Response to reviewer

We gratefully thank the reviewer for the constructive comments and suggestions to improve the manuscript. Below are the detailed point-to-point responses to the reviewer's comments. For clarity, the reviewer's comments are listed below in *black italics*, while our responses and changes in the manuscript are shown in blue and red, respectively. The changes in the revised manuscript and supporting materials are also highlighted.

Anonymous Referee #2 (RC3)

This paper explores the significant role of long-range transported dust in influencing ice-nucleating particles (INPs) and cloud formation within the North China Plain. The authors analyzed precipitation samples from Mount Tai, a background site, over several months in 2021, revealing that INP concentrations were highest in spring. This seasonal increase was primarily linked to mineral dust transported from distant arid regions, a finding supported by satellite data and chemical analyses using a Positive Matrix Factorization model. The study concludes that this transported dust is a dominant factor in INP abundance, driving large-scale ice cloud formation and impacting aerosol-cloud interactions in continental areas.
This reviewer has several major and minor comments. While the study addresses an important topic relevant to the journal, the paper currently lacks sufficient empirical evidence, methodological detail, and analytical rigor to fully support its central claims regarding the dominant role of long-range transported dust in INP concentrations and cloud formation. Significant revisions, particularly in addressing the methodological gaps, substantiating claims with clearer data correlations, and refining the seasonality analysis, are deemed necessary before considering publication in ACP.

Response: We sincerely thank the reviewer for the thorough evaluation and constructive comments. We fully acknowledge the importance of providing sufficient empirical evidence, methodological detail, and analytical rigor to substantiate our conclusions. Accordingly, we have made substantial improvements in the revised manuscript. In the Methods section, we added details on the sampling procedures for rainwater and blank samples and recalculated the data after subtracting field blanks. We also expanded the description of the INP characterization method and clarified the uncertainty associated with the use of CWC. In the Results and Discussion section, we refined the PMF model analysis to distinguish two types of dust. Combined with backward trajectory analysis, we separated long-range transported mineral dust from local road dust, thereby further highlighting the role of long-range transported mineral dust in INPs. We believe these revisions have significantly improved the quality of the manuscript. Below are the detailed point-to-point responses.

Major comments:
[1] How do the authors assess the background contribution of aerosols dry and wet depositions in precipitation samples during each sampling interval? How about the influence of evaporation? Is it accounted in total rainwater amount and estimation in Ninp? This reviewer finds the Milli-Q water background spectrum of the authors' freezing assay in the supplemental material, but not the field blank background. These background data need to be presented in

the main manuscript.

Response: Thanks for the comment. We provide detailed sample collection processes in the revision. During dry periods without precipitation, the bucket was stored indoors with a lid to prevent contamination. At the onset of precipitation, a new clean polyethylene bag was immediately mounted on the bucket, and the sampling time was recorded. Once precipitation ceased, the polyethylene bag containing the sample was promptly retrieved to minimize contamination from dry deposition. Samples were then transferred to pre-sterilized polycarbonate bottles and stored at -20℃ until further analysis. During sampling, the total precipitation amount was measured directly with a rain gauge, and only precipitation events with amounts greater than 1.0 mm were analyzed in this study. The sampling procedures have been described previously (Liu et al., 2023). Evaporation during precipitation was considered negligible.

We poured 100 ml Milli-Q water into a clean polyethylene bag as the field blank, and the measured concentrations were subtracted from those of the precipitation samples at each freezing temperature (Figure R1 and revised Figure S3).

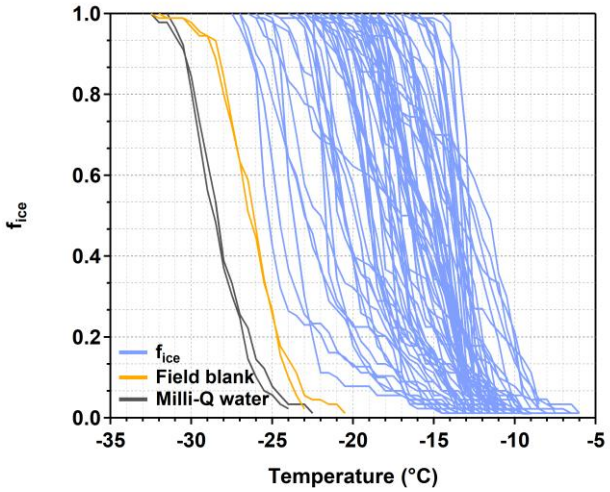

Figure R1. Frozen fractions of rainwater samples (f_{ice}), Milli-Q water and filed blanks as a function of temperature.

The following revisions have been made in the revised manuscript.

A polyethylene bucket (50 cm in diameter) equipped with a pre-cleaned polyethylene bag was positioned 1.5 m above ground level for sample collection. During dry periods without precipitation, the bucket was stored indoors with a lid to prevent contamination. At the onset of precipitation, a new clean polyethylene bag was immediately mounted on the bucket, and the sampling time was recorded. Once precipitation ceased, the polyethylene bag containing the sample was promptly retrieved to minimize contamination from dry deposition. Samples were then transferred to pre-sterilized polycarbonate bottles and stored at −20 ℃ until further analysis (Liu et al., 2023). During sampling, the total precipitation amount was measured directly with a rain gauge, and only precipitation events with amounts greater than 1.0 mm were analyzed in this study. A total of 67 precipitation samples were collected across four seasons: 11 in spring, 29 in summer, 25 in autumn, and 2 in winter. Two field blanks were prepared by pouring 100 ml Milli-Q water into a clean polyethylene bag, and the measured concentrations

were subtracted from those of the precipitation samples at each freezing temperature (Figure S3). All precipitation samples analyzed in this study were rainfall. The two winter samples are reported only in terms of their concentrations and were excluded from further analysis.

[2] Does the precipitation type (or intensity) have any influences on Ninp? Some previous studies of precipitation INP report it. Discussion should be provided in the main manuscript.

Response: Thanks for the comment. In this study, the precipitation intensity ranged from 1 mm to 156 mm. We analyzed the relationship between precipitation intensity and N_{INP}. As shown in Figure R2, no clear pattern was observed between precipitation intensity and N_{INP}.

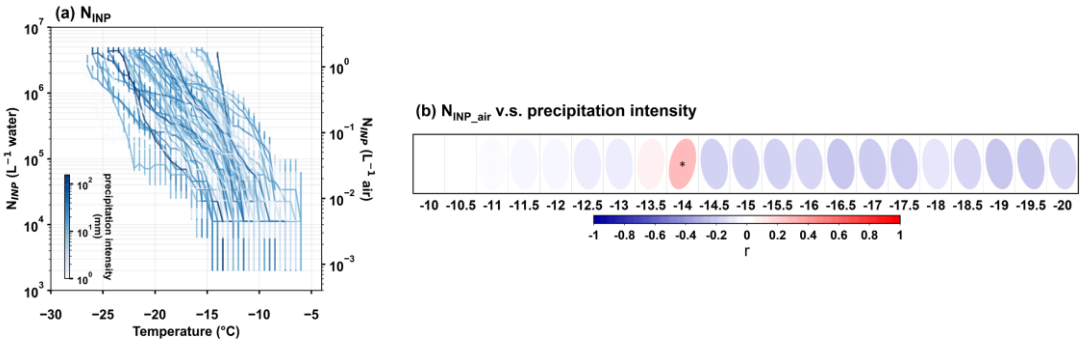

Figure R2. (a) N_{INP} spectra per unit volume of air (N_{INP_air}, left axis) and per unit volume of water (N_{INP_water}, right axis) as functions of temperature. Line colors indicate rainfall intensity, and error bars represent the 95% confidence intervals. (b) Relationships between precipitation intensity and N_{INP_air}.

We have added the following statement in the revised manuscript:

Precipitation intensity had little influence on INP concentrations (Figure S4).

[3] Seasonality analysis is thin in winter as there are only two samples analyzed, and this reviewer does not feel comfortable to see anyone claims seasonality with such a low number of samples.

Response: Thanks. We agree and have remove the winter samples from the analysis in the revised manuscript.

We have added the following statement in revision.

The two winter samples are reported only in terms of their concentrations and were excluded from further analysis.

[4] How does the authors segregate the local dust aerosol contribution from long-range ones? The approach should be explicitly clarified in the method section.

Response: We thank the reviewer for the valuable comments. We segregate the local dust from long-range transported dust primarily based on the PMF analysis results combined with the HYSPLIT backward trajectories, as detailed below:

(1) After excluding the winter samples, we re-conducted the PMF analysis. The resulting source profiles are shown in Figure R3, where two types of dust were identified. Factor 1

exhibited high loadings of mineral elements such as Al, Fe, Mn, Ti, Mg^{2+} and Ca^{2+}, and was identified as mineral dust (Yuan et al., 2008; Huang et al., 2014). Factor 2 was characterized by exhibited elevated levels of water-soluble Mg^{2+} and Ca^{2+}, but lower concentrations of Al, Mn and Fe. In addition, several pollution-related elements, including Zn, Pb, Ni, Cu, and Cl^-, exhibited moderately high loadings. These features suggest that Factor 2 represents road dust, mainly originating from the re-suspended dust and surface soil from unpaved roads (Hien et al., 2001; Yuan et al., 2008).

(2) We calculated the 24-hour backward trajectories corresponding to the different source factors. Trajectories characterized by high concentrations of Al, Mn, Fe (top 15th percentile) were assigned to Factor 1 (mineral dust), whereas those elevated Ca^{2+} and Mg^{2+} (top 15th percentile) but without high Al, Mn, Fe were classified as Factor 2 (road dust). As shown in Figure R4, the air masses associated with Factor 1 mainly originated from long-range transport from the northwest, while those dominated by Factor 2 were dispersed across various directions over short distances, implying a more local origin compared with Factor 1.

Overall, these evidences support the interpretation that Factor 1 corresponds to long-range transported mineral dust, while Factor 2 reflects locally generated road dust.

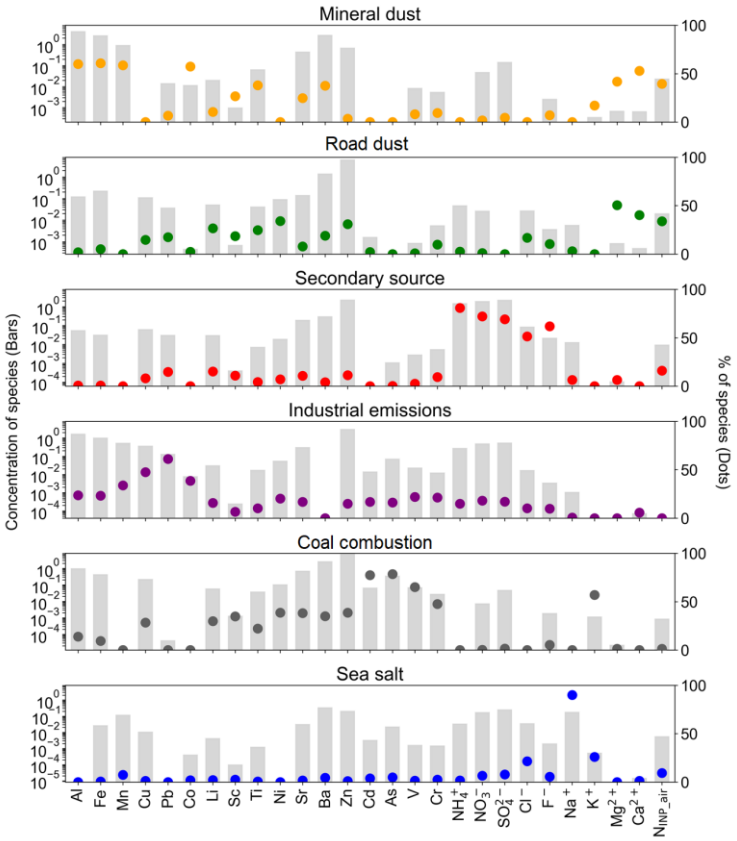

Figure R3. Source apportionment of 17 metallic elements, 9 water-soluble ions, and $N_{INP\text{-}air}$ (-16 °C) using the positive matrix factorization (PMF) model. Bars represent the concentration of individual species, and dots indicate their percentage contributions. The units are μg/ml for inorganic ions and ng/ml for metal elements.

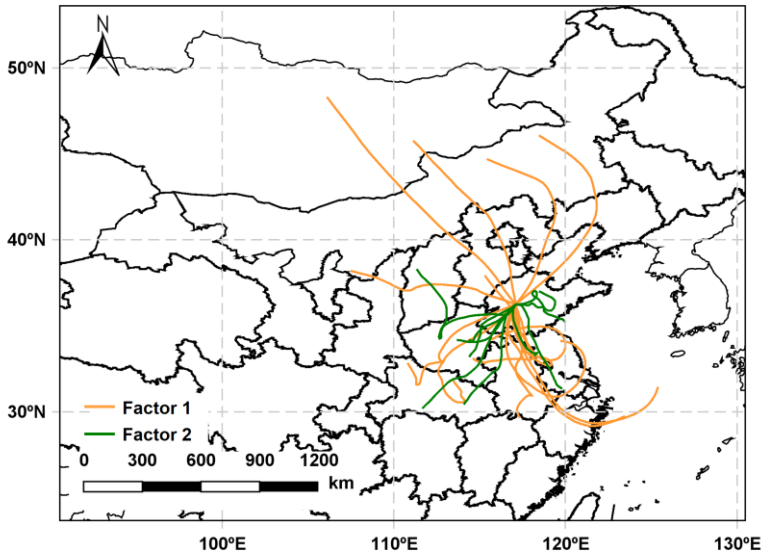

Figure R4. 24-hour backward trajectories of air mass associated with Factor 1 and Factor 2.

[5] L196-201: This part sounds very speculative. What evidence that the authors have to specify it's biological? Heat labile INP analysis? Then, why heat sensitivity is not the highest in spring? Also, what evidence can the authors offer to refer to the feldspar involvement?

Response: We thank the reviewer for the valuable comments. We agree that discussing the possible reasons for the high N_{INP} values in spring is confusing and speculative here, and therefore we have moved this part to section 3.2.

The following revisions have been made in the revised manuscript.

The elevated N_{INP_air} in spring is likely attributed to the combined influence of multiple types of INP, which will be discussed in Section 3.2.

In Section 3.1, we observed a seasonal enhancement of N_{INP_air} during spring compared to other seasons. Given the substantial contribution of biological sources at Mount Tai, this springtime enhancement may be partially attributed to biogenic emissions. However, the proportions of HS-N_{INP_air} in the summer and fall were higher than in spring (Figure 2c-e), despite generally lower N_{INP_air} values in these two seasons. Figure 2c-e showed that the HR-N_{INP} in spring was significantly higher than in summer and fall across all freezing temperatures, with particularly pronounced differences at -20 °C, where springtime HR-N_{INP} exceeded that of the other seasons by more than a factor of two. Building on this, we propose that the enhanced ice-nucleating activity observed in spring may be influenced by heat-resistant components, for example, mineral dust (Conen et al., 2011).

[6] L242-244: If dust is truly responsible for INP propensity, present the correlation between (long-range-transported) dust conc and Ninp in some ways. The authors can keep everything simple this way… If they cannot, please explain why not.

Response: We appreciate the comments. Since direct measurements of long-range transported dust concentrations were not available, we used two proxy indicators: the observed PM_{10}-$PM_{2.5}$ concentration and satellite-retrieved Dust Aerosol Optical Depth (DAOD) at 550 nm. We examined the relationship between PM_{10}-$PM_{2.5}$ and N_{INP}, as well as between DAOD and N_{INP}.

A weak positive correlation was observed at lower freezing temperatures, but the results were not statistically significant (Figure R5). We attribute this lack of correlation to several factors: (1) dust events are episodic and may not coincide with precipitation events, such that simple concentration metrics cannot fully capture the effective INP fraction; (2) the ice-nucleating efficiency of dust is highly dependent on mineralogical composition, which is not reflected in bulk dust mass concentrations; and (3) daily satellite retrievals of DAOD involve substantial uncertainties, further complicating the correlation analysis.

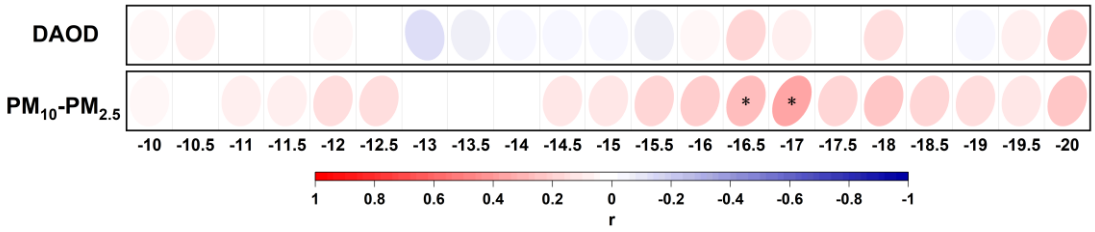

Figure R5. Relationships between N_{INP_air} and (a) DAOD and (b) PM_{10}-$PM_{2.5}$.

[7] Figure 3: This paper does not describe how the composition of INPs was measured. L279-states the inclusion of mineral INP active at -16 dC. It could be biogenic or organic that come with minerals, right? Isn't it misleading to cite Tobo et al.?

Response: Thanks for the comments. Chemical analysis of precipitation is described in section 2.3, including water-soluble ions (F^-, Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) and trace metal elements (Li, Al, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Sr, Cd, Ba, and Pb). The characteristics of INPs were determined using wet-heat treatment analyses, as detailed in Section 2.1. We agree that the citation of Tobo et al. (2020) may have been misleading in this context and have removed it in the revised manuscript.

We have added the following statement in revision.

Wet-heat treatment was applied to identify the thermally distinct fractions of INPs. Rainwater samples in sealed vessels were immersed in a boiling water bath and heated at 95 ℃ for 30 min, followed the procedure described in Daily et al. (2022). The samples were cooled to room temperature, after which the ice nucleation activity experiment was repeated.

[8] L276-280: This reviewer agrees with the statements. Then, the tile could be misleading. It could be local biogenic materials mixed with long-range-transported dust act as INPs, correct? What proof the authors have to explicitly say it's long-range-transported dust in the title and elsewhere in the manuscript? The same applies to L313-315 – the authors may need to rephrase these parts.

Response: Thanks for the comments. We have revised the PMF analysis results and clarified that Factor 1 originates from long-range transported dust (see our response to Comment [4]). The contribution of Factor 1 to INPs is 39.4% on annual average, increasing to 44.8% in spring.

We acknowledge that local biogenic materials mixed with long range transported dust can enhance the ice-nucleating activity, however, this aspect is beyond the focus of the present study.

We have revised these statements as follows:

We therefore speculate that the elevated N_{INP_air} at warmer temperatures may result from the

combined effects of biogenic aerosols and mineral dust. Such interactions have also been shown to enhance the ice-nucleating activity of mineral dust (Yahya et al., 2019; O'sullivan et al., 2016). Nonetheless, we did not perform a detailed analysis on such interactions in the present study.

[9] Sect. 3.4 is speculative and substantial revision seems necessary.

Response: Thanks for the comments. We have revised these paragraphs as follows:

Figure 4 illustrates the monthly average cloud-related parameters in the NCP region (blue dashed box in Figure S1, covering the region between 33°N-42°N latitude and 112°E-121°E longitude). TCC exhibited a bimodal distribution, with peaks in March and July (Figure 4a). In comparison, winter months showed relatively lower TCC, suggesting weaker aerosol-cloud interactions. Cloud cover increased with altitude (Figures 4b-4d), consistent with distinct roles of INPs and cloud condensation nuclei (CCN), with CCN dominating warm cloud formation at lower altitudes and INPs govering cold and mixed-phase cloud development at mid-upper troposphere (Kuba and Fujiyoshi, 2006; Morrison et al., 2012; Hogan et al., 2004). The increase in cloud cover with altitude underscores the important role of INPs cloud formation over the NCP.

In spring, the monthly mean CBH and CTH ranged between 2.8-4.0 km a.s.l. and 6.9-8.6 km a.s.l., respectively, and CTT varied between -28.4 ℃ to -32.4 ℃ (Figure 4e-f), indicating favorable conditions for mixed-phase clouds formation. As illustrated in Figure S13b, the mean dust transport height was approximately 1.7 ± 0.5 km, reaching a maximum of 4.2 km in April. These findings suggest that suspended dust particles in spring can reach cloud levels and serve as effective INPs, playing a critical role in modulating cloud microphysics and development.

During summer, CBH was lower, which may have facilitated vertical mixing and enhanced the upward transport of surface-originating INPs, including biological particles such as pollen and microbial fragments. In winter, cloud cover remained relatively low across all altitude (Figure 4b-d), suggesting limited cloud development. The small difference between CBH and CTH further indicates the shallow vertical extent of cloud layers in winter.

Minor comments:

L37-38: A reference is missing.

Response: We have added references. The revised statement reads as follows.

Among these, immersion freezing, initiated by the immersion of ice nuclei within liquid droplets, is widely recognized as the dominant pathway for ice nucleation in mixed-phase clouds (Hoose et al., 2010; Murray et al., 2012; Kanji et al., 2017).

L78-80: Do the author mean aerosol composition? Or INP composition? How do they segregate and measure the INP of different sources, such as organic, biological, and minerals. If it's really INP composition, please clarify the approach in the method section. Also, there are only two winter samples that the authors analyzed, and the sample size is too small to validate seasonality statistically. This reviewer highly recommends the author to rephrase seasonal variations and/or seasonality here and elsewhere applicable.

Response: Thanks for the comment. We measured the chemical composition of precipitation samples, as described in Section 2.3. We have revised the statement to read:

In this study, we analyzed the INPs in precipitation samples collected at the summit of Mount

Tai (1534 m a.s.l.) from February to November 2021. The seasonal variations and potential sources of INPs were analyzed, with a particular focus on assessing the influence of dust events on INP concentrations and associated cloud properties.

We agree that the winter sample size is too small and have therefore excluded these samples from the main analysis in the revised manuscript. The following statement has been added in revision:

The two winter samples are reported only in terms of their concentrations and were excluded from further analysis.

L89-93: The winter sample is poorly represented. Why? Please provide justifications. Less precipitation in winter? Logistics reason? Also, please clarify what the authors used for sampling intervals for individual samples. How did you preserve rainwater without impact of evaporation and dry/wet depositions?

Response: We have removed the analysis of winter samples in the revised manuscript.

We apologize for the confusion regarding the sampling procedure and have provided a more detailed description in the revision. Details are provided in our response to comment [1].

Revised manuscript:

A polyethylene bucket (50 cm in diameter) equipped with a pre-cleaned polyethylene bag was positioned 1.5 m above ground level for sample collection. During dry periods without precipitation, the bucket was stored indoors with a lid to prevent contamination. At the onset of precipitation, a new clean polyethylene bag was immediately mounted on the bucket, and the sampling time was recorded. Once precipitation ceased, the polyethylene bag containing the sample was promptly retrieved to minimize contamination from dry deposition. Samples were then transferred to pre-sterilized polycarbonate bottles and stored at −20 °C until further analysis (Liu et al., 2023). During sampling, the total precipitation amount was measured directly with a rain gauge, and only precipitation events with amounts greater than 1.0 mm were analyzed in this study. A total of 67 precipitation samples were collected across four seasons: 11 in spring, 29 in summer, 25 in autumn, and 2 in winter. Two field blanks were prepared by pouring 100 ml Milli-Q water into a clean polyethylene bag, and the measured concentrations were subtracted from those of the precipitation samples at each freezing temperature (Figure S3). All precipitation samples analyzed in this study were rainfall. The two winter samples are reported only in terms of their concentrations and were excluded from further analysis.

L109-111: This seems a big assumption. Can the authors provide any evidence that CWC can be considered a constant for their sampling location and conditions throughout the year?

Response: Thanks for the comment. A cloud water content (CWC) of 0.4 g m^{-3} has been widely adopted in numerous studies (Niu et al., 2024; Vepuri et al., 2021; Petters and Wright, 2015a; Beall et al., 2020; Petters and Wright, 2015b), covering various site types (urban, rural, coastal and mountain), as well as different seasons. Vepuri et al. (2021) reported INPs in precipitation samples in the Texas Panhandle region over four seasons, from June 2018 to July 2019. They assumed a constant CWC of 0.4 g m^{-3} for the following reasons: (1) Petters and Wright (2015) and references therein showed typical values of CWC for different cloud types could narrowly range within a factor of 2 from 0.4 g m^{-3}; (2) variations in N_{INP} with CWC values for different

cloud types in the atmosphere would typically be limited within a factor of 2, whereas the measured uncertainties of N_{INP} could be larger than that; and (3) Zhang et al. (2006) suggests that evaporation does not contribute to NINP bias for both strong convective systems and persistent rain events with cloud base heights of ≈ 3 km. In our study, the observed CWC value was not available, and we therefore followed Vepuri et al. (2021) in adopting a constant CWC of 0.4 g m^{-3}. We acknowledge that this assumption introduces uncertainty and have clarified this in the revised manuscript.

The cloud water content (CWC) was assumed to be 0.4 g m^{-3}, a value widely adopted in previous studies (e.g., (Chen et al., 2021; Chen et al., 2024; Niu et al., 2024; Vepuri et al., 2021; Petters and Wright, 2015a)). For cloud droplets with a volume of 1 pL dispersed in 1 m^3 of air, the corresponding cloud water volume per unit air volume ($F_{cloud\text{-}air}$) is 4×10^{-7} m^3 water/m^3 air. Seasonal variability in CWC was not considered in this study (Vepuri et al., 2021).

L115-116: How about water insoluble components? Most INPs are assumed to be water insoluble, providing surface for ice embryo to form. This circles back to the reviewer's question listed above – have the authors looked into aerosol or INP composition?

Response: Thanks for the comment. The metal elements in the precipitation samples were analyzed, as our response to Comment [7].

L189 & Figure 1 caption: Please rephrase urban to rural here and elsewhere the revision applies. The study location in Vepuri et al. is indeed a rural area. Also, rephrase "our measurements" to our Ninp measurements.

Response: Thanks. We have revised accordingly.

L213-: The method of heat application test needs to be described in Sect. 2.2.

Response: Thanks for the comment. We have moved the method to Sect. 2.2.

Wet-heat treatment was applied to identify the thermally distinct fractions of INPs. Rainwater samples in sealed vessels were immersed in a boiling water bath and heated at 95 ℃ for 30 min, followed the procedure described in Daily et al. (2022). The samples were cooled to room temperature, after which the ice nucleation activity experiment was repeated.

L217- & Figure 2a: It's hard to interpret the data since Figure 2a looks so busy. +/- 2.4 dC seems pretty substantial uncertainties. Absolutely, no exception is involved? The authors might revisit their data (hard to assess from the figure).

Response: Thanks for the comment. Figure 2a has been revised to display only the HR-N_{INP_air} data. After carefully checking the data, two outlier samples we removed, resulting in a smaller standard deviation. In revision, we removed the statement regarding the onset freezing temperature and the revised statement read as follows:

As shown in Figure 2a, the wet-heat treatment led to a reduction in N_{INP_air} by one to two orders of magnitude.

L226-227: Why in summer not spring? Sounds contradicting to L192-198 about the spring Ninp peak.

Response: Thanks for the comment. We revised this paragraph as follows:

Given the substantial contribution of biological sources at Mount Tai, this springtime enhancement may be partially attributed to biogenic emissions. However, the proportions of HS-N_{INP_air} in the summer and fall were higher than in spring (Figure 2c-e), despite generally lower N_{INP_air} values in these two seasons. Figure 2c-e showed that the HR-N_{INP} in spring was significantly higher than in summer and fall across all freezing temperatures, with particularly pronounced differences at -20 °C, where springtime HR-N_{INP} exceeded that of the other seasons by more than a factor of two. Building on this, we propose that the enhanced ice-nucleating activity observed in spring may be influenced by heat-resistant components, for example, mineral dust (Conen et al., 2011).

L276-280 (& L315): Polysaccharides or polymers (insoluble lignin and cellulose INPs)? Cite proper references if the latter is the case.

Response: We appreciate the reviewer's valuable comments. We have revised to "polymers" and added the relevant references. The revised manuscript as follows:

However, the PMF model does not account for biogenic source contributions due to the absence of biological tracers (e.g., proteins and polymers) (Hiranuma et al., 2019; Bogler and Borduas-Dedekind, 2020).

Figure 4 & L286: The authors might want to offer individual sample interval specific data (definitely not monthly average unless there is strong justification that conditions are stable through a month).

Response: We appreciate the reviewer's suggestion. We carefully examined the cloud parameter in the precipitation days, as shown in Figure R6. However, due to their limited temporal resolution and considerable uncertainties, these data cannot robustly support our conclusions at the daily scale. Dust, precipitation and related processes are episodic, and satellite overpasses do not always coincide with the precipitation events we analyzed. Therefore, we opted to use monthly averages to reduce noise and improve representativeness. Moreover, aerosol–cloud interactions typically occur on longer timescales, as dust particles can remain suspended in cloud layers for hours to several days, depending on their size, density, and the atmospheric conditions, before being removed by precipitation. Since our study highlights the important role of long-range transported dust, particularly in spring, we consider seasonal variations to be more representative for characterizing aerosol–cloud interactions.

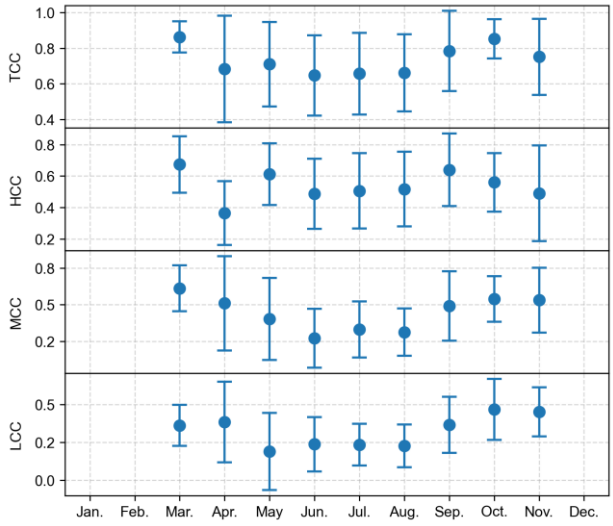

Figure R6. Averaged total cloud cover (TCC), high cloud cover (HCC), medium cloud cover (MCC), and low cloud cover (LCC) over the NCP region (32°N-42°N, 112°E-121°E) on precipitation days in 2021. Error bars represent the standard deviations.

Reference:

Beall, C. M., Lucero, D., Hill, T. C., DeMott, P. J., Stokes, M. D., and Prather, K. A.: Best practices for precipitation sample storage for offline studies of ice nucleation in marine and coastal environments, Atmos. Meas. Tech., 13, 6473-6486, 10.5194/amt-13-6473-2020, 2020.

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Whiteside, C. L., Yamashita, K., Zelenyuk, A., and Möhler, O.: A comprehensive characterization of ice nucleation by three different types of cellulose particles immersed in water, Atmos. Chem. Phys., 19, 4823-4849, 10.5194/acp-19-4823-2019, 2019.

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