

Dear Editor:

Thank you for your comments, and please find author's response as below.

Regards,

1. Page 5: "In order to ensure the reliability of measurements, only dataset, of which acquisition rate higher than 50%, were used during all analysis procedures." Please, clarify this sentence since its meaning is not clear. Probably, replace that sentence with something like "collected, validated data spanned at least 50% of the day". In any case, say more explicitly what has been done.

Authors' response: In revised manuscript, to clarify, we modified the sentence on Page 5 Line 9-10, as:

"To ensure the reliability of measurements, only validated data that spanned at least 50% of one day were used for analysis."

* highlighted in yellow in the 'marked-up' manuscript version

2. Page 13, line 9: "The daily mean kappa values range between 0.07 and 2.19, with a mean of 0.41 ± 0.10 ." What is the utilized symbol here? Obviously, you mean k, not "kappa", right? Please use the correct symbol.

Authors' response: You are right. In the sentence, we mentioned k values not "kappa". Thus, we changed the symbol on Page 12 Line 25- Page 13 Line 1. As:

"The daily mean k_T values range between 0.07 and 2.19, with a mean of 0.41 ± 0.10 ."

* highlighted in yellow in the 'marked-up' manuscript version

3. In any case, be careful to avoid any confusion between the fitting coefficient k (Eq. 1) and the hygroscopicity parameter "kappa". Symbols must be clearly different to each other. I suggest to use another symbol for the fitting coefficient, not "k", in order to avoid confusion.

Authors' response: To avoid confusion between fitting coefficient, k, and hygroscopicity

parameter, kappa, it is good idea to use another symbol for the fitting coefficient. In the revised manuscript, we used another symbol for the fitting coefficient, " k_T ", and the symbol for the hygroscopicity parameter was not used except Equation (2).

* highlighted in yellow in the 'marked-up' manuscript version

4. Add one or few sentences in the Introduction, emphasizing what is the novelty of the present study compared to previous ones and what is the new knowledge brought to the readers.

Authors' response: We added the following sentence in the introduction on Page 4 Line 9-13.

"Although various studies have been performed in Antarctica, research on the seasonal variations of CN, CCN, and the size distribution of aerosol particles has not been considered in the Antarctic Peninsula. Furthermore, studies based on long-term observations are rare. Multi-year observations involving an analysis of the physical characteristics of CCN are conducted for the first time in this study."

* highlighted in yellow in the 'marked-up' manuscript version

5. Finally, take care of improving the use of english throughout the text. If possible, let the paper be read and corrected by a native english speaking person.

Authors' response: Following the editor's comment, the revised manuscript has been edited by an English native speaker.

** These grammatical changes are marked as 'RED' in the 'marked-up' manuscript version

Seasonal variations in physical characteristics of aerosol particles at the King Sejong Station, Antarctic Peninsula

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Abstract

The seasonal variability of the physical characteristics of aerosol particles sampled from the King Sejong Station in the Antarctic Peninsula was investigated over the period between March 2009 and February 2015. Clear seasonal cycles for the total particle concentrations (CN) were observed. The monthly mean CN_{2.5} concentrations of particles with a particle size larger than 2.5 nm were highest during the austral summer with a mean of $1080.39 \pm 595.05 \text{ cm}^{-3}$, and were the lowest during the austral winter with corresponding values of $197.26 \pm 71.71 \text{ cm}^{-3}$. A seasonal pattern in the cloud condensation nuclei (CCN) concentrations coincided with the CN concentrations, where the both concentrations were at a minimum in the winter and maximum in the summer. The measured CCN spectra were approximated by fitting a power law function relating the number of CCN at a given supersaturation (SS) to each SS value, with the fitting coefficients C and k_T . The values of C varied from 6.35 cm^{-3} to 837.24 cm^{-3} , with a mean of $171.48 \pm 62.00 \text{ cm}^{-3}$. The values of k_T ranged between 0.07 and 2.19, with a mean of 0.41 ± 0.10 . In particular, the k_T values during the austral summer were higher than those during the winter, indicating that aerosol particles are more sensitive to SS changes during the summer than they are during the winter. Furthermore, the annual mean hygroscopicity parameter, kappa, was estimated as 0.15 ± 0.05 , for a SS of 0.4%. The effects of the origin and the pathway travelled by the air mass on the physical characteristics of the aerosol particles were also determined. The modal diameter of aerosol particles that originated from the South Pacific Ocean showed a seasonal variations, being equal to from $0.023 \mu\text{m}$ in the winter and to $0.034 \mu\text{m}$ in the summer, for the Aitken mode and $0.086 \mu\text{m}$ in the winter and $0.109 \mu\text{m}$ in the summer for the Aitken and accumulation modes, respectively.

1. Introduction

Aerosol particles in the atmosphere ~~may be~~ emitted either directly from various natural and anthropogenic sources (~~i.e.~~, primary aerosol particles) or produced by gas-to-particle conversion processes (~~i.e.~~, secondary aerosol particles). ~~They~~ These particles influence the local and global climates directly by scattering and absorbing radiation and indirectly by acting as cloud condensation nuclei (CCN) or ice nuclei (~~HN~~) (IPCC, 2013). The physical, chemical, and optical properties of atmospheric aerosol particles determine their impact on climate change. Although various studies on the effects of aerosol particles on climate change have been ~~carried out~~ conducted, ~~the specific~~ direct and indirect climate effects are still highly uncertain (IPCC, 2013). Moreover, in order to understand the where aerosol particles are sources sourced and the how they are processes processed of in the atmospheric aerosol particles atmosphere, ~~there should be a need to have~~ long-term observations for different regions at different regions are required because aerosol particles vary temporally and spatially.

The Antarctic region is highly sensitive to climate changes due to complex interconnected environmental systems (~~e.g. such as~~ snow cover, land ice, sea-ice, and ocean circulation) (Chen et al., 2009). Previous studies ~~show have shown~~ that the Antarctic Continent and ~~the Antarctic~~ Peninsula have experienced ~~noticeable~~ significant climate changes (Rignot et al., 2004; Steig et al., 2009; Pritchard et al., 2012; Schneider et al., 2012). The Antarctic Peninsula, in particular, ~~has a~~ is warming at a rate of more than 5 five times faster than that of the other regions ~~on earth~~ (Vaughan et al., 2003; IPCC, 2013). The Antarctic climate system can be linked ~~with to~~ aerosol particles by complex feedback processes that involve aerosol-cloud interactions. ~~In addition~~ Also, because there are less anthropogenic emission sources in Antarctica, it is a suitable place to study the formation and growth processes of ~~the~~ natural aerosol particles. For these reasons, ~~the observation of their~~ monitoring the physical properties of aerosol particles in Antarctica, ~~e.g. such as~~ total particle concentrations, size ~~distributions~~ distribution, and concentrations of black carbon and activated CCN, is necessary.

Over ~~the years, measurements of~~ time aerosol particles measurements have been ~~widely~~ conducted

at various research stations in Antarctica; notably: Aboa (Koponen et al., 2003; Virkkula et al., 2007; Kyrö et al., 2013), Amunsen-Scott (Arimoto et al., 2004; Park et al., 2004), Concordia (Järvinen et al., 2013), Halley (Rankin and Wolff, 2003; Roscoe et al., 2015), Kohnen (Weller and Wagenbach, 2007; Hara et al., 2010), Maitri (Pant et al., 2011), Mawson (Gras, 1993), McMurdo (Hansen et al., 2001; Mazzera et al., 2001), Neumayer (Weller et al., 2015), Syowa (Ito, 1985; Hara et al., 2011b), and Troll (Fiebig et al., 2014). ~~The~~ Antarctic aerosol particles have been investigated ~~with regard to~~regarding their size distributions (Koponen et al., 2003; Belosi et al., 2012), optical properties (Shaw, 1980; Tomasi et al., 2007; Weller and Lampert, 2008), chemical compositions (Virkkula et al., 2006; Weller and Wagenbach, 2007; Asmi et al., 2010; Hara et al., 2011a), and mass concentrations (Mazzera et al., 2001; Mishra et al., 2004). ~~Some-Other~~ studies have also focused on aerosol transport in the upper atmosphere (Hara et al., 2011b) and new particle formation (Järvinen et al., 2013; Kyrö et al., 2013; Weller et al., 2015). Although various studies have been performed in Antarctica, researches on the seasonal variations of CN, CCN, and the size distribution of aerosol particles has not been considered were less conducted in the Antarctic Peninsula. Furthermore, published papers obtained by studies based on long-term observations were are rare. Based on the mMulti-year observations in Antaretica, especially, involving an analysis on of the physical characteristics of CCN has been are conducted for the first time in the presentthis study.

~~In this study, we~~The continuously monitored-monitoring of the physical characteristics of aerosol particles at the Korean Antarctic station (King Sejong Station) ~~in the Antaretic Peninsula~~ from March 2009 to February 2015 are used as the basis for this study. Measurements ~~for of the~~ aerosol size distribution and concentrations of the total aerosol number, black carbon (BC), and CCN were ~~carried out~~conducted using various instruments. The main aim ~~of this study was is~~ to determine the seasonal variations of the physical properties of aerosol particles in the Antarctic Peninsula. ~~In addition~~Also, the physical characteristics of aerosol particles that originated from the ocean and continent of the Antarctic region ~~were are also~~ investigated with using air mass back-trajectory analysis.

2 Methods

2.1 Sampling site and instrumentation

5 Continuous observations of the physical properties of aerosol particles ~~have been~~was carried out ~~since between~~ March 2009 ~~and February 2015~~ at ~~the~~ King Sejong Station (62.22°S, 58.78°W) in the Antarctic Peninsula. Detailed information ~~of the sampling about sampling~~ site is ~~given~~provided by Choi et al. (2008). In brief, ~~the~~ King Sejong Station is located on the Barton Peninsula of King George Island ~~(KGI), which has a~~ The population density ~~of KGI is~~ higher than ~~that of~~ other regions in
10 Antarctica due to the ~~various~~ research activities ~~carried out from~~conducted on eight permanent on-site stations. The observatory is located approximately 400 m southwest of the main buildings, ~~which and~~ includes ~~the a~~ power generator and crematory ~~of King Sejong Station~~. Thus, the northeastern direction (355°-55°) ~~was is~~ designated as ~~the a~~ local pollution sector because of ~~the~~ emissions from the power generator and crematory ~~at the station~~. ~~We therefore discarded d~~Data from the local pollution
15 ~~sector collected from this sector is discarded~~ to improve data quality, ~~and along with data where BC concentrations were are~~ higher than 100 ng m⁻³ ~~were also discarded~~. In order tTo ensure the reliability of measurements, only validated data that spanned at least 50% of the one day were used during all analysis procedures for analysis. In this study, ~~we present the analysed results of~~ the physical characteristics of aerosol particles ~~obtained during~~collected between March 2009 ~~to and~~ February 2015
20 are analysed.

The physical characteristics of aerosol particles were continuously observed with various instruments ~~that included~~including two condensation particle counters (CPCs), an aethalometer, a cloud condensation nuclei counter (CCNC), and a scanning mobility particle sizer (SMPS). ~~The~~These observation methods are shown in Fig. 1.

25 Based on Global Atmosphere Watch (GAW) aerosol measurements guidelines and

recommendations, ~~we installed~~ a cylindrical stainless common sample inlet was installed. The common inlet was placed on the roof of the observatory (Fig. 1). The diameter and length of the ~~common~~-inlet were 0.1 m and 5.2 m, respectively. In order to understand flow condition in the common inlet, the Reynolds number was calculated. We used mean values of air temperature and pressure measured over the period from March 2009 to February 2015. The mean values of temperature and pressure were - 2.4 °C and 98.8 kPa, respectively. The total flow rate of ~~total-the~~ sampled air was maintained as 150 ~~lpm~~LPM. The Reynolds number in the common inlet was 2388. ~~It, which~~ represents ~~that the flow in the common inlet is a~~ laminar-turbulent transition regime-flow ($2000 < Re < 4000$). For sampling, a short L-bend ~~tube made of~~ stainless steel tube was placed at the center-centre of the common inlet. Sampling ~~in~~ instruments were connected ~~with to~~ the ~~common~~-inlet using conductive tubing to minimize ~~the~~ particle losses. ~~The~~ Diameter and length of the conductive tubing connecting the stack with the sampling devices are 3/8 inches and 0.6 m, respectively. To maintain ~~the~~ ambient conditions, ~~any drying system~~ drying was not ~~used~~ conducted during sampling. The total counting efficiency of the ~~measurement systems~~ sampling equipment was estimated to be 0.92 for 2.5 nm particles, and 1 for particles larger than 10 nm, respectively at the common inlet. ~~The total counting efficiency of 2.5 nm particles was 0.92 in the common inlet system used in this study, whereas the value was calculated as 1 for particles larger than 10 nm (calculated after:~~ (Hind, 1999; Baron and Willeke, 2011). All sampling lines, ~~expect for the common inlet,~~ was/were connected with conductive tubing to minimize ~~the known~~ particle losses.

~~Total~~ The total particle number concentrations ~~were~~ was examined with two CPCs: a TSI model 3776 that ~~measured~~ measures particles >greater than 2.5 nm in diameter; and a TSI model 3772 that ~~measured~~ measures particles >larger than 10 nm. Sample aerosol flow rates ~~of for~~ CPC 3776 and CPC 3772 were 1.5 ~~lpm~~LPM and 1.0 ~~lpm~~LPM, respectively.

~~The~~ An aethalometer (Magee Scientific, AE16) was used to measure the concentration of light ~~absorption~~ particles at for 880 nm wavelengths. The flow rate of the sample was constant at 5.0

~~10~~ ~~pmLPM~~. The main purposes of measuring ~~the~~ BC concentrations ~~were was~~ to investigate ~~the~~ long-range transport ~~of~~ aerosol particles and ~~to~~ assess the ~~influence exerted by impact of~~ local pollution.

To measure the CCN concentrations, a CCNC (DMT CCN-100) was used ~~at for~~ five different supersaturation (~~SS~~) ratios (~~SS~~) (0.2, 0.4, 0.6, 0.8, and 1.0-%) and ~~a~~ total flow rate of 0.5 ~~10~~ ~~pmLPM~~.

5 The CCN concentrations ~~were was~~ determined by exposing aerosol particles at supersaturated conditions and then counting ~~only~~ the number of activated droplets with a detector. The sampling duration was set at ~~approximately~~ 5 min for each SS value (except ~~the for~~ 0.2-% SS) before it was changed to the next ~~SS~~ value. For ~~a~~ 0.2-% SS, ~~the~~ CCN concentrations ~~were was~~ measured for 10 min because ~~it required the~~ additional time ~~was required~~ to achieve stability after ~~completing conducting~~ measurements at ~~a~~ 1-% SS.

10 Aerosol size distributions were continuously measured with ~~the a~~ SMPS, which ~~consisted of comprises~~ a differential mobility ~~analyser analyzer~~ (DMA; HCT ~~inc. Inc.~~, ~~Model~~: LDMA 4210), a CPC (TSI 3772), a control unit, an aerosol neutralizer (soft x-ray), and a data logging system. The length, inner diameter, and outer diameter of the DMA were 44.42 cm, 0.953 cm, and 1.905 cm, respectively. The resolution of ~~the~~ scanning time was set to 120 s for mobility particle diameters ~~from between~~ 0.01 ~~to and~~ 0.30 μm . A closed sheath-air loop with a diaphragm pump in the control unit was used to maintain the sheath flow of ~~the~~ DMA. The flow rate of sheath air of ~~the~~ DMA was 10 ~~10~~ ~~pmLPM~~. The ratio of aerosol flow to sheath flow of ~~the~~ DMA was 1:10.

15 ~~In addition Also~~, meteorological parameters including temperature, relative humidity (RH), wind speed (WS), wind direction (WD), pressure, ~~and as well as~~ UV and solar radiation were also continuously monitored over the entire observation period.

2.2 Back-trajectory analysis

20 ~~In order to associate To relate~~ the physical properties of aerosol particles to their source ~~areas~~ for the sampling periods, ~~the an~~ air mass back trajectory analysis was conducted using ~~the a~~ Hybrid Single-

Particle Lagrangian Integrated Trajectory (HYSPPLIT) model (Stein et al., 2015) (<http://www.arl.noaa.gov/HYSPLIT.php>). For every 6 h period, 120-h air mass back trajectories were analysed, ending at heights of 100m, 500m, and 1500m above ~~the~~ ground level ~~of at~~ the sampling site. ~~The r~~Results ~~where for~~ the origin and pathway of the air masses ~~– that were similar~~ for at least 12 h ~~were similar at for the~~ three different heights were used ~~for the analysis in this study. Based on this analysis, we have classified t~~The air masses ~~were then classified~~ into four groups according to their origin and pathway: ~~–, including~~ two continental regions (South America and Antarctica) and two oceanic areas (South Atlantic and South Pacific Ocean), as ~~are~~ shown in Fig. 2.

10 3 Results and Discussion

3.1 Meteorological conditions

Fig. 3 depicts ~~the~~ monthly variations of ~~the~~ meteorological parameters measured from an automatic weather system (AWS) ~~during the whole over the entire~~ observation period. The temperature varied between $-19.5\text{ }^{\circ}\text{C}$ and $+5.8\text{ }^{\circ}\text{C}$, with a mean of $-2.4 \pm 2.1\text{ }^{\circ}\text{C}$ and ~~the relative humidity (RH)~~ was ~~between 60% and 100%~~, with a mean of $87.9 \pm 3.3\%$. As mentioned previous studies (Kwon and Lee, 2002; Mishra et al., 2004), the observation site was relatively humid and warm compared to inland Antarctic stations due to ~~the effect of a~~ marine environment. The solar radiation varied from 2.3 W m^{-2} to 375.4 W m^{-2} , with a mean of $81.2 \pm 38.9\text{ W m}^{-2}$. No clear annual trends ~~of for~~ temperature ~~are were~~ observed during ~~a the~~ six-year period due to a relatively short observation period. In this ~~manuscript paper, we focused on an analysis of the~~ correlation ~~analysis~~ between temperature (or solar radiation) and ~~the~~ CN concentration ~~is the focus~~.

3.2 Seasonality in the physical characteristics of aerosol particles

3.2.1 Total particle number concentrations

25 Fig. 4 shows the monthly mean particle number (CN) ~~condensations concentration~~ measured with

two types of instruments (TSI CPC 3776 and 3772) over the period ~~from-between~~ March 2009 ~~to-and~~ February 2015. All ~~the~~ seasons mentioned in this study are austral seasons. As can be seen in Fig. 4, there is ~~an-evidenta~~ clear seasonal cycle ~~of-for~~ CN concentrations, which ~~are-the-maximum-in-the~~ summeris highest in the summer (~~from-~~December to February, (DJF)) and ~~minimum-lowest~~ in the winter (~~from-~~June to August, (JJA)). The maximum concentrations of particles larger than 2.5 nm (CN_{2.5}) and larger than 10 nm (CN₁₀) ~~were-was~~ approximately 2000 cm⁻³ in December 2012 and about 800 cm⁻³ in December 2009, ~~respectively~~. The minimum values of CN_{2.5} and CN₁₀ concentrations ~~were~~ was approximately 110 cm⁻³ and 90 cm⁻³, both in August 2013, respectively. ~~Our-These~~ results ~~were~~ are in good-agreement with ~~the-results-of~~ previous studies from other Antarctic stations (Jaenicke et al., 1992; Gras, 1993; Virkkula et al., 2009; Weller et al., 2011). For instance, Virkkula et al. (2009) reported a long-term daily average CN concentrations over the period from between November 2003 to-and January 2007 from observationsmonitoring conditions at Aboa, the Finnish Antarctic research station at a in coastal region in-of Antarctica. The maximum monthly average CN concentrations were ~~observed-measured~~ in February and the minimum ~~concentrations-were-measured~~ in July, which is the darkest period of the year. The cause of the clear seasonal cycle of CN concentrations may be attributed to the formation ~~process-~~ of aerosol particles. ~~The-h~~High CN_{2.5} concentrations during the austral summer season (DJF) should be related primarily to non-sea-salt sulphate and methanesulphonate (MSA) derived from the oxidation of dimethyl sulphide (DMS) produced by phytoplankton (Fattori et al., 2005; Weller et al., 2011). The DMS concentrations ~~increases~~ increases sharply when biological activity ~~is~~ was enhanced due to increasing temperatures and solar radiation (Virkkula et al., 2009). Since ~~our-the~~ the sampling site for this study was in the Antarctic Peninsula, ~~ocean-the~~ biological activity the ocean was considered ~~to-beas~~ an important factor in-for the ~~particle-~~formation and growth of the aerosol particles. ~~Since-the-h~~Higher differences in concentration occur summer rather than winter ~~differences-betweenfor~~ CN_{2.5} and CN₁₀ concentrations ~~correlate~~ correlate with a higher biological activity in summer than winter ~~biological activity, and-as-well-as~~ solar radiation and temperature, ~~our-hypothesis-is~~ It is hypothesized

that ~~the~~ trends ~~of the for each difference should be~~ related to secondary aerosol formation caused by biological activity. To better understand the effect of temperature and solar radiation intensity on the CN_{2.5} concentrations, ~~we compared~~ the relationship between the monthly mean CN_{2.5} concentrations and solar radiation intensity, and the monthly mean CN_{2.5} concentrations and temperature was compared. The correlation coefficient between CN_{2.5} and the solar radiation intensity (opened circle; R²=0.621) was slightly higher than ~~that between for the mean~~ CN_{2.5} concentration and temperature (opened triangle; R²=0.419), as shown in Fig. 5. ~~Our~~ These results suggest that the CN_{2.5} concentrations may be more closely coupled with solar radiation intensity than ~~with~~ temperature.

Unique results ~~of for the~~ CN_{2.5} concentrations were observed as shown in Fig. 4. The CN_{2.5} concentrations in the summer ~~season~~ of 2013-2014 were much lower than other years. Unfortunately, the reason for the lower CN_{2.5} concentrations ~~could can~~ not be explained by the solar radiation intensity ~~and or~~ temperature ~~because the solar radiation and the temperature did not show~~ as there was not any distinctive variation in these values compared with other years. ~~The~~ A possible reason is the type of air masses reached ~~to~~ the sampling site. Although the air mass that originated ~~from in~~ the South Pacific Ocean (Case IV: ~~of the~~ descriptions ~~of the Cases I, II, III and IV are described~~ in section 3.3) was dominant in ~~the~~ summer, based on the air mass back trajectory analysis ~~as~~ explained in Section 2.2, the frequency of the air mass ~~originated~~ from the South Atlantic Ocean (Case II) in the summer of 2013-2014 was higher than other years and the frequency of air mass ~~originated~~ originating from Case IV was lower than other years. Unfortunately, neither CN₁₀ ~~nor~~ SMPS data ~~are is~~ available for the austral summer season of 2013-2014 because of mechanical failures. Thus, it is not possible to directly explain the low concentrations ~~of for~~ CN_{2.5} ~~for in~~ this season ~~in terms of regarding~~ the potential effects ~~of on the~~ air mass characteristics ~~on and~~ the concentration of particles sized between 2.5- and 10 nm size particles. Nevertheless, it is likely that ~~the an~~ increased frequency of air mass originating from the South Atlantic Ocean (Case II) might have resulted in ~~the a~~ lower CN_{2.5} concentration ~~of in~~ the austral summer season of 2013-2014. These CN concentrations ~~were is~~ comparable ~~to the~~ with results from

~~the~~ Aboa Station, which is located in the coastal area of Antarctica and is mainly affected by ~~south~~ South Atlantic air masses (Virkkula et al., 2009). ~~They~~ These results showed ~~the a~~ daily CN concentrations from December 2003 to January 2007. Although there was a variation of the CN concentrations ~~year by year~~ between each year, the daily CN concentrations during ~~the austral~~ the austral summer ~~period~~ were $\sim 600 \text{ cm}^{-3}$.

A more detailed comparison of the monthly trends ~~in~~ for the $\text{CN}_{2.5}$ and CN_{10} concentrations is presented in Fig. 6. The monthly mean CN concentrations increased from September to February ~~mainly~~ during the austral spring and summer periods (Bigg et al., 1984; Jaenicke et al., 1992; Gras, 1993). The CN concentrations sharply decreased from March and remained stable from April to August. In particular, the $\text{CN}_{2.5}$ concentrations during the summer period increased sharply compared to the CN_{10} concentrations. ~~the~~ This increase was probably due to new particle formation. High solar radiation and temperature, and low RH values during the summer are conducive to ~~the~~ new particle formation (Hamed et al., 2007).

3.2.2 Cloud condensation nuclei (CCN) concentrations

Fig. 7(a) shows the monthly mean CCN concentrations at ~~the a~~ SS value of 0.4 % over the period from March 2009 to February 2015. There is a ~~long large~~ gap in data from July 2011 to December 2013 ~~because data were not collected~~ due to a faulty CCN counter. Anttila et al. (2012) measured cloud droplet number concentration (CDNC) and CCN concentrations ~~at for~~ five SS values (0.2, 0.4, 0.6, 0.8, and 1.0%) during the third Palls Cloud Experiment (PaCE-3). ~~They~~ These results showed a correlation between ~~the~~ CDNC and CCN concentrations ~~at for~~ each supersaturation. The relationship between ~~the~~ CDNC and CCN concentrations at ~~the a~~ SS value of 0.4% was approximately linear. ~~Furthermore,~~ while CCN concentrations were lower than ~~the~~ CDNC, when the SS value was lower than 0.4%; and ~~CCN concentrations at upper 0.4%~~ higher than ~~the~~ CDNC, when the CCN concentration was above 0.4%. Based on ~~this these~~ results, ~~in this study, the~~ supersaturation of 0.4% was chosen to investigate

seasonal variations of the CCN. Monthly variations in the CCN concentrations was found, with the a maximum values being observed during the summer periods (DJF) and the a minimum concentrations were observed during the winter periods (JJA). The monthly mean CCN concentrations were in the range of ranged between 20.63 cm⁻³ in July 2009 and 227.52 cm⁻³ in January 2014, with a mean of 112.80 ± 39.05 cm⁻³. Fig. 7(b) also shows seasonality in for the CCN concentrations at an with a SS value of 0.4%. The CCN concentrations gradually decreased from February and remained stable during the winter, while the CCN concentrations from September increased sharply, as is shown in Fig. 7(b). The maximum CCN concentration in January was 199.89 ± 37.07 cm⁻³ and the minimum CCN concentration in August was 42.13 ± 14.51 cm⁻³. This clear seasonality of for the CCN concentrations follows the seasonal variation march of the CN concentrations. As shown in Fig. 6, the CN₁₀ concentrations as well as CN_{2.5} concentrations increased during the summer. In addition, the aerosol size distributions measured by the SMPS showed that the concentrations of for accumulation mode particles in the range of 100 and 300 nm, as well as Aitken mode particles during the summer, increased significantly, as can be seen in Fig. 8. Accumulation mode particles can easily act as CCN (Dusek et al., 2006), hence CCN concentrations increase during the summer and decrease during the winter.

In order to investigate the seasonal variations of fractions of the CCN concentrations at for each SS value in for the CCN concentrations at a SS of 1.0%, the CCN data were examined in more detail. An analysis of the cumulative CCN concentrations shown as a fraction of the CCN concentration measured at the a SS of 1.0% was carried out, and the The results are shown in Fig. 9. Here, fractions of the CCN concentrations were estimated by dividing the CCN concentrations at each SS value by the total CCN concentrations at the a SS of 1.0%. Although a clear seasonal trend of for CCN concentrations with a maximum during the summer and a minimum during the winter was presented, as mentioned earlier, the fraction of CCN concentrations at the a SS value of 0.2% in for the activated CCN concentrations showed a different pattern with a maximum value in July and a minimum value in December, as shown in Fig. 9. The numbers at the top of Fig. 9 represent the mean CCN concentrations

at ~~the a~~ SS values of 1.0%. The fraction of particles activated to CCN at ~~the a~~ SS value of 0.2% during ~~the~~ summer and ~~the~~ winter was 0.49 ± 0.07 and 0.62 ± 0.06 , respectively. The fraction at ~~the a~~ SS value of 0.2% during ~~the~~ winter (JJA) was similar to ~~those that~~ (~ 0.52) measured ~~in at~~ Mace Head Station, which is a representative site of a marine environment (Paramonov et al., 2015).

5 ~~The s~~Seasonal variations in the mean activation ratio of ~~the~~ CCN concentrations at a SS of 0.4% to the CN concentrations measured ~~from by the~~ two CPCs (TSI 3776 and 3772), ~~as are~~ shown in Fig. 10. The mean values of ~~the~~ activation ratios of CCN/CN_{2.5} and CCN/CN₁₀ were ~~about~~ 0.33 ± 0.10 and 0.40 ± 0.08 , respectively. Although clear changes were observed in the monthly variation ~~in for~~ the CN and CCN concentrations as shown in Fig. 6 and Fig. 7(b), it was seen that the activation ratio
10 (CCN/CN₁₀) was similar regardless of seasonality. The reason that no clear change is observed in the activation ratios at ~~the~~ King Sejong Station ~~in the Antarctic Peninsula~~, might be the variation ~~of in~~ the concentrations of accumulation mode particles, as can be seen in Fig. 8. The lower activation ratios in September and November ~~are mainly is~~ because of the size and chemical properties of ~~the~~ aerosol particles. Both, the size and chemical ~~components composition~~ of ~~the~~ aerosol particles ~~may can~~ have
15 a large impact on the activation ratio (Dusek et al., 2006; Leena et al., 2016). The concentrations of Aitken mode aerosol particles increased sharply compared to ~~their the~~ concentrations in August. Thus, the activation ratio decreased dramatically. Unfortunately, ~~we did not confirm the~~ aerosol size distribution ~~was not confirmed at the because our~~ aerosol size distribution data ~~in for~~ November ~~was~~ ~~not un~~reliable due to malfunctioning instruments.

20 The CCN concentrations ~~at for different~~ SS values can be represented by ~~a the~~ power-law function, defined by Twomey (1959):

$$N_{CCN} = C \cdot (SS)^{k_T} \quad (1)$$

25 where N_{CCN} is the concentration of CCN at given a supersaturation values (SS), C and k_T are coefficient

constants estimated from CCN spectra. The correlation coefficient between the measured CCN concentrations at the a SS of 0.4% and the ~~power-law fitted~~ values of the power law function, r , was 0.978. The values of C varied from 6.35 cm⁻³ to 837.24 cm⁻³, with a mean of 171.48 ± 62.00 cm⁻³. The daily mean k_T values range between 0.07 and 2.19, with a mean of 0.41 ± 0.10. The monthly variations of k_T and C values are ~~also~~ summarized in Fig. 11. A comparison with CCN concentrations indicated that the values ~~of for~~ k_T during the austral winter (June) were also the lowest (0.29 ± 0.06), while during ~~the~~ summer (December) they were ~~the~~ highest (0.55 ± 0.13). Based on this result, aerosol particles activated to CCN during ~~the~~ summer are expected to be more sensitive to SS changes than those aerosol particles activated during ~~the~~ winter.

It is useful to infer the hygroscopic properties of aerosol particles with ~~a the~~ hygroscopicity parameter, kappa. ~~The k~~Kappa values varied from 0 for insoluble particles to larger than 1 for water-soluble salt particles (Petters and Kreidenweis, 2007). ~~The k~~Kappa values can be defined ~~by Petters and Kreidenweis (2007)~~ as

$$\kappa = \frac{4A^3}{27D_{act}^3 \ln^2 SS}, \quad A = \frac{4\sigma_w M_w}{RT\rho_w} \quad (2)$$

where σ_w is surface tension of water, M_w is the molecular weight of water, R is the universal gas constant, T is the temperature, and ρ_w is the density of water. SS is the supersaturation applied in the CCNC. The critical diameter, D_{act} , was estimated following method of Furutani et al. (2008)

$$\frac{\int_{D_0}^{D_{act}} n(D) dD}{N_{tot}} = 1 - \frac{CCN}{CN} \quad (3)$$

where N_{tot} is total number concentrations of aerosol particles measured by SMPS. D is the electric

mobility diameter observed by SMPS. In this calculation, ~~10 nm was applied for~~ D_0 was assumed to be 10 nm, at beginning of the ~~where~~ SMPS scan ~~starts~~. The CCN/CN ratio indicates the fraction of CCN-active aerosols ~~among of the~~ total particle concentrations. In this study, the kappa values were estimated using ~~the a~~ monthly mean CCN concentrations at ~~the a~~ SS of 0.4%, the monthly mean CN concentrations measured by ~~the~~ CPC₂ and the monthly mean size number distribution results obtained from ~~the~~ SMPS data. The annual mean kappa value was calculated to be 0.15 ± 0.05 . This value is comparable to the previous studies from ~~the~~ Arctic and ~~subarectic~~ Subarctic areas. For example, Latham et al. (2013) who measured the CCN activity ~~at in~~ the Arctic ~~by~~ using aircraft measurements, reported ~~the a~~ kappa value of 0.32 ± 0.21 . Martin et al. (2011) inferred ~~a~~ total kappa value of 0.33 ± 0.13 during ~~a~~ cruise observation in Longyearbyen, Svalbard. Kammermann et al. (2010) reported ~~the~~ kappa values ~~that~~ varied between 0.07- ~~and~~ 0.21 ~~in the period of over an~~ 18 days period within the Arctic Circle in Sweden. Jaatinen et al. (2014) also showed ~~the a~~ kappa value of 0.13 ± 0.07 using ~~a~~ 13-day ~~set of~~ data dataset from the at subarectic Subarctic area ~~in of~~ Finland (Pallas-Sodankylä station).

3.2.3 Black carbon (BC) concentrations

Fig. 12 shows ~~the~~ variations of monthly mean BC concentrations over the whole sampling period. To eliminate ~~effect~~ of local pollution on ~~observations, in this study, from this study, data, where~~ BC concentrations ~~were~~ higher than 100 ng m^{-3} , were discarded. The daily mean BC concentrations varied between 1.07 ng m^{-3} and 75.97 ng m^{-3} , with a mean of $27.43 \pm 4.98 \text{ ng m}^{-3}$. The BC concentrations observed at ~~our station~~ King Sejong Station ~~were was~~ higher than those at other stations in Antarctica (Bodhaine, 1995; Wolff and Cachier, 1998; Pereira et al., 2006; Weller et al., 2013). For instance, the annual mean BC concentrations at the South Pole, Halley, Neumayer, and Ferraz station were 0.65, 1.0, 2.6, and 8.3 ng m^{-3} , respectively. The reason for the higher BC concentrations might be related to location of sampling site. There are nine permanent on-site stations ~~on in~~ the Baton Peninsula of King George Island. In particular, six stations are located within a 10-km radius ~~from the of~~ King Sejong

Station. The presence of these stations should affect the measured BC concentrations ~~at the King Sejong Station~~, causing a positive bias. ~~Additionally~~Also, no clear seasonal patterns were observed in ~~our~~this study throughout the entire observation period. However, clear seasonal patterns were observed in previous studies ~~were observed~~ at other stations in Antarctica (Wolff and Cachier, 1998; Weller et al., 2013). Wolff and Cachier (1998) showed seasonal cycles of BC measured at the ~~Halley station~~Station and the South Pole with a Aethalometer. They found that although BC concentrations varied depending on the sampling site, ~~the~~ BC concentrations decreased during the austral winter (JJA) and increased during the austral summer (DJF). ~~Contrarily, according~~Contrary to Pereira et al. (2006), although BC concentration during ~~the~~ summer ~~increased~~increases slightly, no clear seasonal trends were observed unlike the results measured by Wolff and Cachier (1998). This suggests that the BC concentrations are dependent on the sampling site and the long-range transport of air masses.

3.3 Effect of air mass trajectory on the physical properties of aerosol particles

In this section, the effect of the origin and pathway of the air mass on the physical characteristics of the aerosol particles is presented. As mentioned earlier in See Section 2.2, ~~we classified~~ air masses are classified into four groups based on air mass back trajectory analysis. The wind data and aerosol characteristics ~~with of~~ the four types of air masses ~~during over~~ the entire observation period ~~are is~~ listed in Table 1. The very few cases of air masses originating from ~~the continent of~~ South America (Case I) showed the highest BC and CCN concentrations (~~Table 1~~). This ~~might~~may be due to anthropogenic influences at the source and the aging of aerosol particles. The CN₁₀ concentrations ~~were~~was similar regardless of the origin and pathway of air masses, whereas an enhancement of the CN_{2.5} concentrations was observed when the air mass originated from the ocean (Case II and IV). This is probably due to the high biological activity in the South Atlantic and South Pacific Oceans during ~~the~~ summer (DJF) ~~period~~. A more detailed comparison, excluding the results of Case I ~~of from~~ the CN concentrations based on ~~the~~an air mass analysis is shown in Fig. 13. It should be noted that the mean

values of CN concentrations in this analysis fall within the range of standard deviations ~~of each case for~~ the mean values of each case. This is probably because of new particle formation, causing a bias, as new particle formation ~~results in mainly contributes~~ a sudden increase ~~of in~~ aerosol ~~number~~ concentrations (Kulmala et al, 2004; Pierce and Adams, 2009). ~~The M~~minimum concentrations of aerosol particles ~~for~~ (CN_{2.5} and CN₁₀) ~~originating~~ from the ocean (Case II and IV). ~~These are were~~ observed from April to September, whereas the concentrations of aerosol particles (CN_{2.5}) originating from the South Atlantic (Case II) and the South Pacific (Case IV) Oceans ~~were the are~~ highest in November and February, respectively. ~~Here we It was~~ found that the peak month ~~of for~~ the CN_{2.5} concentrations had discrepancies ~~in accordance~~ with the air mass history. This is probably due to a difference in the chemical compounds that contributed to aerosol formation processes and/or ~~in~~ variations ~~of in~~ biogenic activity according to the origin and transport pathway of the air masses. To verify this, further studies on the chemical compositions of aerosol particles ~~need to be carried out in the future are required~~. ~~When For~~ air masses ~~were~~ transported from the South Pacific Ocean to ~~the~~ King Sejong Station (Case IV), the seasonality of the aerosol size distribution was also investigated. ~~The A~~ aerosol size distribution parameters ~~were~~ fitted to a log-normal distribution ~~were~~ derived for each season. The seasonally averaged log-normal aerosol size distributions ~~are is~~ shown in Fig. 14. The computed modal diameters along with the standard deviation and number concentrations are summarized in Table 2. It is ~~obvious clear~~ that the modal diameters during ~~the~~ summer are larger than ~~those~~ during ~~the~~ winter for both Aitken (0.034 against 0.023 μm, respectively) and accumulation modes (0.109 against 0.086 μm, respectively). The number concentrations for ~~the~~ summer are also higher than those for the winter for the (304.36 ± 20.10 against 49.16 ± 3.88 cm⁻³, respectively) and accumulation (140.25 ± 10.64 against 44.78 ± 14.24 cm⁻³, respectively) modes. The enhancement of the number concentrations for the Aitken mode during ~~the~~ summer should be linked to new particle formation over the oceans as a product of biological activity. The spring and autumn seasons shows intermediate values. ~~Our~~ These results are similar to those of previous laboratory and field experiments

(Sellegrì et al., 2006; Yoon et al., 2007). O'Dowd et al. (2004) suggested that primary formation processes play a significant role in marine aerosol production in the North Atlantic Ocean. In addition, the contribution of biological organic compounds to the marine aerosol distribution might be dominant (Kim et al., 2015).

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4 Summary and conclusions

~~The s~~Seasonal variations in the physical characteristics of aerosol particles at ~~the~~ King Sejong Station (62.22°S, 58.78°W) in the Antarctic Peninsula were investigated based on ~~the~~ in-situ measured aerosol data ~~for the period from~~ between March 2009 ~~to and~~ February 2015. ~~An obvious~~ A clear seasonal variation of particle number concentrations (CN) exists, with ~~the~~ maximum concentrations in the austral summer (DJF) and ~~the~~ minimum concentrations in the austral winter (JJA). The maximum CN concentrations of particles larger than 2.5 nm (CN_{2.5}) and 10 nm (CN₁₀) ~~were was~~ approximately 2000 cm⁻³ in December 2012 and about 800 cm⁻³ in December 2009 and February 2015, respectively. In particular, the CN_{2.5} concentrations increased sharply during the summer compared to CN₁₀ concentrations, suggesting that ~~the~~ particle formation processes ~~were are probably likely~~ driven by ~~the~~ high biological activity ~~during the season~~.

In addition, ~~we presented the~~ clear seasonal trends of CCN concentrations at ~~the a~~ supersaturation (SS) of 0.4% ~~were presented~~. The maximum mean CCN concentration of $199.89 \pm 37.07 \text{ cm}^{-3}$ was measured in January and the minimum mean CCN concentration was $42.13 \pm 14.51 \text{ cm}^{-3}$ in August. The activation ratio (CCN/CN₁₀) of aerosol particles at ~~the~~ King Sejong Station (0.40 ± 0.08) in the Antarctic Peninsula was lower than those at the Arctic area (0.52) (Latham et al., 2013). ~~We also estimated~~ C and k_T values from the measured CCN results at for each SS value were also estimated. The measured CCN spectra were approximated by fitting a power law ~~fitting~~ function relating the number of CCN at a given SS to the SS, with the fitting coefficient, C and k_T (Twomey 1959). The values of C varied between 6.35 cm^{-3} and 837.24 cm^{-3} , with a mean of $171.48 \pm 62.00 \text{ cm}^{-3}$. The values

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of k_T ranged between 0.07 and 2.19, with a mean of 0.41 ± 0.10 . The k_T values during the austral summer periods (DJF) were higher than those during the winter periods (JJA). Furthermore, the annual mean hygroscopicity parameter, kappa, was estimated as 0.15 ± 0.05 , for a SS of 0.4%.

Based on the a backward trajectory analysis, ~~we classified the~~ air masses ~~were classified~~ into four groups according to their origin and pathway ~~including~~: two continental regions (South America and Antarctica) and two oceanic areas (South Atlantic and South Pacific Ocean). ~~We~~ It was found that most air masses originated from the oceanic areas. The very few cases of air masses originating from the South American continent (Case I) showed the highest BC and CCN concentrations. The CN₁₀ concentrations were analogous regardless of origin, whereas CN_{2.5} concentrations showed differing values. The CN_{2.5} concentrations that originated from oceanic areas (Case II and IV) were higher than those from continental regions (Case III). ~~i~~ In particular, the CN_{2.5} concentrations showed clear seasonal variations; with minimum concentrations from April to September and maximum concentrations in November from the South Atlantic Ocean (Case II) and in February from the South Pacific Ocean (Case IV). Furthermore, ~~in terms of~~ regarding Case IV, an analysis of aerosol size distributions in the 0.01 to 0.3 μm range was performed. The modal diameters also showed seasonal variations of; 0.023 μm in the winter and 0.034 μm in the summer for the Aitken mode and 0.086 μm in the winter and 0.109 μm in the summer for the accumulation mode.

Overall, this study is the first of its kind to ~~analyze~~ analyse seasonal variations in ~~the~~ physical characteristics ~~of for~~ aerosol particles in the Antarctic Peninsula. The aerosol particle formation process is still not fully understood, and thus, more studies should be necessary to determine seasonal variations in the chemical characteristics of atmospheric aerosols.

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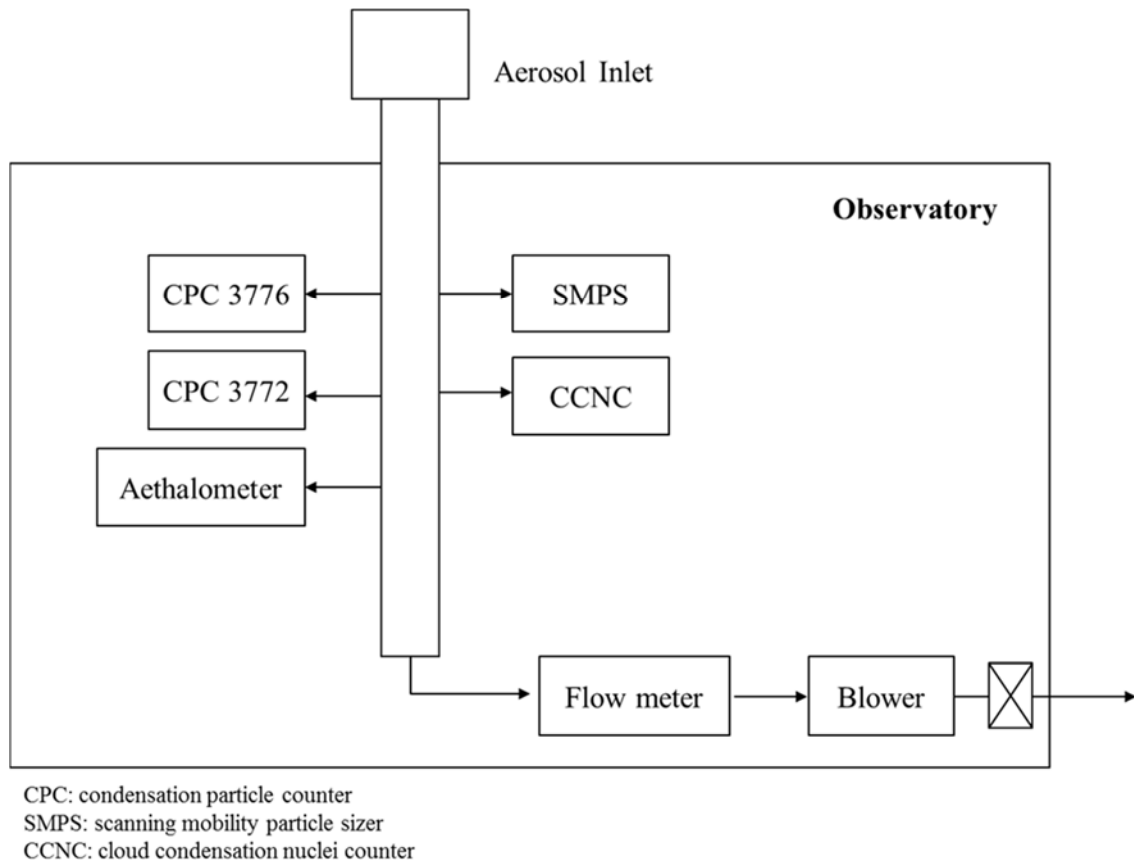
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5 Figure 1. A schematic diagram for the observation methods used in this study.

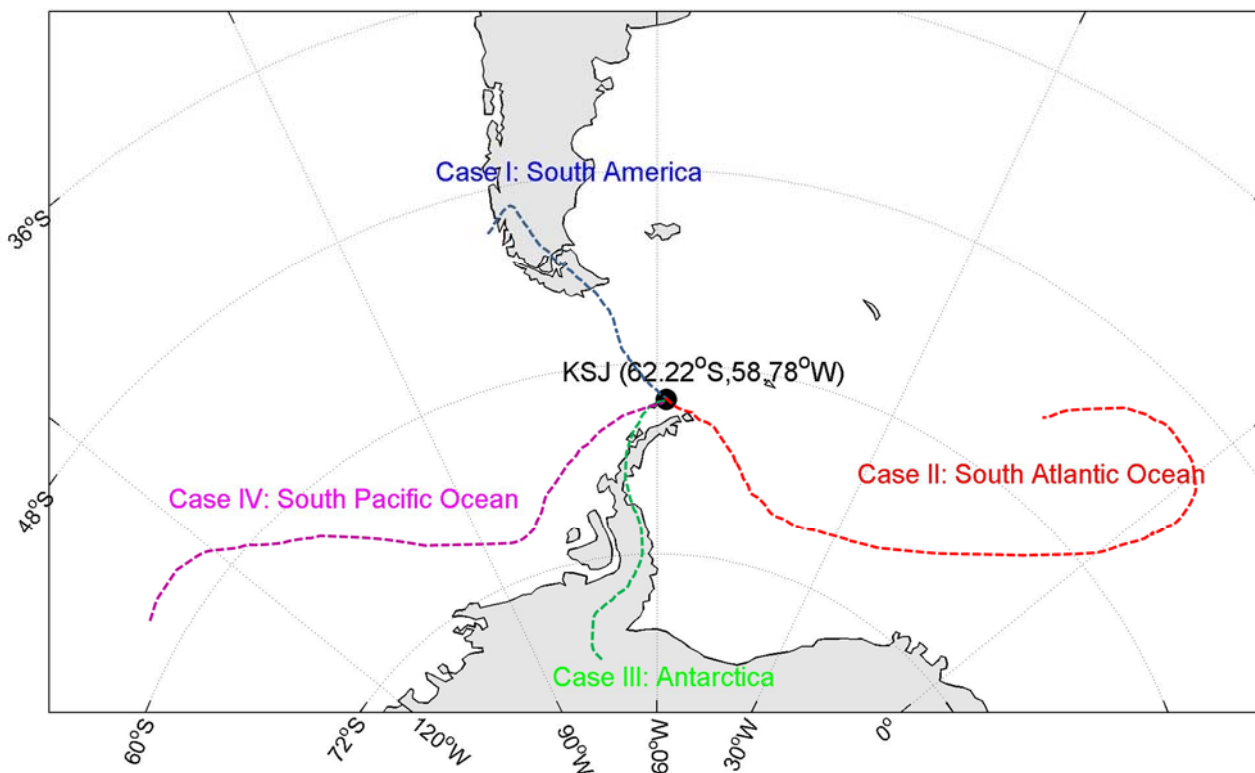


Figure 2. Map of the sampling site (62.22°S, 58.78°W; black circle) and classification of the four cases according to the origin and pathway of the air masses. Dot lines represent example of back trajectories according to cases.

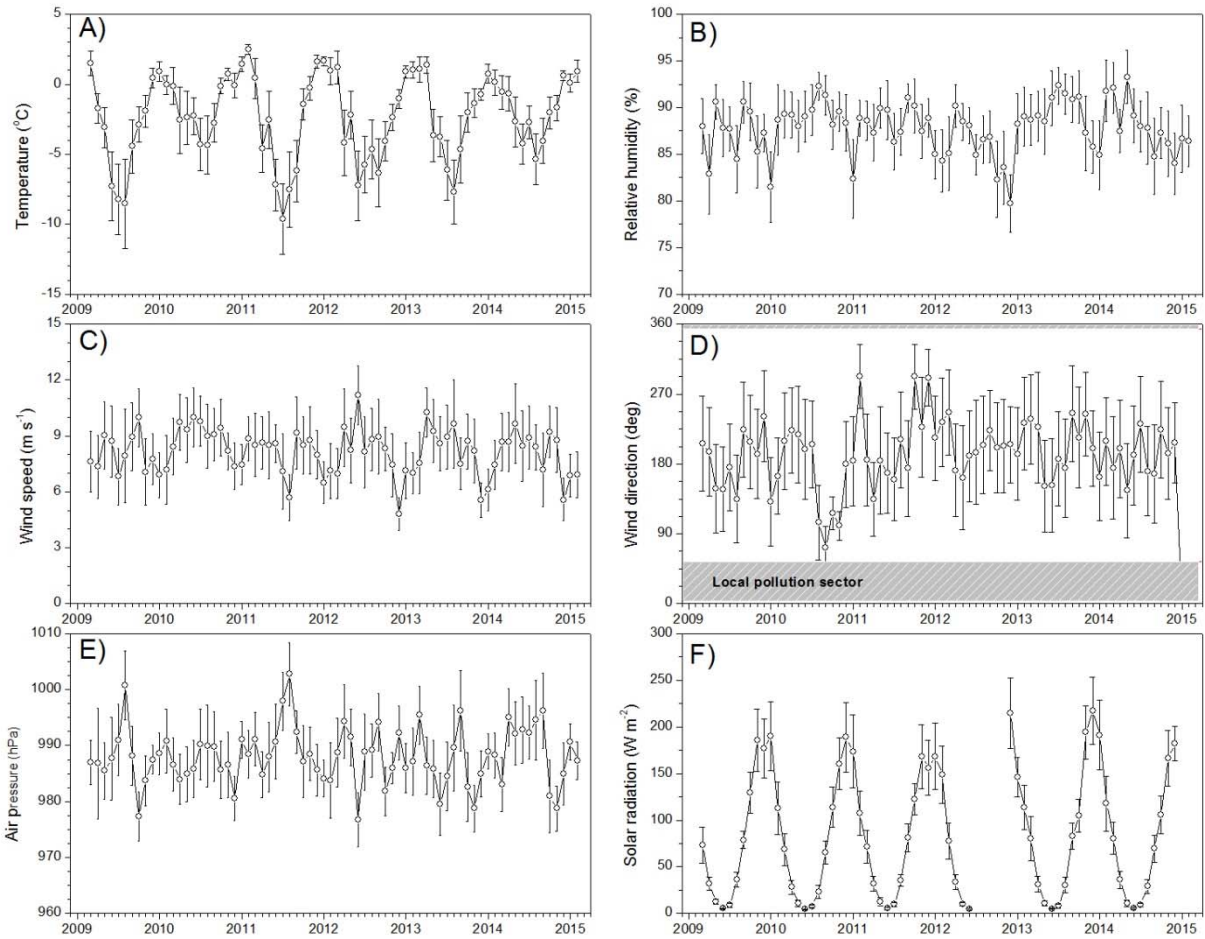


Figure 3. Monthly mean variation of (a) temperature, (b) relative humidity, (c) wind speed, (d) wind direction, (e) air pressure, and (f) solar radiation over the period from March 2009 to February 2015. The shaded area in Figure 3(d) represents the wind direction for the local pollution sector.

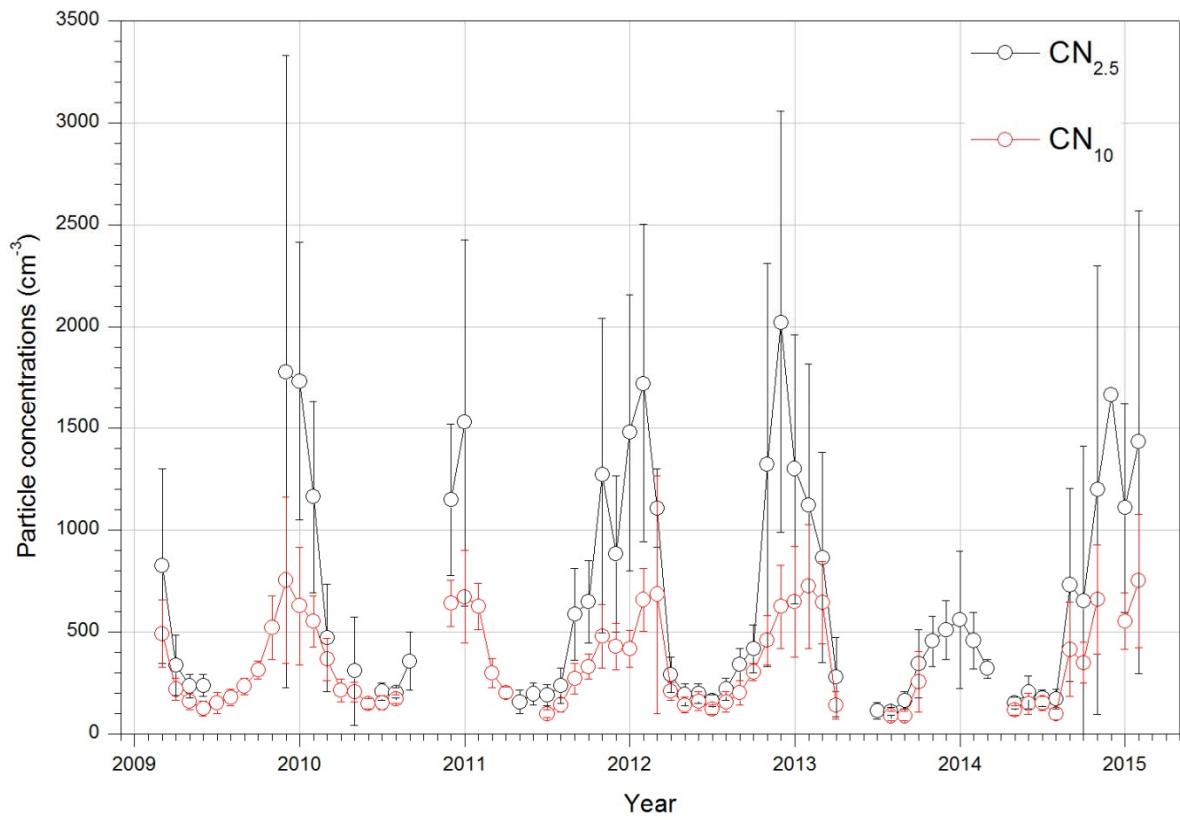


Figure 4. Monthly variations of mean CN_{2.5} (black opened circle) and CN₁₀ (red opened circle) concentrations with a standard deviation from March 2009 to February 2015. The error bars represent the standard deviation of the measurements from the mean value.

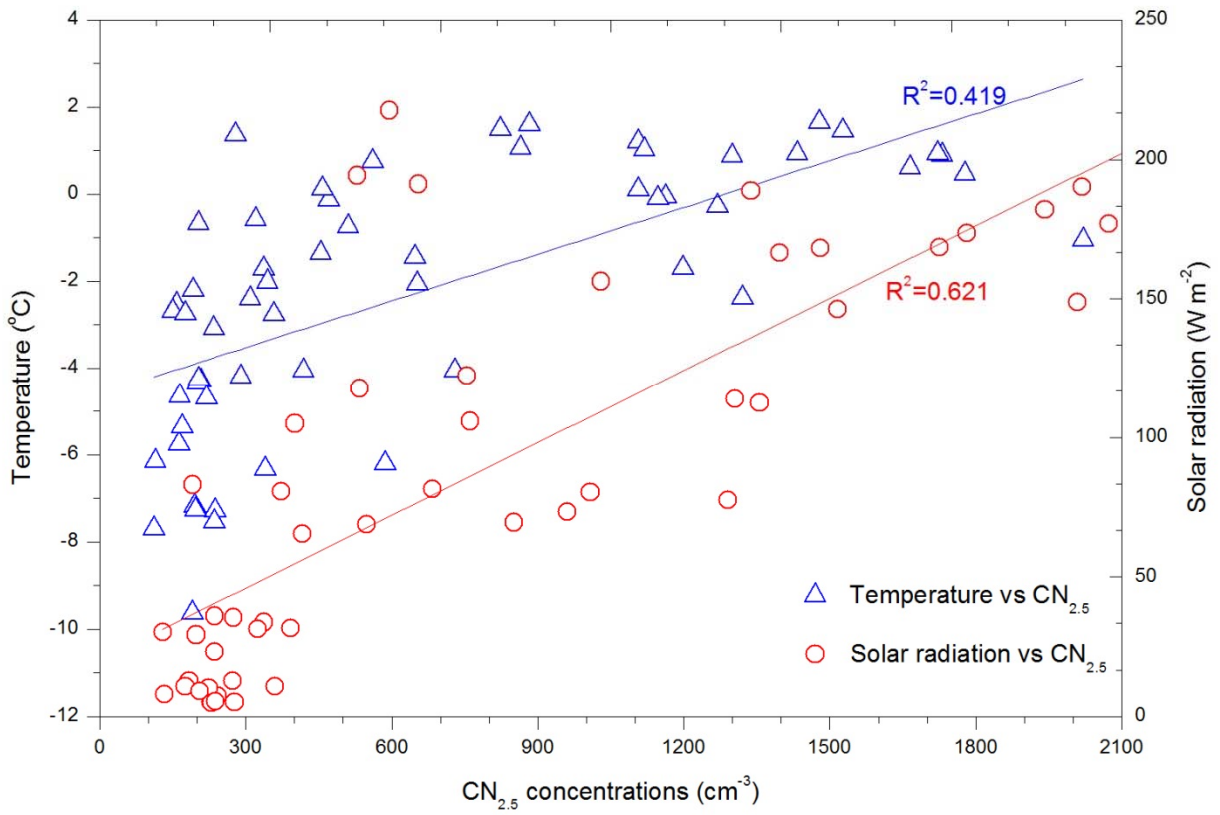


Figure 5. Scatterplot diagram of monthly mean CN_{2.5} concentrations and monthly mean temperature (blue opened triangle) or monthly mean solar radiation intensity (red opened circle). Blue and red solid

5 lines are a regression lines.

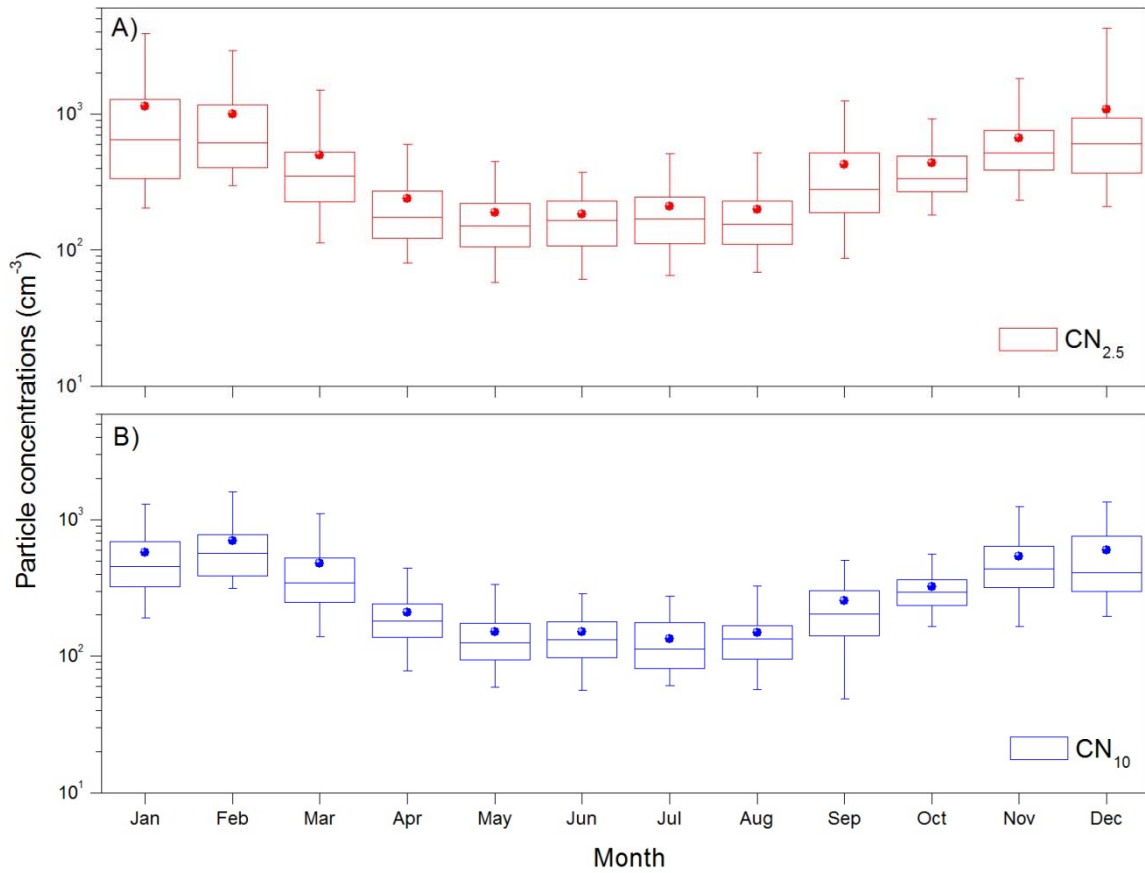


Figure 6. Box plots of seasonality of (a) CN_{2.5} and (b) CN₁₀ concentrations. Lines in the middle of the boxes indicate sample medians (mean value is closed circle), lower and upper lines of the boxes are the 25th and 75th percentiles, and whiskers indicate the 5th and 95th percentiles.

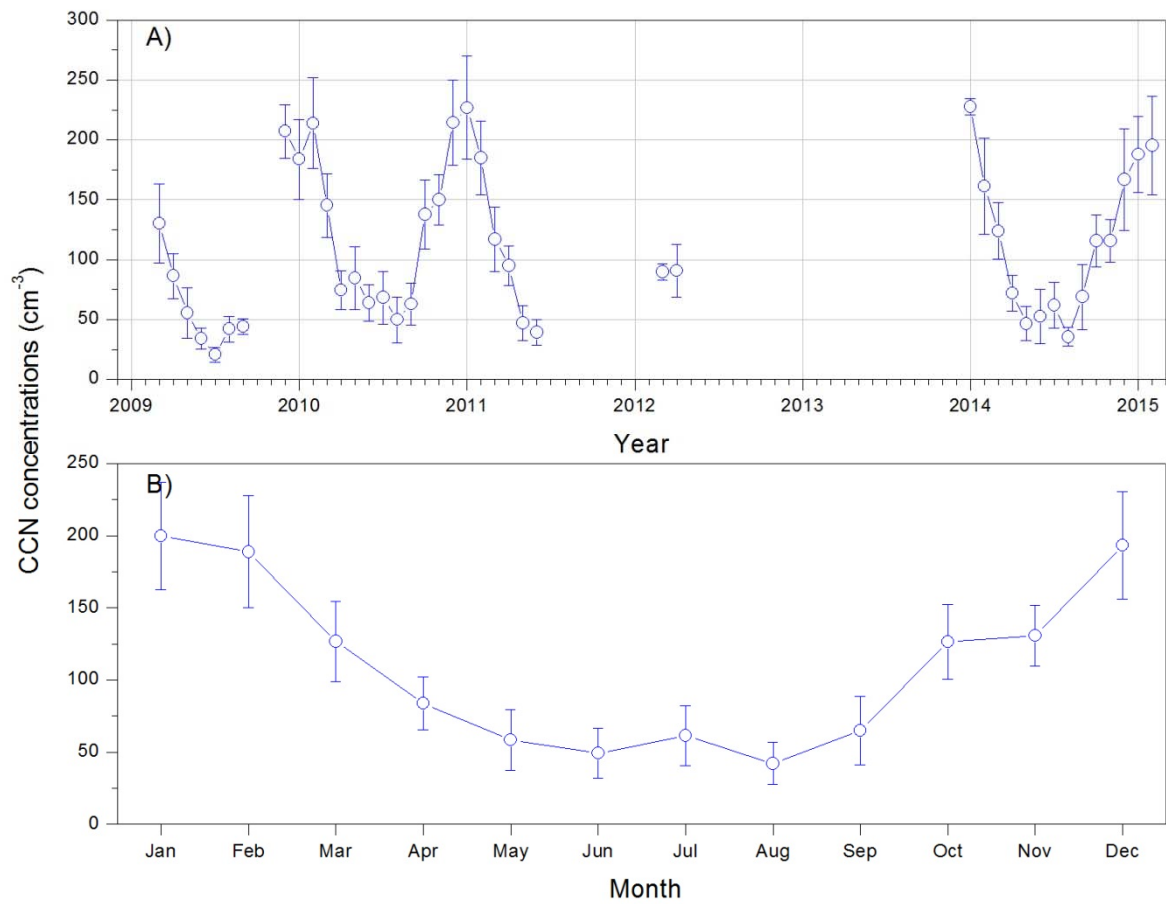


Figure 7. (a) Monthly mean CCN concentrations at the SS of 0.4 % with a standard deviation from March 2009 to February 2015 (b) Seasonal variation of mean CCN concentrations at the SS of 0.4 % with a standard deviation.

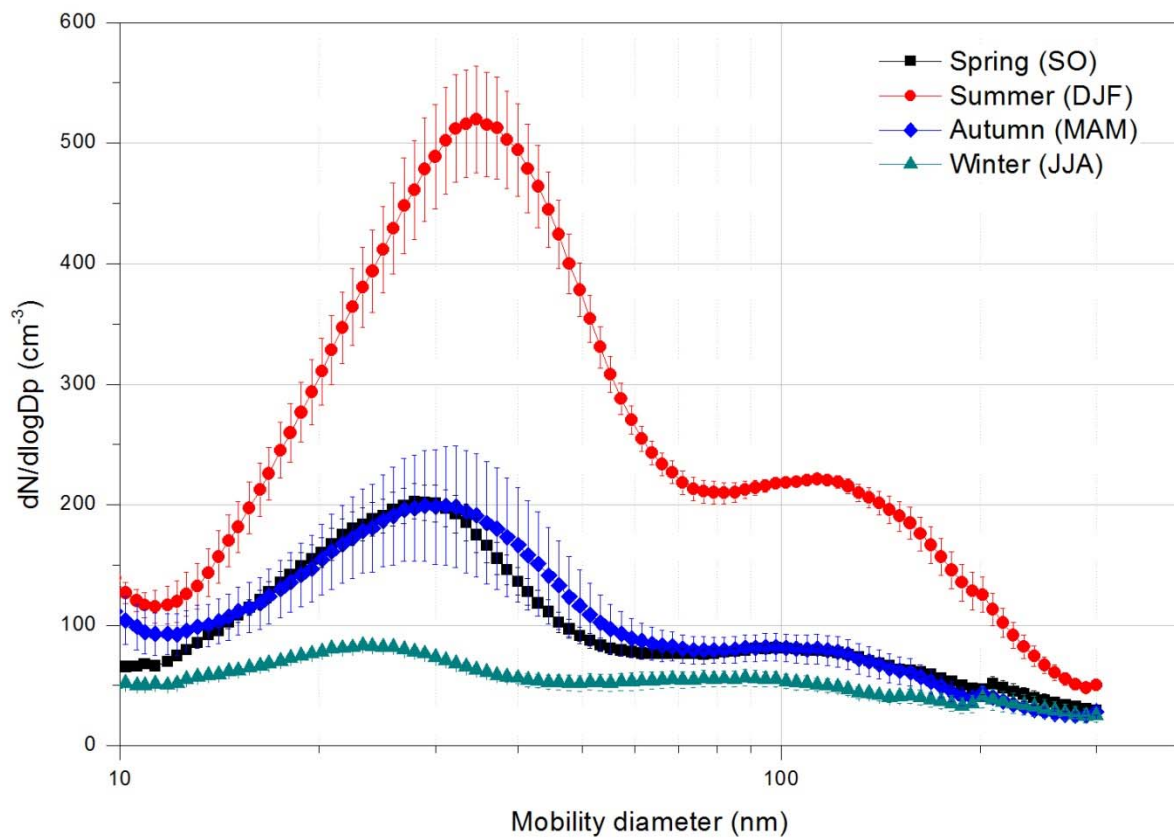


Figure 8. Seasonal mean aerosol size distribution measured by the SMPS at the King Sejong research station over the period from March 2009 to February 2015. The error bars represent the standard deviation of the measurements from the mean value.

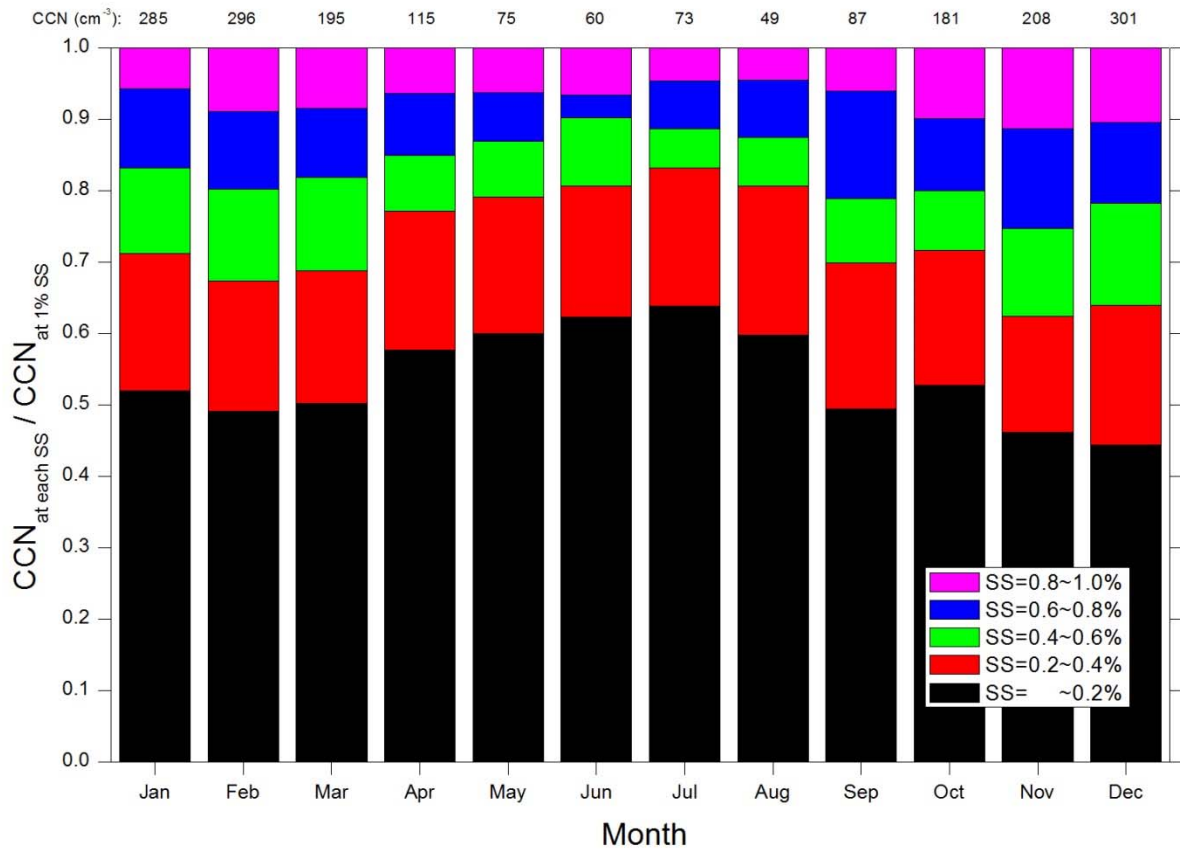


Figure 9. Monthly mean cumulative CCN concentrations shown as fractions of the CCN concentration at the SS of 1.0 %. Colours indicate the SS bins. The number at top of figure represents monthly mean

5 CCN concentrations at the SS values of 1.0 %.

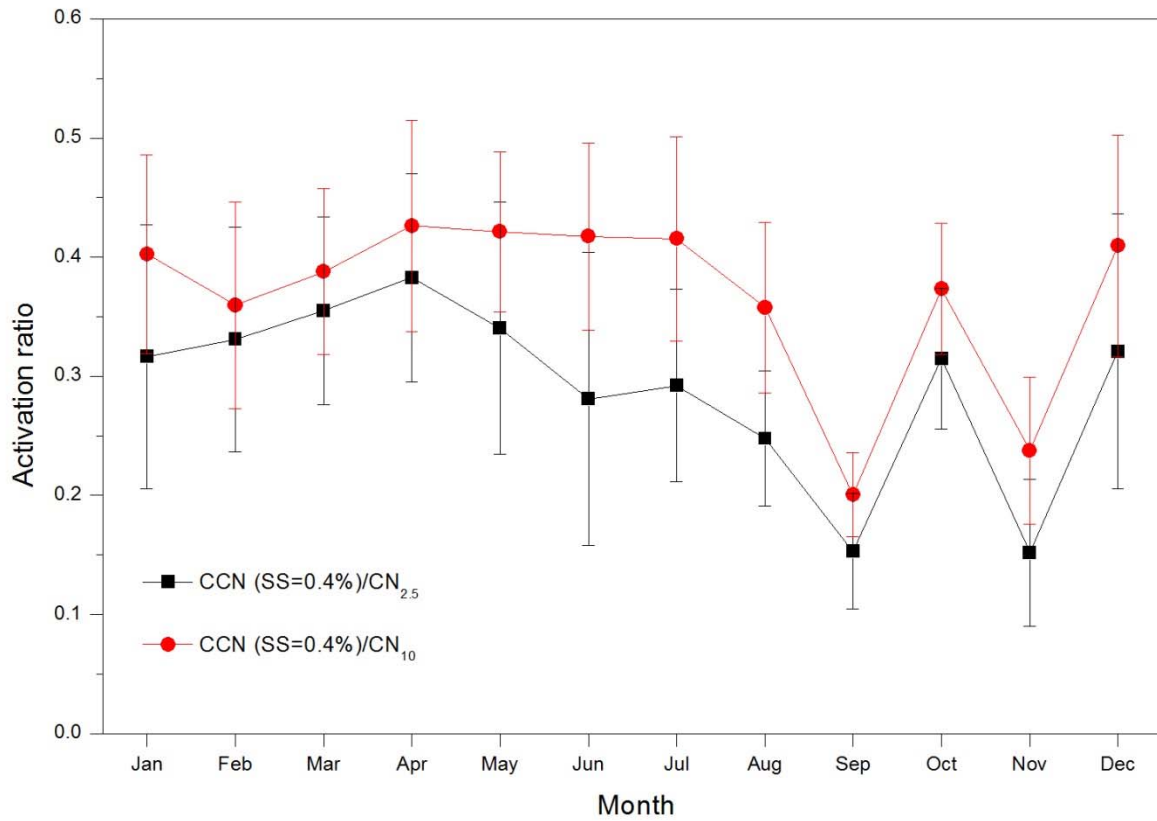


Figure 10. Comparison of the seasonal mean variation of the activation ratio between measurements (CPC 3776 and CPC 3772) by two CPCs. The error bars represent the standard deviation of the measurements from the mean value.

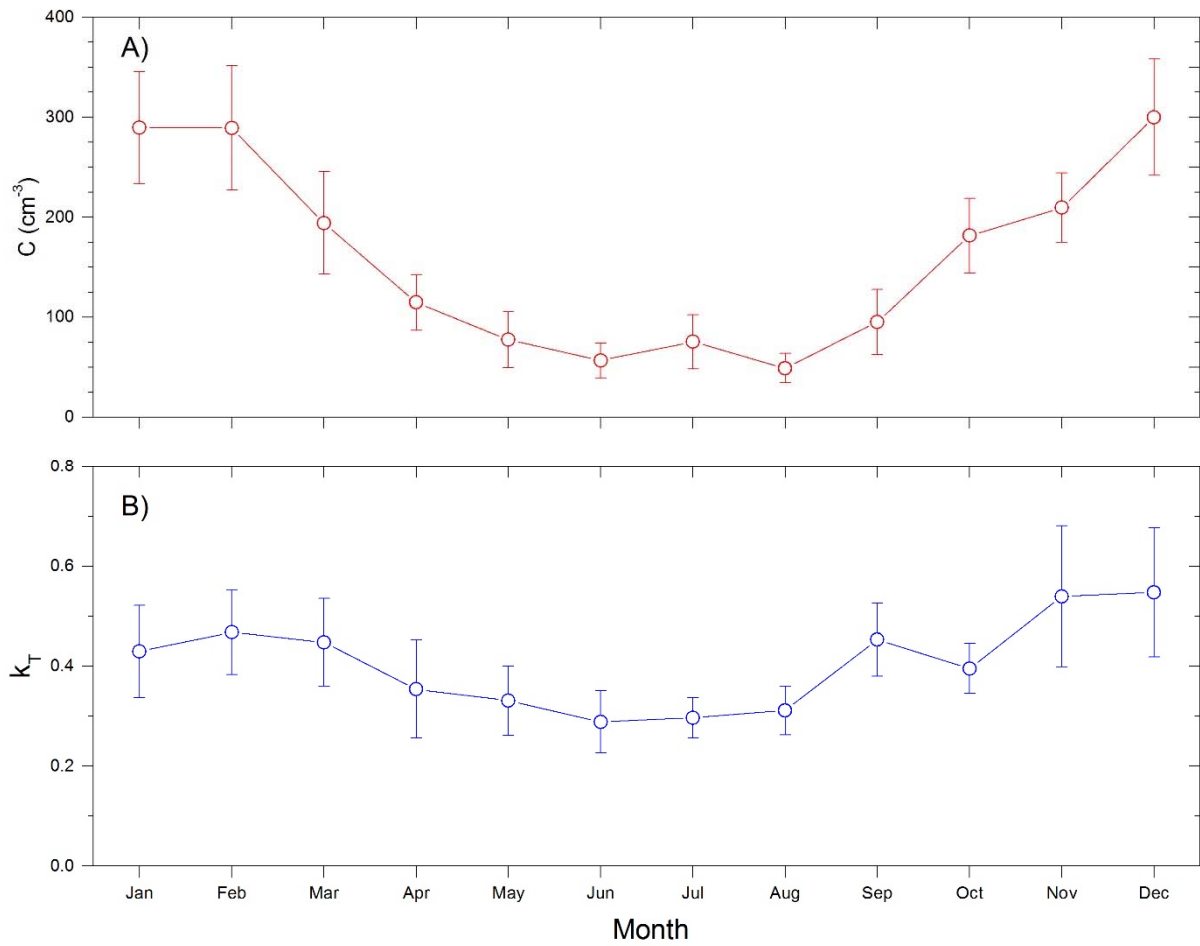


Figure 11. Seasonality of monthly mean values of (a) C and (b) k_T over the whole observation periods.

5 The error bar represents the standard deviation of the measurements from the mean value.

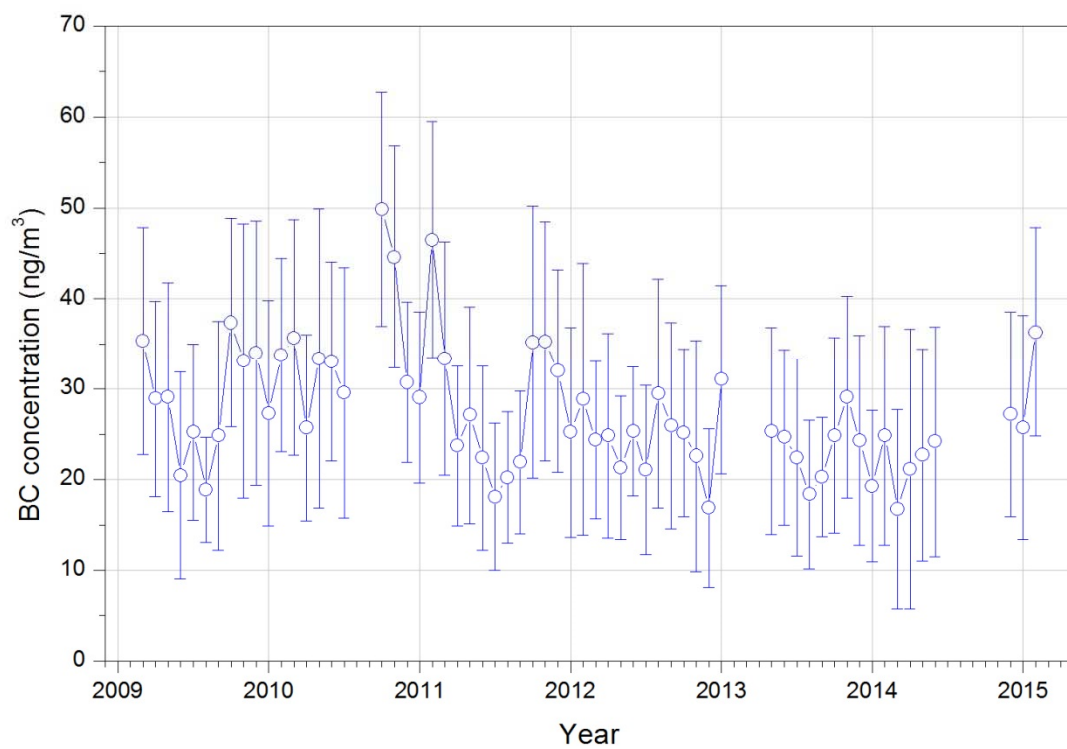


Figure 12. Monthly mean concentrations of black carbon over the period from March 2009 to February 2015. The error bars represent the standard deviation of the measurements from the mean value.

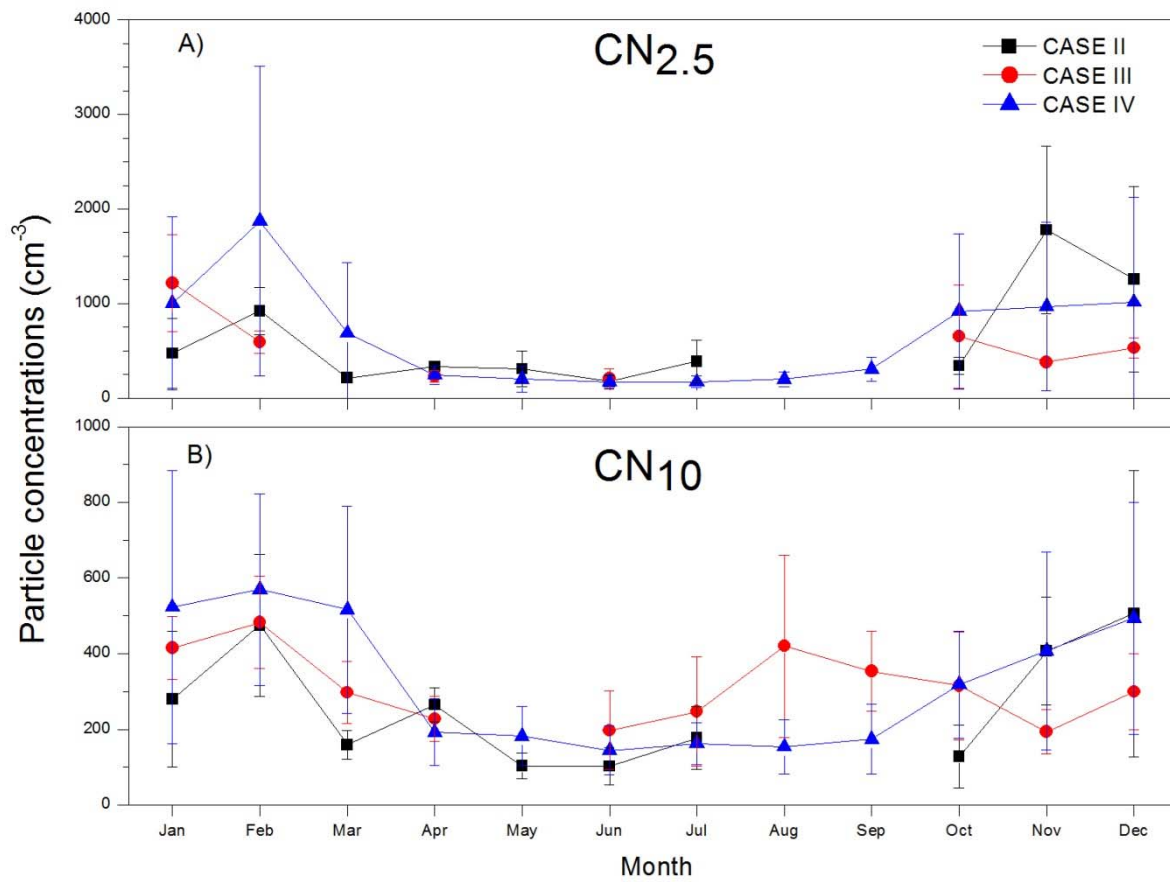


Figure 13. Seasonal variation of mean (a) CN_{2.5} and (b) CN₁₀ concentrations with a standard deviation depending on the air mass origin. The error bars represent the standard deviation of the measurements

5 from the mean value.

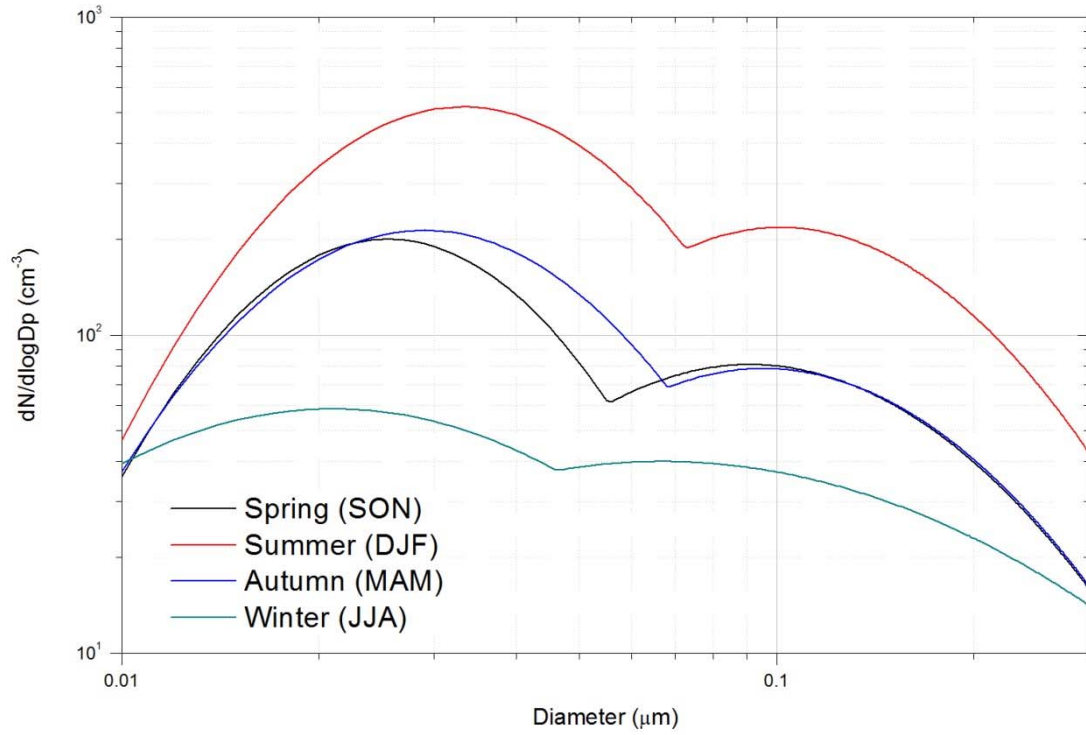


Figure 14. Seasonal lognormally fitted size distribution of aerosol particles originating from the South Pacific Ocean, ranging from 0.01 to 0.3 μm (Case IV).

Table 1. Summary of meteorology and aerosol data according to the origin and transport pathway of aerosol particles. Case I, Case II, Case III, and Case IV refer to the origin and pathway of the air masses from South America, South Atlantic Ocean, Antarctica and South Pacific Ocean, respectively.

	Overall	Case I	Case II	Case III	Case IV
Wind speed (m s⁻¹)	8.4 ± 1.8	2.6 ± 1.1	6.0 ± 1.5	6.7 ± 1.7	8.6 ± 1.8
Wind direction (deg)	237.2 ± 55.8	186.2 ± 20.7	155.9 ± 50.3	206.9 ± 52.3	242.7 ± 55.3
BC concentrations (ng m⁻³)	65.1 ± 29.2	122.2 ± 10.6	36.7 ± 14.2	65.6 ± 30.0	66.5 ± 29.5
CCN concentrations (cm⁻³)	129.7 ± 50.5	212.8 ± 50.2	146.0 ± 50.3	128.9 ± 34.9	128.7 ± 50.8
CN_{2.5} concentrations (cm⁻³)	737.3 ± 849.4	374.9 ± 64.4	605.3 ± 517.6	578.9 ± 377.3	751.2 ± 877.1
CN₁₀ concentrations (cm⁻³)	347.8 ± 229.1	358.8 ± 61.2	268.8 ± 173.9	331.9 ± 133.0	352.2 ± 234.9
Frequency		3	113	118	2407

Table 2. Seasonal size distribution lognormal fitting parameters for the Aitken and Accumulation mode of aerosol particles originating from a Case IV scenario. N, σ , and D_g refer to the number concentrations, a standard deviation, and the geometric mean diameter, respectively.

	Aitken mode			Accumulation mode		
	N (cm ⁻³)	σ	D_g (μ m)	N (cm ⁻³)	σ	D_g (μ m)
Spring (SON)	112.010	1.655	0.026	53.873	1.939	0.094
Summer (DJF)	304.359	1.727	0.034	140.250	1.823	0.109
Autumn (MAM)	118.643	1.764	0.028	50.934	1.901	0.092
Winter (JJA)	49.164	2.296	0.023	44.780	2.827	0.086

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