



Identification of Cloud Droplets, Ice Crystals, and Aerosols Using Ground-based Fog and Aerosol Spectrometer: Chamber Experiments and Field Observations

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Abstract. Simultaneously identifying micron-scale aerosols, cloud droplets, and ice crystals within mixed-phase clouds is essential for advancing cloud microphysical research, yet it presents substantial technical challenges. This study proposes a
15 novel phase identification method based on the Ground-based Fog and Aerosol Spectrometer (GFAS; DMT Inc.), which is rigorously validated through cloud chamber experiments and in-situ observations. Cloud chamber experiments conducted at -14°C , -10°C , and -6°C yielded distributions of the polarization ratio ($\delta = (S-P)/(S+P)$) and total scattering counts ($\text{TotalCounts} = S+P$) for highly non-spherical ice crystals. Based on these measurements, quantitative thresholds were established to classify three distinct particle regimes: ice crystals ($\delta > -0.33$, $\text{TotalCounts} < 1.68 \times 10^2$), water droplets
20 ($\delta < -0.82$, high TotalCounts), and a transition zone ($-0.82 \leq \delta \leq -0.33$) corresponding to weakly non-spherical ice or aerosols. Applied to ground-based observations at Mt. Liupan in Northwest China from January to April 2024, the threshold method was applied to a complete in-cloud glaciation cycle. The GFAS effectively distinguishes highly non-spherical ice crystals from pure water droplets, while the TotalCounts threshold achieves a separation efficiency exceeding 98 % in distinguishing pure ice crystals from aerosols. Although aerosol δ values primarily fall within the transition zone (-0.82 and -0.33) and overlap
25 with weakly non-spherical ice crystals, preliminary discrimination remains achievable using a joint constraint of δ and TotalCounts . This method provides a temperature-independent diagnostic tool for quantifying the glaciation degree in mixed-phase clouds, offering critical observational support for understanding cloud microphysical processes.

1 Introduction

Aerosols and hydrometeor particles constitute fundamental components of the atmospheric system, exerting profound
30 impacts on global climate through modulating radiative transfer, precipitation development, and energy budgets (Twomey, 1974; Ramanathan et al., 2001; Bellouin et al., 2020; Wang et al., 2024). Clouds are macroscopic ensembles of hydrometeor



particles, whose microphysical properties are strongly modulated by aerosols serving as cloud-condensation and ice-nucleating particles (see overview in Korolev et al., 2017). Among various cloud regimes, mixed-phase clouds, which globally cover vast atmospheric regions, play a pivotal role in the climate system (Wylie and Menzel, 1999). Within mixed-phase clouds, ice crystals exhibiting diverse morphological habits coexist with supercooled liquid droplets. Their complex microphysical interactions, phase partitioning, and phase-transformation behaviours govern cloud optical properties and precipitation formation, thereby presenting major challenges for robustly quantifying global radiation budgets and hydrological cycles (Wendisch et al., 2005; Pinto, 1998; Korolev, 2007). Despite their pervasive global distribution, state-of-the-art climate models tend to underestimate the fraction of supercooled liquid water in mixed-phase clouds. Because liquid-dominated clouds reflect substantially more solar radiation than fully glaciated clouds, this biased phase partitioning introduces marked uncertainties into climate projections, suggesting that the rate and magnitude of anthropogenic global warming require critical re-evaluation (Tan et al., 2016). Furthermore, cloud phase is extraordinarily sensitive to atmospheric dynamics, particularly in-cloud vertical velocity (Korolev and Isaac, 2003), highlighting the urgent requirement for high-resolution observational constraints on phase transition processes.

Liquid water content (LWC) and ice water content (IWC) serve as core microphysical variables governing the cloud phase of mixed-phase clouds (Korolev et al., 2003; Zhao et al., 2023). However, substantial biases and uncertainties persist in their numerical representation within current atmospheric models (Storelvmo et al., 2008; Morrison et al., 2012; Forbes and Ahlgrim, 2014). Resolving these model discrepancies requires accurate discrimination of particle categories and thermodynamic phases through direct in-situ observations (Field et al., 2004; Shupe et al., 2006). Although coexisting liquid droplets, ice hydrometeors, and background aerosols exhibit distinct physical and optical signatures, classifying these species over narrow micron-scale size intervals remains a primary challenge. This limitation hinders the mechanistic interpretation of key microphysical evolution, including ice nucleation and secondary ice production, which are critical for predicting precipitation initiation and cloud-radiative feedbacks (Lohmann, 2002; Jackson et al., 2012; Field et al., 2017; Wang et al., 2026). Conventional in-situ probes struggle to achieve refined measurements of single-particle optical properties, particularly for fine particles with diameters below 50 μm (McFarquhar et al., 2007). While optical imaging instruments such as the two-dimensional stereo (2D-S) probe provide valuable morphological details for large hydrometeors, optical diffraction limits severely constrain their capability to unambiguously classify smaller, highly irregular particles or distinguish heterogeneous aerosols from small ice crystals (Lawson et al., 2006). Consequently, simultaneously identifying micron-scale aerosols, cloud droplets, and ice crystals stands as both a critical technical bottleneck and an indispensable prerequisite for advancing cloud microphysical research.

In-situ measurement represents the primary and reliable approach for quantifying mixed-phase cloud microphysics. In particular, ground-based cloud and fog observations provide continuous, high-temporal-resolution time-series at fixed sites, facilitating in-depth mechanistic investigations into cloud microphysical evolution (Korolev, 2017; Wang et al., 2023, 2025). Over past decades, a comprehensive suite of in-situ instrumental systems has been developed to observe mixed-phase clouds. These encompass icing detectors (e.g., Rosemount Icing Detector; Baumgardner and Rodi, 1989), thermal bulk sensors



including hot-wire probes (e.g., Nevzorov probe; Korolev et al., 1998) and evaporative sensors (e.g., TWC evaporator; Nicholls et al., 1990), conventional single-channel optical scattering probes (e.g., Forward Scattering Spectrometer Probe, FSSP; Knollenberg, 1981; Cloud Droplet Probe, CDP; Lance et al., 2010; Cloud Aerosol Spectrometer, CAS; Baumgardner et al., 2003), polarization-enhanced scattering probes (e.g., CAS with Polarization, CAS-POL; Baumgardner et al., 2014; Back-
70 scatter Cloud Probe with Polarization Detection, BCPD; Freer et al., 2014), and particle imaging probes (e.g., 2D-C optical array probe; Knollenberg, 1970; Cloud Imaging Probe, CIP; Baumgardner et al., 2001; 2D-S; Lawson et al., 2006). Despite these technological advancements, each instrument category exhibits inherent limitations in robust phase discrimination: icing detectors offer only qualitative indications of supercooled liquid water presence (Mazin et al., 2001); thermal bulk sensors retrieve bulk LWC and IWC without single-particle attributes; imaging probes capture valuable morphological features for
75 large hydrometeors but lack particle flux constraints and suffer from optical diffraction limits for fine particles below 50 μm ; and conventional single-channel scattering probes cannot distinguish scattering signals originating from ice crystals and water droplets (Korolev et al., 2017). Furthermore, while polarization-enhanced scattering probes significantly improve phase classification by detecting particle depolarization, unambiguously separating small, weakly non-spherical ice crystals from background aerosols remains challenging due to overlapping polarization signatures.

80 Theoretically, polarization detection discriminates particle phases by exploiting morphologically induced scattering differences. Surface tension maintains liquid droplets in an approximately spherical geometry, preserving the linear polarization of incident radiation, whereas non-spherical ice crystals alter the polarization state and generate cross-polarized backscatter (Clauss et al., 2013; Baumgardner et al., 2014). However, as noted above, small ice crystals and background aerosols often exhibit overlapping single-particle polarization ratios, necessitating advanced observational frameworks. The
85 Ground-based Fog and Aerosol Spectrometer (GFAS) used in this study is a polarization-enabled instrument developed from the third-generation Cloud Aerosol Spectrometer with Polarization-Depolarization (CAS-DPOL). Utilizing a 660 nm linearly polarized laser, the GFAS illuminates particle streams and simultaneously records forward-scattering light over 3.5° – 12° to retrieve optical-equivalent diameters across 0.5 to 40 μm . Concurrently, two photodetectors capture the perpendicular (S) and parallel (P) polarized components of backscattered light over 183.5° – 192° , from which the single-particle polarization ratio (δ
90 $= (S-P)/(S+P)$) is derived to discriminate particle thermodynamic phase states. By coupling single-particle optical sizing with polarization-based phase classification, the GFAS facilitates the refined characterization of size distributions individually for aerosols, droplets, and ice crystals.

Despite the potential of polarization detection, robust phase discrimination in complex cloud environments still faces notable operational challenges. First, conventional classification criteria often rely on single-parameter depolarization
95 thresholds or simplified temperature-dependent assumptions, which fail under supercooled mixed-phase conditions where liquid droplets and ice crystals dynamically coexist (Baumgardner et al., 2014; Glen and Brooks, 2013). Second, heterogeneous aerosols and fine ice crystals often exhibit overlapping δ signatures, making unambiguous aerosol–ice separation challenging. Crucially, the reliability of current polarization threshold criteria would benefit from further cross-validation that bridges controlled laboratory experiments and multi-environment field observations. Consequently, the comprehensive multiphase



100 identification capability of the GFAS has not been systematically established. To address these gaps, this study develops a refined multiphase particle classification framework for GFAS measurements. By integrating temperature-controlled cloud chamber experiments with in-situ field observations at Mt. Liupan, we establish empirical dual-parameter thresholds based on the polarization ratio (δ) and the total backscattering signal (TotalCounts) to distinguish aerosols, supercooled cloud droplets, and ice crystals. This work provides a transferable observational methodology for accurate single-particle phase identification, offering reliable technical support for future cloud microphysical field campaigns.

2 Data and Methods

2.1 Cloud Chamber Experiments

Controlled cold cloud chamber experiments were conducted on 28–29 January 2026 at the cloud chamber facility of Chengdu University of Information Technology to acquire calibrated particle microphysical datasets under controlled sub-zero conditions using the GFAS. The cloud chamber generates artificial cloud and fog environments via integrated temperature control using cooling condensation tubes, pressure reduction using vacuum pumps, and aerosol/catalyst injection systems. In this experiment, cloud formation was induced through synchronized humidification and cooling, with continuous observations performed at three target temperatures: -6°C , -10°C , and -14°C .

To trigger heterogeneous ice nucleation above the homogeneous freezing threshold, Snomax, a freeze-dried inactivated bacterial protein exhibiting high ice-nucleation activity, was injected into the chamber as an ice nucleating particle surrogate (Kanji et al., 2017). Under the specified thermal and microphysical regimes, Snomax seeding induced complete cloud glaciation at -10°C . In contrast, glaciation remained incomplete at -6°C , yielding a quasi-steady mixed-phase environment composed of coexisting supercooled liquid droplets and ice crystals. Consequently, these three temperature scenarios provide well-constrained reference datasets representing both mixed-phase and fully glaciated cloud regimes.

The particle morphology under these experimental conditions was further examined using a holographic fog droplet spectrometer; ice crystals observed at -6°C exhibited highly non-spherical, irregular structures, while coexisting liquid droplets remained near-spherical (Figure S1 in the Supplementary). The resulting chamber-derived polarization signatures were subsequently used to establish empirical thresholds for the polarization ratio and TotalCounts, providing the foundation for phase classification in subsequent field observations at Mt. Liupan.

2.2 Mountain in-situ Cloud Field Campaign

A 70-day continuous cloud field campaign was conducted at the Liupan Mountain observational station in northwestern China ($35^{\circ}39'\text{N}$, $106^{\circ}12'\text{E}$; 2845.2 m a.s.l.) from 23 January to 2 April 2024. Geographically situated in the transitional zone between semi-arid and humid climatic regions, the station serves as a representative platform for orographic cloud observations. The local climate is characterized by frequent fog events, averaging 153 fog days annually. Driven by Asian monsoon influences, the region maintains abundant atmospheric moisture, with spring and winter minimum



temperatures reaching as low as -15°C . These meteorological conditions favor the frequent formation of supercooled fog and orographically forced mixed-phase clouds.

During the observational period, 10 valid cloud and fog events were recorded, each defined by horizontal visibility below 1 km and a duration exceeding 1 h. This study focuses on the microphysical characteristics of aerosols, liquid water droplets, and ice crystals across these observed events. Concurrent in-situ meteorological parameters, including ambient air temperature, wind speed, and wind direction, were collected at the Liupan Mountain Meteorological Station to contextualize the microphysical observations. All times in this study are reported in Beijing Time (BJT; UTC+8). Photographs of the cloud chamber setup and of the GFAS deployment at the Mt. Liupan station are provided in Figure S2 in the Supplementary.

2.3 GFAS Instrument Specifications

The GFAS is a multi-functional particle spectrometer designed to retrieve the optical-equivalent diameter of atmospheric aerosols, liquid water droplets, and ice crystals, covering a size range of $0.5\text{--}40\text{ }\mu\text{m}$. Integrating the technical features of the Fog Monitor (FM-120) and the CAS-DPOL, the instrument employs a 660 nm polarized laser for single-particle detection. As shown in Figure 1, particle sizing is based on Mie scattering theory under the fundamental assumption of spherical particle geometry. As individual particles traverse the laser detection volume, scattered light is collected by three separate photodetectors: a forward scattering detector covering $3.5^{\circ}\text{--}12^{\circ}$ to retrieve the optical-equivalent diameter, and two sideways/backscattering detectors positioned at a nominal scattering angle of 90° to capture the perpendicular (S) and parallel (P) polarized components over $183.5^{\circ}\text{--}192^{\circ}$. The GFAS derives a single-particle polarization ratio (δ) defined as $\delta=(S-P)/(S+P)$. This formulation is consistent with the scheme adopted by the CPSPD (Baumgardner et al., 2014), but differs from the conventional CAS-DPOL definition of $S/(S+P)$ (Glen and Brooks, 2013).

Particle-laser interactions modulate the polarization state of incident radiation, with the degree of depolarization governed primarily by particle microphysics (size, morphology, and refractive index) and scattering geometry. The polarization ratio quantifies the relative difference between the two orthogonal backscattering components. Under the GFAS-specific formulation, non-spherical and irregular particles enhance perpendicular scattering, leading to elevated δ values that reflect a higher degree of shape irregularity. Conversely, ideal spherical particles (e.g., liquid water droplets) produce minimal perpendicular scattering, yielding δ values approaching -1 .

In addition, this study defines a supplementary parameter, TotalCounts, as the sum of the peak intensities of the two polarized backscattered components ($S+P$). Because both signals originate from backscattered radiation captured by the GFAS, TotalCounts serves as a practical proxy for the total backscattering intensity of an individual particle, providing complementary optical constraints to refine particle morphological and phase discrimination.

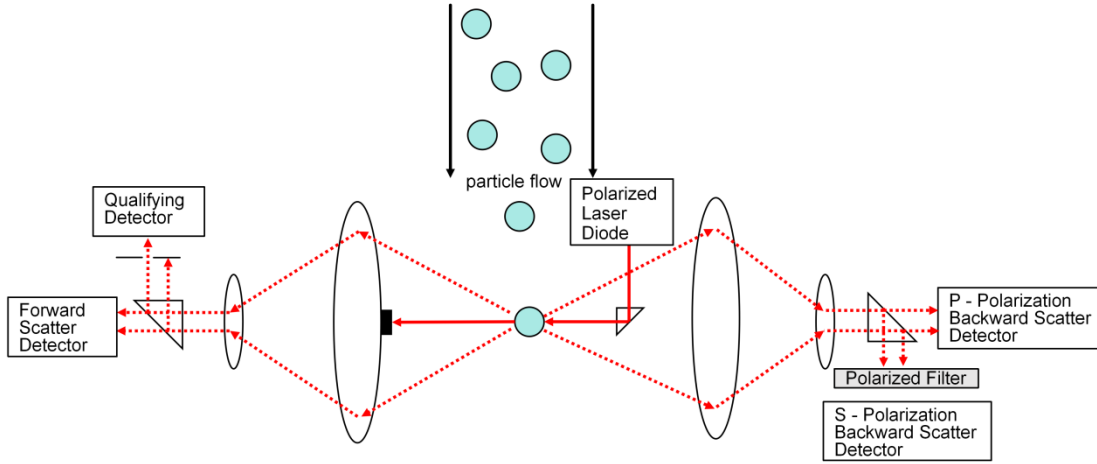


Figure 1. Schematic illustration of the GFAS instrument.

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2.4 Glaciation Index Calculation

In this study, phase classification of field-observed mixed-phase cloud particles is implemented based on empirical thresholds of polarization ratio and TotalCounts, calibrated against reference datasets of pure ice crystals and mixed-phase clouds obtained from cloud chamber experiments. Following particle classification, the particle equivalent optical diameters retrieved from forward-scattering measurements are used to calculate the liquid water content (LWC) and the ice water content (IWC), enabling the calculation of the cloud glaciation index.

In established parameterisation schemes, the mass of an individual ice crystal is commonly estimated using a power-law diameter–mass relationship:

$$IWC = \alpha D^\beta \quad (1)$$

where D is the optical-equivalent diameter, and the empirical parameters α and β are defined as temperature-dependent functions of the temperature T (in Kelvin; Fontaine et al., 2014):

$$\begin{aligned} \alpha &= 0.0020 \cdot e^{0.0037 \cdot T} + 5.1 \times 10^{-17} \cdot e^{0.1213 \cdot T} \\ \beta &= 7.13 \cdot e^{-0.0053 \cdot T} + 5.4 \times 10^{-7} \cdot e^{0.0435 \cdot T} \end{aligned} \quad (2)$$

Particles classified as liquid water droplets based on low δ values are identified as spherical liquid droplets for LWC calculation, whereas IWC is computed by integrating the mass over the classified ice crystals size distribution. Finally, the glaciation index (μ) is determined as follows:



$$\mu = \text{IWC} / (\text{IWC} + \text{LWC}) \quad (3)$$

Although IWC retrievals for small ice crystals ($< 50 \mu\text{m}$) inherently carry quantitative uncertainties due to optical resolution limits and morphological variability, the temporal evolution of μ derived from GFAS provides a physically robust metric for characterizing ice–water phase transitions in mixed-phase clouds.

3 Results and Discussions

3.1 Threshold Determination from Cloud Chamber Experiments

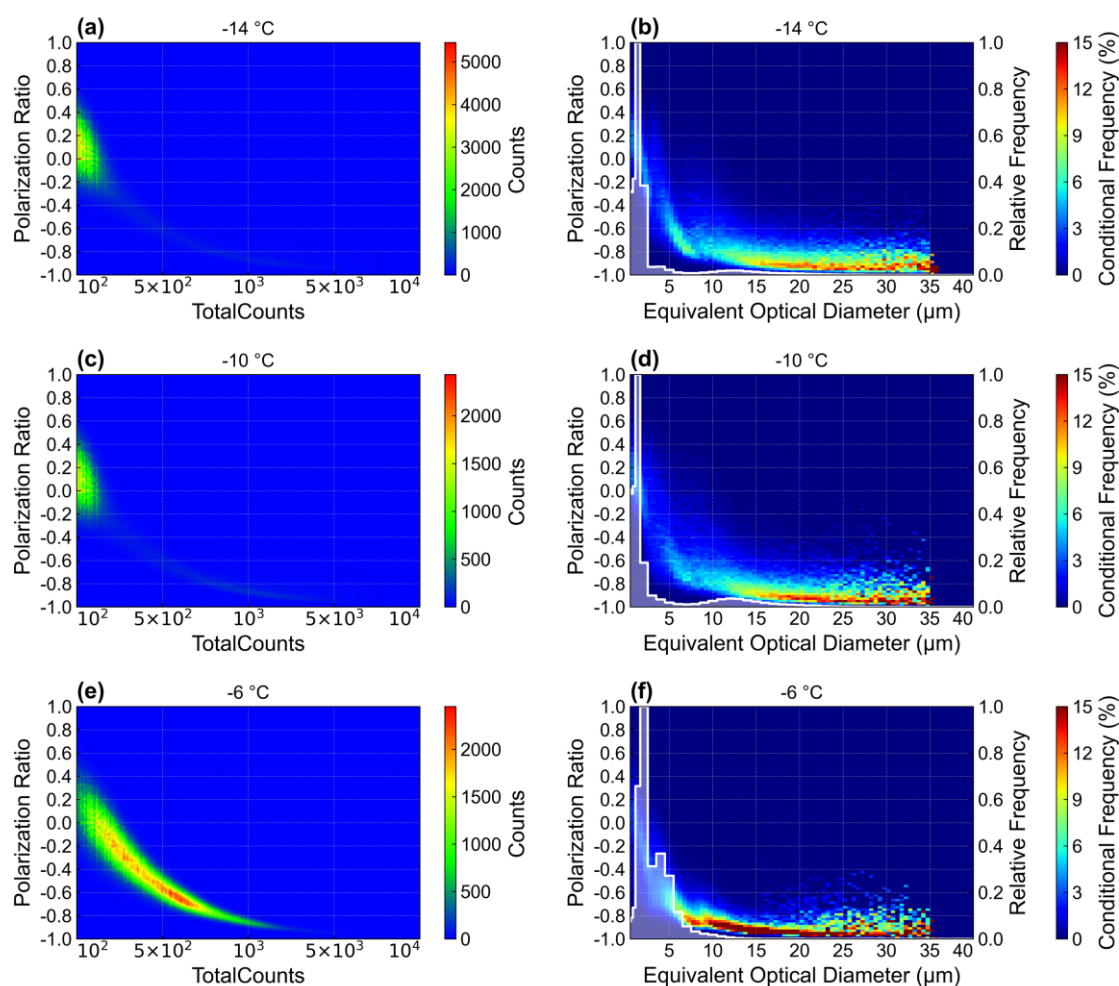


Figure 2. Particle polarization characteristics and size-resolved polarization ratio distributions measured by the GFAS during controlled cloud chamber experiments at different temperatures. Panels (a), (c), and (e) illustrate the joint distributions of particle polarization ratio and TotalCounts at $-14 \text{ }^{\circ}\text{C}$, $-10 \text{ }^{\circ}\text{C}$, and $-6 \text{ }^{\circ}\text{C}$, respectively, while panels (b), (d), and (f) present the corresponding size-dependent polarization ratio distributions and normalised particle relative frequencies. All results are derived from 30 min of continuous



observations at each target temperature.

Figure 2 presents the particle polarization characteristics from the cloud chamber experiments. Figure 2a shows the polarization signatures of ice crystals generated via Snomax seeding at -14°C . Driven by the strong ice nucleation activity of Snomax and high supersaturation within the chamber environment, abundant ice crystals rapidly form at this temperature. The corresponding pure ice crystal dataset exhibits a distinct and concentrated δ distribution in the high-value range. The δ of ice crystals predominantly ranges from -0.4 to 0.4 , with negligible data below -0.82 ; particles with $\delta > -0.33$ account for 90.34 % of the total population. This distribution demonstrates that highly glaciated or fully frozen ice crystals possess highly irregular, non-spherical morphologies, with polarization ratios substantially higher than the near -1 characteristic of ideal spherical particles. Accordingly, polarized light scattered by these irregular ice crystals yields a considerably stronger perpendicular scattering component relative to the parallel component.

Furthermore, the ice crystals measured at -14°C show consistently low TotalCounts values with nearly all values below 2×10^2 , as shown in Figure 2a. Although TotalCounts is an instrument-specific scattering intensity metric, it closely correlates with the backscattering properties of the particle under polarized illumination. The low TotalCounts signature of ice crystals indicates markedly weaker backscattering compared with liquid droplets and aerosols, whose polarization features are presented in subsequent analyses. A threshold of 1.68×10^2 for TotalCounts captures 90.46 % of the ice crystals in Figure 2a. This distinct behaviour can be physically interpreted by morphological differences between hydrometeors. Spherical liquid droplets generate prominent backscattering peaks via diffraction and internal multiple reflection effects. In contrast, ice crystals have extremely complex non-spherical structures, including hexagonal plates, columns, dendrites, aggregates, and even hollow structures (Schmitt et al., 2016). Such irregularity breaks the spherical symmetry, causing the scattered energy to be dispersed over a wider angular range rather than concentrated in the backscattering direction. Additionally, the porous and cavity-containing internal structure of ice crystals induces extensive multiple scattering and diffuse reflection, dissipating photon energy or redirecting it away from the backscattering angle and further weakening detector-received signals (Schmitt et al., 2016). These microphysical properties collectively account for the unique polarization signatures of ice crystals.

Size-resolved polarization characteristics at -14°C are displayed in Figure 2b. The polarization ratio tends to decrease with increasing particle size, indicating that the ice crystals generated in the cloud chamber are relatively small, with a size peak below $5\text{ }\mu\text{m}$. Larger particles ($>5\text{ }\mu\text{m}$) constitute only a minor fraction and tend to have much lower polarization ratios, mostly ranging from -0.6 to -1 . These large particles likely correspond to residual unfrozen droplets or incompletely glaciated ice crystals.

Figure 2c shows the particle polarization characteristics obtained from cloud chamber experiments at -10°C . Note that the ice-nucleating activity of Snomax increases steeply with decreasing temperature between -5 and -10°C , with its ice-active site density rising by several orders of magnitude across this interval (Wex et al., 2015; Kanji et al., 2017). Thus, at -10°C the majority of Snomax active sites were activated and complete glaciation was achieved, whereas at -6°C only a small fraction of the active sites was effective, which accounts for the quasi-steady mixed-phase state observed at this



temperature. This temperature produces highly glaciated and completely frozen ice crystals with strong non-sphericity, yielding high polarization ratios analogous to those observed at -14°C . The ice crystal population at -10°C is characterised by high δ values and low TotalCounts, with 88.25 % of particles satisfying $\delta > -0.33$. Adopting the TotalCounts threshold of 1.68×10^2 enables the selection of 88.37 % of the ice crystal particles in this dataset. The size-dependent polarization trend at -10°C (Figure 2d) is broadly consistent with the -14°C results. However, low δ values are more concentrated for particles larger than $10\text{ }\mu\text{m}$, accompanied by a reduced fraction of particles with $\delta > -0.6$. This discrepancy implies that large particles experience less complete glaciation at -10°C than at -14°C , resulting in lower and more tightly constrained δ .

The particle polarization characteristics observed at -6°C (Figure 2e) differ substantially from the fully glaciated conditions at -10°C and -14°C . The -6°C experiment produces abundant particles with low morphological irregularity, together with residual unfrozen Snomax aerosol particles. In contrast to typical fully frozen ice crystals ($\delta > -0.33$, TotalCounts $< 1.68 \times 10^2$), many particles fall within the parameter range of $\delta < -0.5$ and TotalCounts $> 3 \times 10^2$. Since ambient temperatures near -6°C frequently occur in natural mixed-phase cloud environments, these observations provide representative polarization characteristics for mixed-phase cloud conditions. Notably, a distinct particle population with $\delta < -0.82$ and extremely high TotalCounts ($> 10^3$) is identified at -6°C , consistent with the canonical optical signatures of spherical liquid droplets. Spherical droplets barely alter the polarization state of incident radiation and produce negligible perpendicular scattering, resulting in polarization ratios approaching -1 . The stronger backscattering ability of liquid droplets relative to ice crystals is consistent with established findings in cloud microphysics and radar meteorology (Battaglia et al., 2020). At the relatively warm temperature of -6°C , the transition between liquid and ice phases presents continuous polarization variations rather than discrete states. In addition to typical ice crystal and liquid droplet populations, abundant particles occupy an intermediate transition zone, demonstrating continuous evolution of polarization ratio and TotalCounts during ice–water phase transformation. Incompletely frozen droplets retain high sphericity under insufficient freezing durations, whereas prolonged freezing and kinetic anisotropic growth gradually enhance ice crystal irregularity. Partially frozen ice crystals with low non-sphericity therefore exhibit transitional polarization features between pure ice and pure liquid states.

Size-resolved polarization distributions at -6°C (Figure 2f) reveal a primary high-polarization peak for particles smaller than $5\text{ }\mu\text{m}$, consistent with the fully non-spherical ice crystal populations observed at -10°C and -14°C . Unlike the colder temperature conditions, a distinct secondary particle peak emerges in the $3\text{--}10\text{ }\mu\text{m}$ size range, characterised by lower polarization ratios and higher particle concentrations. This secondary peak corresponds to unfrozen supercooled droplets and newly formed, low-irregularity ice crystals generated under incomplete freezing conditions.

Quantitatively, a polarization ratio of $\delta = -0.82$ corresponds to a theoretical S/P ratio of approximately 0.1, whereas $\delta = -0.33$ yields an S/P ratio of 0.50. Although ideal spherical droplets theoretically exhibit extremely low polarization ratios ($\delta = -1$), minor deviations from perfect sphericity induced by surface tension oscillations and aerodynamic flow-field perturbations result in slightly elevated, yet consistently low δ values for natural liquid droplets. Conversely, the high morphological variability of ice crystals and their random orientations relative to the incident laser beam naturally generate a



250 broad range of δ values for non-spherical hydrometeors. Based on the cloud chamber experimental datasets, two single-parameter polarization thresholds are established for phase discrimination: particles with $\delta > -0.33$ are classified as ice crystals, while particles with $\delta < -0.82$ are categorized as liquid droplets. The upper threshold ($\delta > -0.33$) targets highly non-spherical ice hydrometeors, enabling effective separation from aerosols, albeit with the potential exclusion of weakly irregular or early-stage ice particles. The lower threshold ($\delta < -0.82$) ensures strict screening for spherical liquid hydrometeors. Particles falling

255 within the transitional range ($-0.82 < \delta < -0.33$) are interpreted as incompletely frozen hydrometeors, including partially frozen liquid-ice aggregates and early-stage ice crystals prior to substantial riming or secondary ice multiplication. In addition to the polarization ratio, the total scattering counts (TotalCounts = $S+P$) provide an auxiliary constraint. Chamber statistics show that 90.46 % of the highly non-spherical ice crystals exhibit TotalCounts below 1.68×10^2 , whereas liquid droplets yield markedly higher counts owing to their strong backward scattering. Accordingly, the complete classification criteria are: (i) ice

260 crystals, $\delta > -0.33$ and TotalCounts $< 1.68 \times 10^2$; (ii) liquid droplets, $\delta < -0.82$ (with high TotalCounts); (iii) transition zone, $-0.82 \leq \delta \leq -0.33$, within which TotalCounts assists in separating weakly irregular ice crystals from aerosols.

3.2 Field Application of Thresholds in Mixed-phase Clouds

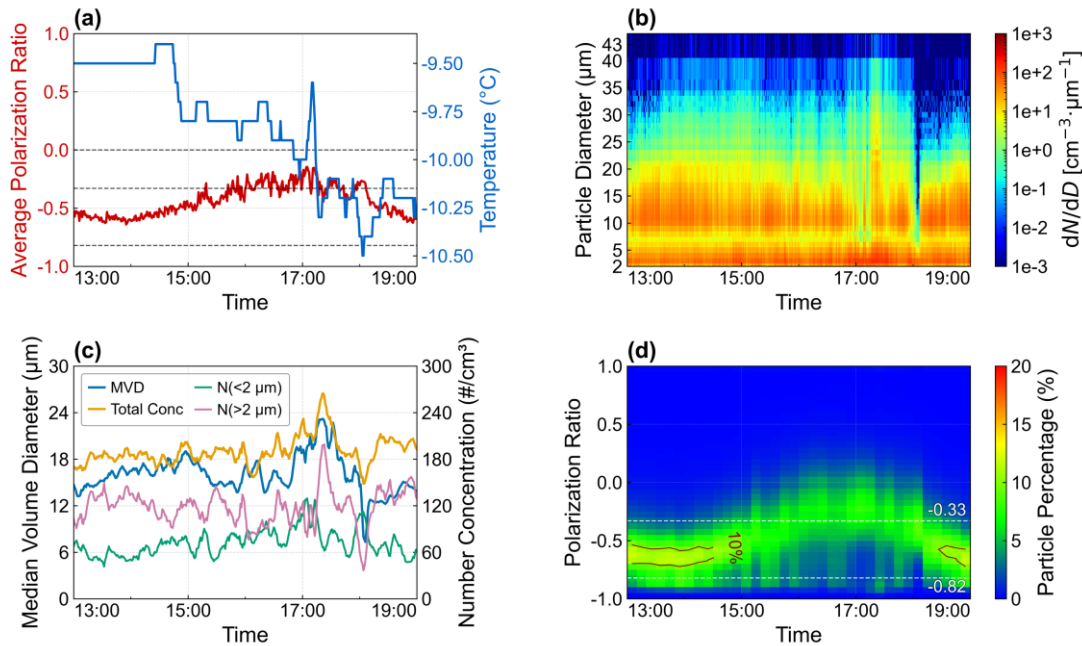


Figure 3. Temporal evolution of cloud microphysical properties during a mixed-phase event on 26 February 2024 at Mt. Liupan. (a) Average particle polarization ratio (δ) and ambient temperature. (b) Particle number size distribution (dN/dD) for diameters 2–50 μm . (c) Mean volume diameter (MVD) and number concentration of total particles (N_{total}), particles larger than 2 μm , and particles smaller than 2 μm . (d) Probability density function of single-particle δ . All times are in Beijing Time (UTC+8).



The thresholds derived from cloud chamber experiments were further applied to in-situ mixed-phase cloud observations at Mt. Liupan. Figure 3 displays the temporal evolution of key microphysical and optical variables during a representative mixed-phase cloud event on 26 February 2024. This event exhibited large-amplitude variations in the average δ , serving as a clear signature of active ice–water phase transformation. As shown in Figure 3a, the average δ increased by more than 0.4, from approximately -0.7 to -0.3 , between 13:00 and 16:00 BJT, and subsequently decreased, forming a single prominent peak that spans a full glaciation cycle. This sustained elevation in δ was accompanied by a subtle ambient cooling of $\sim 1^\circ\text{C}$, suggesting that cooling likely triggered droplet freezing. The temporal evolution of the cloud microphysics is shown in Figure 3b, which presents the cloud droplet number size distribution across the 2 to 50 μm range. Throughout the event, the spectrum maintained a persistent bimodal structure, with a fine mode centred at 2–5 μm and a coarse mode at 10–15 μm . Notably, the modal diameters remained virtually stationary; instead, spectral evolution was characterized by a progressive broadening driven by a marked concentration increase in particles larger than 10 μm . This spectral broadening directly reflects the generation and growth of larger hydrometeors.

Quantitative changes in bulk microphysical parameters are summarized in Figure 3c. The total particle number concentration increased from approximately 180 cm^{-3} during the initial stage to about 240 cm^{-3} near the peak of glaciation. This increase was predominantly contributed by cloud particles with diameter $>2\text{ }\mu\text{m}$, while the concentration of interstitial particles (diameter $<2\text{ }\mu\text{m}$) remained relatively stable. Concurrently, the mean volume diameter (MVD) increased from 12 μm to 24 μm , confirming that the newly generated ice crystals contributed to larger particle sizes. The synchronous elevation of the cloud particle concentration and MVD indicates that activation of new cloud particles and their subsequent growth occurred in tandem with the glaciation process. The most direct evidence of phase transformation is provided by Figure 3d, which documents the temporal evolution of the probability density function of single-particle δ . During the initial stage, δ values were concentrated between -1.0 and -0.5 , signalling a population dominated by near-spherical liquid droplets. As glaciation intensified, the distribution shifted markedly toward higher δ values, with a substantial proportion of particles appeared in the high- δ regime ($\delta > -0.33$), reflecting the rapid accumulation of irregular ice crystals. After 17:00 BJT, the distribution gradually returned to lower δ values, marking the dissipation of the glaciated cloud. Collectively, these synchronized variations, including the rise and fall of average δ , spectral broadening without modal shifts, elevated cloud particle concentrations alongside doubling MVD, and the temporal shift of the δ PDF, provide consistent, multi-angle evidence that a complete glaciation episode was successfully resolved by the GFAS.

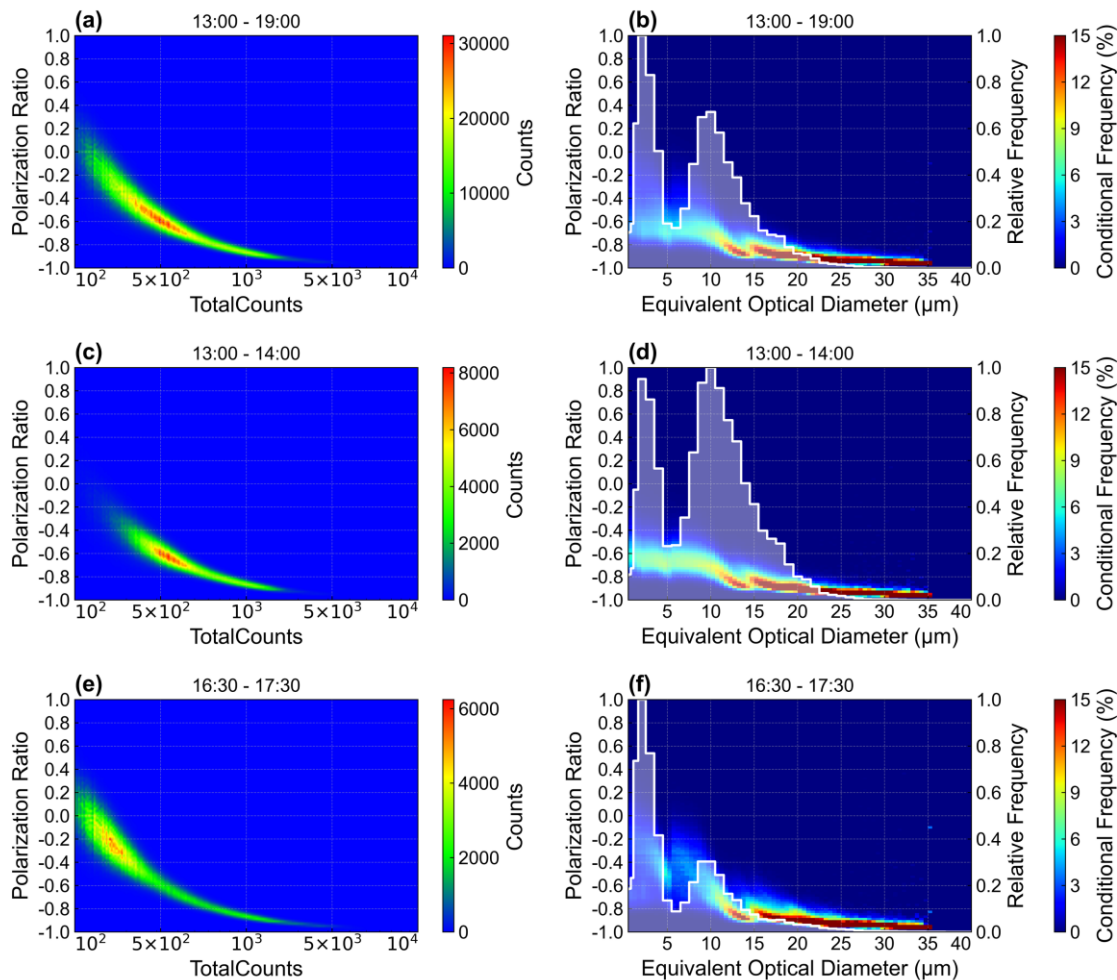


Figure 4. Particle polarization characteristics during different evolutionary stages of the observed mixed-phase cloud event on 26 February 2024. (a, c, e) show the distributions of polarization ratio and TotalCounts for three different time intervals, while (b, d, f) present the corresponding size-resolved polarization ratio distributions and normalized relative frequencies of particles. (a,b) show statistics for the full cloud–fog event, while (c,d) and (e,f) represent the one-hour interval before the rise and around the peak of the average polarization ratio, respectively.

Figure 4 illustrates the particle polarization properties at different evolutionary stages of the mixed-phase cloud event on 26 February 2024. As shown in Figure 4a, particles spanning ice, liquid, and transitional phases coexist throughout the entire cloud event. These polarization features are highly consistent with the mixed-phase characteristics observed under the
295 -6°C cloud chamber experiment (Figure 2e), confirming that the sampled cloud event corresponded to a typical mixed-phase cloud process. Figure 4b indicates that the particle size distribution during the event presents a dominant bimodal structure, with a primary peak at $2\text{--}5\text{ }\mu\text{m}$ and a secondary peak at $10\text{--}15\text{ }\mu\text{m}$. Particles with high polarization ratios are mostly concentrated below $10\text{ }\mu\text{m}$, while low-polarization-ratio particles primarily occupy the size range above $10\text{ }\mu\text{m}$, which are



dominated by large liquid droplets actively involved in ice–water phase transition. In the initial one-hour period of the fog
event (Figure 4c), particles generally exhibit low polarization ratios. A large number of particles fall within the typical liquid
droplet regime with $\delta < -0.82$ and $\text{TotalCounts} > 5 \times 10^2$, whereas few particles appear in the typical ice crystal regime with
 $\delta > -0.33$ and $\text{TotalCounts} < 1.68 \times 10^2$. Most particles are concentrated in the transitional zone between pure liquid and pure
ice states. The corresponding size-resolved polarization distribution (Figure 4d) shows that low-polarization droplets peak at
10–15 μm , while high-polarization particles are mainly confined to sizes smaller than 5 μm . These results indicate that well-
developed, highly irregular ice crystals had not yet formed in the initial stage. The cloud was dominated by large supercooled
droplets, while small particles with intermediate polarization ratios likely represented low-irregularity partially frozen ice
crystals, suggesting the initiation of subtle ice–water transformation at the early stage.

As the temperature decreases, the glaciation of the cloud intensified continuously. Figure 4e shows the particle
polarization signatures during the period of maximum average δ , representing the most vigorous stage of phase transition and
the highest glaciation degree throughout the event. Comparison with the initial-stage results (Figure 4c) reveals distinct
microphysical variations. First, a considerable number of particles occurred within the typical ice crystal regime, forming a
prominent particle concentration peak in this parameter space, which indicates massive and intensive ice crystal generation.
Second, the abundance of pure liquid droplets decreased substantially, accompanied by a systematic increase in overall particle
 δ . During this stage, supercooled liquid droplets continuously froze into ice crystals, the proportion of low-irregularity
transitional ice crystals declined, and the population of highly non-spherical ice crystals increased significantly, further
enhancing cloud glaciation. The size-resolved distribution at the peak glaciation stage (Figure 4f) shows that newly generated
ice crystals were predominantly smaller than 5 μm , while particles larger than 10 μm remained mostly unfrozen liquid droplets.
Comparing with the initial-stage size-resolved polarization characteristics in Figure 4d, the peak of large low-polarization-
ratio droplets at 10–15 μm decreased markedly, while the concentration peak of high-polarization small particles below 5 μm
increased sharply. This contrasting variation provides direct observational evidence for continuous droplet freezing and in-
cloud glaciation progression.

The phase discrimination framework established in this study enables quantitative diagnosis of ice–water
transformation processes. First, the evolutionary features of in-situ phase partitioning can be intuitively characterised by the
temporal variation in the number fraction of different particle types. Figure 5a shows the time series of the number
concentration proportions of different phase particles. Combined with the stage-dependent polarization features in Figure 4,
the glaciation process is characterised by a persistent increase in ice crystal number fraction and a gradual reduction in
transitional partially frozen particles, while the liquid droplet fraction shows minimal variation. This trend indicates that the
accumulation of mature, non-spherical ice crystals stems primarily from the continuous evolution and freezing of transitional
particles during phase transformation. Nevertheless, relying solely on particle number fractions presents inherent limitations
for quantifying phase transformation, as it overlooks variations in particle size and mass contribution. To overcome this
constraint, we further evaluated the temporal evolution of phase-resolved liquid and ice water contents and their relative mass
contributions, i.e., the glaciation index μ (Figure 5b). Specifically, particles with $\delta < -0.82$ are categorized as spherical droplets



to calculate LWC, whereas particles satisfying the ice crystal criteria with $\delta > -0.33$ and $\text{TotalCounts} < 1.68 \times 10^2$ are used to derive IWC via Equation 1. The cloud glaciation index μ is further calculated via Equation 3. As shown in Figure 5b, IWC increases continuously while LWC declines throughout the phase transition, demonstrating persistent mass transfer from the liquid phase to the ice phase. Consistent with the microphysical variations in Figure 3d, the glaciation index exhibits a similar trend with the average polarization ratio.

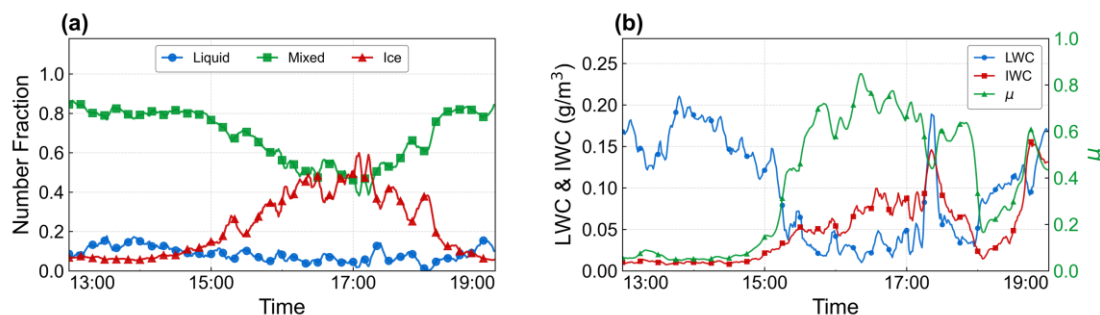


Figure 5. Phase classification of cloud particles based on polarization ratio thresholds. (a) Temporal variations in the number fractions of liquid, ice, and transitional mixed-phase particles; (b) time-series evolution of in-situ liquid water content (LWC) and ice water content (IWC), together with the corresponding glaciation index (μ).

3.3 Polarization Characteristics of Background Aerosols Under Cloud-free Conditions

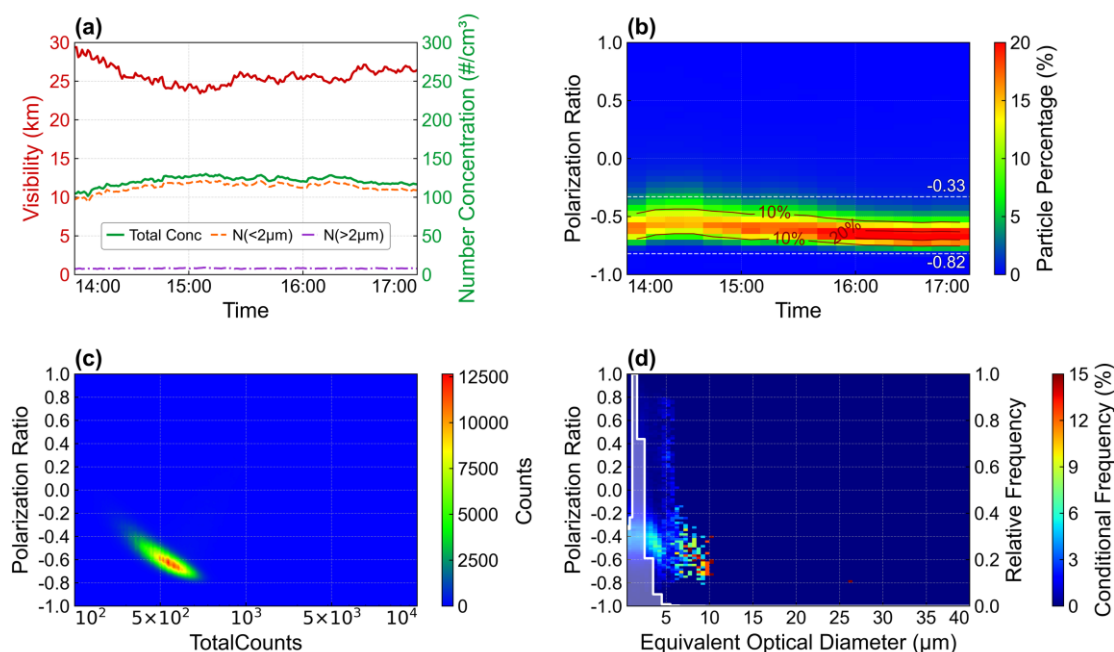


Figure 6. Polarization characteristics of the cloud-free aerosol event observed on 22 February 2024. (a) Time series of visibility and particle number concentration; (b) temporal evolution of the particle polarization ratio



distribution; (c) overall distribution characteristics of polarization ratio and TotalCounts; (d) size-resolved polarization ratio distribution and corresponding particle relative frequency distribution.

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To characterize the polarization characteristics of ambient aerosol particles, a representative cloud-free period from 14:00 to 17:00 on 22 BJT February 2024 was selected for detailed analysis. This interval was dominated by elevated aerosol loading in the complete absence of cloud conditions. As shown in Figure 6a, atmospheric visibility consistently exceeded 10 km, the particle size distribution peaked below 2 μm , and the total particle number concentration remained above 100 cm^3 .
345 These meteorological and microphysical criteria effectively eliminate potential contamination from cloud hydrometeors, providing a robust sample set for evaluating baseline aerosol polarization properties.

Figure 6b shows the temporal evolution of the particle polarization ratios during the cloud-free period. The average polarization ratio of ambient aerosols at the Mt. Liupan site is approximately -0.54 , with most particles lying between -0.82 and -0.33 and the full distribution spanning -0.8 to -0.2 (Figure 6c). Within this range, spherical cloud droplets ($\delta < -0.82$)
350 are essentially absent, so the polarization ratio by itself separates the aerosol population clearly from liquid droplets. Relative to the chamber-generated ice crystals, however, the aerosol distribution is much closer to the ice domain: the frequency peak near -0.6 lies just below the ice-crystal threshold of -0.33 , and the high- δ tail extends into the region $\delta > -0.33$, overlapping the optical signatures of weakly to highly non-spherical ice crystals — a behavior qualitatively consistent with previous in-situ polarization measurements of ambient mineral dust, which showed that single-particle polarization ratios vary
355 systematically with particle size and source region (Tian et al., 2020), and with laboratory polarization measurements demonstrating that non-spherical aerosol particles exhibit polarization ratios spanning wide ranges dependent on particle type (Glen and Brooks, 2013). Therefore, the polarization ratio alone cannot distinguish aerosols from ice crystals. Introducing TotalCounts resolves most of this ambiguity: about 90 % of the highly non-spherical ice crystals exhibit TotalCounts below 1.68×10^2 , whereas aerosols generally produce markedly higher counts, so the joint criterion ($\delta > -0.33$ and TotalCounts $<$
360 1.68×10^2) separates ice crystals from aerosols with an efficiency exceeding 98 %. TotalCounts alone, however, is not a clean discriminator either: weakly non-spherical ice crystals, whose backward scattering is enhanced by their residual sphericity, can fall within the same TotalCounts range as aerosols. This residual overlap is precisely why the classification framework retains an intermediate regime ($-0.82 < \delta < -0.33$) instead of forcing a binary decision, and why the joint δ -TotalCounts constraint, rather than either parameter alone, is required for robust phase discrimination.

365 Notably, a threshold classification based solely on $\delta > -0.33$ would misidentify a fraction of aerosol particles as ice crystals. However, aerosols and ice crystals exhibit distinctly different TotalCounts magnitudes: aerosols produce considerably higher backscattering signals, with the majority displaying elevated TotalCounts values that contrast sharply with the ice crystal criterion (TotalCounts $< 1.68 \times 10^2$). Statistical analysis reveals that 98.31 % of aerosol particles exceed this TotalCounts upper boundary. This verifies that coupling constraints from both the polarization ratio and TotalCounts
370 effectively separate aerosols from fully glaciated, highly non-spherical ice crystals under cold atmospheric conditions.



Nevertheless, the unambiguous separation between aerosols and low-irregularity ice crystals remains challenging due to their overlapping optical signatures.

Overall, ambient aerosol particles exhibit unique polarization characteristics, displaying moderate polarization ratios and backscattering intensities that fall between the typical regimes of pure liquid droplets and pure ice crystals. This empirical finding further corroborates the co-existence of both liquid water particles and ice crystal particles identified in the field observations discussed in Section 3.2. The size-resolved aerosol polarization features are illustrated in Figure 6d. The aerosol size distribution peaks below 2 μm , with corresponding polarization ratios predominantly spanning from -0.7 and -0.2 , which are distinctly higher than those of spherical water droplets. Notably, certain aerosol subsets present polarization signatures that directly overlap with those of highly non-spherical ice crystals. This overlap provides compelling evidence that single-parameter polarization thresholding is inherently insufficient for accurate aerosol–ice discrimination, underscoring the necessity of the dual-parameter classification framework established in this study.

4 Conclusions and Atmospheric Implications

However, polarization-based phase discrimination schemes for fine particles are typically calibrated against either controlled laboratory conditions or a single field campaign alone, and systematic cross-validation that couples multi-temperature cloud chamber experiments with in-situ field observations remains rare, particularly for the synchronous discrimination of aerosols, supercooled liquid droplets, and ice crystals smaller than 50 μm in mixed-phase clouds. Targeting these research gaps, this study establishes a temperature-independent dual-parameter phase discrimination framework based on the particle polarization ratio (δ) and the total scattering counts (TotalCounts) measured by the Ground-based Fog and Aerosol Spectrometer (GFAS). The threshold criteria were comprehensively derived by synthesizing cloud chamber experiments with winter–spring field observations at Mt. Liupan in Northwest China.

These thresholds enable robust classification of fine cloud and aerosol particles with distinct microphysical properties. Highly non-spherical ice crystals are characterised by elevated polarization ratios ($\delta > -0.33$, satisfied by 90.34 % of ice crystal particles) and low backscattering intensity, with 90.46 % yielding TotalCounts values lower than 1.68×10^2 . Spherical supercooled liquid droplets exhibit distinctly low polarization ratios ($\delta < -0.82$) and strong backscattering signals. The transitional parameter range ($-0.82 < \delta < -0.33$) accommodates low-irregularity, incompletely frozen ice crystals and ambient aerosol particles. Crucially, the auxiliary TotalCounts upper threshold of 1.68×10^2 effectively separates over 90 % of non-spherical ice crystals from aerosols. When applied to field mixed-phase cloud observations, the proposed dual thresholds demonstrate high operational applicability. Throughout the in-cloud glaciation process, the average particle polarization ratio co-varied tightly with the glaciation index derived from the phase-resolved water contents. The corresponding scatter relationship is illustrated in Figure S3 in the Supplementary. Because the glaciation index is itself constructed from the δ -based classification, this consistency is best interpreted as the internal coherence of the framework rather than an independent



validation; the classification is anchored by the chamber calibration, while the concurrent increases in particle number concentration, MVD, and the observed cooling provide independent support for the diagnosed glaciation.

405 These findings provide novel microphysical observational evidence for ice–water phase transformation in mixed-phase clouds and advance the application of polarization detection technology for fine particle phase identification within the sub-50 μm size range. Compared with conventional temperature-dependent phase discrimination schemes, the joint constraint of polarization ratio and TotalCounts proposed in this study effectively overcomes the inherent defects of temperature threshold methods, particularly the inability to distinguish coexisting ice and liquid particles under supercooled conditions. This approach achieves synchronous in-situ classification of ice crystals, liquid droplets, and transitional partially frozen particles, 410 filling a key methodological gap in fine-scale mixed-phase cloud microphysical observations. In terms of cloud microphysical process research, this method enables continuous tracking of ice–water phase evolution based on real-time single-particle optical properties. The close consistency between the temporal evolution of the polarization ratio and the classified glaciation index provides a temperature-independent diagnostic framework to quantitatively constrain the initiation timing, transformation rate, and evolution magnitude of in-cloud glaciation. In the typical mixed-phase cloud temperature range of 415 $-10\text{ }^{\circ}\text{C}$ to $-20\text{ }^{\circ}\text{C}$, phase identification for particles in the 0.5–50 μm size range is a key bottleneck limiting the accurate estimation of cloud glaciation rate (Costa et al., 2017). Systematic underestimation of ice crystal concentration in this size range induces substantial biases in simulated cloud albedo and life cycle. Furthermore, investigations of Amazonian convective clouds have confirmed that precise characterization of the sub-50 μm initial droplet spectrum has a decisive impact on evaluating subsequent Bergeron process efficiency, whose activation largely depends on accurate quantification of the relative 420 abundance of micron-scale ice crystals and supercooled liquid droplets (Braga et al., 2017). The dual-parameter discrimination method established in this study achieves full-spectrum identification of 0.5–40 μm ice crystals and unfrozen supercooled droplets, providing a direct observational tool for validating Bergeron growth mechanisms and in-cloud ice–water transformation processes. Practically, this study provides a feasible operational particle classification scheme for ground-based in-situ observations of clouds, fogs, and aerosols, offering technical support for long-term observations at mountainous, polar, 425 and other remote observational stations.

Despite these promising findings, several limitations warrant further investigation. First, low-irregularity partially frozen ice crystals and ambient aerosols exhibit overlapping optical signatures within the transitional polarization interval, leading to incomplete separation between these two particle types. Second, the retrieval accuracy of ice water content for sub-50 μm small ice crystals is constrained by uncertainties in existing diameter–mass parameterization schemes, which introduces 430 quantitative errors to microphysical calculations. Third, the field observations were conducted at a single mountain site during winter and spring, limiting the geographical and seasonal universality of the established thresholds. Future work will extend observational durations and expand deployment across diverse regions and seasons to optimize the general applicability of these thresholds. Moreover, synergistic observations combining GFAS with complementary remote sensing and in-situ instruments will be implemented to integrate multi-source microphysical and optical data, enhancing particle phase



435 classification reliability and advancing the quantitative understanding of ice--water transformation mechanisms in mixed-phase clouds.

Code and data availability.

Data used in this study are available from Jiang and Wang (2026) or <https://doi.org/10.5281/zenodo.22908180>.

Competing interests.

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

Acknowledgements.

We thank Hao Zhang, Tao Wang, and Pingshun Wang for carefully maintaining the in-site measurements.

Financial support.

This study was supported by the NSFC Regional Innovation and Development Joint Fund (U24A20604), the National Natural
445 Science Foundation of China (42575074), and the Fundamental Research Funds for the Central Universities (lzujbky-2026-jd zx07).

Author contribution

YJ, JL, and Yuan Wang designed the research. YJ, Yuan Wang, ZJ, PZ, QX, and ZS conducted field observations and data
collection. YJ, Yang Wang, and HW performed cloud chamber experiments. YJ and Yuan Wang analyzed the data and drafted
450 the manuscript. All co-authors contributed to result interpretation, manuscript review and editing.

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