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Sink, Source or Something In-Between? Net Effects of Precipitation on Aerosol Particle Populations

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Abstract Interactions between atmospheric aerosols, clouds, and precipitation impact Earth's radiative balance and air quality, yet remain poorly constrained. Precipitating clouds serve as major sinks for particulate matter, but recent studies suggest that precipitation may also act as a particle source. The magnitude of the sources versus sinks, particularly for cloud condensation nuclei (CCN) numbers, remain unquantified. This study analyzes multi-year in situ observations from tropical and boreal forests, as well as Arctic marine environment, showing links between recent precipitation and enhanced particle concentrations, including CCN-sized particles. In some cases, the magnitude of precipitation-related source equals or surpasses corresponding removal effect. Our findings highlight the importance of cloud-processed material in determining near-surface particle concentrations and the value of long-term in situ observations for understanding aerosol particle life cycle. Robust patterns emerge from sufficiently long data series, allowing for quantitative assessment of the large-scale significance of new phenomena observed in case studies.

Plain Language Summary Atmospheric aerosols, clouds, and precipitation play a significant role in Earth's temperature regulation and air quality. However, understanding their interactions is still a challenge. While clouds and precipitation help remove particles from the atmosphere, recent research suggests rain could also introduce new particles. The extent of this particle source and its impact on climate are still unknown. In this study, we analyzed years of observational data from clean environments, including tropical and boreal forests and the Arctic marine boundary layer. We discovered that after precipitation, new particles were sometimes added to the surface atmosphere. In some cases, rain introduced as many or even more particles than it removed. Our findings highlight the importance of considering how clouds and rain recycle particles when studying air quality and climate. Long-term, real-world observations help us understand atmospheric particle life cycles and identify consistent patterns, ultimately improving our knowledge of the complex interactions between aerosols, clouds, and precipitation.

Théodore Khadir and Ilona Riipinen contributed equally to this work.

Key Points:

- Precipitation can act as a source for particles of varying sizes depending on the environment, reflecting diverse underlying mechanisms
- Recycling cloud-processed material influences near-surface particle concentrations, emphasizing its relevance for climate model implementation
- Studying the time-dependent instead of total accumulated precipitation elucidates direct versus indirect effects on aerosol populations

Supporting Information:

Supporting Information may be found in the online version of this article.

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1. Introduction

Understanding the impacts anthropogenic aerosol perturbations on cloud and precipitation properties is critical to improve climate model projections (Intergovernmental Panel on Climate Change, 2014a, 2014b; Seinfeld et al., 2016). Consequently, substantial efforts have been focused on resolving the sources of aerosol and cloud condensation nuclei (CCN) in the present and preindustrial atmospheres (Almeida et al., 2014; Bianchi et al., 2021; Carslaw et al., 2017; Kirkby et al., 2016), and their impact on cloud optical properties and lifetime (Lohmann & Feichter, 2005; Malavelle et al., 2017; Quaas et al., 2020; Stevens & Feingold, 2009; Yli-Juuti et al., 2021). Depending on their composition and ambient conditions, the diameters of CCN span from some tens of nanometers upwards. In the present atmosphere, about half of CCN-sized particles originate from secondary sources that is, new particle formation (NPF) from oxidation and condensation of gaseous-phase precursors (Dunne et al., 2016; Merikanto et al., 2009; Spracklen et al., 2008). The corresponding fraction of secondary CCN number in the preindustrial atmosphere is around 70% (Gordon et al., 2017).

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Wet scavenging by clouds and precipitation are the most important sinks for atmospheric particulate mass and the number concentration of accumulation mode (about 100 nm in diameter) particles (Moran-Zuloaga et al., 2018; Ohata et al., 2016; Pöhlker et al., 2018; Seinfeld & Pandis, 2006). The net effect (i.e., the magnitude and sign) of clouds and precipitation on aerosol populations can, however, vary due to the complex response of atmospheric aerosol dynamics to changes in meteorological conditions. Compared to secondary aerosol production associated with atmospheric vapors, much less focus and effort has been put onto understanding the net impacts of clouds and precipitation on atmospheric aerosol and CCN loadings. This lack of consideration has left one half of the feedback related to the life cycle of secondary aerosol particles more poorly constrained than the other (i.e., impacts of aerosol populations on clouds and precipitation, see Figure 1) (Isokääntä et al., 2022; Tunved et al., 2013; Yli-Juuti et al., 2021). Clouds can indirectly enhance concentrations of ultrafine aerosol in nucleation (below about 25 nm in diameter) and Aitken (between about 25 and 100 nm in diameter) modes by removing the accumulation mode particles, which are a major sink for nucleation gaseous precursors and for particles formed by nucleation (Dal Maso, 2002; Riipinen et al., 2011; Westervelt et al., 2013; Zhang et al., 2012). Targeted observations indicate direct production of new particles in the vicinity of clouds (Braga et al., 2022; Clarke, 1992; Clarke et al., 1999; Ekman et al., 2006; Hegg et al., 1990; Krejci, 2003; Murphy et al., 2015; Wehner et al., 2015) and transport of particles and vapors into the boundary layer (BL, Poschl et al., 2010; Prenni et al., 2013; Wang et al., 2016; Andreae et al., 2018; Kompalli et al., 2020; Wang, Liu, et al., 2022; Wang, Gordon, et al., 2022). While some prior studies have reported clouds and precipitation as a source of new aerosol particles and secondary CCN, to our knowledge, none have systematically examined the magnitude of this feedback over extended periods and in various environments. This limits quantitative assessment of the importance of this phenomenon. Statistical analysis of long-term observations is the only route toward assessing the potential of these phenomena in climate-relevant scales—given the high variability and short time atmospheric lifetime of aerosol particles.

The relative magnitudes of the source versus sink processes related to clouds and precipitation in different environments are likely driven by regional scale meteorology including the type of precipitation and clouds. Different dominant sources of aerosols and their precursors adds to the complexity of the studied system. If the CCN production associated with clouds and precipitation is significant compared with the NPF during cloud-free and pristine conditions (Kerminen et al., 2012; Riipinen et al., 2012), then the atmospheric system is expected to be less sensitive to secondary aerosol production than previously thought (Gordon et al., 2017). Here we study the key environmental drivers behind changes in aerosol number size distributions and concentrations of CCN-sized particles, focusing on the overall impact of precipitation in tropical, boreal and Arctic environments. The atmospheric composition in these three remote sites is substantially influenced by natural sources. We contrast the scavenging effects by precipitation and clouds to (a) the potential role of clouds and precipitation to enhance BL aerosol concentration; and (b) the magnitude of the photochemical gas-to-particle aerosol source and consequent growth. Finally, we provide quantitative estimates of the net effects of clouds and precipitation on aerosol particle numbers at the three sites, and discuss the implications of the findings for understanding the key drivers of surface-level aerosol observations.

2. Materials and Methods

2.1. Measurement Sites and Instrumentation

Three relatively pristine measurement sites were selected to analyze particle number size distribution (PNSD) data: Amazon Tall Tower Observatory (ATTO) in the Amazon Rainforest, SMEAR II (Station for Measuring Ecosystem–Atmosphere Relations) at Hyytiälä in the boreal forest (HYY), and Zeppelin Observatory in Ny-Ålesund (ZEP), which represents marine Arctic conditions (Hari et al., 2013; Andreae et al., 2015; C. Pöhlker et al., 2019; Platt et al., 2022). ATTO measurements cover the particle size range from 10 to 400 nm with a temporal resolution of 5 min, obtained from a scanning mobility particle sizer (SMPS) sampling from a mast at 60 m a.g.l above the forest canopy. The data spans from March 2014 to January 2019. HYY measurements cover the size distribution between 3 and 1,000 nm with a temporal resolution of 10 min, obtained from differential mobility particle sizer (DMPS) measurements sampled at the surface level. The data spans from January 2005 to December 2019. ZEP measurements cover the size range from 5 to 809 nm with a time resolution of 15 min, obtained from custom made DMPS. The data spans from January 2010 to December 2019. Only measurements between the 10–400 nm size range were used for multi-site comparison of PNSDs and PNSDs were resampled hourly.

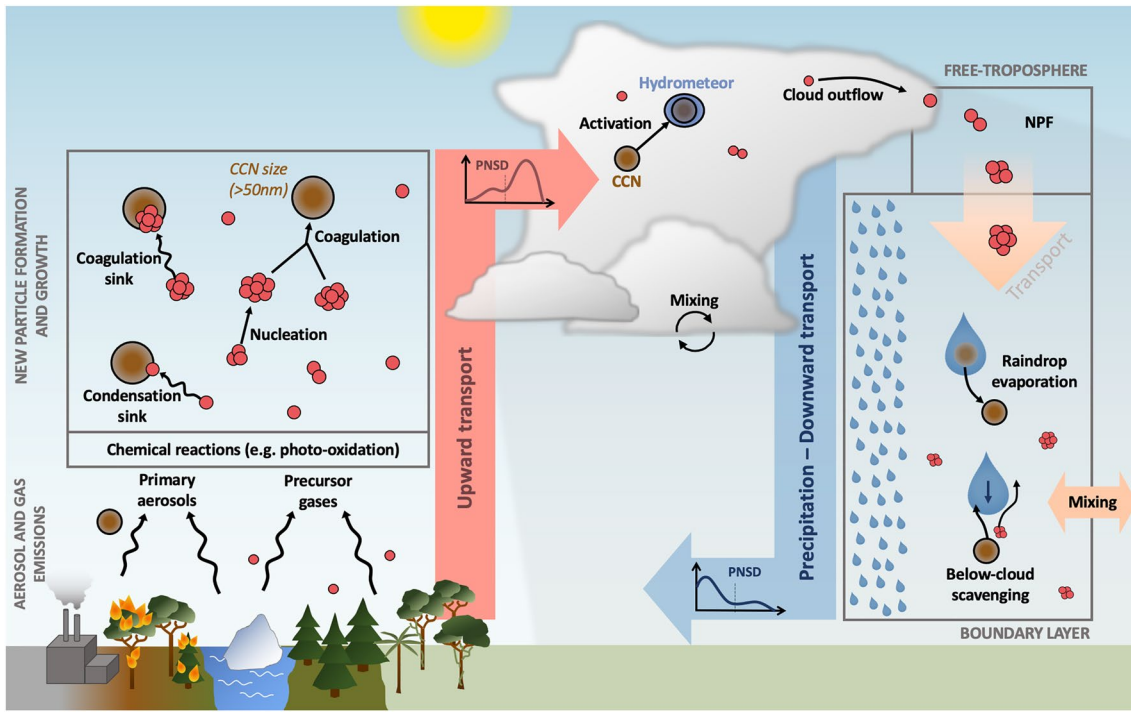

Figure 1. Conceptual model of the aerosol life cycle in the presence of precipitating clouds (not in scale). New particle formation (NPF) and consecutive growth depend mainly on the amount of solar radiation, precursor gases, condensation and coagulation sinks. Clouds scavenge CCN-sized particles that activate into hydrometeors or collide with cloud- and raindrops and ice crystals. Cloud hydrometeors and precipitation can evaporate and release cloud-processed particles and precursor gases. As precipitation is associated with downward transport, cloud-processed material and interstitial aerosol can be transported into the boundary layer and mix with pre-existing aerosol populations.

The atmospheric composition at ATTO varies with seasons due to different air mass origins, precipitation patterns, and biomass burning. Wet season (Feb-May) brings clear air masses that experience heavy precipitation and originate from the Northeast over unaltered rainforests, followed by a wet to dry transition period (Jun-Jul), then dry season (Aug-Nov) with strongly polluted air masses from Southeast due to less precipitation (Figure S1 in Supporting Information S1) and biomass burning. Concentrations of particles smaller than 50 nm are consistently low throughout the year at ATTO due to infrequent new particle formation events in BL.

The particle concentration and NPF frequency at HYY show strong seasonality (Dal Maso et al., 2005) (Spring: Mar-May, Summer: Jun-Aug, Autumn: Sep-Nov and winter: Dec-Feb). Accumulation mode is present all-year around, and Aitken mode is also usually present. Spring has the largest NPF frequency (Nieminen et al., 2014), and winter has less than 10% NPF days. Gas phase precursors from natural sources, especially monoterpenes (Hakola et al., 2012), dominate in HYY during spring and summer. NPF frequency is also influenced by air mass sources (Maso et al., 2007), with NPF events increasing when air comes from cleaner regions due to reductions in coagulation sink and CS.

ZEP follows the typical Arctic aerosol seasonality, characterized by (a) the Haze period (Mar-May) dominated by long-range transported accumulation mode particles, a phenomenon controlled by the reduced wet scavenging (Garrett et al., 2011); (b) summer (Jun-Aug), characterized by increased photochemistry and increased precipitation, frequent NPF events and an abundance of Aitken and ultrafine aerosol (Lee et al., 2020) and a decline in the accumulation mode (Tunved et al., 2013); (c) the slow-build up season (Oct-Feb), a transition phase, with the arrival of the polar night and diminishing photochemistry, combined with a low, but increasing accumulation mode and reduced precipitation, potentially allowing precursor gases to build-up (Boyer et al., 2023; Tunved et al., 2013).

2.2. Hysplit Backward Trajectories

The Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) was used to calculate the backward trajectories (BTs) of air masses at each station every hour during the PNSD measurement period.

Input meteorological data from the Global Data 5 Assimilation System (GDAS1, 1° × 1° resolution) (Pöhlker et al., 2019; Stein et al., 2015) were used for the BT calculations. The starting points of the BTs used for ATTO, HYY and ZEP, were respectively: (longitude = 59.0, latitude = −2.14, height = 60 m.a.g.l), (longitude = 24.3, latitude = 61.8, height = 100 m.a.g.l) and (longitude = 78.906, latitude = 11.888, height = 250 m.a.g.l). These altitudes are all located within BL.

We employed 4-day (96-hr) BTs, aiming to capture meteorological influences on aerosol dynamics. Direct effects, like wet scavenging and vertical transport, happen rapidly, while indirect effects, such as the decrease in CS and subsequent NPF, manifest over extended periods (see e.g., Dal Maso et al., 2002). The chosen 4-day span ensures the encapsulation of these processes while minimizing uncertainties in trajectory calculations for distant points from our monitoring stations.

2.3. Precipitation and Solar Radiation History Along Air Mass Transport

To assess aerosol exposure to precipitation and solar radiation during transport, we integrated data from hourly trajectory points leading up to the air mass arrival. In this study the term "precipitation" refers to the liquid equivalent of total precipitation. This data, along with solar radiation rates, was sourced from GDAS1. Recognizing the prominence of convective precipitation at ATTO and the reanalysis data limitations, we strategically collocated hourly trajectory points with the TRMM 3B42 V7 satellite product (0.25° × 0.25°, 3-hr resolution) (Michot et al., 2018). This method offers a more precise depiction of the precipitation in the tropics.

2.4. Correlation Analysis Along Backward Trajectories

Spearman correlation coefficients were calculated to evaluate the strength and direction of the monotonic relationship between particle number concentration and precipitation rate at given time steps along the air mass backward trajectories as a function of the particle diameter. p-value of 0.02 was used to assess the significance of the strength of the relationship.

2.5. Comparison of the Impact of Recent Precipitation Versus Solar Radiation on PNSDs

To investigate how precipitation and solar radiation (used here as a proxy for photochemically induced NPF) affect particle concentrations and potential particle sources, we examined variables accumulated 6 hours before particle concentration measurements. This approach limits indirect (longer-term) precipitation effects related to larger particle removal. Median concentrations were calculated for particle size ranges of 10–50 nm ($N_{<50}$), 50–400 nm ($N_{>50}$), and 100–400 nm ($N_{>100}$) across bins of accumulated precipitation and solar radiation variables in their 0–99th percentile range. Bin-median concentrations were then normalized by the median concentration of the entire data set to estimate deviations from typical concentrations when precipitation and solar radiation vary. Linear least-squares regression was used to determine the strength and significance of the relationships and compare parameters across locations. The impact of changes in precipitation and solar radiation on particle concentration was assessed by comparing the concentration at zero parameters to the concentration at the 99th percentile based on linear regression.

3. Results

3.1. Response of Aerosol Concentration to Precipitation History

Figures 2a–2c show the relationship between aerosol number size distribution and 96-hr accumulated precipitation along air mass back trajectories, and compares it with the median aerosol population at the three sites (see also Figure S1 in Supporting Information S1 for the transport patterns and source regions). A common feature across all sites and seasons is the well-known effect of precipitation and the associated clouds in scavenging the larger end of the aerosol size distribution. High values of accumulated precipitation led to anomalously low concentrations of accumulation mode particles. The threshold size for this effect is dependent on the environment (about 60 nm for ATTO, about 40 nm for HYY depending on season, and about 70 nm for ZEP, see also Figure S2 in Supporting Information S1), and influenced by a combination of processes that impact the aerosol size distribution and cloud formation, including the main particle sources, and atmospheric conditions such

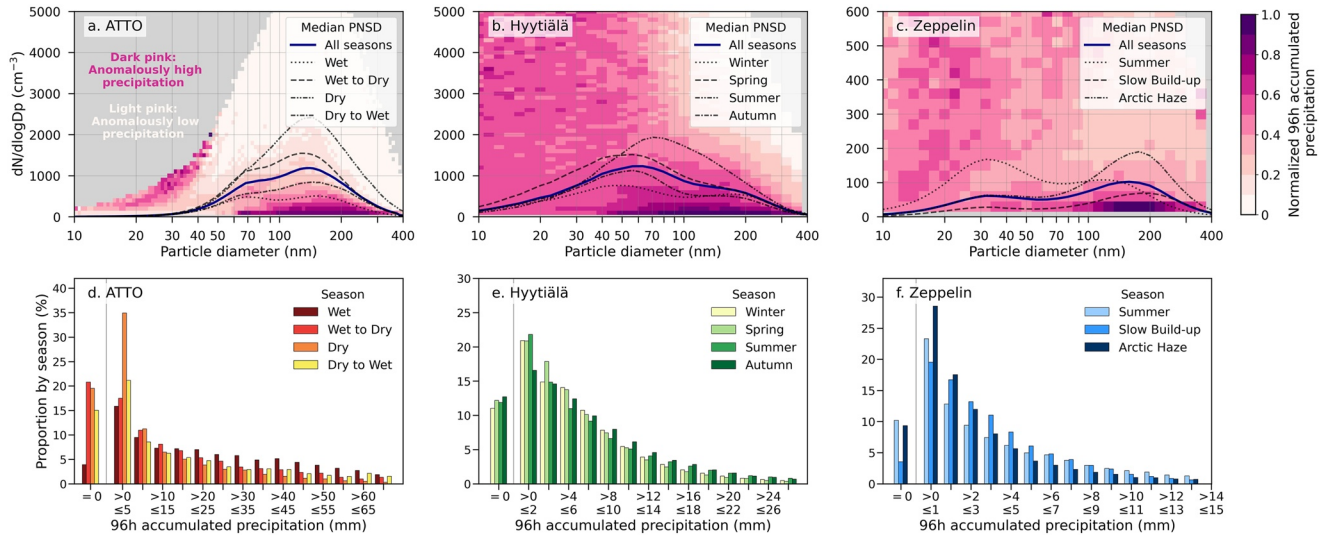

Figure 2. The relationship between aerosol particle number size distribution and 96 hr accumulated precipitation (a–c), and the distribution of 96 hr accumulated precipitation values for trajectories (d–f) arriving at each of the measurement sites (ATTO, HYY, ZEP). The blue solid lines represent the median size distributions over the whole year, the dotted, dashed and dash-dotted lines represent the different seasons (see legends for details, and Figures S2 and S3 in Supporting Information S1 for seasonality). The median accumulated precipitation values during the last 96 hr for a given size distribution value are depicted using the color scale, normalized to the maximum of each site. Note the different *x*-axis ranges for the sites, indicative of the unique precipitation regimes in these regions. Relationships with accumulated solar radiation can be found in Figures S9 and S10 in Supporting Information S1.

as updraft velocities and aerosol concentrations (Abdul-Razzak & Ghan, 2000; McFiggans et al., 2006). These conditions affect the ambient supersaturation, which in turn impacts cloud formation, precipitation intensity and type (Figures 2d–2f and Figure S3 in Supporting Information S1). Interestingly, for both dry and wet seasons at ATTO, and for the slow build-up season in ZEP, high accumulated precipitation values are linked to anomalously high abundances of nucleation and Aitken mode particles (< about 60 nm for ATTO, < about 50 nm for ZEP, Figure S2 in Supporting Information S1).

3.2. Direct and Indirect Effects of Precipitation on PNSD

The effects of recent precipitation differ from the effects of precipitation taking place further upwind from the measurement location (Figure 3). The signature of recent precipitation (12–24 hr before the observation site and closer) is clearly different from precipitation further away (>24 hr) for all three observation sites. The signature of the "older" precipitation is strikingly similar between the three environments: high rain intensities at >12–24 hr before air masses arrivals to the station correlate with a lower abundance of the accumulation mode, and a higher abundance of Aitken mode (about 40–70 nm and smaller) particles. The latter can be explained by the removal of larger particles (as seen in Figure 2), that leads to a reduction in CS and coagulation sink, and an increase in the lifetime of Aitken and nucleation mode particles. At all investigated sites, recent precipitation taking place in close vicinity (<12hr of air mass transport time) is associated with elevated particle concentrations. The sizes of the particles that correlate positively with recent precipitation vary between the sites, being smaller than about 50 nm for ATTO, between 150 and 400 nm for HYY, and smaller than about 30 nm for ZEP—for the latter, however, the positive correlation seems to be present only during the slow build-up season when precipitation is higher and CS is low.

The varying size ranges of particles associated with recent precipitation in the different environments indicate differences in composition, meteorological conditions, and sources and/or chemical ages of the particles. The positive correlation is strongest at ATTO, where Aitken mode particle source related to a downward transport of particles during rain events (or their precursors) from higher altitudes has been previously identified based on shorter-term observations (J. Wang et al., 2016; Andreae et al., 2018; Machado et al., 2021; Franco et al., 2022). For HYY, the observed positive correlation between rather large accumulation mode particles and recent precipitation can be linked to cloud-processed aerosol entrainment, as HYY often encounters air masses from SO_2-rich polluted areas undergoing cloud processing, as detailed by Isokääntä et al. (2022). For ZEP, while Tunved

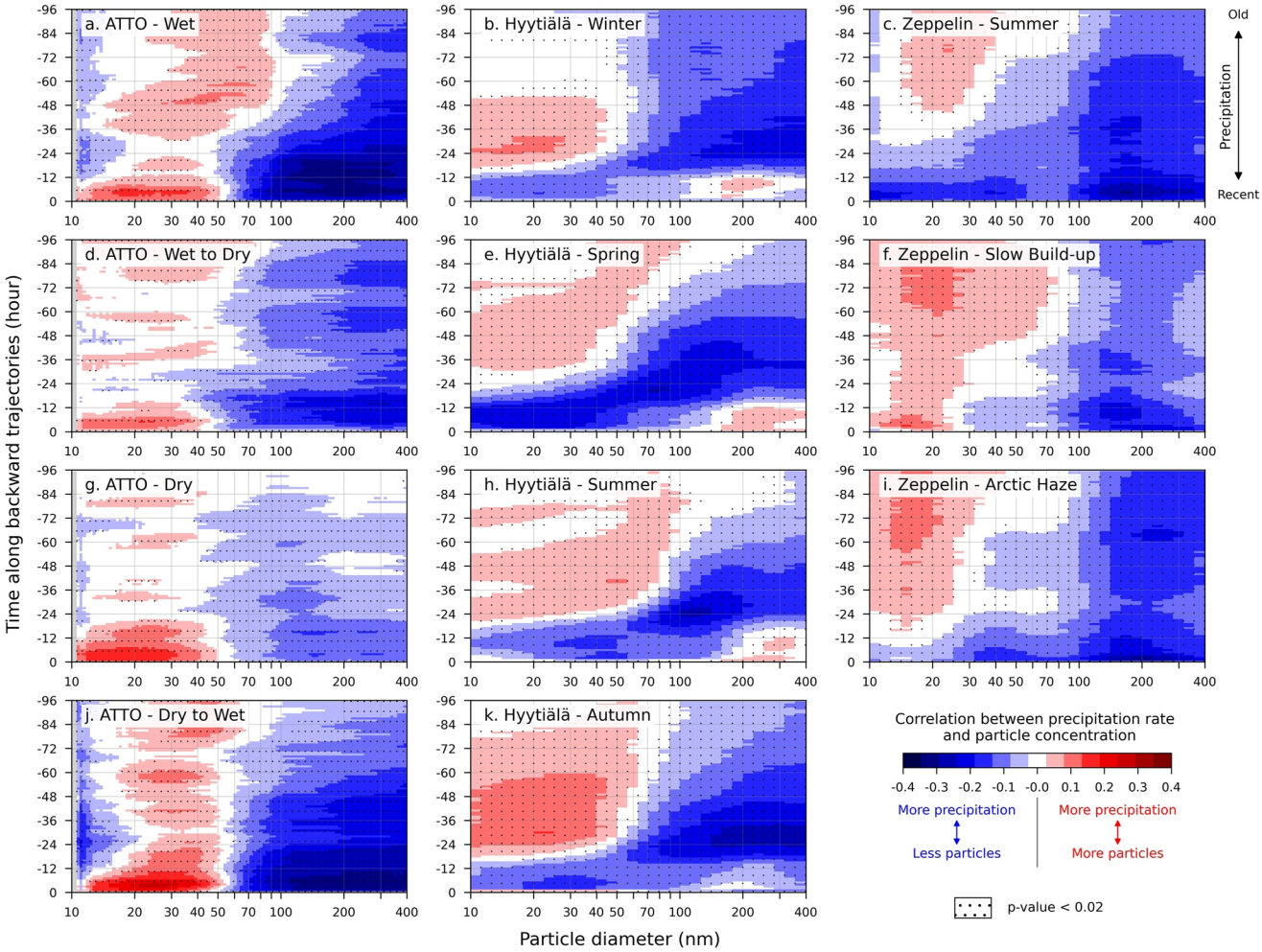

Figure 3. Correlation between precipitation intensity and particle number size distribution at ATTO (a, d, g, j), HYY (b, e, h, k) and ZEP (c, f, i). The color scale depicts the Spearman correlation coefficient between precipitation intensity at a given point of the 96 hr trajectories and the aerosol particle number size distribution. Stippling indicates statistical significance to the 98% level.

et al., 2013 identified the indirect source effect on smaller particles, to our knowledge, no study has reported the direct effect observed in our results. However, the importance of solar radiation for new particle formation and consecutive growth to Aitken mode particles has been identified and reported for HYY, ZEP and many other sites (Dal Maso, 2002; Größ et al., 2018; Pietikäinen et al., 2014), and it is well-established that photochemistry plays a crucial role in forming low-volatility gaseous precursors that can participate in NPF (Kulmala, 2003; McMurry & Friedlander, 1979). This begs to investigate the relative magnitude of the precipitation-related particle source and compare it with (a) scavenging of CCN-sized particles by precipitation; (b) photochemically-driven particle source—can the former be considered a non-negligible source of aerosol and CCN at some sites?

For $N_{>50}$ and $N_{>100}$ at ATTO and ZEP (b–c), concentrations in air masses experiencing precipitation are consistently lower as compared with concentrations without precipitation, and demonstrate a negative slope as precipitation increases. At HYY, if compared with no-precipitation conditions, the relative net removal effect of precipitation on $N_{>50}$ and $N_{>100}$ is weaker than at ATTO and ZEP (b–c). Interestingly, concentrations slightly increase under light precipitation compared to no-precipitation (first two points in panel c). These observed enhancements are robust, as the majority of data points in HYY is concentrated within this precipitation range (Figures S4 and S5 in Supporting Information S1). The effect is more pronounced under conditions of high recent light exposure (Wang, Liu, et al., 2022; Wang, Gordon, et al., 2022) and during spring (Figures S6 and S11 in Supporting Information S1). This might indicate a source of cloud-processed aerosol contributing to the number concentrations as reported in Isokääntä et al. (2022), occurring simultaneously with the known removal mechanisms. At ATTO,

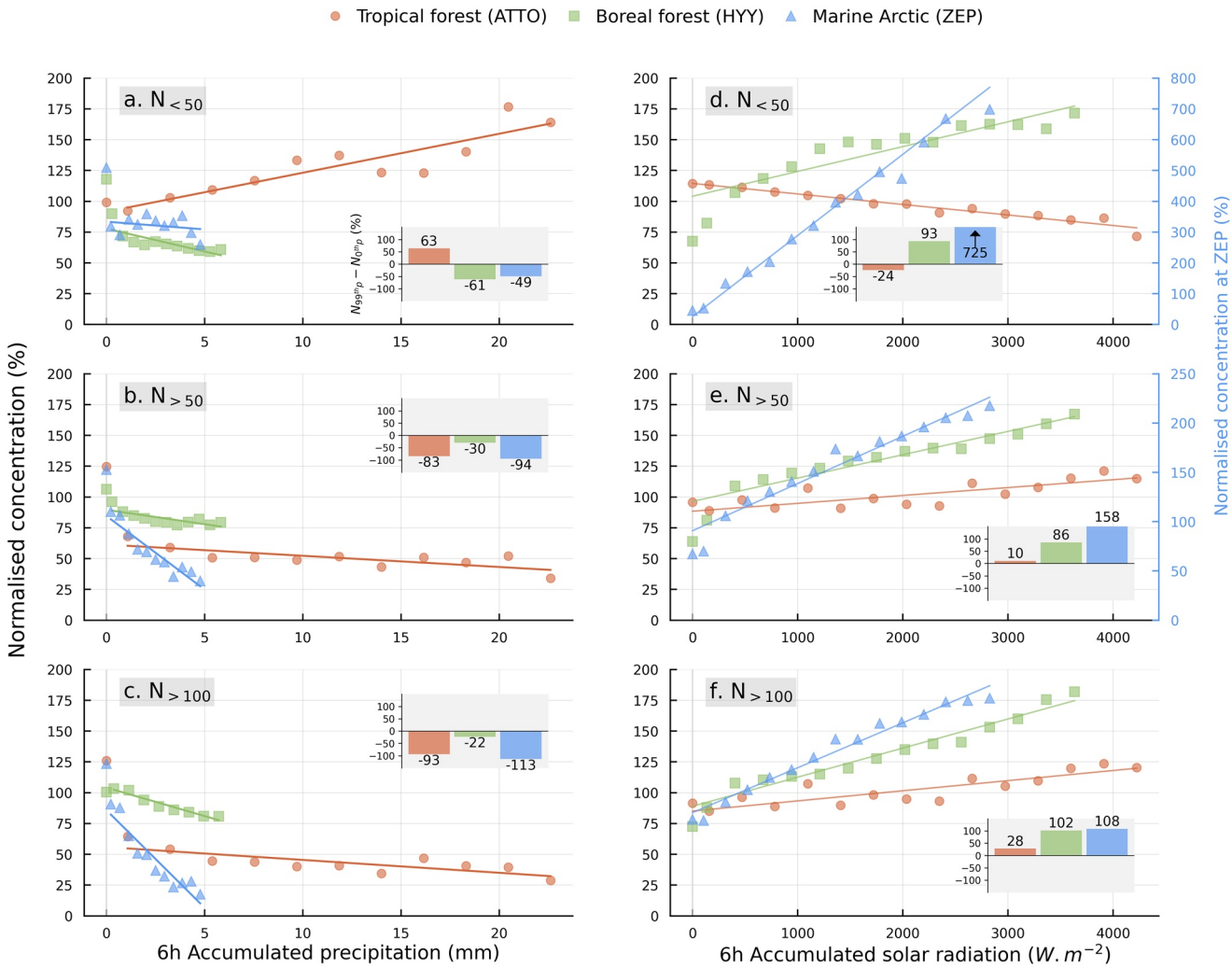

Figure 4. Normalized particle number concentrations of all data points, as a function of 6-hr accumulated (a–c) precipitation and (d–f) solar radiation, for particles smaller than 50 nm ($N_{<50}$), larger than 50 nm ($N_{>50}$) and larger than 100 nm ($N_{>100}$). Markers represent normalized median concentrations in bins of accumulated variables, and the bar plots show normalized concentration variabilities in the 0th–99th percentiles range of the 6-hr accumulated variables (difference between start and end points). Solid lines representing linear regressions, are included for trend visualization. For insights into seasonal relationships, consult Figures S6–S8 in Supporting Information S1. Furthermore, Figure S16 in Supporting Information S1 delves into variables accumulated over 96-hr.

the positive response to solar radiation is outweighed by the negative response to precipitation, but solar radiation response dominates at ZEP and HYY for $N_{>50}$ and $N_{>100}$ (panels b–c vs. e–f). The results are drastically different for $N_{<50}$ at ATTO, where consistent increase in $N_{<50}$ particles with increasing precipitation during last 6 hr explains about +60% variability (panel a, and Table S1). There is thus an important sub-50 nm particle source associated with precipitation. Consecutive condensation of vapors in boundary layer can then further enhance their contribution to CCN numbers (Stolzenburg et al., 2018; Zaveri et al., 2022; Zhou, 2002). The positive response of $N_{<50}$ to recent precipitation exceeds the negative response to solar radiation (see panel (a) versus (d)).

Contrasting dark versus light (for precipitation) and precipitating versus non-precipitating (for solar radiation) conditions illustrates that the simultaneous variation of precipitation and solar radiation cannot solely account for the aerosol responses observed in Figure 4 (see also Figures S11 and S12 in Supporting Information S1). Specifically, for ATTO $N_{<50}$ does not exhibit a significant or positive response to decreased CS, suggesting an immediate influence from the precipitation rather than the secondary effects (Figures S3–S15 in Supporting Information S1). For HYY and ZEP $N_{<50}$ shows a decline with recent accumulated precipitation. The dominant positive correlation with solar radiation at these sites underscores the significant role of photochemically-induced NPF, which is notably less prevalent at ATTO. Finally, while we have used a linear regression model to compare the magnitudes of the various sink and source effects, it should be noted that the dependencies arising from the plethora of physical

and chemical processes are not strictly linear. These dependencies can be further explored with robust theoretical knowledge of the key processes and additional, larger, observational data sets. While our results demonstrate the immense value of long-enough data series collected in situ from various environments, even more data is critical for more robust statistics given the large scatter in the data (see Figures S4 and S5 in Supporting Information S1).

4. Discussion

In this study, we have shown that besides the well-known removal effect of aerosol particles by clouds and precipitation, there are also signs of a direct source of boundary layer particles associated with recent (<12–24 hr) accumulated precipitation. A positive correlation between particles of a specific size range and precipitation in the air masses arriving at the measurement station was present at all three studied sites (ATTO, HYY and ZEP), and the effect was most pronounced for sub-50 nm particles at ATTO. Our method examines the effects of precipitation based on their distance along the trajectory, emphasizing the importance of the timing of these events. Traditional approach of focusing on accumulated precipitation might inadvertently blend effects where precipitation serves as both a source and a sink for aerosol concentrations. While a shorter-term and case-based analysis of the links between precipitation, downdrafts and particle numbers have been reported before (Franco et al., 2022; Machado et al., 2021; Wang et al., 2016), to our knowledge this is the first time the frequency and relative magnitude of such precipitation-related particle concentration enhancement has been presented. By studying the impact of precipitation along air mass back trajectories, we were able to subtract the indirect effect of the condensation sink reduction by precipitation on the number of small particles. The magnitude of the precipitation-related particle source is generally small when compared with the removal effect of CCN-sized particles, but not insignificant. For instance, at ATTO, the removal of accumulation particles and the addition of sub-50 nm particles with increasing recent precipitation is comparable, and in HYY, the precipitation-related particle source seems to compensate for a large fraction of the removal effect of CCN-sized particles in boundary layer.

Our results have implications for example, (a) identifying a potential new replenishing source of CCN observed at the ground-level, especially in the tropics; (b) the interpretation of in situ observational data when it comes to the source and sink processes driving the observed atmospheric aerosol number size distributions; and (c) the representation of aerosol and precursor gas scavenging, processing and recycling by clouds and precipitation within atmospheric models. For the latter, our results suggest that the scavenging and cloud-processing parametrizations in models should ideally account also for the replenishment of boundary layer aerosol population due to cloud processing and precipitation. Since precipitation patterns are predicted to change in the future (Trenberth, 2011) the importance and regional distribution of this mechanism is also likely to change, hence potentially changing the atmospheric aerosol populations and aerosol-cloud interactions in the future. The exact theoretical formulation and prediction of such process should, however, be based on a process-level understanding of the phenomenon. The presented data and analysis does not allow for an ambiguous separation between numerous processes and effects, for example, distinguishing impacts of clouds versus precipitation, phase of precipitation, downward transport of particles formed at higher altitudes (Andreae et al., 2018; Wang et al., 2016), "recycled" material originating from the evaporation of processed cloud hydrometeors (Braga et al., 2022; Isokääntä et al., 2022; Krejci, 2003), in situ NPF and processing through atmospheric chemistry in the boundary layer associated with enhanced concentrations of reactive species during rain events (Gerken et al., 2016), raindrop impaction with surfaces (Joung et al., 2017; Wang et al., 2016), and ion-induced processes (Junninen et al., 2008; Kolarž et al., 2012; Parts et al., 2007; Wimmer et al., 2018). We have demonstrated the value of using long-term observational data sets of aerosol size distributions, especially when coupled to meteorological data and remote sensing. To constrain the key processes and their importance within the full loop of gas-aerosol-cloud-precipitation interactions, more targeted observations of vertical motion in the atmosphere, type of precipitation, vertical profiles and conditions close to clouds are needed. The observations must be coupled with detailed cloud-resolving models that account for the relevant gas, aerosol and cloud processes on relevant temporal and spatial scales.

Data Availability Statement

The observational and model data used for the statistical analyses in the study are available through the following link: https://doi.org/10.5281/zenodo.7907474 (Khadir, 2023a). The Python analysis and visualisation codes are compiled in a Jupyter Notebook, accessible through the following link: https://doi.org/10.5281/zenodo.8305875 (Khadir, 2023b).

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Erratum

In the originally published version of this article, the third and fourth paragraphs of Section 3.2 were published in the reverse of their correct order, owing to a typesetting error. The error has been corrected, and this may be considered the authoritative version of record.