Seasonal Variations of Atmospheric Aerosol Particles Focused on Cloud Condensation Nuclei and Ice Nucleating Particles from Ground-Based Observations in Tsukuba, Japan

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Abstract

Since March 2012, multi-year ground-based observation of atmospheric aerosol particles has been carried out in Tsukuba, Japan to characterize the aerosol particle number concentrations (NCs), air mass origin relevance, and specifically, their cloud condensation nuclei (CCN) and ice nucleating particle (INP) characteristics. The CCN NCs at any water supersaturation (SS) exhibit strong seasonality, being higher in winter and lower in summer; this pattern is similar in the polluted urban environment in East Asia and contrary to that in the Pacific coastal region. The hygroscopicity (κ) is generally high in early autumn and low in early summer, likely due to the seasonal difference of synoptic-scale systems. In contrast, the INP NCs and ice nucleation active surface site density (ns) at defined temperature (~15 to ~35°C) and SS (0%–5%) lack clear seasonal influence. The average INP NCs and ns in this study were comparable at warmer temperatures and approximately one order of magnitude lower at colder temperatures, compared with those in other urban locations under limited dust impact. Moreover, the ns values were one to four orders of magnitude lower and exhibited weaker temperature dependence than previous parameterizations on mineral dust particles.

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1. Introduction

Atmospheric aerosol particles (APs) can act as cloud condensation nuclei (CCN) and/or ice nucleating particles (INPs) under suitable conditions. These particles significantly impact climate change and weather forecasting by modulating cloud albedo (Twomey 1974) and by modulating precipitation efficiency, spatial distribution and lifetime of cloud systems (Albrecht 1989), causing regional or global energy and hydrological cycles alterations. To assess the influence of APs on climate and precipitation, it is necessary to comprehend the temporal and regional behavior of the APs with CCN and INP characteristics. As air in the convective boundary layer flows into the low-level or deep convective clouds, the APs involved are vital to cloud formation. Thus, understanding the behavior of APs, especially in the upper convective boundary layer, is crucial.

In general, continuous in-situ observations of APs and their physicochemical properties (including CCN and INP characteristics) in the upper convective boundary layer is challenging. Although aircraft observations produce vertical profiles, the data are commonly discontinuous. However, a previous study suggested that AP data, including the CCN, from aircraft measurements in the boundary layer were comparable to ground-based measurements with insignificant nearby aerosol sources (Yamashita et al. 2015).

To estimate the impact of APs on climate change and weather forecasting using numerical models, including aerosol physicochemical properties as prognostic variables, determining the CCN and INP characteristics of aerosols (e.g., dust, sea salt, sulfate, black carbon, organic carbon, etc.) is imperative. Moreover, obtaining the long-term observation data for APs physicochemical properties, including the CCN and INP characteristics, is relevant for validating and improving the numerical modeling of aerosol-cloud-precipitation interactions.

Many studies are available on long-term observations of the physicochemical properties of APs and CCN activation spectra. However, as far as we know, no study focuses on the seasonal and interannual variabilities of APs including the CCN and INP properties. A possible reason for this absence of studies is that online measurement techniques (e.g., the continuous-flow diffusion-chamber type; CFDC-type) and offline approaches (such as filter sampling) for deriving INP properties are not fully automated. Based on the design by Rogers et al. (1988), MRI built the CFDC-type ice nucleus counter (INC) (Saito et al. 2011), that introduced the automation of ice coating of cylinder walls and measuring the INP temperature and supersaturation spectra. This device is suitable for long-term automatic INP characteristics measurements.

In this study, we analyze multi-year APs, CCN, and INP datasets measured in Tsukuba, Japan. The aims of this study are: to characterize seasonal and interannual variations in the APs, CCN, and INP properties data; to investigate the relationships between these properties; to assess their dependence on the origin of air masses in the area; and to establish the similarities and differences between previous studies and our results.

2. Methods

2.1 Site and instrumentation

Multi-year ground-based continuous observation has been conducted since March 2012 on the MRI campus in Tsukuba (hereafter TKB, 36.056°N, 140.125°E, 24 m above mean sea level) which is located 50–60 km north-east of Tokyo. The location, schematic diagram, and configuration of the monitoring system are shown in Fig. S1. The APs in ambient outside air were sampled using a PM10 inlet (11 m above ground level) at a flow rate of 120 L min⁻¹ via a stainless steel line with an internal diameter of 7.0 cm. The branching method, which transfers inlet sample air to each instrument through an L-shaped stainless steel tube, was applied to satisfy the isokinetic sampling condition. The air was dried using a diffusion dryer filled with silica gel (Yamashita et al. 2015).
INC measurements were derived as described in Kuo et al. (2019). (e.g., Connolly et al. 2009) density ($n_s$ at $−35°C$ and INP NCs and ice nucleation active surface site (INAS) data for each supersaturation scanning at $−15$, $−20$, $−25$, $−30$, and $−35°C$ were processed for the duration at each SS. We processed INC resolution. The CCNC data for the stepwise SS setting column (SD) from each instrument were analyzed using the finest time resolution of measurements by each instrument are presented in Table S1. The CCNC was operated at a constant water supersaturation (SS) of 0.5% for one column, and cyclically at 0.1, 0.2, 0.5, 0.8, and 1.0% SS over 30 min for the other. The operation procedure for automatic INC measurements are described in Saito et al. (2011). Ice nucleation spectra were obtained at five temperatures from $−15$ to $−35°C$ (5°C intervals) and in the humidity range of an ice saturation to SS of $−15%$. Here, we utilized INC data limited to an SS range of 0%−5%, representing exclusively ice nucleation via the condensation/immersion freezing mode. The entire procedures required approximately 4 h and was typically performed two times from 9 AM to 5 PM on weekdays. Double impactors with 50% cut-off diameter of 1 μm (~1 L min$^{-1}$ flow rate) were installed upstream of the INC inlet for APs in the supermicron size range removal.

In this study, data from March 2012 to December 2018 were utilized, although the INC and POPC data start from April 2013 and April 2015, respectively. The total sampling periods of SMPS, standard OPC, POPC, CCNC, and INC are 48,720 (2,091), 50,235, 39,360 (2,091), and 3,087 (983) hours (the number of days), respectively.

### 2.2 Data analyses

For the data processing of SMPS, standard OPC, POPC, and CCNC, the number concentration (NC) and size distribution (SD) from each instrument were analyzed using the finest time resolution. The CCNC data for the stepwise SS setting column were processed for the duration at each SS. We processed INC data for each supersaturation scanning at $−15$, $−20$, $−25$, $−30$, and $−35°C$, and INP NCs and ice nucleation active surface site (INAS) (e.g., Connolly et al. 2009) density ($n_s$) measured in SS range of 0%−5% are used in this study. After estimating the critical dry diameter from the measured SD of the APs and the CCN NC at 0%−5% are used in this study. After estimating the critical dry diameter from the measured SD of the APs and the CCN NC at 0%−5%, representing exclusively ice nucleation via the condensation/immersion freezing mode. The entire procedures required approximately 4 h and was typically performed two times from 9 AM to 5 PM on weekdays. Double impactors with 50% cut-off diameter of 1 μm (~1 L min$^{-1}$ flow rate) were installed upstream of the INC inlet for APs in the supermicron size range removal.

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### 3. Results

#### 3.1 Overall statistics on the NCs of the APs, CCN, and INP, and origin of air masses

The time-series of monthly-averaged NCs of the APs for several size ranges are shown in Fig. 1, with the overall statistics of the NCs presented in Table S2. The CCN NCs at 0.5% SS are within the NCs range between those for APs > 0.1 and > 0.3 μm, while the INP NCs at $−25°C$ are comparable to those for APs > 5 μm. The latter does not indicate that APs > 5 μm were mostly INPs.

Figure 2a shows the monthly-averaged relative distribution of the origin of air masses from the three-day backward trajectories, and the 11 regions produced are displayed in Fig. 2b. Evidently, air masses are dominant in the JS and NW regions between June-August and October-March, respectively. Overall, the mean distribution is ~40% in the JN and JS regions surrounding Japan, ~30% in the NW, ~10% in the CN, and ~10% in the Pacific Ocean regions, ME and SE.

#### 3.2 CCN NCs and κ seasonal variations

Monthly variations of CCN NCs and κ at different SSs are displayed in Fig. 3. Considering the median values for all years, the CCN NCs are generally high in winter (November-February) and low in summer (June-September excluding July) at all SSs compared to other seasons. Their interannual variations are minor, whereas monthly fluctuations are significant, as depicted by the error bars in Figs. 3a−3e. In contrast, the κ is characterized by sizable interannual variability and high monthly fluctuations, as shown in Figs. 3f−3j. The κ values were generally high in August-September and low in May-June and in December. Other local maxima emerge at SSs of 0.5% and 0.8% in January.

The frequency distribution of the κ manifests the difference between June and September, displayed in Fig. 4. The distribution shifts to more hygroscopic particles in September, especially at lower SSs, with a significant contribution from κ > 0.5.
3.3 INP NCs and \( n_s \) seasonal variations

The monthly INP NCs and \( n_s \) variations are shown in Fig. 5, with both parameters characterized by considerable interannual variability and large monthly fluctuations. In fact, the months showing maximum and minimum values differ as the temperature varies. According to the arithmetic mean for all years, the INP NCs are highest in June and lowest in February, although local maxima are present in September \((T = −15°C)\), April \((T = −25°C)\), and December \((T = −35°C)\), while local minima emerge in March and November \((T = −15°C)\) and in October \((T = −25°C)\). Similarly, the \( n_s \) reaches its peak value in September and least value in February, with local maximum in June \((T = −30°C)\) and December \((T = −35°C)\).

4. Discussion

4.1 Comparison with seasonal variations of CCN NCs and hygroscopicity in other regions

Seasonal variations of CCN NCs and \( \kappa \) parameters with APs physicochemical properties have been reported at regionally representative locations such as high alpine site, Amazon rain forest, boreal forest, coastal, rural, or urban areas (e.g., Schmale et al. 2018). In Schmale et al. (2018), the Noto Peninsula site (NOT; Iwamoto et al. 2016) in the Pacific coastal area and the Seoul site (SEL; Kim et al. 2014) in urban environment are included from the East Asia region. For TKB, whose location is categorized by a rural background, the APs are controlled by two major sources: long-range transport by continental or maritime air masses and local pollution, although the latter influence may be transient. The CCN NCs at an SS of 0.2% for TKB are comparable to those for NOT and about half those for SEL. For NOT, the CCN NCs are highest in spring and lowest in winter, a trend similar to that of another coastal site near Taipei in Taiwan (TPE; Cheung et al. 2020). For TKB and at SEL, the seasonal variations exhibit similar trends, with the highest values in winter and lowest in autumn at all SSs. This behavior is attributed primarily to the closeness of the inversion layer to the ground in winter and secondarily to local pollution. For SEL, the CCN NCs for all SSs display strong negative correlations with the planetary boundary layer height, without seasonal dependence (Kim et al. 2014). A coastal site such as NOT is considered not directly influenced by the inversion layer height in winter, but likely by a convectively mixed boundary layer depth.

According to Andreae and Rosenfeld (2008), a \( \kappa \) value of 0.3 reflects polluted continental environments. The median \( \kappa \) values in this study ranged from 0.08 (1.0% SS) to 0.38 (0.1% SS), and these are significantly lower than those for corresponding SSs for the coastal TPE (from 0.18 at 0.8% to 0.56 at 0.12%). The mean \( \kappa \) value in this study of 0.17 at 0.5% SS falls between the categories for rural (0.48) and urban (0.1) areas (Schmale et al. 2018). The lower \( \kappa \) values for TKB relative to the average continental environment value suggest a comparable or higher contribution of organics compared to other sites in East Asia (e.g., Meng et al. 2014; Iwamoto et al. 2016; Cheung et al. 2020).

As shown in Fig. 2, the air masses display no clear difference in origin in June and September. However, in early summer, TKB is typically located north of the Baiu front, whereas in early autumn, the autumnal rain front exerts a major influence over the main island of Japan, with tropical cyclones coming from the south. Therefore, continental polluted air masses may contribute to the lower \( \kappa \) values in June, while maritime polluted air masses from the Pacific become dominant and contribute to the higher \( \kappa \) values in September. The air masses are characterized by the monthly-average SDs of the APs measured by the SMPS for both months displayed in Fig. S2, with fewer particles in September than in June.

4.2 Comparison of INP NCs and \( n_s \) seasonal variations with other regions

There are a limited number of studies on the seasonal variations of measured INP. Regarding the free troposphere (FT) measurements, Conen et al. (2015) reported INP NCs at ~8°C using an offline technique throughout the year, and the seasonal trends for the high alpine station, Jungfraujoch (JFJ), in Switzerland, were attributed to boundary layer influence. In contrast, Lacher et al. (2018) presented background FT INP NCs at ~31 to ~32°C from an online technique involving nine field campaigns at JFJ, with the influences of the local boundary layer, dust and marine events excluded. Despite higher background FT concentrations in some seasons due to increased dust and marine events, no seasonal trend is apparent. The median and mean INP NCs measured at ~30°C in this study are comparable to those for the background FT.

Notably, most INP data involve studies in North America and Europe without locations strongly affected by air pollution (Petters and Wright 2015; Mason et al. 2016). Chen et al. (2018) investigated the INP NCs down to ~25°C for urban air pollution in Beijing, China, in winter, concluding no direct air pollution impact on these for the temperature range investigated. The average INP NCs in this study are comparable to the values of \( 10^{-10} \) L⁻¹ for temperatures ranging from ~10 to ~25°C reported in Chen et al. (2018).

Comparison of the INP NCs to the AP NCs is displayed in Fig. 6, along with the parameterization of “global” average INPs proposed by DeMott et al. (2010). No correlations between the INP NCs and the AP NCs were found in any month, and neither
Fig. 3. Monthly variations of (a–e) CCN NCs and (f–j) \( \kappa \) at SS values of: 0.1% (a, f), 0.2% (b, g), 0.5% (c, h), 0.8% (d, i), and 1.0% (e, j). The filled squares and red open circles denote the monthly median values for each year and for all years, respectively. The error bars cover the 10th and 90th percentiles for each year.
were the NCs nor surface areas of AP larger than 0.5 μm classified as aspherical particles (including mineral dust) by POPC. Despite the significant scatter in Figs. 6a–6c, the DeMott parameterization sometimes overpredicts the measured INP by an order of magnitude in winter and spring whereas the average prediction is satisfactory or involves underprediction by an order of magnitude in autumn.

The $n_s$ calculated from the five INC operating temperatures for all data in this study are shown in Fig. 7, along with the parameterizations of dust and/or surrogate particles by Niemand et al. (2012), Atkinson et al. (2013), and Broadly et al. (2012). The $n_s$ values range from one to four orders of magnitude lower, with weaker temperature dependence than those from the previous parameterizations. One of the main reasons for the relatively low $n_s$ values is the difference in particle populations; the AP in this study should be largely composed of various types of aerosols with lower ice-nucleating abilities than those of dust particles. For instance, sea spray aerosols may partly contribute to the low $n_s$ values in this study, as indicated in DeMott et al. (2016).

We also compared of the $n_s$ values in this study to those of several field campaigns at other locations from previous studies (not shown). The $n_s$ values at −30°C are approximately 1.5 and 2.5 orders of magnitude lower than those from JFJ under the background FT and Saharan dust events conditions, respectively (Lacher et al. 2018). Gong et al. (2019) reported the $n_s$ values between −15 and −20°C from a spring field campaign in Cyprus, in the Mediterranean region, were mainly influenced by the long-range transport of anthropogenic emissions from Europe and the Middle East. The reported $n_s$ values are comparable to those in this study, likely because of the insignificant dust-impact on the sampling data.

5. Conclusions

To characterize the CCN and INP characteristics of atmospheric APs, we conducted the ground-based observations in Tsukuba, Japan, representing a rural polluted environment in East Asia. From our multi-year datasets, the CCN NCs exhibit strong seasonality, with high values in winter and low values in summer, consistent with the results for urban polluted sites such as SEL, and in contrast to those of Pacific coastal sites such as NOT and TPE. Although the $κ$ displayed no clear seasonal trend, high values were common in early autumn and low values in early summer. This pattern is attributed to the seasonal difference of the synoptic-scale systems around the study site. The mean $κ$ of 0.17 at 0.5% SS was lower than the representative value of 0.3 for polluted continental environments, suggesting a higher contribution of weakly hygroscopic materials such as organics. The INP NCs and $n_s$ data also lack clear seasonality, although high values were frequent in June and September, with low values common in February. The mean $n_s$ values were comparable to or approximately an order of magnitude lower than data for urban locations when dust-impact periods were excluded.

In this study we presented the seasonal and interannual variations of the CCN and INP under complex influences of rural and urban polluted environments associated with continental and maritime air masses. The chemical composition and mixing state of APs from concurrent measurements, however, were not involved in this study. These will be addressed in future studies to clarify parameters of APs that are linked with their CCN and INP characteristics.

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Supplements

The supplementary information includes the size ranges and finest time resolutions of measurements by each instrument (Table...
Fig. 5. Monthly variations of (a–e) INP NCs and (f–j) $n_i$ at the following temperatures: $-15^\circ$C (a, f), $-20^\circ$C (b, g), $-25^\circ$C (c, h), $-30^\circ$C (d, i), and $-35^\circ$C (e, j). The INP parameters herein only cover values for SS of 0%–5%. Monthly median values are plotted as in Fig. 3. Note that certain median values are not shown as they were below the limit of quantification. The blue asterisks and open triangles represent the monthly arithmetic mean and maximum values for all years, respectively.
S1), the summarized overall statistics on the monthly average values of APs, CCN and INP (Table. S2), the location and configuration of the MRI’s aerosol monitoring system (Fig. S1), and the seasonal variations of averaged SDs of APs measured by the SMPS (Fig. S2).

Fig. 6. Relationship between the INP NCs and the parameters of APs > 0.5 μm by the POPC in February (a, d), May (b, e), and September (c, f) at −25°C for the following: (upper) NCs of all particles and NCs of aspherical particles, which are compared with the parameterization of DeMott et al (2010); (lower) total surface area of aspherical particles and its ratio to that of all particles.

Fig. 7. Plot comparing the $n_s$ values from previous studies on dust parameterization (solid lines) and the measured ranges of APs using MRI’s CFDC-type INC. The red asterisks and open triangles denote the median and maximum values at five temperatures (−15, −20, −25, −30, and −35°C), respectively, with error bars of 10th and 90th percentiles.

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