how spatial scale modulates
171 the sensitivity of CER to AOD (S_{CER}) and the sensitivity of N_d to AOD (S_{N_d}) over eastern China during
172 the two periods; and (2) clarify the regulatory role of LWP in aci by examining the LWP-stratified
173 responses of S_{CER} and S_{N_d} to spatial scale and AOD variations. This study reveals the spatial
174 scale-dependent sensitivity of aerosol indirect effects, supporting the development of scale-adaptable
175 parameterization modules and improved regional aerosol effect assessment under different model grid
176 resolutions.

177 **2 Method**

178 **2.1 Study area**

179 Eastern China (30°N-40°N, 112°E-122°E; Figure 1) has undergone remarkable economic expansion over
180 the past three decades, which was accompanied by a substantial increase in AOD. Eastern China presents
181 a unique atmospheric laboratory due to its complex aerosol composition - featuring both anthropogenic
182 pollutants from industrial emissions and natural mineral dust transported from Central Asian deserts,
183 particularly during the spring (Proestakis et al., 2018; Liu et al., 2021). The multitude of sources and the
184 persistent nature of these aerosol particles, which can remain suspended for days to weeks and be
185 transported over long distances in the absence of precipitation (Costantino et al., 2013; Li et al., 2021;
186 Leung et al., 2023), makes eastern China an ideal study area for investigating aci. Our research leverages
187 satellite observations to systematically evaluate the sensitivity of cloud properties (S_{CER} and S_{Nd}) to the
188 AOD variation, thereby revealing the scale-sensitive patterns of aerosol indirect effects and clarifying
189 the regulatory role of LWP in aci over this region.



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191 **Figure 1. Map of annual averaged MODIS/AQUA level 2 AOD for all years during the period from 2008 to**
192 **2022. The red rectangle (30°–40°N, 112°–122°E) indicates the study area.**

193 **2.2 Data used**

194 Data used in this study were acquired by the MODIS instrument aboard NASA’s Aqua satellite, which
195 features an extensive swath width of approximately 2300 km and comprehensive spectral coverage across
196 multiple bands (King et al., 2003). The satellite’s equator crossing time is 13:30 (Local time, i.e. in the

197 early afternoon, coinciding with optimal development conditions for continental warm cloud systems
198 (Wang et al., 2014; Liu et al., 2024). For aerosol characterization, we utilized the MODIS Collection 6.1
199 aerosol product (MOD04), generated from cloud-screened pixels with a native resolution of 500 m at
200 nadir and subsequently aggregated to 10 km grid cells (Remer et al., 2005; Levy et al., 2010). AOD
201 retrieval over land uses radiances measured at the top of the atmosphere (TOA) at wavelengths of 0.47,
202 0.66, and 2.13 μm (Remer et al., 2005). The MODIS AOD (at 550 nm) Level 2 product (10 km \times 10
203 km) has been validated against ground-based remote sensing data and the results show that 69.40% of
204 the MODIS AOD data fall within the expected uncertainty of $\pm (0.05 + 15 \%)$ over land (Levy et al.,
205 2013). In this study, AOD larger than 1.5 was excluded from further analysis to mitigate potential
206 retrieval overestimation. This threshold was selected based on two key considerations: (1) Christensen
207 et al. (2017) used MOD06 C6 data (1 km \times 1 km) and reported that “large aerosol optical depths remain
208 in the MODIS-observed pixels near cloud edges, due primarily to 3D effects (Varnáí and Marshak, 2009)
209 and the swelling of aerosols by higher relative humidity”; (2) the threshold of 1.5 aligns with widely
210 adopted thresholds in regional aci studies over eastern China, where high AOD often coincides with
211 complex surface conditions (e.g., urbanization, heterogeneous land cover) that exacerbate retrieval biases
212 (Wang et al., 2015; Liu et al., 2017, 2021).

213 The cloud properties used in this study, including CER, LWP, COT, CTP, and cloud phase infrared (CPI)
214 index, were derived from the Collection 6.1 MODIS Level 2 cloud product (MYD06) (King et al., 2003).
215 The retrieval of these cloud characteristics utilizes six spectral channels spanning wavelengths from the
216 visible to the near-infrared (0.66, 0.86, 1.24, 1.64, 2.12, and 3.75 μm) as described by King et al. (1997).
217 Uncertainties in the MODIS C6.1 cloud parameters over land originate from instrument calibration,
218 atmospheric correction, land surface properties, and model assumptions (Platnick et al., 2017, 2018). For
219 COT, these include scene-dependent Level 1B data errors (1.5%–30%), land surface albedo errors
220 ($\pm 15\%$), and atmospheric correction errors ($\pm 20\%$). The C6.1 algorithm addresses some prior limitations
221 by inheriting C6’s optimized lookup table design, which reduces interpolation errors to 0.1%–0.2% for
222 near-nadir views and corrects C5’s overestimation of thin-cloud COT (Platnick et al., 2017). CER
223 uncertainties, stemming from solar irradiance error ($\sim 4\%$ at 3.7 μm), atmospheric correction, and
224 scattering differences, are mitigated as C6.1 retains C6’s separate multi-band reporting, thereby
225 eliminating C5’s systematic bias (Platnick et al., 2017). LWP uncertainty is linked to COT/CER retrieval

226 errors and cloud-phase classification accuracy; the latter is improved by C6's voting-based phase
227 algorithm (preserved in C6.1), which reduces misclassification over complex surfaces like vegetation
228 and deserts (Marchant et al., 2015; Platnick et al., 2017). For CTP (1 km resolution), uncertainties from
229 viewing angles and cloud structure are partially countered in C6.1 by assigning fill values when the 1 km
230 retrieval fails, avoiding surface parameter defaults. For land clouds above 3 km, CTP accuracy reaches
231 ~50 hPa (Baum et al., 2012). Finally, CPI adopts C6's weighted voting logic (replacing C5's sequential
232 tree), with C6.1 maintaining an enhanced Phase Agreement Fraction against CALIOP/POLDER data,
233 which reduces uncertainties from weak thin-cloud signals and complex land interference (Marchant et
234 al., 2015; Platnick et al., 2017).

235 Following the methodology of Platnick et al. (2017), CER and COT measurements at 3.7 μm were used
236 to estimate N_d through adiabatic approximation principles (Quaas et al., 2006). Previous investigations
237 have demonstrated that implementing filters based on cloud adiabaticity produced minimal effects on
238 S_{Nd} estimates while significantly reducing the available dataset by up to 63.00% (Gryspeerd et al., 2022).
239 Therefore, such filtering procedures were not adopted in the current analysis. Instead, N_d calculations are
240 initially performed at the native pixel resolution (approximately 1 km) prior to spatial aggregation,
241 thereby avoiding potential biases associated with deriving N_d from nonlinear combinations of CER and
242 COT at coarser resolutions (Feingold et al., 2022). To maintain data quality, the analysis incorporated
243 several quality control measures: only single-phase liquid clouds (CPI = 1) with CTP exceeding 700 hPa
244 and LWP smaller than 200 g m^{-2} are considered, consistent with the typical atmospheric distribution of
245 aerosols in the lower troposphere (Michibata et al., 2014). Pixels with CER values smaller than 4 μm or
246 COT values smaller than 4 were excluded due to increased retrieval uncertainties (Sourdeval et al., 2016).
247 Additionally, observations were restricted to solar zenith angles $<65.00^\circ$ and sensor zenith angles
248 $<41.40^\circ$. This constraint was intended to reduce the influence of well-documented biases, as elaborated
249 in Grosvenor et al. (2018).

250 CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) operates within the A-
251 Train constellation alongside the Aqua satellite and other NASA Earth-observing platforms. The primary
252 instrument aboard CALIPSO is the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP).
253 CALIOP is a two-wavelength, polarization-sensitive lidar specifically designed to provide high-
254 resolution vertical profiles of aerosols and clouds on a global scale (Winker et al., 2009). The mission

255 and its lidar instrument are described in Winker et al. (2009), and the associated Level 1 data products
256 are detailed in Winker et al. (2010). This advanced sensor features an exceptionally narrow ground
257 footprint of 70 m diameter for each laser pulse. The vertical resolution of CALIOP's products varies with
258 altitude: 30 m within 0-8.2 km, 60 m between 8.2-20.2 km, and 180 m from 20.2-30.1 km, while
259 maintaining a consistent 5 km horizontal resolution along the track direction (Liu et al., 2009).
260 The coordinated A-Train configuration ensures near-simultaneous observations (within 1-2 minutes)
261 between MODIS/Aqua and CALIOP/CALIPSO for identical atmospheric targets (Stephens et al., 2002).
262 This temporal synchronization guarantees data consistency when extracting coincident measurements,
263 avoiding interferences such as aerosol diffusion and cloud evolution caused by observational time lags—
264 an advantage unparalleled by positioning methods like random grid points and ground-based stations.
265 For spatial compatibility, we resampled the higher-resolution MODIS cloud products (CER, LWP, and
266 N_d at 1 km native resolution) to match CALIOP's 5 km along-track scale, while directly utilizing the 5
267 km-resolution CTP and CPI parameters. In cases where CALIOP detected aerosol presence, we
268 computed spatial averages of MODIS aerosol and cloud retrieval products across multiple observation
269 scales (detailed in Section 2.4) centred on CALIOP targets. This approach assumes reasonable
270 homogeneity of aerosol properties between adjacent clear and cloudy regions (Anderson et al., 2003;
271 Quaas et al., 2008). Table 1 provides a comprehensive overview of the aerosol and cloud datasets
272 including the parameters used from each product, the resolution, and the data source, used in this study.

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285 **Table 1. Aerosol and cloud products, parameters, horizontal resolutions, and their sources applied in the**
 286 **present study.**

Product	Parameters	Horizontal resolution	Data source		
Aerosol (MYD04 Level 2 Collection 6)	Latitude	10 km	MODIS		
	Longitude	10 km			
	Scan_Start_Time	10 km			
	AOD at 550 nm	10 km			
Cloud (MYD06 Level 2 Collection 6)	Latitude	5 km			
	Longitude	5 km			
	Scan_Start_Time	5 km			
	CER at 3.7 um and 2.1 um	1 km			
	LWP at 3.7 um	1 km			
	COT at 3.7 um	1 km			
	Cloud multi-layer flag	1 km			
	Cloud_Phase_Infrared_Day	5 km			
	Cloud_TOP_Pressure_Day	5 km			
	Sensor_Zenith_Day	5 km			
	Solar_Zenith_Day	5 km			
	Aerosol (05kmALay)	Latitude		5 km	CALIOP
		Longitude		5 km	
Profile_Time		5 km			

287 **2.3 Calculation of sensitivities**

288 Variations in aerosol loading significantly influence cloud optical properties (such as COT) and
 289 microphysical parameters (such as CER). Under specific environmental conditions, aerosol particles can
 290 transform into CCN or INP, a process primarily determined by their chemical composition and ambient
 291 temperature (Bellouin et al., 2020). When these nuclei are activated, water vapor condenses on their
 292 surfaces to form cloud droplets or ice particles. As the concentration of aerosol particles increases, the
 293 number of CCN or INP may rise correspondingly, leading to an increase in the number of cloud droplets.
 294 Notably, under conditions where the liquid water content in clouds remains constant (i.e., LWP), the same
 295 amount of water vapor is distributed across more cloud droplets, resulting in a reduction in the size of
 296 individual droplets. Specifically, as aerosol concentration increases, the CER decreases, while cloud
 297 albedo increases. On the basis of findings of Kaufman and Fraser (1997), Feingold et al. (2001) pointed
 298 out that the sensitivity of cloud microphysical properties (e.g., CER) to changes in the number
 299 concentration of aerosol particles (e.g., using AOD as a measure) can be described by the following
 300 formula:

301
$$S_{\text{CER}} = \left. \frac{d \ln r_e}{d \ln \alpha} \right|_{\text{LWP}} \quad -0.33 < S < 0 \quad (1)$$

302 Where r_e represents the CER and α represents the AOD. Following Andreae (2009), AOD and CCN
 303 are correlated and AOD varies with CCN following a power law relationship. Eq. (1) describes the
 304 relative change of CER with the relative change of the AOD for constant LWP. It is noted that this
 305 formulation differs from that used in recent studies (e.g., Bellouin et al., 2020) where S is expressed in
 306 terms of N_d with no restriction in LWP. The sensitivity S of CER to AOD can be determined as the slope
 307 of a linear fit to a log-log plot of CER versus AOD. The effect of aerosols on CER is analyzed by
 308 comparing the difference in S_{CER} and correlation coefficients between AOD and CER under different
 309 spatial scales (Section 2.4) and LWPs (Section 3.2).

310 In this study, the variation in N_d with CCN is referred to as the susceptibility S_{N_d} . Following the method
 311 of Gryspeerdt et al. (2023), the sensitivity, S_{N_d} , of a cloud property, N_d , to α is defined here as

312
$$S_{N_d} = d \ln N_d / d \ln \alpha \quad 0 < S < 1 \quad (2)$$

313 Relations between CER and N_d and AOD are determined through Eq. 1 and Eq. 2 and correlation
 314 coefficients R. The significance of these relations is determined by using the student's t test, i.e. the
 315 results are statistically significant when the p value is smaller than 0.01, where p is defined as the
 316 probability of obtaining a result equal to or "more extreme" than what was actually observed.

317 This method quantifies the sensitivity of CER and N_d to AOD variations via linear regression in log-log
 318 space, using Eq. 1 and Eq. 2, respectively. Its core assumptions, uncertainties, and limitations are highly
 319 consistent: both rely on AOD as an aerosol proxy variable, assume constant cloud liquid water content
 320 and a linear sensitivity relationship, and depend on the reliability of satellite-retrieved parameters
 321 (Feingold et al., 2001; Gryspeerdt et al., 2023). However, AOD cannot distinguish aerosol size and
 322 hygroscopicity, retrieval errors are substantial in clean conditions, and linear fitting fails to capture
 323 nonlinear/non-monotonic responses. Both methods are constrained by satellite retrieval biases, limited
 324 scenario applicability (only valid for specific homogeneous clouds and aerosol types), the omission of
 325 key modulating factors (dynamical conditions, aerosol type) and feedback processes, and can only assess
 326 first-order direct effects. Reliability requires scenario constraints and uncertainty analysis; the only
 327 nuances come from the target variable (CER vs. N_d), which do not alter the shared methodological
 328 limitations.

329

330 2.4 Research design for scale effects analysis

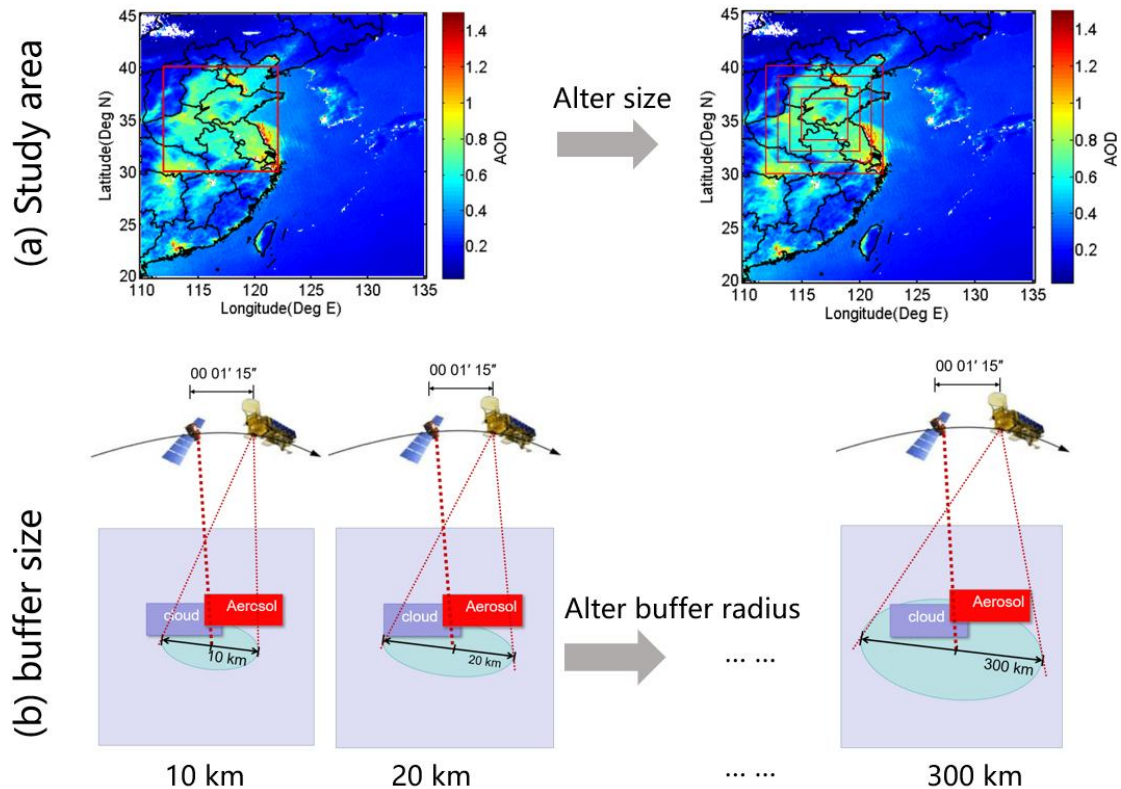
331 This study was conducted at multiple spatial scales to examine the scale dependence of S_{CER} and S_{Nd} in
332 delineating aci (Fig. 2). Here, the spatial scales are described by two parameters: study area size (the
333 geographic scope of the analysis) and buffer size (the local spatial extent around each observation point
334 for aggregating aerosol and cloud data). To this end, the study area was divided into four congruent square
335 research areas all centered at the same geographical location (35°N, 117°E) over Eastern China. Hence,
336 spatial extent varies from the whole study area as defined in Section 2.1 (30°N-40°N, 112°E-122°E) to
337 successively smaller areas simulated by decreasing the study area in steps of 2° to 4°×4° as illustrated in
338 Figure 2a.

339 Buffer size refers here to a circular spatial domain centered at each point in the study area where CALIOP
340 detected the presence of aerosols. Within this circular domain, MODIS-retrieved cloud and aerosol data
341 (AOD, CER, N_d , LWP) are spatially averaged to construct matched aerosol-cloud datasets at different
342 local scales. As previously noted, this approach relies on the assumption that aerosol properties are
343 reasonably homogeneous between adjacent clear and cloudy regions (Anderson et al., 2003; Quaas et al.,
344 2008), and this premise is supported by the short-range transport of aerosols (e.g., 10–300 km) and the
345 near-simultaneous observations (1–2 minutes) by MODIS and CALIOP within the A-Train constellation.
346 Buffer zones with sizes increasing from 10 to 300 km (10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 120, 140,
347 150, 160, 180, 200, 250, and 300 km) were determined within the whole study area by using CALIOP
348 data. Previous observations indicate that the typical horizontal scale of cloud clusters ranges from tens
349 to hundreds of kilometers (Zhang et al., 2024; Cai et al., 2022), supported by CloudSat/CALIPSO satellite
350 data showing power-law distributed cloud scales (10-1000 km fitting range) covering major cloud types
351 (Zhang et al., 2024) and regional evidence of consistent multi-season, multi-latitude cloud extents (Cai
352 et al., 2022). Meanwhile, aerosol spatial homogeneity varies with distance: local-scale aerosols (≤ 50 km)
353 exhibit high homogeneity due to consistent sources and stable diffusion, while regional-scale aerosols ($>$
354 100 km) show enhanced heterogeneity from multi-source mixing and atmospheric transport (Hassan et
355 al., 2024; Mohebalhojeh et al., 2026). Thus, the 10–300 km buffer range covers both cloud characteristic
356 scales and the aerosol homogeneity transition range, ensuring that MODIS data averaging effectively
357 captures aci. This range avoids insufficient MODIS pixel coverage due to excessively small buffer sizes
358 (< 10 km). It also prevents conflation between regional meteorological variations and local aci signals

359 arising from overly large buffer sizes (> 300 km), as synoptic-scale circulation and other regional
360 meteorological changes may interfere with local aci signals (Quaas et al., 2010). Meanwhile, this range
361 aligns with the 50 – 150 km buffer sizes widely adopted in regional aci studies (Wang et al., 2015; Liu et
362 al., 2017; 2024), enabling cross-validation of results and ensuring that MODIS data averaging effectively
363 captures aci.

364 MODIS-retrieved cloud and aerosol data were averaged over a buffer area around each CALIOP data
365 point with a radius varying from 10 to 300 km. Thus, a dataset including aerosol and cloud properties
366 was constructed with different buffer sizes. The effect of buffer size on the sensitivity of CER and N_d to
367 variations in AOD was determined in each study area varying from $4^\circ \times 4^\circ$ to $10^\circ \times 10^\circ$. To this end, for
368 each buffer size, the averaged AOD and cloud parameters were paired to calculate the sensitivities S_{CER}
369 (Eq. 1) and S_{N_d} (Eq. 2), as well as their correlation coefficients (R) between cloud properties (e.g., CER,
370 N_d) and AOD. The optimal buffer size for each study area is defined as the one maximizing the R. This
371 definition is adopted based on two core considerations. Firstly, it aligns with the statistical principle that
372 a higher R value indicates a stronger linear correlation between the two variables in log-log space,
373 minimizing the interference of random noise and non-aerosol confounding factors on the sensitivity
374 estimation (Quaas et al., 2006; Gryspeerdt et al., 2022). This ensures that the derived S_{CER} can reliably
375 reflect the intrinsic relationship between aerosol loading and cloud droplet effective radius, rather than
376 spurious correlations caused by inappropriate spatial scales. Secondly, this definition also facilitates
377 comparability with existing literature, as it aligns with the methodological framework of satellite-based
378 aci studies (Saponaro et al., 2017; Liu et al., 2021). In these studies, the optimal spatial scale is typically
379 identified by maximizing the statistical robustness of variable correlations.

380 The dataset was used to study the characteristics of aerosol indirect effects as function of buffer size and
381 study area, for two different periods: one with a high aerosol content (2008-2014) and another one with
382 a decreasing aerosol content (2015-2022). This approach determined the optimal buffer size for aerosol
383 indirect effect estimations as a function of study domain size, providing key observational constraints for
384 constructing scale-adaptive parameterization frameworks suitable for different domain sizes over eastern
385 China.



386

387 **Figure 2. (a) Schematic diagram of study area and buffer size patterns applied in this study. (b) scheme of**
 388 **CALIPSO-MODIS coincidence methodology. When CALIPSO detects the presence of aerosol and cloud fields,**
 389 **we look for MODIS retrievals within a buffer size from the CALIPSO target. The temporal coincidence is**
 390 **insured by the coordinated satellite orbits.**

391 **3 Results**

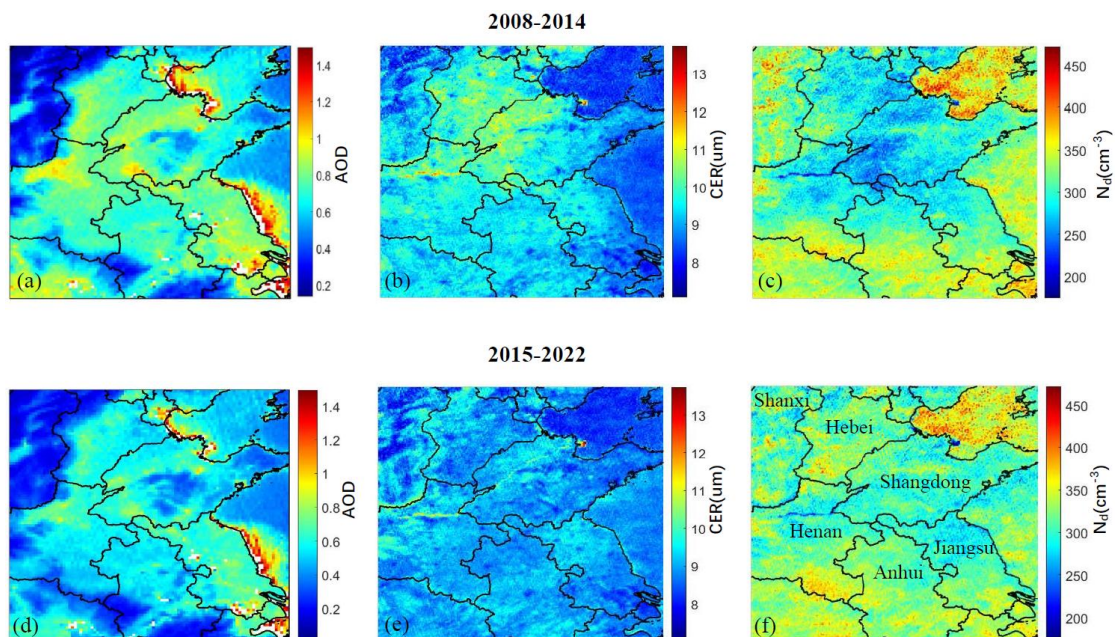
392 **3.1 Spatial variations of aerosol and cloud parameters**

393 Figure 3 illustrates the spatial distributions of AOD and cloud properties (CER and N_d) across the study
 394 region, averaged for the periods 2008-2014 and 2015-2022. The AOD spatial patterns (Fig. 3a,d) show
 395 similar spatial distributions during both periods, but with notably reduced values during the latter.
 396 Pronounced spatial gradients in AOD are evident during both periods. The lowest AOD values occur over
 397 the mountainous regions of Shanxi province in the northwest, while elevated concentrations appear in
 398 the southeastern areas encompassing the Hebei and Shandong provinces. This geographical contrast
 399 arises from the mountain ranges that demarcate the heavily industrialized, densely populated North China
 400 Plain (NCP) in the east - characterized by substantial anthropogenic emissions - from the relatively
 401 cleaner western regions. Under prevailing southeasterly wind conditions, these topographic barriers
 402 effectively block transport of atmospheric pollutants which accumulate along their windward slopes

403 (Sundström et al., 2012). The concentration of heavy industries and power generation facilities in the
 404 NCP are primarily responsible for the observed high AOD concentrations, together with meteorological
 405 and geographical conditions. Additionally, lower AOD values appear in southern Anhui and central
 406 Shandong relative to the surrounding regions.

407 The CER spatial distributions (Fig. 3b,e) reveal distinct differences between the two periods. During
 408 2008-2014, larger cloud droplets predominated in the northern sectors, particularly throughout Hebei and
 409 western Shandong. Notably, the spatial correspondence between AOD and CER maxima aligns with the
 410 anti-Twomey effect, suggesting that the high aerosol loading promoted cloud droplet growth rather than
 411 suppression - consistent with findings from Wang et al. (2014) and Liu et al. (2018). The 2015-2022
 412 period shows markedly reduced CER values (typically $<10 \mu\text{m}$) with enhanced spatial homogeneity.

413 Similarly, N_d exhibits contrasting spatial patterns between the two periods (Fig. 3c,f). The earlier
 414 timeframe shows depressed N_d values in central regions surrounded by elevated concentrations
 415 peripherally. This pattern reverses during 2015-2022, with increases of N_d in the central area
 416 accompanied by overall reduction of the cloud droplet concentrations in the surrounding regions.

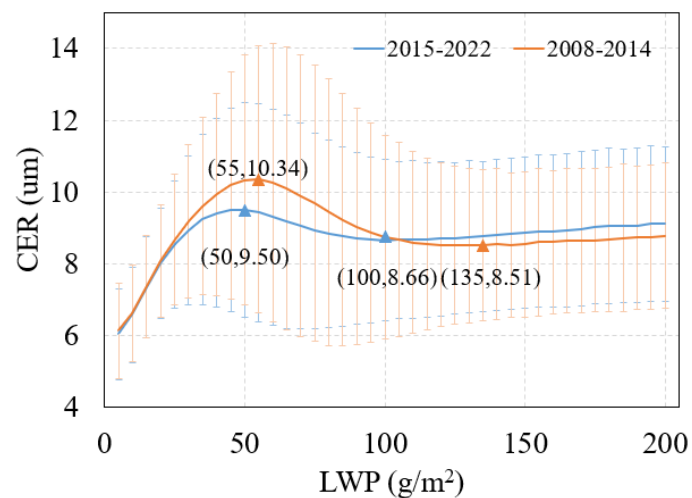


417
 418 **Figure 3. Spatial distributions of AOD (a, d), CER (b, e) and N_d (c, f), averaged over the years 2008-2014 (top**
 419 **row) and 2015-2022 (bottom row) over the study area. The lines are provincial borders and the names of**
 420 **provinces mentioned in the text are indicated in Fig. (f).**

421 3.2 Sensitivity of CER to AOD stratified by LWP

422 Before analyzing the influence of AOD on CER, the relationship between CER and LWP should be

423 investigated. The values of the LWP were divided into 40 subsets with a width of 5 g/m^2 , and then the
 424 average value of CER in each subset was calculated and plotted as function of LWP (Figure 4).
 425 The variation of CER with LWP shows three regimes. For LWP smaller than 55 g/m^2 (period 1) or 50
 426 g/m^2 (period 2), CER increased rapidly with the increase of LWP. This first LWP regime is referred to as
 427 a rapid growth regime (LWP1). The second LWP regime, referred to as a decreasing regime (LWP2),
 428 applies to the LWP range from 55 g/m^2 to 135 g/m^2 (period 1) or $50\text{-}100 \text{ g/m}^2$ (period 2) and CER
 429 decreased with the increase of LWP. When LWP was greater than 135 g/m^2 (period 1) or 100 g/m^2 (period
 430 2), CER increased with increasing LWP but at a much slower rate than during the first regime; the third
 431 LWP regime is therefore referred to as a slow growth regime (LWP3). These results show that CER is
 432 very sensitive to the changes in LWP, which is consistent with the study of Liu et al. (2021). Specifically,
 433 CER exhibited a three-stage variation with LWP: rapid growth when $\text{LWP} < 50 \text{ g/m}^2$ (with the fastest
 434 change rate), a stable state during $50\text{-}150 \text{ g/m}^2$, and slow growth when $\text{LWP} > 150 \text{ g/m}^2$ (at a rate much
 435 lower than the first stage). This highlighted the necessity of fixing LWP conditions to accurately
 436 investigate the impact of AOD on CER. To separate the effects of changing LWP on CER from those of
 437 changing AOD on CER, relations between CER and AOD were evaluated for constant LWP
 438 (McComiskey et al., 2012), for each of the three regimes mentioned above, by using double-logarithmic
 439 plots of AOD versus CER. The number of CER observations in the third regime is too small to achieve
 440 statistically meaningful results, therefore the sensitivity of CER to AOD was only analyzed for the rapid
 441 growth and decreasing CER regimes.



442
 443 **Figure 4. Variation of CER with LWP over the study area. Here all CER data were averaged in LWP bins,**
 444 **from 0 to 200 g/m^2 with a width of 5 g/m^2 . The red line is a fit to the data during 2008-2014 and the blue line**
 445 **for the data during 2015-2022. The numbers in parentheses indicate the (LWP, CER) values for the inflection**

446 points between the regimes. The error bars (the vertical lines) denote the standard deviations, indicating the
447 variability of CER around the average at each LWP value.

448 3.2.1 Rapid CER growth regime

449 For the first LWP regime, the S_{CER} is negative (as shown in Figure 5). This aligns with the Twomey effect
450 (Twomey, 1977): an increase in aerosols raises the number of CCN, and with constant LWP, less water
451 vapor is available per cloud droplet, reducing CER, increasing cloud albedo, and ultimately cooling the
452 atmosphere. This indicates that, in the rapid growth regime, the interaction between AOD and CER in
453 the target region is in agreement with the Twomey effect. The data in Figure 5a show that during period
454 1, S_{CER} varies with buffer size and that the variations are different for different study areas. The value of
455 $|S_{CER}|$ is smallest for the largest study area ($10^\circ \times 10^\circ$) and decreases with buffer size to a minimum for
456 buffer size of 150 km and then increases. For the smallest study area ($4^\circ \times 4^\circ$), the sensitivity exhibits a
457 much stronger negative correlation, with its magnitude decreasing as the buffer size expands, especially
458 for $50 \text{ km} < \text{buffer size} < 100 \text{ km}$. For the 2 intermediate study areas, the sensitivities are initially similar
459 (except for the smallest buffer size) and diverge for $\text{buffer size} > 100 \text{ km}$. The data show that the value of
460 $|S_{CER}|$ overall becomes smaller as study area increases. The decrease of $|S_{CER}|$ with increasing study area
461 is mechanistically tied to scale-dependent aerosol indirect effect theory and meteorological confounding
462 (Quaas et al., 2009; McComiskey & Feingold, 2012). The correlation coefficients R (bottom of Figure 5
463 a) are similar for all four study areas at small buffer sizes, increase fast with buffer size to a maximum
464 for a buffer size of about 50 km and then decrease and diverge. The largest decrease is observed for the
465 study area of $10^\circ \times 10^\circ$. In this study, the optimal scale for each study area is indicated in Figure 5 with a
466 red solid square. A plot of the optimal scale versus the size of the study area in Figure 6 (curve LWP1-
467 period1) shows that, as the study area size increased from $6^\circ \times 6^\circ$ to $10^\circ \times 10^\circ$, the optimal scale decreased
468 from 100 km to 30 km.

469 As compared with period 1, in period 2 (Figure 5b) the value of the $|S_{CER}|$ also decreases as the buffer
470 size increases. However, the scale sensitivity analysis for period 2 reveals two distinct characteristics
471 different from period 1: (1) the four S_{CER} curves for different study areas are much closer than during
472 period 1; (2) with the exception of the study area of $10^\circ \times 10^\circ$, the values of the $|S_{CER}|$ for the other three
473 study areas are significantly reduced (closer to zero), indicating a corresponding weakening of aci
474 intensity against the background of decreased aerosol concentrations. Particularly noteworthy is that

475 during period 1, the R values for the $10^\circ \times 10^\circ$ study area show a sharp declining trend when the buffer
476 size exceeds 60 km, while in period 2 this happens for buffer size larger than 110 km and the R value
477 curves for all study areas significantly expand their high-value ranges. Also for period 2, the R values for
478 the study areas of $10^\circ \times 10^\circ$ and $8^\circ \times 8^\circ$ are very similar, in contrast to period 1 when only the R values for
479 the study areas of $10^\circ \times 10^\circ$ decreases fast. Across different study areas, the optimal scale (Curve LWP1-
480 peirod2) shows a behavior opposite to that during period 1: as the study area size increases from $6^\circ \times 6^\circ$
481 to $10^\circ \times 10^\circ$, the optimal scale increases from 20 km to 80 km. It is noted that for a study area of $4^\circ \times 4^\circ$
482 the optimal scale is 50 km in both periods. For the first LWP regime, S_{CER} estimates and correlation
483 coefficients R by period and optimal buffer size across $4^\circ \times 4^\circ$ to $10^\circ \times 10^\circ$ study areas are given in
484 Appendix A1.

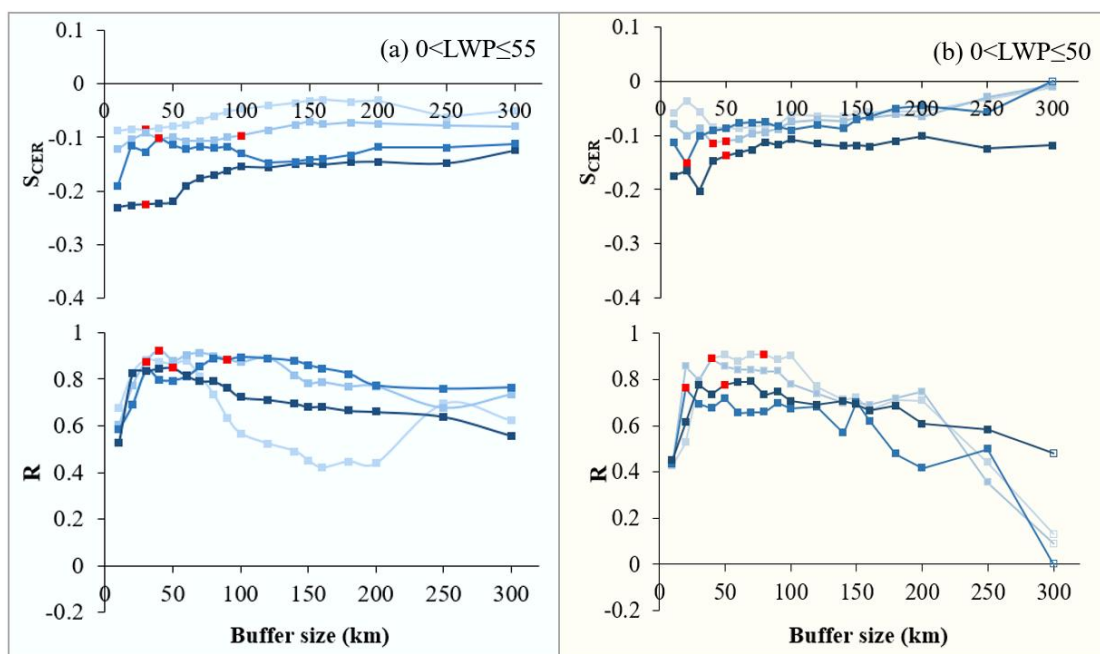
485 **3.2.2 Decreasing CER regime**

486 During the second LWP regime (Figure 7), the AOD and CER is also negatively correlated ($S_{CER} < 0$)
487 during both periods, but the values of S_{CER} vary stronger with the size of study area: the curve dispersion
488 increases in the second LWP regime, the values of $|S_{CER}|$ are larger overall, and the relatively concentrated
489 trend in the period 1 shifts to more scattered fluctuations in the period 2. During period 1, for the largest
490 ($10^\circ \times 10^\circ$) study area: the value of $|S_{CER}|$ (Figure 7a) decreases with increasing buffer size, nearing zero
491 at ~150 km, briefly turning slightly positive, then slowly falling below zero by 220 km. For the three
492 smaller study areas, the S_{CER} values are all negative and $|S_{CER}|$ overall decreased with increasing buffer
493 size. Through the sensitivity of S_{CER} to buffer size across varying study areas during period 1, the value
494 of $|S_{CER}|$ for a given buffer size becomes smaller with increasing study area size. Additionally, across all
495 study areas, $|S_{CER}|$ initially increases with the increase of the buffer size, then decreases, and gradually
496 stabilizes thereafter. The data in Figure 6 (Curve LWP2-peirod1), show that, the optimal size varies
497 between 30 km and 60 km with no clear dependence on the size of the study area.

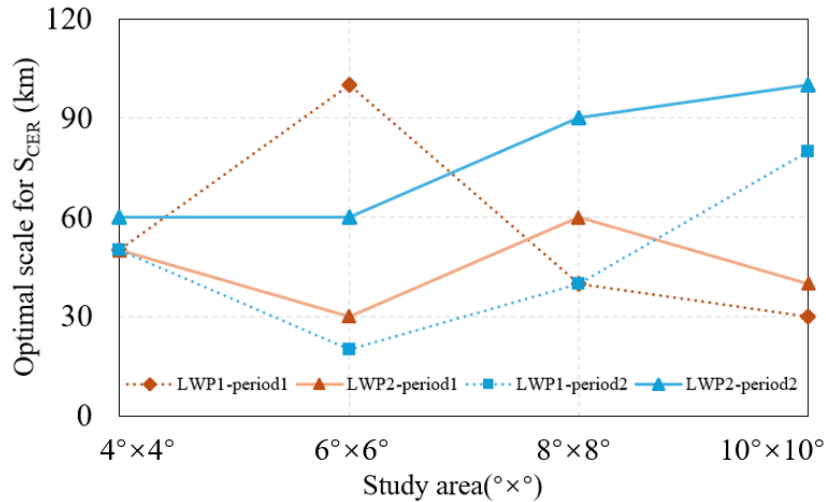
498 Compared with period 1, the value of $|S_{CER}|$ in period 2 (Figure 7b) also decreased overall with increasing
499 buffer size, and at the same buffer size, $|S_{CER}|$ decreased as study area increased. However, the scale
500 sensitivity analysis for period 2 revealed two distinct characteristics: (1) the four curves for different
501 study areas were closer than during period 1; (2) the $|S_{CER}|$ for the study area of $10^\circ \times 10^\circ$ was overall
502 larger, while the values of the S_{CER} curves for the study areas of $4^\circ \times 4^\circ$ and $6^\circ \times 6^\circ$ were significantly

503 reduced and that for the study area of $8^\circ \times 8^\circ$ was slightly reduced. The reduction may be attributed to
 504 weakened aci resulting from decreased regional aerosol concentrations (Jia et al., 2022; Li et al., 2024;
 505 Zhao et al., 2025). The variation of the optimal scale (Curve LWP2-period2) with the size of the study
 506 area is similar to that during period 1 in the rapid growth LWP regime: as the study area size increased
 507 from $6^\circ \times 6^\circ$ to $10^\circ \times 10^\circ$, the optimal scale increased from 60 km to 100 km. For the second LWP regime,
 508 S_{CER} estimates and correlation coefficients R between CER and AOD by period and optimal buffer size
 509 across $4^\circ \times 4^\circ$ to $10^\circ \times 10^\circ$ study areas are given in Appendix A1.

510 Through comparative analysis of S_{CER} data distribution across different LWP regimes under different
 511 aerosol conditions (i.e. high AOD and decreasing AOD), we found that the $|S_{CER}|$ in the second LWP
 512 regime was significantly larger than that in first LWP regime except for the study area of $10^\circ \times 10^\circ$ for
 513 buffer size >100 m, where S_{CER} curves corresponding to different study areas showed greater dispersion.
 514 This pattern highlights the dominant role of LWP in regulating aci sensitivity, with AOD variations
 515 further modulating the magnitude of such differences. Sample sizes of CER and AOD for the first LWP
 516 regime across both study periods, all buffer sizes and study areas, are presented in Appendices A3–A6.

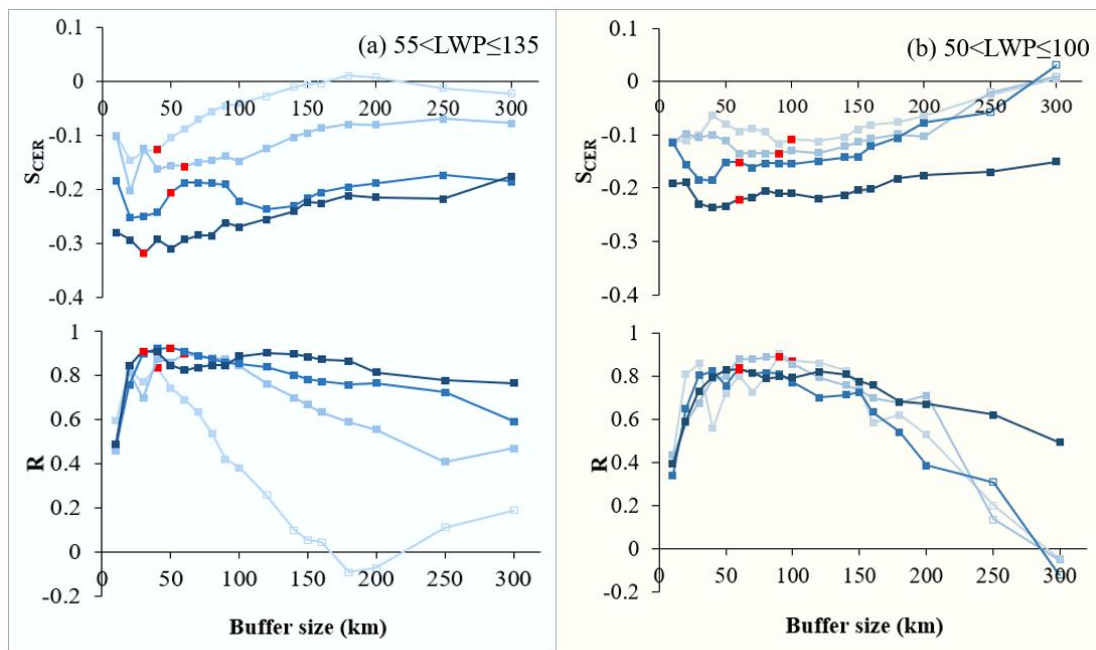


517 Study area $10^\circ \times 10^\circ$ $8^\circ \times 8^\circ$ $6^\circ \times 6^\circ$ $4^\circ \times 4^\circ$
 518 **Figure 5. Variation of S_{CER} (top) and correlation coefficient R (bottom) with buffer size for different study**
 519 **areas (see legend at the bottom) for (a) the LWP regime with $0 < LWP \leq 55$ g/m^2 over the years 2008-2014 and**
 520 **(b) the LWP regime with $0 < LWP \leq 50$ g/m^2 over the years 2015-2022. Filled squares indicate that the results**
 521 **are significant at the 0.01 level and open squares indicate that the results are not statistically significant. Red**
 522 **solid squares indicate the optimal buffer sizes for each study area, as shown in Appendix A1.**



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Figure 6. Relationship between optimal scale for SCER and the size of the study area. Here LWP1-period1 and LWP2-period1 indicate the optimal scale in period 2008-2014 for the first LWP regime and that for the second LWP regime, respectively. LWP1-period2 and LWP2-priod2 indicate the optimal scale in period 2015-2022 for the first LWP regime and that for the second LWP regime, respectively.



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Figure 7. Variation of SCER (top) and correlation coefficient R (bottom) with buffer size for different study areas (see legend at the bottom) for (a) the LWP regime with $55 < LWP \leq 135$ g/m² over the years 2008-2014 and (b) the LWP regime with $50 < LWP \leq 100$ g/m² over the years 2015-2022. Filled squares indicates that the results are significant at the 0.01 level and the open squares indicates that the results are not statistically significant. Red solid squares indicate the optimal buffer sizes for each study area, as shown in Appendix A1.

535 3.3 Sensitivity of N_d to AOD

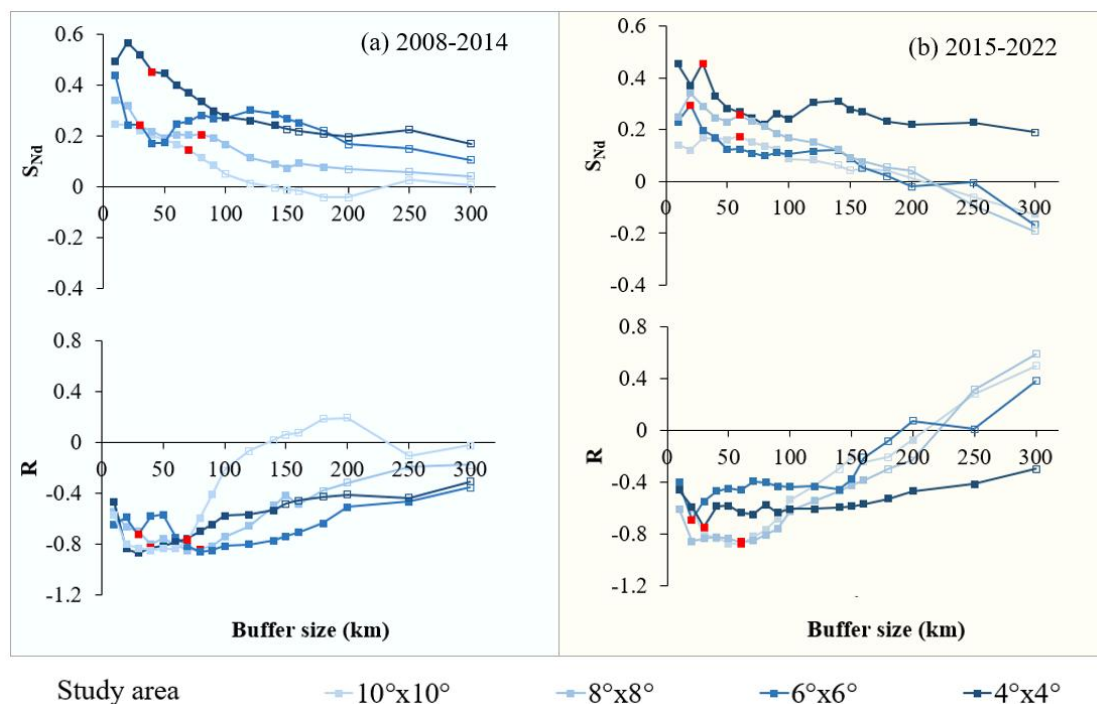
536 The sensitivity S_{Nd} is defined as the slope of a linear fit to a log-log plot of S_{Nd} versus AOD (Eq. 2). For

537 each period, we binned the data into AOD intervals of 0.02 and averaged N_d within each bin. Figure 8
538 shows the variation of S_{Nd} with buffer size for different study areas. In contrast to S_{CER} , S_{Nd} is
539 predominantly positive ($p < 0.01$) in both periods and decreases with increasing buffer size. During period
540 1 (2008–2014, high AOD), S_{Nd} for the $6^\circ \times 6^\circ$ study area decreases rapidly to a minimum at buffer sizes
541 of 40–50 km, then increases to a maximum at a buffer size of 120 km. For buffer size ≥ 120 km, the two
542 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
543 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
544 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
545 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
546 minimum for a buffer size of 80 km, followed by an increase to a maximum for a buffer size of 140 km.
547 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
548 $8^\circ \times 8^\circ$ and $10^\circ \times 10^\circ$ areas (Fig. 9), reflecting different aci characteristics under varying AOD conditions.
549 Estimates of S_{Nd} and R, stratified by optimal buffer size, for study areas ranging from $4^\circ \times 4^\circ$ to $10^\circ \times 10^\circ$
550 during the two periods are presented in the Appendix A2.

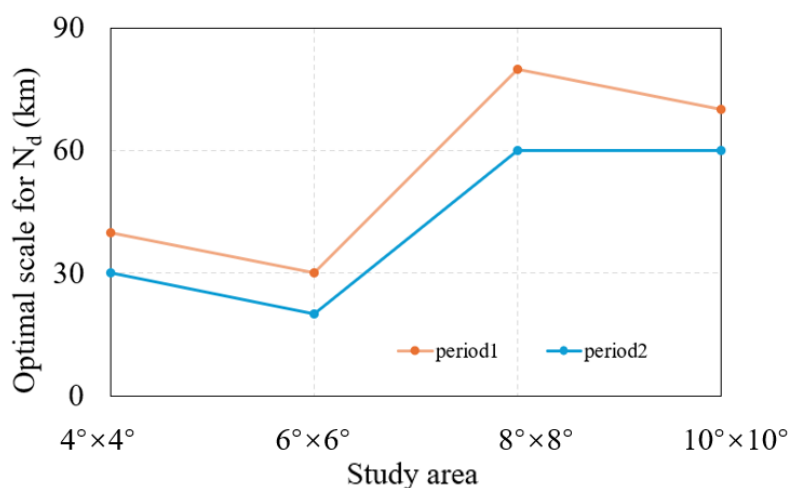
551 Expanding the study area inevitably increases spatial heterogeneity in aerosol loading, meteorology, and
552 cloud type, while also introducing larger satellite retrieval uncertainties. This spatial mixing blends clean
553 and polluted air masses, thereby introducing a statistical dilution effect on the estimated S_{Nd} . Therefore,
554 the decrease in S_{Nd} with increasing study area size (Fig. 8) reflects not only a genuine weakening of
555 aerosol-cloud interactions but also the combined effects of meteorological confounding, cloud regime
556 transitions, and retrieval limitations. It is noted that S_{CER} and S_{Nd} exhibit distinct anomalies in large buffer
557 zones, which may be associated with all these factors.

558 Changes in aerosol chemical composition between the two periods may also modulate S_{Nd} . During 2008–
559 2014, aerosols over eastern China were dominated by sulfate (30%–40% of $PM_{2.5}$ mass; Huang et al.,
560 2014; Zheng et al., 2018). Given the strong hygroscopicity of sulfate-dominated aerosols (Zhang et al.,
561 2012; Liu et al., 2023), their CCN activation efficiency was likely high, providing a favorable physical
562 basis for aci (Lee et al., 2009). In contrast, during 2015–2022, policy interventions (e.g., the Air Pollution
563 Prevention and Control Action Plan; Zheng et al., 2018) drove a structural transition: the sulfate mass
564 fraction dropped sharply to 15%–25% (an absolute reduction of $>50\%$), while less hygroscopic
565 components (nitrate, carbonaceous aerosols, and secondary organic aerosols) increased in relative

566 proportion (Huang et al., 2014; Zheng et al., 2018). As these components generally exhibit weaker
 567 hygroscopicity compared with sulfate (Zhang et al., 2012; Liu et al., 2023), this compositional shift may
 568 have reduced CCN activation efficiency under the same AOD, thereby weakening the sensitivity of N_d
 569 to AOD and altering aci intensity (Lee et al., 2009).



570 Study area $10^{\circ} \times 10^{\circ}$ $8^{\circ} \times 8^{\circ}$ $6^{\circ} \times 6^{\circ}$ $4^{\circ} \times 4^{\circ}$
 571 **Figure 8. Variation of S_{Nd} (top) and correlation coefficient R (bottom) with buffer size for different study areas**
 572 **(see legend at the bottom) for the LWP regime with $0 < LWP \leq 200 \text{ g/m}^2$ over the years 2008-2014 (a) and 2015-**
 573 **2022 (b). Filled squares indicates that the results are significant at the 0.01 level and open squares indicate**
 574 **that the results are not statistically significant. Red solid squares indicate the optimal buffer size for each**
 575 **study area, as shown in Appendix A2.**



576 **Figure 9. Relationship between optimal scale for N_d and the size of the study area. Here period1 and period2**
 577 **indicate the optimal scale in period 2008-2014 and the optimal scale in period 2015-2022, respectively.**
 578

579 **4 Discussion**

580 **4.1 The importance of liquid water path constraint**

581 LWP is a critical parameter governing cloud radiative properties (Murray-Watson et al., 2022). The
582 quantification of albedo effects strongly depends on the spatial scale and the LWP. Neglecting LWP
583 constraints in aci studies can weaken microphysical signals, leading to underestimation of radiative
584 forcing (McComiskey et al., 2012). To address this, we first systematically investigated the dynamic
585 relationship between CER and LWP before analyzing CER sensitivity to AOD. The results demonstrate
586 pronounced CER sensitivity to LWP variations, which can be categorized into three distinct regimes
587 (Figure 4):

588 In the first LWP regime, CER increases rapidly with LWP, i.e. the evolution of CER is predominantly
589 driven by changes in LWP. This dominance may lead to overestimation of the influence of the AOD on
590 CER (Liu et al., 2021).

591 In the second LWP regime, CER decreases with increasing LWP. In this regime, the regulatory effect of
592 LWP on CER weakens significantly, and CER variations become increasingly governed by aerosol-
593 related processes, indicating the growing dominance of aerosol indirect effects.

594 The third regime contains an insufficient number of CER observations to yield statistically significant
595 results, which excludes the analysis of the sensitivity of CER to AOD.

596 Comparative analysis of scale-conditioned S_{CER} across LWP regimes in periods 1 and 2 revealed
597 markedly enhanced sensitivity of S_{CER} to AOD in the second LWP regime. A trade-off exists between
598 AOD and LWP under conditions of insufficient water vapor. This trade-off leads to smaller CER values.

599 As suggested by Costantino et al. (2013), the LWP response to aerosol invigoration is influenced by two
600 competing mechanisms: a drying effect caused by enhanced entrainment of dry air at cloud top (dominant
601 in optically thin clouds) and a moistening effect from precipitation suppression (dominant in optically
602 thick clouds). For larger LWP, the supply of cloud water is sufficient. The increase in aerosol number
603 concentrations significantly affects the distribution of cloud droplet number concentrations and sizes.

604 This further enhances the sensitivity of CER to AOD. For small aerosol concentrations, the values of
605 $|S_{CER}|$ (Figure 5b, 7b) decreased overall with expanding buffer size within the same study area. For fixed
606 buffer size, $|S_{CER}|$ decreased as the study area increased. The ranges of $|S_{CER}|$ values across different study
607 areas showed a convergent pattern. These values typically remained small and close to zero. During the

608 high AOD period (2008–2014), anthropogenic emissions and dust transport provided abundant CCN.
609 This laid the material foundation for strong aci. It enhanced the synergistic effect of “sufficient liquid
610 water + abundant CCN” in the second LWP regime. This synergistic effect amplified the difference in
611 S_{CER} between the two LWP regimes. In the period of decreasing AOD (2015–2022), CCN concentration
612 decreased following the implementation of clean air policies (de Leeuw et al., 2021; 2023). This
613 reduction weakened the direct impact of aerosols on CER. However, the LWP-driven microphysical
614 differences persisted. Thus, S_{CER} in the second regime remained significantly smaller than that in the first
615 regime, albeit with a smaller difference. Additionally, the complexity of aerosol types during the high
616 AOD period (e.g., mixing of anthropogenic pollutants and natural dust) may have adjusted the value of
617 S_{CER} . However, it did not alter the dominant role of LWP. This aligns with the theory that “aerosol indirect
618 effects are jointly regulated by concentration and type” (Liu et al., 2017).

619 The larger S_{CER} observed at larger spatial scales (Figures 5 and 7) may be attributed to meteorological
620 confounding effects. In addition, clouds with larger LWP are usually associated with strong updrafts
621 (such as convective clouds), and stronger turbulence and vertical transport will bring more aerosols into
622 the clouds, increasing CCN concentration and a decrease in particle size, making them more sensitive to
623 changes in AOD (Jones et al., 2009; Han et al., 2022; Fan et al., 2025). Therefore, this phenomenon is
624 the result of the combined action of cloud microphysical processes (CCN activation, cloud droplet
625 competition growth) and dynamic processes (updrafts, turbulent mixing). If the characteristics of aerosols
626 (such as composition) change in the second LWP regime, this sensitivity may be further amplified.
627 Consequently, the LWP-stratified S_{CER} quantification framework enables precise characterization of
628 scale-dependent aci, providing robust physical insights for climate effect assessments and effectively
629 reducing uncertainties in future climate projections.

630 The central hypothesis of this study—that LWP is relatively consistent between the two periods (2008–
631 2014 and 2015–2022), supporting valid comparisons of the spatial sensitivity of AOD-CER
632 relationships—is well-supported by the following analysis. The differences in the mean, median, 25th,
633 and 50th percentiles of LWP between the two periods are all less than 5%, indicating a stable overall
634 water vapor supply level. The spatial patterns of high-LWP regions (e.g., southeastern areas) and low-
635 LWP regions (e.g., the mountainous areas in northern Shanxi) remained stable across the two periods
636 (see Appendix B1), demonstrating that LWP spatial distribution characteristics are highly consistent. The

637 sample proportions of LWP in the rapid growth regime are 59.30% (period 1: 0–55 g/m²) and 55.36%
638 (period 2: 0–50 g/m²), while those in the decreasing regime are 29.64% (period 1: 55–135 g/m²) and
639 24.59% (period 2: 50–100 g/m²), suggesting that there is no systematic temporal shift in the LWP
640 distribution. Meanwhile, short-term fluctuations are smoothed by multi-year averaging and large-sample
641 statistics, resulting in a weak indirect impact of aerosols on LWP (LWP only increased by 5.6%, much
642 smaller than the 24% decrease in AOD). Additionally, LWP-stratified analysis (i.e., binning LWP into
643 5 g m⁻² intervals) further isolates interference. The validation of the core hypothesis provides a reliable
644 premise for accurately quantifying the impact of aerosol concentration changes on the sensitivity of cloud
645 parameters and their spatial scale dependence.

646 **4.2 Scale dependence of cloud parameters sensitivities to aerosol variations**

647 Extensive studies have demonstrated a significant spatial scale dependence of aerosol indirect effects
648 (McComiskey et al., 2012; Possner et al., 2016; Glotfelty et al., 2020; Ekman et al., 2023). Failure to
649 explicitly define the scale-dependent behavior of aerosol indirect effects may introduce systematic biases
650 and inconsistencies in subsequent process analyses. Based on satellite observations, this study confirms
651 statistically significant negative correlations between CER and AOD, as well as positive correlations
652 between N_d and AOD during periods with different aerosol concentrations, aligning with classical aci
653 theory (Quaas et al., 2009). Analysis of scale-conditioned S_{CER} and S_{N_d} reveals that for fixed buffer size,
654 an increase in the size of the study area leads to a systematic reduction in S_{CER} (less negative) and S_{N_d},
655 corroborating the nonlinear attenuation of aerosol signals with spatial domain expansion (Quaas et al.,
656 2009). The results from this study suggest that AOD-cloud property correlations in large study areas are
657 susceptible to meteorological confounding effects (Quaas et al., 2010; Boucher and Quaas, 2012;
658 Gryspeerdt et al., 2014; Liu et al., 2024). Theoretically, aerosol regulation of cloud microphysics is
659 strongly local: smaller domains (e.g., 4°×4°) feature homogeneous meteorological conditions (humidity,
660 updrafts), preserving undiluted aci signals and yielding larger |S_{CER}| (pronounced Twomey effect). In
661 contrast, expanded domains (e.g., 10°×10°) encompass heterogeneous meteorological conditions
662 (circulation differences, boundary layer variability) that independently modulate cloud droplet growth.
663 For example, strong updrafts enhance liquid water supply, offsetting aerosol-induced radius reduction
664 (Altaratz et al., 2014), weakening aerosol-CER correlations and reducing |S_{CER}|. Consistent with Grandey

665 & Stier (2010), large-scale domains introduce “dilution bias” via non-target meteorological variability.
666 This scale-dependent confounding mechanism elucidates uncertainties in aerosol indirect effect
667 assessments at regional scales.
668 Multi-scale spatial analysis identifies different optimal buffer sizes for S_{CER} and S_{Nd} in different periods.
669 These findings align closely with satellite-based aerosol indirect effect studies (Wang et al., 2015; Liu et
670 al., 2017), providing critical scale benchmarks for satellite product validation. Wang et al. (2015) reported
671 an inverse “Twomey” effect between aerosols and CER in eastern China by analyzing aerosol and CER
672 within a 50 km buffer zone around CALIOP samples. Similarly, Liu et al. (2017) systematically examined
673 the response mechanisms of warm cloud macro- and microphysical parameters to increasing AOD in the
674 Yangtze River Delta region, also using CALIOP samples within a 50 km buffer zone. The present study
675 further shows that, as aerosol concentrations decrease, S_{CER} values across different study areas with the
676 same buffer size exhibit convergence characteristics, with generally smaller S_{CER} (closer to zero). This
677 indicates a significant weakening of aci intensity and reduced spatial extent dependency in low aerosol
678 loading conditions. This phenomenon is consistent with the simulated behavior of aerosol-limited cloud
679 regimes, where aci are quantitatively modulated by moisture availability and lose their sensitivity to
680 large-scale dynamical stability, leading to a weaker and more homogeneous effect (Zhao et al., 2025).
681 By systematically quantifying the scale-response characteristics of aerosol indirect effects, this work not
682 only elucidates the dynamic scale behavior of aci, but also, more critically, establishes criteria for
683 determining optimal buffer size in regional aerosol indirect effect studies. These results provide
684 quantitative observational constraints for refining scale-dependent parameterization modules in climate
685 models, particularly for representing the modulation of ACI by spatial scale and LWP, thereby improving
686 model predictive reliability.

687 **4.3 Contrasting sensitivity patterns of cloud parameters in response to AOD**

688 A comprehensive comparison of the sensitivity S_{CER} and S_{Nd} reveals that the responses of CER and N_d to
689 AOD exhibit distinct yet inherently interconnected characteristics. These characteristics are jointly
690 modulated by spatial scale and LWP regimes (Figs. 5, 7, 8; Appendices A1–A2), which profoundly reflect
691 the core microphysical mechanisms of aci. Details are elaborated as follows:

692 **4.3.1 Core differences in response modes between S_{CER} and N_d to AOD**

693 S_{CER} is consistently negative across both periods and all LWP regimes ($-0.33 < S_{CER} < 0$) (Figs. 5, 7;
694 Appendix A1), indicating that an increase in AOD leads to a decrease in CER. This aligns with the core
695 principle of the Twomey effect (Twomey, 1977; Feingold et al., 2001). The values of $|S_{CER}|$ are larger in
696 the second LWP regime than in the first regime, reflecting stronger aerosol modulation of cloud
697 microphysical properties when liquid water is abundant (McComiskey & Feingold, 2012). In contrast,
698 S_{Nd} maintains a significant positive correlation with AOD across all scenarios ($0 < S_{Nd} < 1$) (Fig. 8;
699 Appendix A2), confirming that higher AOD directly promotes CCN activation and thereby increases
700 cloud droplet number concentration (Andreae, 2009).

701 **4.3.2 Synergistic modulation of AOD and spatial scale**

702 Using the LWP interval corresponding to S_{Nd} ($0 < LWP \leq 200 \text{ g/m}^2$) as a benchmark, comparisons
703 between the two periods (incorporating average values of S_{CER} across two LWP regimes) reveal distinct
704 characteristics:

705 For the small-scale study area ($4^\circ \times 4^\circ$): In period 1, the average $|S_{CER}|$ across two LWP regimes is 0.271
706 (0.2232 for the 0–55 g/m^2 LWP regime, 0.3189 for the 55–135 g/m^2 LWP regime) and $S_{Nd}=0.4496$, both
707 significantly higher than those in period 2 (average $|S_{CER}|=0.154$, with 0.0863 for the 0–50 g/m^2 LWP
708 regime and 0.2212 for the 50–100 g/m^2 LWP regime; $S_{Nd}=0.2903$). The negative correlation between
709 AOD and CER is more significant in period 1, as sufficient CCN in small-scale areas amplifies both
710 cloud droplet number increase and size reduction, enhancing the Twomey effect.

711 For the medium-to-large scale study areas ($6^\circ \times 6^\circ$, $8^\circ \times 8^\circ$, $10^\circ \times 10^\circ$): In period 1, the average $|S_{CER}|$ across
712 the two LWP regimes is 0.1683 (0.1305 for 0–55 g/m^2 , 0.2061 for 55–135 g/m^2), 0.13065 (0.1026 for 0–
713 55 g/m^2 , 0.1587 for 55–135 g/m^2), and 0.1067 (0.0858 for 0–55 g/m^2 , 0.0885 for 55–135 g/m^2),
714 respectively, all higher than the corresponding values in period 2 (0.1516, 0.1246, 0.0985). However, S_{Nd}
715 in period 1 (0.2430, 0.2050, 0.1430) is lower than that in period 2 (0.2960, 0.2680, 0.1740), with no
716 significant difference in the negative correlation between AOD and CER between the two periods.

717 This characteristic indicates that meteorological confounding effects are enhanced at larger scales,
718 weakening the regulation of S_{Nd} by aerosols, while at small scales the aci is directly driven by AOD levels.

719 **4.4 Limitations and future perspectives**

720 This study has three significant limitations. Firstly, similar to most previous studies (Wang et al., 2015;
721 Liu et al., 2021), this study only utilized MODIS data with a resolution of 10 km to explore scale effects,
722 ignoring finer or coarser resolution data. Therefore, using a 10 km buffer size as the minimum
723 observation unit, this limitation makes the indirect effects of aerosols on smaller scales still unknown,
724 which may lead to inaccurate evaluation of aerosol indirect effects. Therefore, future research can
725 improve the sensitivity of aerosol indirect effects to scale changes by using observation data with higher
726 accuracy or model simulations. Secondly, the current research focuses on the influence of buffer size and
727 study areas, the potential impact of spatial aggregation methods (especially zoning directionality) on the
728 quantitative results of aerosol indirect effects has not been systematically evaluated. Future research
729 should further investigate the sensitivity of aerosol indirect effects to zoning direction. Moreover, the
730 current study employs a uniform buffer size for both aerosol and cloud parameters, failing to account for
731 potential interaction effects arising from discrepancies of buffer size between them. Therefore, clarifying
732 scale dependence will avoid directly extrapolating local observation results to a larger study area when
733 downscaling climate models or formulating regional environmental policies.

734 **5 Conclusions**

735 Based on MODIS and CALIOP satellite observations of aerosol and cloud parameters over land in eastern
736 China, a comparative analysis was conducted of the sensitivity of cloud microphysical parameters (CER
737 and N_d) to variations in AOD during two characteristic periods: 2008–2014 (period 1, high AOD) and
738 2015–2022 (period 2, decreasing AOD). Through systematic analysis of the relationship between CER
739 and LWP and their response mechanisms to AOD across these two periods, the following key conclusions
740 were drawn:

741 Firstly, CER exhibited three distinct regimes with varying LWP. During the rapid growth regime (LWP
742 $< 55/50$ g/m²), CER showed significant negative sensitivity to AOD (S_{CER}), consistent with the Twomey
743 effect; during the decreasing regime (LWP = 55-135/50-100 g/m²), S_{CER} remained negative but with
744 enhanced sensitivity; and during the slow growth regime (LWP $> 135/100$ g/m²), the rate of CER change
745 significantly decreased. These findings confirmed that LWP is a crucial, regulatory factor influencing the
746 CER response to AOD.

747 Secondly, scale dependence analysis led to the conclusion that characteristics in aci are significantly
748 scale-dependent, with systematic variations of both S_{CER} and S_{Nd} with changes in buffer size and study
749 area. The results of the study show that S_{CER} systematically increased as buffer size increased and became
750 negligible for buffer sizes larger than 150-200 km, while the optimal buffer sizes varied substantially
751 with the size of the study area in the range from $6^\circ \times 6^\circ$ to $10^\circ \times 10^\circ$: increased as study area increased
752 during period 2, but decreased in period 1 for regime 2. Particularly noteworthy is the enhanced
753 consistency of S_{CER} across different study areas and the significant increase in S_{CER} during period 2,
754 reflecting weaker aci due to declining regional aerosol concentrations.

755 Finally, the sensitivity of N_d to AOD (S_{Nd}) exhibited distinct characteristics compared to S_{CER} : S_{Nd} showed
756 a significant positive correlation ($p < 0.01$) and demonstrated nonlinear attenuation with increasing buffer
757 size and study area. The optimal buffer size for S_{Nd} was smaller during period 2 than during period 1 and
758 increased with the study area size, being substantially larger for study areas of $8^\circ \times 8^\circ$ and $10^\circ \times 10^\circ$ than
759 for study areas of $4^\circ \times 4^\circ$ and $6^\circ \times 6^\circ$ areas.

760 These observed patterns correspond to the fundamental microphysical pathway of aci, whereby increased
761 aerosol loading enhances N_d and reduces CER under constant LWP conditions. The results emphasize
762 that accurate quantification of ACI must explicitly consider both the phase-dependent characteristics of
763 LWP and spatial scale effects when assessing aerosol indirect effects.

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

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775 **Appendix**

776 **A1. Estimates of S_{CER} , computed using Eq. (1), and correlation coefficients R between CER and AOD,**
 777 **stratified by LWP, and optimal buffer size (see text), for study areas varying from $4^\circ \times 4^\circ$ to $10^\circ \times 10^\circ$, during**
 778 **the periods 2008-2014 and 2015-2022. Statistically significant data points are indicated with * (p value < 0.01).**

Years	LWP (g/m^2)	Study area	Optimal scale (km)	S_{CER}	R
2008-2014	0-55	$10^\circ \times 10^\circ$	30	-0.0858	0.8828*
		$8^\circ \times 8^\circ$	40	-0.1026	0.9220*
		$6^\circ \times 6^\circ$	100	-0.1305	0.8939*
		$4^\circ \times 4^\circ$	50	-0.2232	0.8459*
	55-135	$10^\circ \times 10^\circ$	40	-0.1275	0.8377*
		$8^\circ \times 8^\circ$	60	-0.1587	0.8978*
		$6^\circ \times 6^\circ$	30	-0.2061	0.9245*
		$4^\circ \times 4^\circ$	50	-0.3189	0.9096*
2015-2022	0-50	$10^\circ \times 10^\circ$	80	-0.0885	0.9082*
		$8^\circ \times 8^\circ$	40	-0.1138	0.8886*
		$6^\circ \times 6^\circ$	20	-0.1517	0.7618*
		$4^\circ \times 4^\circ$	50	-0.0863	0.6403*
	50-100	$10^\circ \times 10^\circ$	100	-0.1084	0.8717*
		$8^\circ \times 8^\circ$	90	-0.1354	0.8910*
		$6^\circ \times 6^\circ$	60	-0.1514	0.8384*
		$4^\circ \times 4^\circ$	60	-0.2212	0.8318*

779 **A2. Estimates of S_{Nd} , computed using Eq. (2), and correlation coefficients R between N_d and AOD, stratified**
 780 **by optimal buffer size (see text) for study areas varying from $4^\circ \times 4^\circ$ to $10^\circ \times 10^\circ$, during the periods 2008-2014**
 781 **and 2015-2022. Statistically significant data points are indicated with * (p value < 0.01).**

Years	Study area	Optimal scale (km)	S_{Nd}	R
2008-2014	$10^\circ \times 10^\circ$	70	0.1434	-0.8507*
	$8^\circ \times 8^\circ$	80	0.2045	-0.8646*
	$6^\circ \times 6^\circ$	30	0.2430	-0.8741*
	$4^\circ \times 4^\circ$	40	0.4496	-0.8523*
2015-2022	$10^\circ \times 10^\circ$	60	0.1742	-0.8788*
	$8^\circ \times 8^\circ$	60	0.2682	-0.8638*
	$6^\circ \times 6^\circ$	20	0.2964	-0.6900*
	$4^\circ \times 4^\circ$	30	0.2903	-0.7478*

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792 **A3. Sample sizes of cloud droplet effective radius (CER) and aerosol optical depth (AOD) across different**
 793 **buffer sizes, study areas (LWP regime 1, 2008–2014).**

Buffer sizes	Sample sizes of CER in LWP regime 1				Sample sizes of AOD in LWP regime 1			
	10°×10°	8°×8°	6°×6°	4°×4°	10°×10°	8°×8°	6°×6°	4°×4°
10	25054	16133	8551	3879	47846	32406	18711	8808
20	41667	26507	14077	6376	64878	42949	24462	11377
30	54960	34885	18421	8346	76569	50055	28274	13047
40	66170	42147	22136	9966	86291	56006	31523	14482
50	76207	48769	25738	11593	94726	61227	34272	15664
60	85194	54539	28946	12979	102262	65860	36741	16695
70	93413	59681	31788	14192	108792	69756	38749	17519
80	100728	64370	34330	15317	114357	73103	40498	18246
90	107291	68535	36552	16270	119187	76077	42016	18893
100	113457	72525	38761	17155	123607	78839	43496	19469
120	124793	79882	42945	18964	131449	83807	46217	20534
140	134597	86324	46525	20462	138140	88001	48518	21557
150	138760	89086	48145	21141	141012	89985	49570	21991
160	142693	91808	49602	21734	143725	91767	50570	22436
180	150015	96789	52326	22860	148480	94870	52385	23155
200	156655	101246	54894	23983	153084	97946	53963	23921
250	169423	109475	59449	26008	162530	104346	56987	25388
300	178015	115051	62656	27442	170117	109006	59332	26453

794 **A4. Sample sizes of cloud droplet effective radius (CER) and aerosol optical depth (AOD) across different**
 795 **buffer sizes, study areas (LWP regime 2, 2008–2014).**

Buffer sizes	Sample sizes of CER in LWP regime 2				Sample sizes of AOD in LWP regime 2			
	10°×10°	8°×8°	6°×6°	4°×4°	10°×10°	8°×8°	6°×6°	4°×4°
10	22338	14357	7737	3715	47846	32406	18711	8808
20	35406	22471	11964	5697	64878	42949	24462	11377
30	45473	28805	15162	7187	76569	50055	28274	13047
40	54074	34078	17839	8368	86291	56006	31523	14482
50	61579	38732	20290	9444	94726	61227	34272	15664
60	68173	43017	22553	10424	102262	65860	36741	16695
70	74272	46909	24584	11230	108792	69756	38749	17519
80	79999	50507	26412	11950	114357	73103	40498	18246
90	85339	53806	28177	12654	119187	76077	42016	18893
100	90440	56944	29812	13329	123607	78839	43496	19469
120	99758	62598	32673	14486	131449	83807	46217	20534
140	108723	68273	35600	15795	138140	88001	48518	21557
150	112740	70969	36864	16426	141012	89985	49570	21991
160	116558	73463	38046	16974	143725	91767	50570	22436
180	123653	78031	40203	17917	148480	94870	52385	23155
200	129864	81813	42186	18674	153084	97946	53963	23921
250	142429	89112	46079	20120	162530	104346	56987	25388
300	152460	95213	49452	21369	170117	109006	59332	26453

796 **A5. Sample sizes of cloud droplet effective radius (CER) and aerosol optical depth (AOD) across different**
 797 **buffer sizes, study areas (LWP regime 1, 2015–2022).**

Buffer sizes	Sample sizes of CER in LWP regime 1				Sample sizes of AOD in LWP regime 1			
	10°×10°	8°×8°	6°×6°	4°×4°	10°×10°	8°×8°	6°×6°	4°×4°
10	25054	16133	8551	3879	47846	32406	18711	8808
20	41667	26507	14077	6376	64878	42949	24462	11377
30	54960	34885	18421	8346	76569	50055	28274	13047
40	66170	42147	22136	9966	86291	56006	31523	14482
50	76207	48769	25738	11593	94726	61227	34272	15664
60	85194	54539	28946	12979	102262	65860	36741	16695
70	93413	59681	31788	14192	108792	69756	38749	17519
80	100728	64370	34330	15317	114357	73103	40498	18246
90	107291	68535	36552	16270	119187	76077	42016	18893
100	113457	72525	38761	17155	123607	78839	43496	19469
120	124793	79882	42945	18964	131449	83807	46217	20534
140	134597	86324	46525	20462	138140	88001	48518	21557
150	138760	89086	48145	21141	141012	89985	49570	21991
160	142693	91808	49602	21734	143725	91767	50570	22436
180	150015	96789	52326	22860	148480	94870	52385	23155
200	156655	101246	54894	23983	153084	97946	53963	23921
250	169423	109475	59449	26008	162530	104346	56987	25388

798 **A6. Sample sizes of cloud droplet effective radius (CER) and aerosol optical depth (AOD) across different**
 799 **buffer size, study areas (LWP regime 2, 2015–2022).**

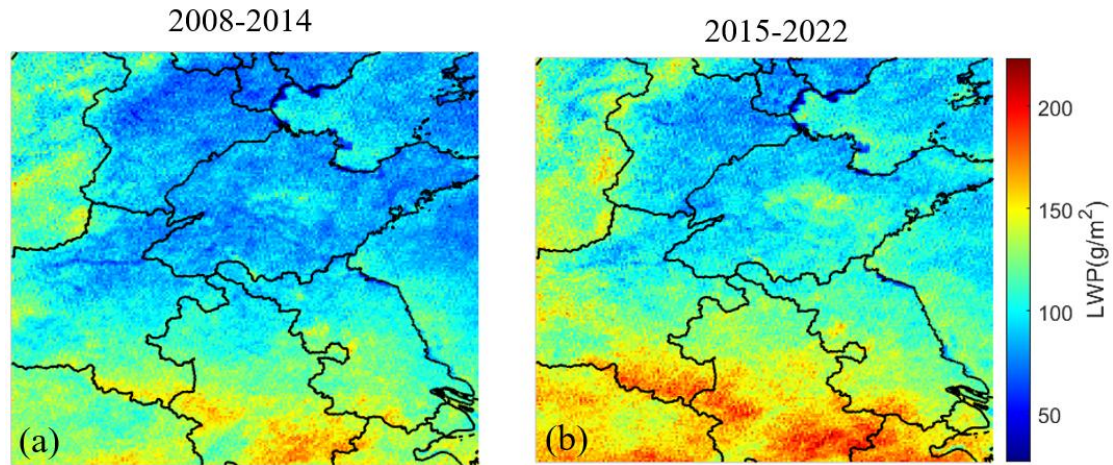
Buffer sizes	Sample sizes of CER in LWP regime 2				Sample sizes of AOD in LWP regime 2			
	10°×10°	8°×8°	6°×6°	4°×4°	10°×10°	8°×8°	6°×6°	4°×4°
10	6548	6548	5449	2803	12892	12892	12892	5961
20	24886	16258	8833	4421	46067	30536	17623	8079
30	32260	20786	11214	5490	55767	36685	20780	9408
40	38784	24739	13284	6440	63457	41493	23310	10474
50	44831	28399	15176	7183	70030	45443	25423	11419
60	50181	31609	16909	7816	75667	48900	27138	12122
70	54865	34476	18456	8415	80872	51807	28559	12723
80	58946	37042	19776	8901	85308	54361	29846	13245
90	62835	39437	21006	9359	89202	56659	31024	13706
100	66623	41757	22165	9796	92939	58931	32143	14126
120	73427	46010	24248	10605	99298	62674	34081	14927
140	79486	49854	26210	11312	104540	65994	36013	15617
150	82336	51650	26988	11633	107089	67545	36876	15939
160	85055	53440	27893	11942	109626	69104	37718	16261
180	89669	56388	29401	12520	114187	72088	39380	16889
200	93898	58986	30700	13034	117969	74508	40596	17401
250	102933	64417	33649	14040	125590	79489	43005	18271
300	109964	68913	35755	14701	132000	83404	45012	18913

801 **A7. Sample sizes of cloud droplet number concentration (N_a) and aerosol optical depth (AOD) across different**
 802 **buffer sizes and study areas during 2008-2014.**

Buffer sizes	Sample sizes of N_a				Sample sizes of AOD			
	$10^\circ \times 10^\circ$	$8^\circ \times 8^\circ$	$6^\circ \times 6^\circ$	$4^\circ \times 4^\circ$	$10^\circ \times 10^\circ$	$8^\circ \times 8^\circ$	$6^\circ \times 6^\circ$	$4^\circ \times 4^\circ$
10	47846	32406	18711	8808	50686	32611	17320	8053
20	64878	42949	24462	11377	70102	45066	23944	11084
30	76569	50055	28274	13047	84442	54334	28822	13435
40	86291	56006	31523	14482	96186	62056	32857	15309
50	94726	61227	34272	15664	106166	68620	36473	16906
60	102262	65860	36741	16695	114950	74328	39638	18292
70	108792	69756	38749	17519	122840	79508	42510	19478
80	114357	73103	40498	18246	129763	84048	45038	20599
90	119187	76077	42016	18893	135810	87931	47225	21548
100	123607	78839	43496	19469	141407	91543	49256	22436
120	131449	83807	46217	20534	151353	98040	53031	24145
140	138140	88001	48518	21557	159758	103516	56167	25517
150	141012	89985	49570	21991	163180	105820	57561	26100
160	143725	91767	50570	22436	166373	108000	58769	26661
180	148480	94870	52385	23155	172007	111773	60981	27665
200	153084	97946	53963	23921	176658	114922	62862	28696
250	162530	104346	56987	25388	185393	120627	66331	30409
300	170117	109006	59332	26453	191138	124592	68823	31481

803 **A8. Sample sizes of cloud droplet number concentration (N_a) and aerosol optical depth (AOD) across different**
 804 **buffer sizes and study areas during 2015-2022.**

Buffer sizes	Sample sizes of N_a				Sample sizes of AOD			
	$10^\circ \times 10^\circ$	$8^\circ \times 8^\circ$	$6^\circ \times 6^\circ$	$4^\circ \times 4^\circ$	$10^\circ \times 10^\circ$	$8^\circ \times 8^\circ$	$6^\circ \times 6^\circ$	$4^\circ \times 4^\circ$
10	37182	23994	13048	6149	31621	21503	12892	5961
20	51313	32990	17755	8252	46067	30536	17623	8079
30	61921	39800	21327	9788	55767	36685	20780	9408
40	70663	45351	24213	11063	63457	41493	23310	10474
50	78059	50080	26808	12118	70030	45443	25423	11419
60	84354	53975	28967	13015	75667	48900	27138	12122
70	89675	57448	30881	13832	80872	51807	28559	12723
80	94620	60671	32636	14556	85308	54361	29846	13245
90	99042	63497	34138	15155	89202	56659	31024	13706
100	102936	65993	35444	15703	92939	58931	32143	14126
120	110590	70884	38002	16787	99298	62674	34081	14927
140	116487	74472	40007	17549	104540	65994	36013	15617
150	119140	76230	41049	17958	107089	67545	36876	15939
160	121506	77846	41958	18265	109626	69104	37718	16261
180	125788	80845	43582	18935	114187	72088	39380	16889
200	129613	83370	44887	19538	117969	74508	40596	17401
250	136752	87967	47756	20784	125590	79489	43005	18271
300	141896	91273	49807	21431	132000	83404	45012	18913



805
 806 **B1. Spatial distributions of LWP averaged over the years 2008-2014 (a) and 2015-2022 (b) over the study area.**
 807 **The lines are provincial borders and the names of provinces mentioned in the text are indicated in Fig. 3(f).**

808 **Data availability**

809 All data used in this study are publicly available. The satellite data from the MODIS instrument used in
 810 this study were obtained from <https://ladsweb.nascom.nasa.gov/search/> (last access: 02 July 2025). The
 811 satellite data from CALIOP were obtained from <https://subset.larc.nasa.gov/calipso/login.php> (last
 812 access: 02 July 2025).

813 **Author contributions**

814 YL, LT and GL designed the research. YL and LT led the analyses. YL and GL wrote the manuscript
 815 with major input from JH, and further input from all other authors. All authors contributed to interpreting
 816 the results and to the finalization and revision of the manuscript.

817 **Competing interests**

818 The authors declare that they have no conflict of interest.

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