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## RESEARCH ARTICLE

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## Seasonal Trends of Atmospheric Ice Nucleating Particles Over Tokyo



### Key Points:

- Characteristics of ice nucleating particle (INP) number concentrations over Tokyo were quantified by continuous year-round measurements
- The INP spectra at temperatures below  $-20^{\circ}\text{C}$  did not show clear seasonality but were rapidly enhanced during an Asian dust event
- The INP spectra at temperatures above  $-20^{\circ}\text{C}$  appeared to show seasonal variations and tended to be enhanced in warm/wet seasons

### Supporting Information:

- Supporting Information S1

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**Abstract** Ice nucleating particles (INPs) originating from Asia are expected to have large impacts on aerosol-cloud-precipitation interactions on local, regional, and global scales. However, their seasonal variability is poorly understood. Here, we present a year-round record of atmospheric INPs measured on Tokyo Skytree, which is the world's tallest broadcasting tower located in the Tokyo Metropolitan area. The INP number concentrations showed relatively small variations in the temperature regime below  $-20^{\circ}\text{C}$ , whereas the values were episodically enhanced by long-range transported Asian dusts. On the other hand, the INP spectra in the temperature regime warmer than  $-20^{\circ}\text{C}$  exhibited measurable seasonal variations. Notably, the INP number concentrations in the temperature regime between  $-15^{\circ}\text{C}$  and  $-10^{\circ}\text{C}$  tended to indicate higher values in warm/wet seasons and lower values in cold/dry seasons. Our results suggest that Asian dust events and seasonal variations in certain particles of biological origin linked to local/regional meteorology might influence the seasonal trends of the INP spectra over the Tokyo Metropolitan area.

## 1. Introduction

Mixed-phase clouds consisting of both ice crystals and supercooled liquid droplets can occur in a wide variety of locations from low to high latitudes in all seasons. Since the ice phase is thermodynamically more stable than the supercooled liquid phase at temperatures below  $0^{\circ}\text{C}$ , it is expected that ice crystals in mixed-phase clouds can grow in size more efficiently than ambient supercooled liquid droplets at the expense of the droplets, thus largely influencing the cloud properties and lifetime (Korolev et al., 2017). Despite their importance, large uncertainties remain in the representation of ice crystal formation processes (e.g., heterogeneous ice nucleation and secondary ice production) in mixed-phase clouds in global climate models (Korolev et al., 2017). As for heterogeneous ice nucleation processes in mixed-phase clouds, it is thought that immersion freezing (ice nucleation by particles immersed in supercooled water) is the dominant heterogeneous ice nucleation process and that only a subset of insoluble particles in the atmosphere can serve as ice nucleating particles (INPs) in the immersion mode (Kanji et al., 2017; Murray et al., 2012).

Aerosol particles in Asian outflow are thought to be one of possible major sources of the global INP load and have large impacts on aerosol-cloud-precipitation interactions on local, regional, and global scales. For example, aircraft measurements around the Japan Islands by Asano et al. (2002) and Gaynet et al. (2002) revealed the occurrence of boundary-layer mixed-phase clouds at temperatures above  $-15^{\circ}\text{C}$  during a wintertime cold-air outbreak event from the Asian continent (Asano et al., 2002; Gaynet et al., 2002), indicating the existence of certain efficient INPs in Asian continental outflow. Ground-based in situ and/or lidar measurements in China (Bi et al., 2019), Japan (Isono et al., 1959; Murayama et al., 2001; Sakai et al., 2004), and the United States (Ault et al., 2011; Creamean et al., 2013; Pratt et al., 2009; Sassen, 2002) have suggested the importance of long-range transported dust particles and/or primary biological aerosol particles (PBAPs) such as fungal spores and bacteria from the Asian continent for heterogeneous ice nucleation and subsequent precipitation processes. On the other hand, recent field studies indicated that the release of urban aerosol particles from anthropogenic sources in Asia might not necessarily induce enhancement of atmo-

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spheric INPs (Bi et al., 2019; Chen et al., 2018; Yadav et al., 2019). However, since available atmospheric INP data are based on intensive field measurements at a specific area for relatively short periods (days to months), the spatial and seasonal variations of atmospheric INPs remain poorly understood.

Recently, we started continuous in situ measurements of clouds and aerosols on Tokyo Skytree, which is the world's tallest television and radio broadcasting tower located in the Tokyo Metropolitan area in Japan (Misumi et al., 2018; Uetake et al., 2019). It is likely that low-level stratiform clouds and aerosols measured over Tokyo are influenced by air masses originating from the Asian continental sources, as well as local/regional terrestrial and oceanic sources, especially from autumn to spring (Misumi et al., 2018; Uetake et al., 2019). It may also be important to note that the occurrence of summertime heavy rain events linked to complex urban land use-weather-climate feedbacks has been reported in large coastal cities like Tokyo (Misumi et al., 2019; Shepherd, 2005). In this regard, recent work suggests that dusts from local sources might serve as efficient INPs or giant cloud condensation nuclei during the formation of urban-induced heavy rain in Tokyo (Uchiyama et al., 2019). For these reasons, we consider that there is a need for a better understanding of INPs over Tokyo throughout the year. Here, we report on a year-round study of INPs active in the immersion mode (i.e., under conditions relevant for mixed-phase cloud formation) based on continuous measurements on Tokyo Skytree for the period from August 2016 to July 2017.

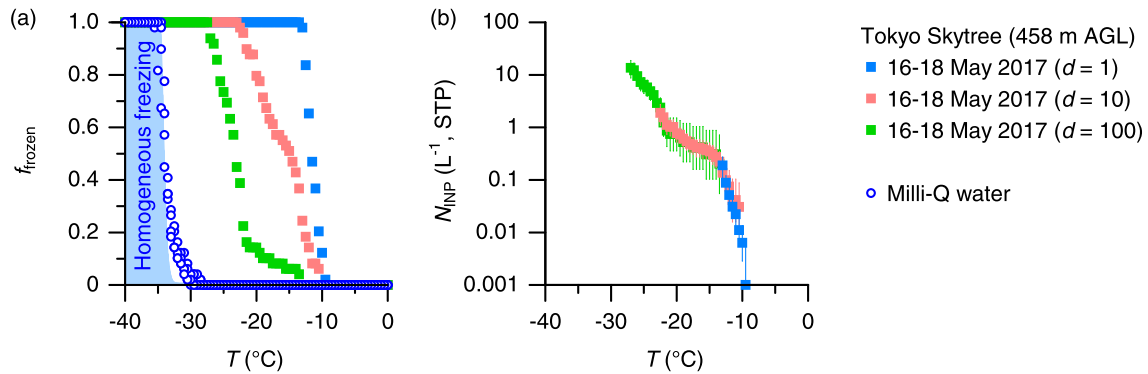
## 2. Materials and Methods

### 2.1. Aerosol Measurements on Tokyo Skytree

Continuous year-round measurements of clouds and aerosols over Tokyo have been conducted at a monitoring site located on the 458-m level floor of Tokyo Skytree (35.71°N, 139.81°E) in Sumida, Tokyo, Japan (Misumi et al., 2018; Uetake et al., 2019). Ambient aerosol particles were carried to instruments installed in a monitoring room through a short length (~1.5 m) of 1/4" conductive silicon tubes. In supporting information Figure S1, we summarize the particle transmission efficiency of the sampling tubing calculated based on theoretical formulae (Kulkarni et al., 2011; Yoshida et al., 2018). Here, we assume that aerosol particles are spherical particles with a density of 1–3 g cm<sup>-3</sup>. The calculations indicate that while most particles with diameters of 1 μm or smaller could pass through the tubing, measurable particle losses would occur in the supermicron size range (e.g., the transmission efficiency for particles with a diameter of 5 μm is expected to be ~60–90%), and most particles larger than 20 μm could not reach the instruments. Aerosol sampling for measuring INPs was conducted continuously and sequentially at 3-day (72-hour) intervals using a 10-line Global Sampler (GS-10N, Tokyo Dylec Corp.). The first sampling was started at 00:00 JST (Japan Standard Time, UTC + 9 hours) on 1 August 2016. Ambient aerosol samples were collected on a precleaned Whatman Nuclepore track-etched membrane filter (47 mm in diameter and 0.2 μm in pore size) supported by a filter cassette screen (part no 59-005147-0010, Thermo Fisher Scientific) and mounted in an NILU inline filter holder system at a flow rate of 2 L min<sup>-1</sup>. The precleaned filters were prepared by soaking in a ~15% H<sub>2</sub>O<sub>2</sub> solution, followed by rinsing with Milli-Q purified water (≥18 MΩ cm) and drying in a clean booth. The total volume of the sample air for each sampling was typically ~8,640 L at standard temperature and pressure (STP, 0°C and 1 atm) conditions. Each filter used for aerosol sampling was placed into a sterile centrifuge tube after collection and stored at ~4°C in the refrigerator. The samples collected for the period from August 2016 to July 2017 (122 samples in total) were used for measuring the number concentrations of INPs (see section 2.2). In addition, we referred to the number concentrations of ambient aerosol particles with equivalent optical diameters larger than 0.5 μm measured using an optical particle counter (KA-03, RION Co., Ltd.) installed in the monitoring room (the values at STP conditions are available at 1-hour intervals).

### 2.2. Freezing Experiments for INP Measurements

To quantify the number concentrations of immersion-mode INPs in the atmosphere, the ice nucleating ability of ambient aerosol particles immersed in supercooled water droplets was measured using the Cryogenic Refrigerator Applied to Freezing Test (CRAFT, Tobo, 2016). The basic procedures for measuring the number concentrations of atmospheric INPs using the CRAFT system are essentially the same as those reported previously (DeMott et al., 2017; Tobo et al., 2019). To prepare a suspension of particles, a filter used for aerosol sampling on Tokyo Skytree was immersed into Milli-Q purified water (≥18 MΩ cm) in a centrifuge tube and shaken with a vortex mixer for ~5 min. After preparing the suspension, the particle-containing water



**Figure 1.** Exemplary spectra derived from freezing experiments with  $5 \mu\text{L}$  droplets. (a) Fraction of the droplets frozen ( $f_{\text{frozen}}$ ) as a function of temperature ( $T$ ). Particle-containing droplets were prepared using a suspension containing aerosol samples collected at the 458-m level of Tokyo Skytree during 16–18 May 2017. The dilution ratio of the suspension relative to the initial suspension ( $d$ ) ranges from 1 to 100. Also shown are the results of freezing experiments with Milli-Q water droplets (four runs) and a classical nucleation theory-based parameterization for homogeneous freezing of  $5 \mu\text{L}$  pure water droplets with a cooling rate of  $1^{\circ}\text{C min}^{-1}$  (Murray et al., 2010). (b) Number concentrations of atmospheric INPs ( $N_{\text{INP}}$ ) calculated using equations 1 and 2. Error bars represent the 95% confidence intervals.

droplets (typically, 49 droplets) with a volume of  $5 \mu\text{L}$  were placed on an aluminum plate coated with a thin layer of Vaseline (petroleum jelly). Then, the temperature was lowered at a cooling rate of  $1^{\circ}\text{C min}^{-1}$  in the CRAFT system, and the number fractions of the droplets frozen ( $f_{\text{frozen}}$ ) and unfrozen ( $f_{\text{unfrozen}}$ ) were counted every  $0.5^{\circ}\text{C}$ . By assuming that ice nucleation by particles immersed in supercooled droplets can be regarded as a temperature-dependent and time-independent process, the cumulative number of ice nucleation active sites per unit volume of water ( $K$ ) at a given temperature ( $T$ ) can be expressed as follows:

$$K(T) = -\frac{\ln(1 - f_{\text{frozen}}(T))}{V_{\text{drop}}} = -\frac{\ln(f_{\text{unfrozen}}(T))}{V_{\text{drop}}} \quad (1)$$

where  $V_{\text{drop}}$  is the volume of a droplet ( $=5 \mu\text{L}$ ). The number concentrations of atmospheric INPs at STP conditions can be described by the equation:

$$N_{\text{INP}}(T) = K(T) \cdot \frac{d}{C_v} \quad (2)$$

where  $C_v$  is the ratio of the total volume of the sample air (typically,  $\sim 8,640 \text{ L}$ ) to the volume of the initial suspension and  $d$  is the dilution ratio of the suspension relative to the initial suspension. In this study, the minimum limit of detection was set at  $\sim 0.002 \text{ L}^{-1}$  for the samples collected during the period from August to December 2016 and  $\sim 0.001 \text{ L}^{-1}$  for those collected during the period from January to July 2017, respectively. In Figure 1, we present an example demonstration of freezing experiments with droplets containing aerosol samples when the total volume of sample air was  $8,640 \text{ L}$ , and the minimum limit of detection was set at  $\sim 0.001 \text{ L}^{-1}$ . In this case, the volume of the initial suspension was adjusted to  $2.1 \text{ mL}$ . Since all droplets made of the initial suspension were frozen before reaching  $-25^{\circ}\text{C}$ , the initial suspension was diluted ( $d = 10$  and  $100$ ) and then the second and third freezing experiments were conducted (Figure 1a). Finally, the  $N_{\text{INP}}$  values over a temperature range down to  $-25^{\circ}\text{C}$  (Figure 1b) were quantified by applying the results of the freezing experiments to equations 1 and 2. Since none of the droplets made of Milli-Q purified water freeze at temperatures warmer than  $-25^{\circ}\text{C}$  in the CRAFT system (Tobo, 2016; see also Figure 1a), it is expected that the derived  $N_{\text{INP}}$  values would be hardly influenced by artifacts related to freezing of pure water droplets.

Some particle suspensions prepared from the samples collected in May 2017 and placed in centrifuge tubes were heated in boiling water for  $\sim 60 \text{ min}$ , cooled to room temperature, and then used for additional freezing experiments. The  $N_{\text{INP}}$  values derived from the additional experiments were used to examine contributions of heat-sensitive materials (e.g., proteins) to atmospheric INP populations.

### 2.3. Lidar Measurements

The vertical profiles of aerosol layers over Tokyo were measured using an automated elastic scattering lidar system installed at a station of the Shinjuku Gyoen National Garden (35.69°N, 139.71°E; ~10 km west-southwest of Tokyo Skytree) in Shinjuku, Tokyo, Japan. The automated lidar system in Tokyo is an Nd:YAG laser-based Mie scattering lidar developed by the National Institute for Environmental Studies (Shimizu et al., 2016) and has been operated in the framework of the Asian Dust and Aerosol Lidar Observation Network (AD-Net). For this study, we used the attenuated backscatter coefficient and depolarization ratio at the second harmonic wavelength (532 nm) of the laser.

### 2.4. Model Simulations

Global model simulations of airborne dusts were performed using the Community Atmospheric Model version 5 with the Aerosol Two-dimensional bin module for foRmation and Aging Simulation version 2 (CAM5-chem/ATRAS2, Matsui, 2017; Matsui & Mahowald, 2017). The model is driven by the Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA2) meteorology and was run at a horizontal resolution of  $1.9^\circ \times 2.5^\circ$ , with 30 vertical levels from the surface to ~40 km. For this study, we referred to the vertical profiles of simulated dust mass concentrations extracted from a grid point closest to Tokyo Skytree. In addition, the origins of air masses arriving at the monitoring site of Tokyo Skytree were investigated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015).

## 3. Results and Discussion

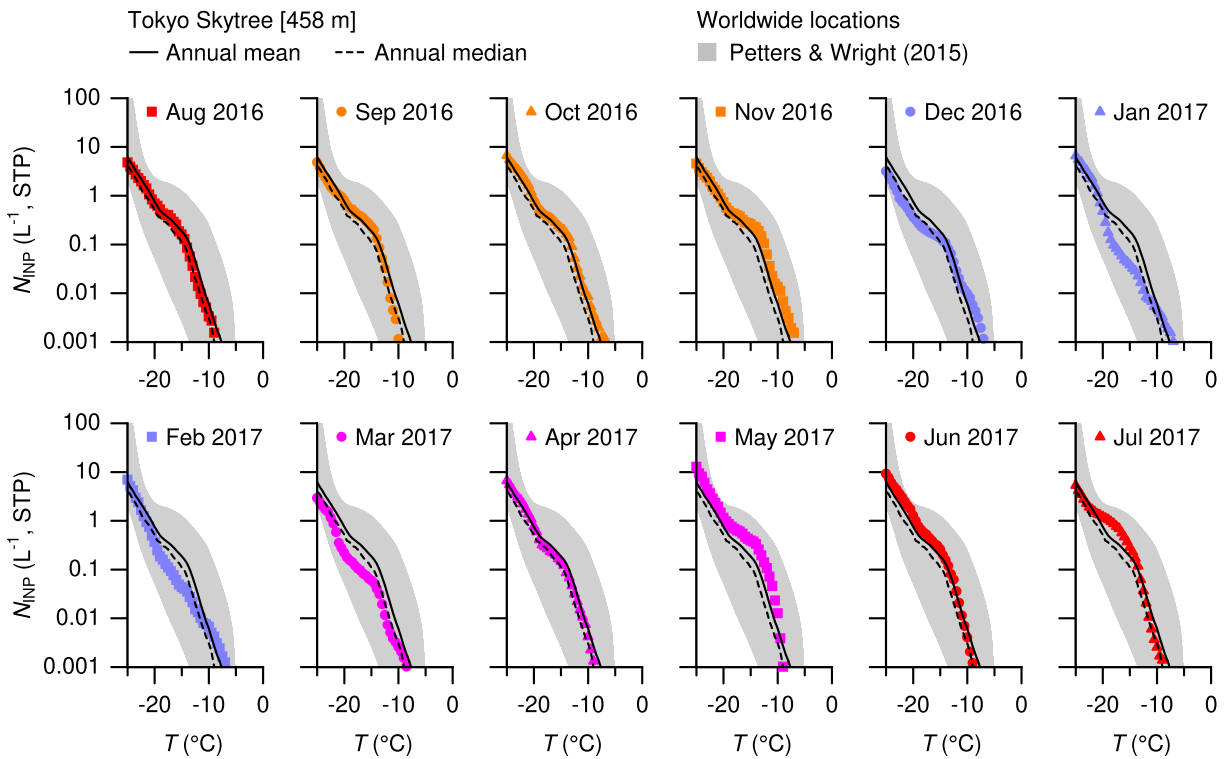
### 3.1. Seasonal Variation of INPs

To quantify the variability of atmospheric INPs over the Tokyo Metropolitan area under conditions relevant for mixed-phase cloud formation, we investigated immersion-mode INPs at a monitoring site on the 458-m level floor of Tokyo Skytree throughout the year. The population of clouds and aerosols at this site was likely to be largely characterized by air masses originating from local/regional terrestrial sources and the Pacific Ocean in mid-summer, while the influence of air masses from the Asia continental outflow was also important in other seasons (Misumi et al., 2018; Uetake et al., 2019; see also Figure S2).

In Figure 2, we summarize the monthly and annual mean INP number concentrations measured at the monitoring site of Tokyo Skytree for the period from August 2016 to July 2017. The INP number concentrations varied in the range of the upper and lower limits of the INP spectra presented by Petters and Wright (2015) and thus were not very different from those of other locations around the world. We note that the INP spectra in November and December 2016 were similar to those of Beijing in the same period reported by Chen et al. (2018) (Figure S3), suggesting that the present results might be typical INP spectra in urban air of East Asia. The seasonal variations of the INP spectra over Tokyo were relatively small in the temperature regime ranging from about  $-25^\circ\text{C}$  to  $-20^\circ\text{C}$ . The monthly mean values were highest in May 2017, and this is mainly because of the influence of an Asian dust event during 7–9 May 2017 (see section 3.2). On the other hand, it seems that the INP spectra in the temperature regime warmer than about  $-20^\circ\text{C}$  showed a relatively high seasonal variability, possibly related to meteorological and biological factors (see section 3.3).

### 3.2. Variation of INPs Associated With Asian Dust Event

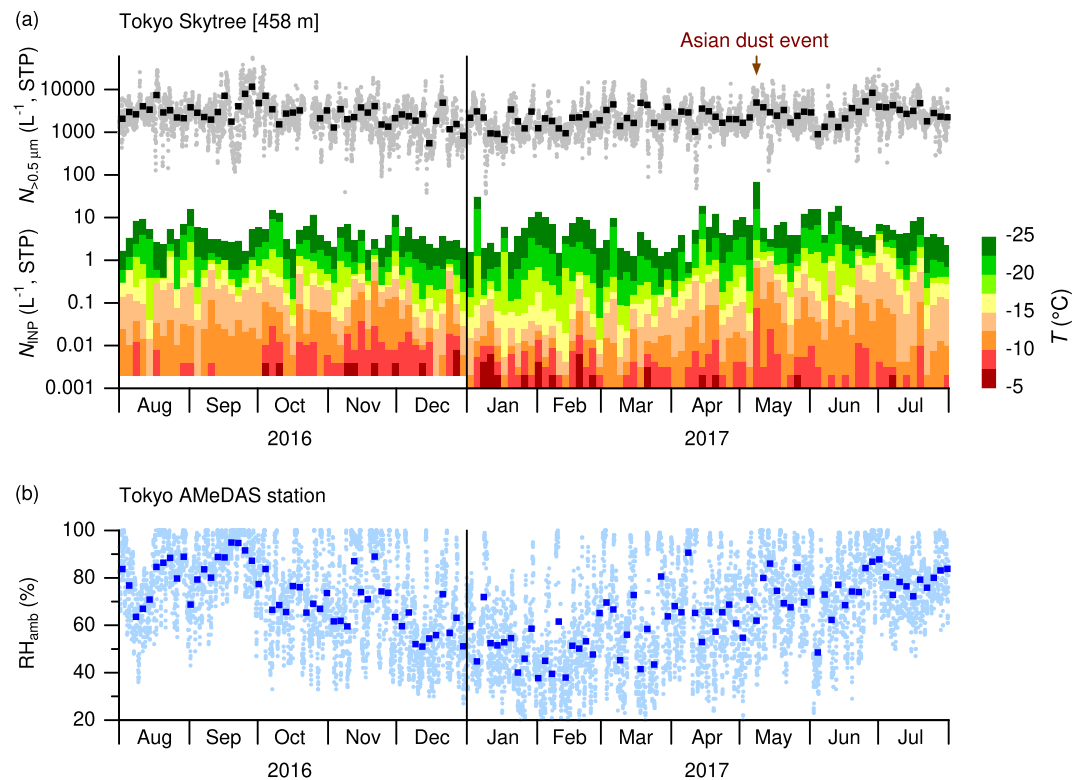
Asian dust is thought to be a major factor in enhancing the INP number concentrations over Tokyo (Isono et al., 1959). As for the period from August 2016 to July 2017, the Japan Meteorological Agency (JMA) officially reported the arrival of Asian dust plumes near the ground level over the Japan Islands only during 7–9 May 2017. The time series of the 3-day mean INP data at the 458-m level of Tokyo Skytree indicated anomalously high INP number concentrations during this period (Figures 3a and 4a). Ground-based lidar measurements in Tokyo showed obviously higher depolarization ratios at altitudes ranging from the ground level to ~3 km during 7–9 May (Figure 4c), indicating the enhancement of nonspherical aerosol particles. The CAM5-chem/ATRAS2 model simulations (Figure 4d) and backward trajectory analyses of air masses (Figure S4) support the idea that the high depolarization ratios were caused by the arrival of Asian dust plumes originating from arid/semiarid regions in northern China and Mongolia. On the other hand, it seems that the changes in the number concentrations of aerosol particles with diameters larger than  $0.5\ \mu\text{m}$  (Figure 3a) and the attenuated backscatter coefficients (Figure 4b) in response to this Asian



**Figure 2.** Monthly mean number concentrations of INPs measured at the 458-m level of Tokyo Skytree during the period from August 2016 to July 2017. Also shown are the annual mean/median values on Tokyo Skytree and the envelope of the INP measurements at various locations in the world presented by Petters and Wright (2015).

dust event were not necessarily clear, probably because these values are sensitive not only to Asian dust particles but also to other types of aerosol particles from local/regional sources (e.g., urban/rural sources and oceanic sources).

The INP number concentrations at the 458-m level of Tokyo Skytree reached a level close to the upper limit of the INP spectra presented by Petters and Wright (2015) during 7–9 May (Figure 4a). It is highly likely that the enhancement of INPs in the temperature regime ranging from about  $-25^{\circ}\text{C}$  to  $-20^{\circ}\text{C}$  was mainly caused by long-range transported Asian dust particles, which are known to serve as efficient INPs in this temperature regime (Boose et al., 2016). However, it remains unclear whether the appearance of the unique INP spectra in the temperature regime warmer than about  $-20^{\circ}\text{C}$  was also attributable to the arrival of Asian dust plumes. There is a possibility that certain PBAPs associated with Asian dust storms could be transported to Japan (Hara & Zhang, 2012), potentially leading to the enhanced INP number concentrations in the warmer temperature regime. Here, we examined the change in the INP spectra before and after heating treatment (Figure S5). The results imply that heat-sensitive biological INPs, such as proteinaceous materials from bacteria and fungi (Garcia et al., 2012; Hill et al., 2016; Pummer et al., 2015), might contribute to the INP population. In addition, heat-stable INPs were detected even in the warmer temperature regime, presumably due to the presence of certain non-proteinaceous materials of biological origin (Hill et al., 2016; McCluskey et al., 2018) and/or highly ice nucleation active mineral components, such as microcline (O’Sullivan et al., 2015). It should be noted here that the unique INP spectra in the warmer temperature regime were found even in the INP data obtained during 10–12 May and 13–15 May (Figure 4a). Asian dust plumes which were not officially reported near the ground level were likely to appear frequently at higher altitudes ( $>1$  km) over Tokyo (Figures 4c and 4d), and the arrival of Asian dust plumes at higher altitudes might have some influence on the INP population at the monitoring site of Tokyo Skytree during 10–12 May and 13–15 May. On the other hand, considering that the INP number concentrations in the temperature regime ranging from about  $-25^{\circ}\text{C}$  to  $-20^{\circ}\text{C}$  were lowered after the dust event of 7–9 May (Figure 4a) and the influence of air masses from the Asian continent to the



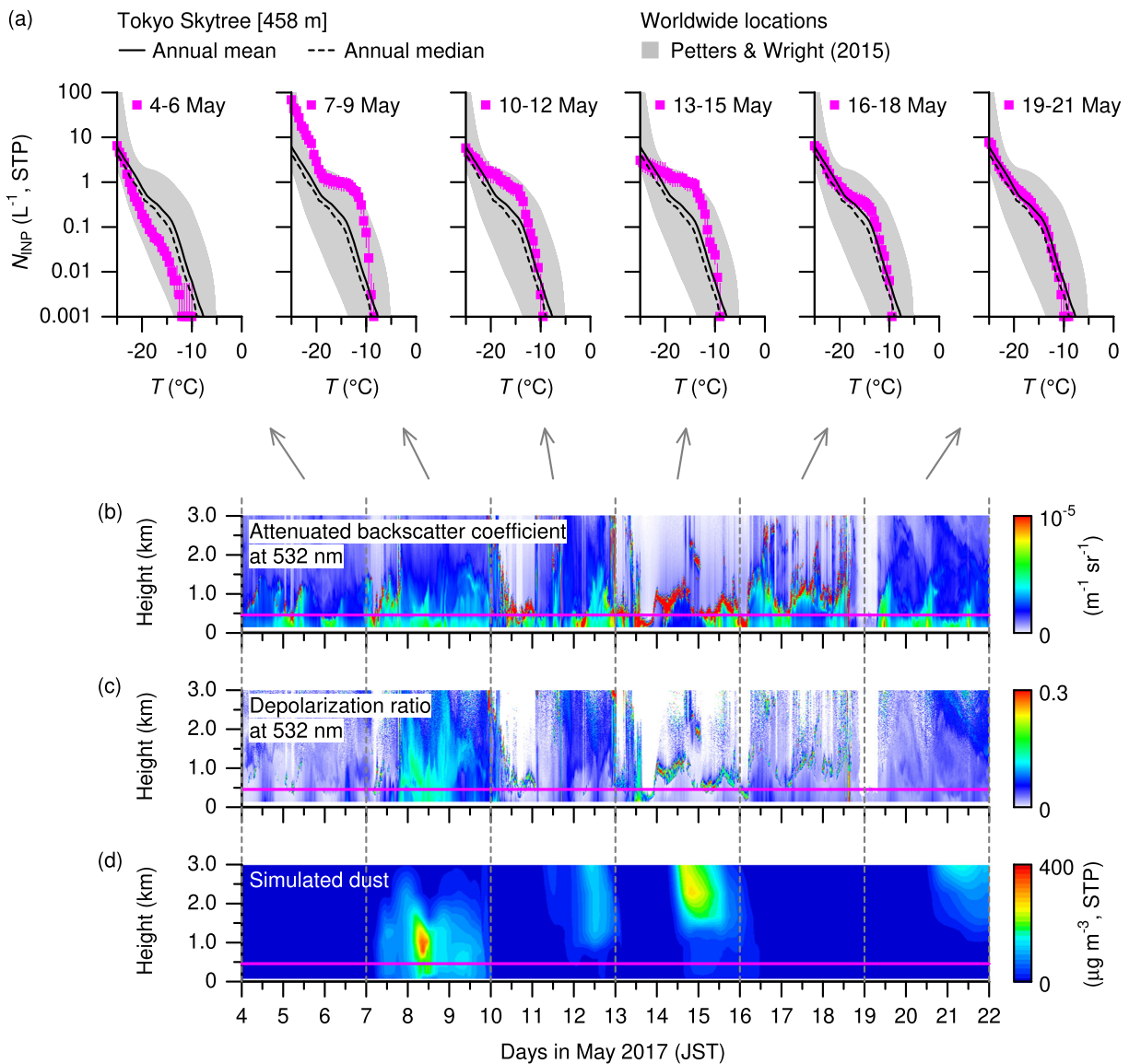
**Figure 3.** Time series of INPs, aerosol particles, and relative humidity ( $RH_{amb}$ ) measured over Tokyo during the period from August 2016 to July 2017. (a) The 3-day mean  $N_{INP}$  and the hourly (gray) and 3-day mean (black) number concentrations of ambient aerosol particles with diameters larger than  $0.5 \mu\text{m}$  ( $N_{>0.5 \mu\text{m}}$ ) at the 458-m level of Tokyo Skytree. Note that the minimum limit of detection of  $N_{INP}$  was set at  $\sim 0.002 \text{ L}^{-1}$  for the period from August to December 2016 and  $\sim 0.001 \text{ L}^{-1}$  for the period from January to July 2017, respectively. (b) The hourly (aqua) and 3-day mean (blue) ambient  $RH_{amb}$  at the Tokyo AMeDAS station.

monitoring site of Tokyo Skytree would be weakened (Figure S4), there remains a possibility that the unique INP spectra in the warmer temperature regime were induced by phenomena other than the Asian continental outflow (see also section 3.3). Further studies are therefore necessary to understand the impact of Asian dust events on the population of atmospheric INPs active in the warmer temperature regime.

### 3.3. Variation of INPs Associated With Other Potential Factors

The INP spectra at the 458-m level of Tokyo Skytree exhibited measurable seasonal variations in the temperature regime warmer than about  $-20^\circ\text{C}$  (Figures 2 and 3a). Earlier field studies at other locations around the world suggest that variations in the number concentrations of INPs active at a given temperature may be related to those of aerosol particles with diameters larger than  $0.5 \mu\text{m}$  (DeMott et al., 2010; Tobo et al., 2013). However, our measurements did not show any clear seasonal variations in the number concentrations of aerosol particles larger than  $0.5 \mu\text{m}$  (Figure 3a), suggesting that the majority of the aerosol particles over Tokyo might have little influence on the variations of INPs active in the warmer temperature regime.

It is important to note that the INP number concentrations in the temperature regime warmer than about  $-15^\circ\text{C}$  tended to show higher values in wet seasons from late spring through to autumn and lower values in dry seasons from winter to early spring in East Asia. Here, we focused on the possible relationship of the INP number concentrations at the 458-m level of Tokyo Skytree with meteorological conditions in the Tokyo metropolitan area. As illustrated in Figure 5, the INP number concentrations in the temperature regime ranging from  $-15^\circ\text{C}$  to  $-10^\circ\text{C}$  were positively correlated with ambient relative humidity near the ground surface measured at the JMA Automated Meteorological Data Acquisition System (AMeDAS) station ( $35.69^\circ\text{N}$ ,  $139.75^\circ\text{E}$ ;  $\sim 7$  km west-southwest of Tokyo Skytree) in Chiyoda, Tokyo, Japan

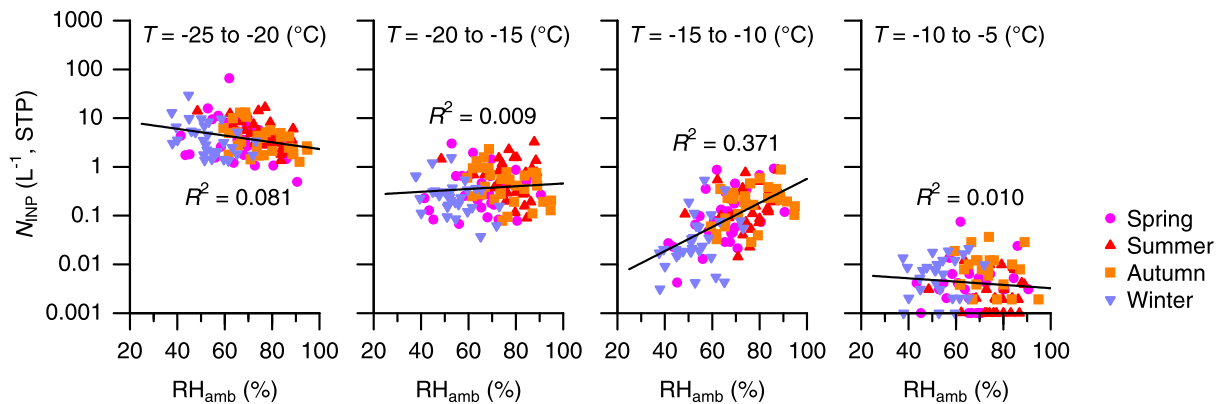


**Figure 4.** Variations of INPs and aerosol particles over Tokyo before, during, and after an Asian dust event of 7–9 May 2017. (a) The 3-day mean  $N_{\text{INP}}$  measured at the 458-m level of Tokyo Skytree. Error bars represent the 95% confidence intervals. Also shown are the annual mean/median values on Tokyo Skytree and the envelope of the INP measurements at various locations in the world presented by Petters and Wright (2015). (b) The attenuated backscatter coefficient at 532 nm measured over Shinjuku, Tokyo using a ground-based lidar system. (c) Same as Figure 4b but for the depolarization ratio at 532 nm. (d) The simulated dust mass concentrations over Tokyo using the CAM5-chem/ATRAS2 model. The pink-colored lines in Figures 4b–4d indicate a height of 458 m AGL.

(see also Figure 3b), while the correlations between the INP data in the other temperature regimes ( $-25^{\circ}\text{C}$  to  $-20^{\circ}\text{C}$ ,  $-20^{\circ}\text{C}$  to  $-15^{\circ}\text{C}$ , and  $-10^{\circ}\text{C}$  to  $-5^{\circ}\text{C}$ ) and humidity were not significant ( $R^2 < 0.1$ ). Similar results were obtained from the comparisons with relative humidity at the 35-, 370-, and 615-m levels of the Tokyo Skytree (Figures S6–S8). The variations in the INP number concentrations might also be related to other meteorological factors, such as ambient temperature and specific humidity (Figure S9). It should also be noted here that the population of INPs active at temperatures warmer than  $-15^{\circ}\text{C}$  is assumed to be dominated by certain particles of biological origin (Kanji et al., 2017; Murray et al., 2012).

Based on these results, we consider that some mechanisms related to meteorological and biological factors might influence the population of certain INPs active at temperatures ranging from  $-15^{\circ}\text{C}$  to  $-10^{\circ}\text{C}$ . For example, field studies in North America suggest that high relative humidity and rain events can trigger the release of PBAPs from vegetated environments, potentially leading to enhancement of INPs in the





**Figure 5.** Comparison of INPs at the 458-m level of Tokyo Skytree with ambient relative humidity at the Tokyo AMeDAS station. The  $N_{\text{INP}}$  data are divided into four groups according to the difference of temperature regime. The  $N_{\text{INP}}$  and  $\text{RH}_{\text{amb}}$  data are the 3-day mean values and categorized by season; spring (March–April–May), summer (June–July–August), autumn (September–October–November), and winter (December–January–February). The equation fitted to the data is  $N_{\text{INP}} = \exp(a \cdot \text{RH}_{\text{amb}} + b)$ , where  $a$  and  $b$  are constants (Table S1).

atmosphere (Huffman et al., 2013; Prenni et al., 2013; Tobo et al., 2013; Wright et al., 2014). In addition, organic matter particles (OMPs) contained in fertile soils may be actively released during rain events (Wang et al., 2016) and serve as efficient INPs (Hill et al., 2016; O’Sullivan et al., 2015; Tobo et al., 2014). The release of PBAPs and OMPs linked to oceanic biological activity may also influence atmospheric INP populations (DeMott et al., 2016; McCluskey et al., 2017; Wilson et al., 2015). A global modeling study by Heald and Spracklen (2009) indicates a pronounced seasonal cycle of the mass concentrations of airborne fungal spores related to the variation of humidity and vegetation cover in Asia, North America, and Europe and shows higher values in warm/wet seasons and lower values in cold/dry seasons. Field measurements of fungal spores in Hualien, Taiwan (Ho et al., 2005) and Tsukuba, Japan (Igarashi et al., 2018) also report a similar seasonality. Unfortunately, we did not measure the amounts of particles of biological origin, such as PBAPs and OMPs, at the monitoring site of Tokyo Skytree, but a DNA sequence-based study at this site for the period from August 2016 to February 2017 indicates that the diversity of airborne microbial community (alpha and beta diversity) could change significantly in response to local meteorological factors, especially relative humidity (Uetake et al., 2019).

We therefore speculate that seasonal variations of certain PBAPs and/or OMPs linked to meteorological and biological factors might largely influence the population of INPs active in the temperature regime between  $-15^{\circ}\text{C}$  and  $-10^{\circ}\text{C}$ , resulting in the unique seasonal variations of the INP spectra over the Tokyo Metropolitan area. We note that there are large uncertainties in INP measurements in the temperature regime warmer than  $-10^{\circ}\text{C}$ , since the INP number concentrations in this regime varied around the minimum limit of detection; however, it seems that their seasonal patterns were quite different from those between  $-15^{\circ}\text{C}$  and  $-10^{\circ}\text{C}$ . These results suggest a need for further studies for quantifying the sources of INPs and their seasonality over urban areas of East Asia.

#### 4. Conclusions

Continuous year-round measurements of INPs active in the immersion mode were performed at a monitoring site of Tokyo Skytree and used to quantify the INP number concentrations over the Tokyo Metropolitan area. Our measurements for the period from August 2016 to July 2017 show that seasonal variations of the INP number concentrations were relatively small in the temperature regime below about  $-20^{\circ}\text{C}$ , whereas the values were episodically enhanced during the arrival of Asian dust plumes. On the other hand, the INP spectra exhibited measurable seasonal variations in the temperature regime warmer than about  $-20^{\circ}\text{C}$ . In particular, the INP number concentrations in the temperature regime between about  $-15^{\circ}\text{C}$  and  $-10^{\circ}\text{C}$  tended to show higher values in warm/wet seasons (late spring through to autumn) and lower values in cold/dry seasons (winter to early spring) in East Asia. The seasonal variations of INPs in this temperature regime might be related to meteorological and biological factors causing the emission of certain INPs of biological origin. This work provides the first comprehensive data set showing detailed seasonal trends of

immersion-mode INPs over an urban area of East Asia and will be useful for studying their impacts on aerosol-cloud-precipitation interactions on local, regional, and global scales.

### Data Availability Statement

We thank Nobuo Sugimoto and Atsushi Shimizu for the provision of the lidar data in Tokyo available at the AD-Net website (<https://www-lidar.nies.go.jp/AD-Net/>), the NOAA Air Resources Laboratory for the provision of the HYSPLIT transport and dispersion model and/or READY website (<https://www.ready.noaa.gov/>), and the Tobu Tower Skytree Co., Ltd. for sharing reference meteorological data on Tokyo Skytree (not publicly available). The INP data on Tokyo Skytree (3-day mean values) are available online (<https://ads.nipr.ac.jp/dataset/A20200727-004>). Other data sets for this research are available at the website (<https://ads.nipr.ac.jp/dataset/A20200915-001>).

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