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

Jaeseok Kim[1], Young Jun Yoon[1,*], Yeontae Gim[1], Hyo Jin Kang[1,2], Jin Hee Choi[1], and Bang Yong Lee[1]

[1]Korea Polar Research Institute, 26 Songdomirae-ro, Yeonsu-gu, Incheon 21990, South Korea

[2]University of Science & Technology (UST), 217 Gajeong-ro, Yuseong-gu, Daejeon 34113, South Korea

*Correspondence to: Y.J. Yoon (yjyoon@kopri.re.kr)

Abstract

Seasonal variability in 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 $CN_{2.5}$ concentration of particles larger than 2.5 nm was highest during the austral summer, with a value of 1080.39 ± 595.05 cm^{-3}, and lowest during the austral winter, with a value of 197.26 ± 71.71 cm^{-3}. A seasonal pattern in the cloud condensation nuclei (CCN) concentration coincides with the CN concentration, where both concentrations are at a minimum in winter and maximum in summer. The measured CCN spectra were approximated by fitting a power law function relating the number of CCN for a given supersaturation (SS) to each SS value, with the fitting coefficients C and k_T. The values for C varied from 6.35 cm^{-3} to 837.24 cm^{-3}, with a mean of 171.48 ± 62.00 cm^{-3}. The values for 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 summer. 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 pathway travelled by the air mass on the physical characteristics of the aerosol particles were also determined. The modal diameter of aerosol particles originating in the South Pacific Ocean showed a seasonal variation, from 0.023 μm in winter to 0.034 μm in summer, and 0.086 μm in winter and 0.109 μm in summer for the Aitken and accumulation modes, respectively.

4

1. Introduction

Aerosol particles in the atmosphere ~~may be~~are 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 ~~(IN)~~ (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 change~~s~~ 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 concentration~~s~~, size ~~distributions~~ distribution, and concentration~~s~~ of black carbon and activated CCN, is necessary.

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

5

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 distribution~~s~~ (Koponen et al., 2003; Belosi et al., 2012), optical propertie~~s~~ (Shaw, 1980; Tomasi et al., 2007; Weller and Lampert, 2008), chemical composition~~s~~ (Virkkula et al., 2006; Weller and Wagenbach, 2007; Asmi et al., 2010; Hara et al., 2011a), and mass concentration~~s~~ (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, research~~es~~ 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 m~~Multi-year observation~~s in Antarctica, especially,~~involving an analysis ~~on~~of the physical characteristics of CCN ~~has been~~are conducted for the first time in ~~the present~~this study.

In ~~this study, we~~The continuous~~ly~~ ~~monitored~~monitoring of the physical characteristics of aerosol particles at the Korean Antarctic station (King Sejong Station) ~~in the Antarctic Peninsula~~from March 2009 to February 2015 are used as the basis for this study. Measurements ~~for~~of the aerosol size distribution and concentration~~s~~ 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 variation~~s~~ of the physical properties of aerosol particles in the Antarctic Peninsula. ~~In addition~~Also, the physical characteristics of aerosol particles that originate~~d~~ 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

Continuous observation~~s~~ 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 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 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 t~~To 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 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.

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 ~~I~~instruments were connected ~~with~~ to the ~~common~~ inlet using conductive tubing to minimize ~~the~~ particle loss~~es~~. The ~~D~~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 condition~~s~~, ~~any drying system~~drying was not ~~used~~ conducted during sampling. The total counting efficiency of the ~~measurement system~~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 line~~s~~, expect for the common inlet, ~~was~~were connected with conductive tubing to minimize ~~the known~~ particle loss~~es~~.

~~Total~~ The total particle number concentration~~s 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

~~lpm~~LPM. The main purpose~~s~~ of measuring the BC concentration~~s 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 concentration~~s~~, 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 ~~lpm~~LPM. The CCN concentration~~s 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 concentration~~s 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.

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 ~~lpm~~LPM. The ratio of aerosol flow to sheath flow of the DMA was 1:10.

~~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

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

Particle Lagrangian Integrated Trajectory (HYSPLIT) 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 mass~~es~~ that were similar for at least 12 h

5   ~~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 mass~~es~~ 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 variation~~s~~ 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 °C and +5.8 °C, with a mean of −2.4 ± 2.1 ºC and ~~the~~ relative humidity (RH) was

15   between 60 ~~%~~ and 100 % , with a mean of 87.9 ± 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 ± 38.9 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

20   ~~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 evident~~a clear seasonal cycle ~~of~~ for CN concentration~~s~~, which ~~are the maximum in the summer~~is highest in the summer (~~from~~ December to February~~,~~ (DJF)) and ~~minimum~~ lowest in the winter (~~from~~ June to August~~,~~ (JJA)). The maximum concentration~~s~~ of particles larger than 2.5 nm ($CN_{2.5}$) and larger than 10 nm (CN_{10}) ~~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 CN_{10} concentration~~s~~ ~~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 concentration~~s over the period from~~ between November 2003 ~~to~~ and January 2007 ~~from observations~~monitoring 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 concentration~~s~~ increase~~s~~ sharply when biological activity ~~is~~ was enhanced due to increasing temperatures and solar radiation (Virkkula et al., 2009). Since ~~our~~ the sampling site for this study was in the Antarctic Peninsula, ~~ocean~~ the biological activity the ocean was considered ~~to be~~as 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 between~~for $CN_{2.5}$ and CN_{10} concentration~~s~~ 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~~are related to secondary aerosol formation caused by biological activity. To better understand the effect of temperature and solar radiation intensity on the $CN_{2.5}$ concentration~~s~~, ~~we compared~~ the relationship between the monthly mean $CN_{2.5}$ concentration~~s~~ and solar radiation intensity, and the monthly mean $CN_{2.5}$ concentration~~s~~ and temperature was

5   compared. The correlation coefficient between $CN_{2.5}$ and the solar radiation intensity (opened circle; $R^2=0.621$) was slightly higher than ~~that between~~for the mean $CN_{2.5}$ concentration and temperature (opened triangle; $R^2=0.419$), as shown in Fig. 5. ~~Our~~These results suggest that the $CN_{2.5}$ concentration~~s~~ may be more closely coupled with solar radiation intensity than ~~with~~ temperature.

Unique results ~~of~~for the $CN_{2.5}$ concentration~~s~~ were observed as shown in Fig. 4. The $CN_{2.5}$

10   concentrations in the summer ~~season~~ of 2013-2014 were much lower than other years. Unfortunately, the reason for the lower $CN_{2.5}$ concentration~~s~~ ~~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 mass~~es~~ reached ~~to~~ the sampling site. Although the air mass that originated ~~from~~in the South Pacific

15   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 CN_{10} ~~n~~or SMPS data ~~are~~is available for the

20   austral summer season of 2013-2014 because of mechanical failures~~,~~. Thus, it is not possible to directly explain the low concentration~~s~~ ~~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

25   summer season of 2013-2014. These CN concentration~~s~~ ~~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 concentration~~s~~ from December 2003 to January 2007. Although there was a variation of the CN concentration~~s~~ ~~year by year~~between each year, the daily CN concentration~~s~~ during the astral summer ~~period were~~was ~ 600 cm^{-3}.

A more detailed comparison of the monthly trends ~~in~~ for the CN$_{2.5}$ and CN$_{10}$ concentration~~s~~ is presented in Fig. 6. The monthly mean CN concentration~~s~~ 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 concentration~~s~~ sharply decreased from March and remained stable from April to August. In particular, the CN$_{2.5}$ concentrations during the summer period increased sharply compared to the CN$_{10}$ concentration~~s,~~. ~~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 concentration~~s~~ 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 concentration~~s~~ ~~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 concentration~~s~~ ~~at~~ for each supersaturation. The relationship between the CDNC and CCN concentration~~s~~ at ~~the~~ a SS value of 0.4% was approximately linear~~,~~. Furthermore, ~~while~~ CCN concentration~~s~~ 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 result~~s~~, ~~in this study, the~~ supersaturation of 0.4% was chosen to investigate

seasonal variation~~s~~ of the CCN. ~~We found m~~Monthly variation~~s~~ in the CCN concentration~~s~~ was found, with ~~the~~ a maximum value~~s being~~ observed during the summer period~~s~~ (DJF) and ~~the~~ a minimum concentration~~s were~~ observed during ~~the~~ winter period~~s~~ (JJA). The monthly mean CCN concentration~~s~~ ~~were in the range of~~ranged between 20.63 cm^{-3} in July 2009 and 227.52 cm^{-3} in January 2014, with a

5   mean of 112.80 ± 39.05 cm^{-3}. Fig. 7(b) also shows seasonality ~~in~~ for the CCN concentration~~s~~ ~~at an~~with a SS value of 0.4~~ ~~%. The CCN concentration~~s~~ gradually decreased from February and remained stable during the winter, while the CCN concentration~~s~~ 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 concentration~~s~~

10   follows the seasonal variation ~~march~~ of the CN concentration~~s~~. As shown in Fig. 6, the CN$_{10}$ concentration~~s~~ as well as CN$_{2.5}$ concentration~~s~~ increased during ~~the~~ summer. In addition, the aerosol size distribution~~s~~ measured by the SMPS showed that the concentration~~s~~ ~~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

15   al., 2006), hence CCN concentrations increase during ~~the~~ summer and decrease during ~~the~~ winter.

       In order to investigate the seasonal variation~~s~~ of fractions of the CCN concentration~~s~~ ~~at~~ for each SS value ~~in~~ for the CCN concentration~~s~~ 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

20   CCN concentrations were estimated by dividing the CCN concentration~~s~~ at each SS value by the total CCN concentration~~s~~ at ~~the~~ a SS of 1.0~~ ~~%. Although a clear seasonal trend ~~of~~ for CCN concentration~~s~~ with a maximum during the summer and a minimum during the winter was presented~~, as mentioned earlier~~, the fraction of CCN concentration~~s~~ at ~~the~~ a SS value of 0.2~~ ~~% ~~in~~ for the activated CCN concentration~~s~~ showed a different pattern with a maximum value in July and a minimum value in

25   December, as shown in Fig. 9. The numbers at the top of Fig. 9 represent the mean CCN concentration~~s~~

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 variation~~s~~ in the mean activation ratio of the CCN concentration~~s~~ at a SS of 0.4 % to the CN concentration~~s~~ measured ~~from~~ by the two CPCs (TSI 3776 and 3772)~~, as~~ are shown in Fig. 10. The mean value~~s~~ of the activation ratios of CCN/CN$_{2.5}$ and CCN/CN$_{10}$ 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/CN$_{10}$) was similar regardless of seasonality. The reason that no clear change is observed in the activation ratio~~s~~ at ~~the~~ King Sejong Station ~~in the Antarctic Peninsula,~~ might be the variation ~~of~~ in the concentration~~s~~ of accumulation mode particles, as can be seen in Fig. 8. The lower activation ratio~~s~~ 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 concentration~~s~~ of Aitken mode aerosol particles increased sharply compared to ~~their~~ the concentration~~s~~ 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):

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

25   where N$_{CCN}$ is the concentration of CCN at given a supersaturation value~~s~~ (SS), C and k_T are coefficient

constants estimated from CCN spectra. The correlation coefficient between the measured CCN concentration~~s~~ at ~~the~~ a SS of 0.4% and the ~~power-law fit~~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 value~~s~~ ~~can be~~are defined ~~by Petters and Kreidenweis (2007)~~ as

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

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)

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

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 concentration~~s~~. In this study, the kappa values were estimated using ~~the~~ a monthly mean CCN concentration~~s~~ at ~~the~~ a SS of 0.4%, the monthly mean CN concentrations measured by the CPC, 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 Artic and ~~subarctic~~ Subarctic areas. For example, Lathem 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 day~~s~~ 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 subarctic~~ Subarctic area ~~in~~ of Finland (Pallas-Sodankylä station).

3.2.3 Black carbon (BC) concentrations

Fig. 12 shows the variation~~s~~ of monthly mean BC concentrations over the whole sampling period. To eliminate effect of local pollution on observation~~s, in this study,~~ from this study, data, where BC concentrations were higher than 100 ng m^{-3}, were discarded. The daily mean BC concentration~~s~~ varied between 1.07 ng m^{-3} and 75.97 ng m^{-3}, with a mean of 27.43 ± 4.98 ng m^{-3}. The BC concentration~~s~~ 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 concentration~~s~~ 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 Hall~~e~~ey ~~station~~ Station and the South Pole with a Aethalometer. They found that although BC concentrations varied depending on the sampling site, ~~the~~ BC concentration~~s~~ 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 mass~~es~~.

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 characteristic~~s~~ of the aerosol particles is presented. As mentioned earlier in ~~Sec.~~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 concentration~~s (Table 1)~~. This ~~might~~ may be due to anthropogenic influences at the source and the aging of aerosol particles. The CN$_{10}$ concentration~~s~~ ~~were~~ was similar regardless of the origin and pathway of air mass~~es~~, whereas an enhancement of the CN$_{2.5}$ concentration~~s~~ 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 concentration~~s~~ based on ~~the~~ an air mass analysis is shown in Fig. 13. It should be noted that the mean

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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

5   aerosol particles for ~~(~~CN2.5 and CN10~~)~~ originat~~eing~~ from the ocean (Case II and IV). These are ~~were~~ observed from April to September, whereas the concentrations of aerosol particles (CN2.5) 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 CN2.5 concentrations had discrepancies ~~in accordance~~ with the air mass history. This is probably due to a

10   difference in the chemical compounds that contribute~~d~~ to aerosol formation processes and/or ~~in~~ variations ~~of~~ in biogenic activity according to the origin and transport pathway of the air mass~~es~~. 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

15   ~~A~~aerosol size distribution parameters were fitted to a log-normal distribution ~~were~~ derived for each season. The seasonally averaged log-normal aerosol size distribution~~s~~ ~~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

20   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 ocean~~s~~ as a product of biological activity. The spring and autumn season~~s~~ show~~s~~

25   intermediate values. ~~Our~~These results are similar to those of previous laboratory and field experiments

(Sellegri 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).

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 concentration~~s~~ (CN) exists, with ~~the~~ maximum concentration~~s~~ in the austral summer (DJF) and ~~the~~ minimum concentration~~s~~ in the austral winter (JJA). The maximum CN concentration~~s~~ of particles larger than 2.5 nm ($CN_{2.5}$) and 10 nm (CN_{10}) ~~were~~ was approximately 2000 cm^{-3} in December 2012 and about 800 cm^{-3} in December 2009 and February 2015, respectively. In particular, the $CN_{2.5}$ concentration~~s~~ increased sharply during the summer compared to CN_{10} concentration~~s~~, 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 concentration~~s~~ at ~~the~~ a supersaturation (SS) of 0.4% were presented. The maximum mean CCN concentration of 199.89 ± 37.07 cm^{-3} was measured in January and the minimum mean CCN concentration was 42.13 ± 14.51 cm^{-3} in August. The activation ratio (CCN/CN_{10}) 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) (Lathem 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 value~~s~~ of C varied between 6.35 cm^{-3} and 837.24 cm^{-3}, with a mean of 171.48 ± 62.00 cm^{-3}. The value~~s~~

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 ~~the~~ summer period~~s~~ (DJF) were higher than those during the winter period~~s~~ (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 mass~~es 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 CN_{10} 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 $CN_{2.5}$ concentration~~s~~ show~~ed~~ 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.

References

Anttila, T., Brus, D., Jaatinen, A., Hyvärinen, A. P., Kivekäs, N., Romakkaniemi, S., Komppula, M., and Lihavainen, H.: Relationships between particles, cloud condensation nuclei and cloud droplet activation during the third Pallas Cloud Experiment, Atmos. Chem. Phys., 12, 11435-11450, 10.5194/acp-12-11435-2012, 2012.

Arimoto, R., Hogan, A., Grube, P., Davis, D., Webb, J., Schloesslin, C., Sage, S., and Raccah, F.: Major ions and radionuclides in aerosol particles from the South Pole during ISCAT-2000, Atmos. Environ., 38, 5473-5484, 10.1016/j.atmosenv.2004.01.049, 2004.

Asmi, E., Frey, A., Virkkula, A., Ehn, M., Manninen, H. E., Timonen, H., Tolonen-Kivimaki, O., Aurela, M., Hillamo, R., and Kulmala, M.: Hygroscopicity and chemical composition of antarctic sub-micrometre aerosol particles and observations of new particle formation, Atmos. Chem. Phys., 10, 4253-4271, 10.5194/acp-10-4253-2010, 2010.

Baron, P. A., and Willeke, K.: Aerosol measurement: principles, techniques, and applications, edited by: 2nd, Wiley-Interscience, New York, 2001.

Bigg, E. K., Gras, J. L., and Evans, C.: Origin of Aitken particles in remote regions of the Southern Hemisphere, J. Atmos. Chem., 1, 203-214, 10.1007/bf00053841, 1984.

Belosi, F., Contini, D., Donateo, A., Santachiara, G., and Prodi, F.: Aerosol size distribution at Nansen Ice Sheet Antarctica, Atmos. Res., 107, 42-50, 10.1016/j.atmosres.2011.12.007, 2012.

Bodhaine, B. A.: Aerosol absorption measurements at Barrow, Mauna Loa and the south pole, J. Geophys. Res., 100, 8967-8975, 10.1029/95jd00513, 1995.

Chen, J. L., Wilson, C. R., Blankenship, D., and Tapley, B. D.: Accelerated Antarctic ice loss from satellite gravity measurements, Nat. Geosci., 2, 859-862, 10.1038/ngeo694, 2009.

Choi, T., Lee, B. Y., Kim, S. J., Yoon, Y. J., and Lee, H. C.: Net radiation and turbulent energy

exchanges over a non-glaciated coastal area on King George Island during four summer seasons, Antarct. Sci., 20, 99-111, 10.1017/s095410200700082x, 2008.

Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Borrmann, S., and Andreae, M. O.: Size matters more than chemistry for cloud-nucleating ability of aerosol particles, Science, 312, 1375-1378, 10.1126/science.1125261, 2006.

Fattori, I., Becagli, S., Bellandi, S., Castellano, E., Innocenti, M., Mannini, A., Severi, M., Vitale, V., and Udisti, R.: Chemical composition and physical features of summer aerosol at Terra Nova Bay and Dome C, Antarctica, J. Environ. Monit., 7, 1265-1274, 10.1039/b507327h, 2005.

Fiebig, M., Hirdman, D., Lunder, C. R., Ogren, J. A., Solberg, S., Stohl, A., and Thompson, R. L.: Annual cycle of Antarctic baseline aerosol: Controlled by photooxidation-limited aerosol formation, Atmos. Chem. Phys., 14, 3083-3093, 10.5194/acp-14-3083-2014, 2014.

Gras, J. L.: Condensation nucleus size distribution at Mawson, Antarctica: Microphysics and chemistry, Atmos. Environ., 27A, 1427-1434, 10.1016/0960-1686(93)90128-l, 1993.

Hamed, A., Joutsensaari, J., Mikkonen, S., Sogacheva, L., Dal Maso, M., Kulmala, M., Cavalli, F., Fuzzi, S., Facchini, M. C., Decesari, S., Mircea, M., Lehtinen, K. E. J., and Laaksonen, A.: Nucleation and growth of new particles in Po Valley, Italy, Atmos. Chem. Phys., 7, 355-376, 2007.

Hansen, A. D. A., Lowenthal, D. H., Chow, J. C., and Watson, J. G.: Black carbon aerosol at McMurdo Station, Antarctica, J. Air Waste Manage. Assoc., 51, 593-600, 2001.

Hara, K., Hirasawa, N., Yamanouchi, T., Wada, M., and Herber, A.: Spatial distributions and mixing states of aerosol particles in the summer Antarctic troposphere, Antarctic Record, 54, 704-730, 2010.

Hara, K., Osada, K., Nishita-Hara, C., Yabuki, M., Hayashi, M., Yamanouchi, T., Wada, M., and Shiobara, M.: Seasonal features of ultrafine particle volatility in the coastal Antarctic troposphere, Atmos. Chem. Phys., 11, 9803-9812, 10.5194/acp-11-9803-2011, 2011a.

Hara, K., Osada, K., Nishita-Hara, C., and Yamanouchi, T.: Seasonal variations and vertical features of aerosol particles in the Antarctic troposphere, Atmos. Chem. Phys., 11, 5471-5484, 10.5194/acp-11-5471-2011, 2011b.

Hinds, W. C.: Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles, edited by: 2nd, Wiley-Interscience, New York, 1999.

IPCC: Climate change 2013: The physical science basis, Intergovernmental panel on Climate Change, Cambridge University Press, New York, USA, 571-740, 2013.

Ito, T.: Study of background aerosols in the Antarctic troposphere, J. Atmos. Chem., 3, 69-91, 10.1007/bf00049369, 1985.

Jaatinen, A., Romakkaniemi, S., Hao, L. Q., Kortelainen, A., Miettinen, P., Mikkonen, S., Smith, J. N., Virtanen, A., Laaksonen, A., Anttila, T., and Hyvärinen, A. P.: The third Pallas Cloud Experiment: Consistency between the aerosol hygroscopic growth and CCN activity, Boreal Environ. Res., 19, 368-382, 2014.

Jaenicke, R., Dreiling, V., Lehmann, E., Koutsenoguii, P. K., and Stingl, J.: Condensation nuclei at the German Antarctic Station "Georg von Neumayer", Tellus Ser. B, 44 B, 311-317, 1992.

Järvinen, E., Virkkula, A., Nieminen, T., Aalto, P. P., Asmi, E., Lanconelli, C., Busetto, M., Lupi, A., Schioppo, R., Vitale, V., Mazzola, M., Petäjä, T., Kerminen, V. M., and Kulmala, M.: Seasonal cycle and modal structure of particle number size distribution at Dome C, Antarctica, Atmos. Chem. Phys., 13, 7473-7487, 10.5194/acp-13-7473-2013, 2013.

Kammermann, L., Gysel, M., Weingartner, E., Herich, H., Cziczo, D. J., Holst, T., Svenningsson, B., Arneth, A., and Baltensperger, U.: Subarctic atmospheric aerosol composition: 3. Measured and modeled properties of cloud condensation nuclei, J. Geophys. Res.: Atmos., 115, 10.1029/2009jd012447, 2010.

Kim, G., Cho, H. J., Seo, A., Kim, D., Gim, Y., Lee, B. Y., Yoon, Y. J., and Park, K.: Comparison of Hygroscopicity, Volatility, and Mixing State of Submicrometer Particles between Cruises over the
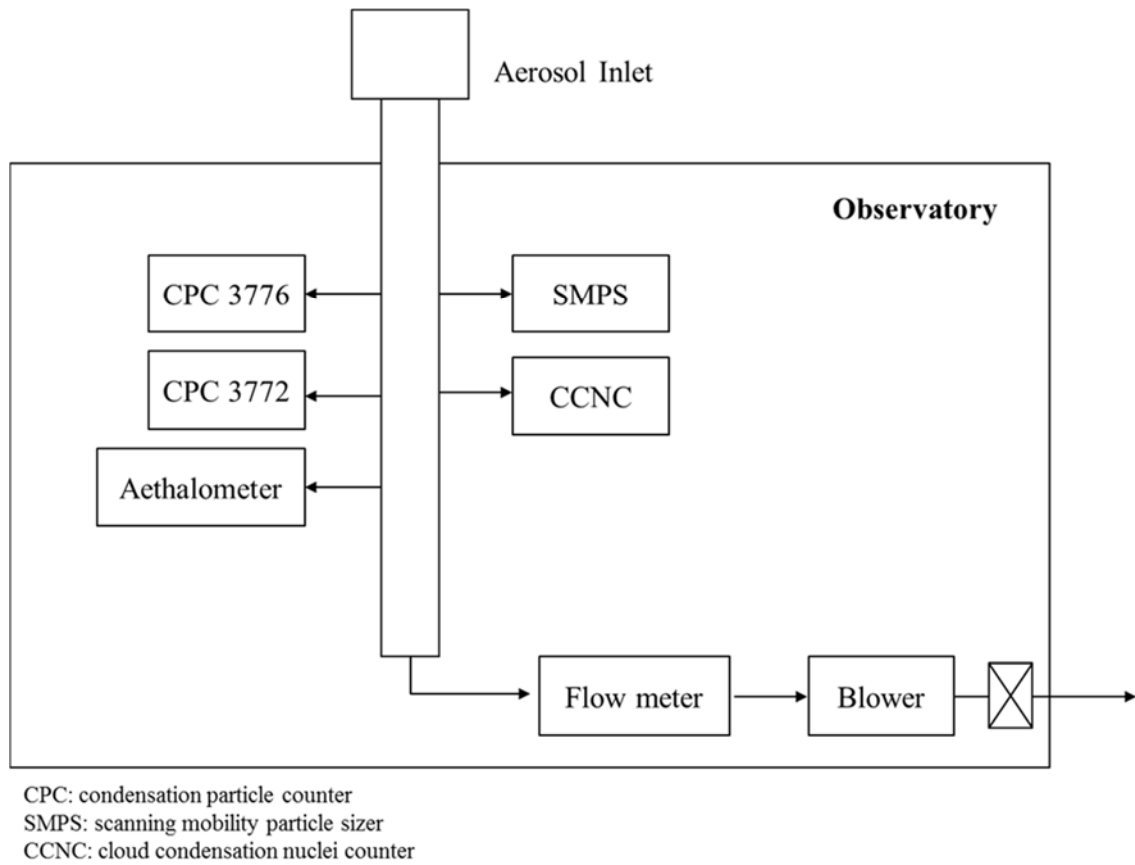
Arctic Ocean and the Pacific Ocean, Environ. Sci. Technol., 49, 12024-12035, 10.1021/acs.est.5b01505, 2015.

Koponen, I. K., Virkkula, A., Hillamo, R., Kerminen, V. M., and Kulmala, M.: Number size distributions and concentrations of the continental summer aerosols in Queen Maud Land, Antarctica, J. Geophys. Res. D: Atmos., 108, AAC 8-1 - AAC 8-10, 2003.

Kwon, T. Y., and Lee, B. Y.: Precipitation anomalies around King Sejong Station, Antarctica associated with El Niño/Southern Oscillation, Ocean and Polar Research, 24, 19-31, 2002.

Kyrö, E. M., Kerminen, V. M., Virkkula, A., Dal Maso, M., Parshintsev, J., Ruíz-Jimenez, J., Forsström, L., Manninen, H. E., Riekkola, M. L., Heinonen, P., and Kulmala, M.: Antarctic new particle formation from continental biogenic precursors, Atmos. Chem. Phys., 13, 3527-3546, 10.5194/acp-13-3527-2013, 2013.

Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V. M., Birmili, W., and McMurry, P. H.: Formation and growth rates of ultrafine atmospheric particles: A review of observations, J. Aerosol. Sci., 35, 143-176, 2004.

Lathem, T. L., Beyersdorf, A. J., Thornhill, K. L., Winstead, E. L., Cubison, M. J., Hecobian, A., Jimenez, J. L., Weber, R. J., Anderson, B. E., and Nenes, A.: Analysis of CCN activity of Arctic aerosol and Canadian biomass burning during summer 2008, Atmos. Chem. Phys., 13, 2735-2756, 10.5194/acp-13-2735-2013, 2013.

Leena, P. P., Pandithurai, G., Anilkumar, V., Murugavel, P., Sonbawne, S. M., and Dani, K. K.: Seasonal variability in aerosol, CCN and their relationship observed at a high altitude site in Western Ghats, Meteorology and Atmospheric Physics, 128, 143-153, 10.1007/s00703-015-0406-0, 2016.

Martin, M., Chang, R. Y. W., Sierau, B., Sjogren, S., Swietlicki, E., Abbatt, J. P. D., Leck, C., and Lohmann, U.: Cloud condensation nuclei closure study on summer arctic aerosol, Atmos. Chem. Phys., 11, 11335-11350, 10.5194/acp-11-11335-2011, 2011.

Mazzera, D. M., Lowenthal, D. H., Chow, J. C., and Watson, J. G.: Sources of PM10 and sulfate aerosol at McMurdo station, Antarctica, Chemosphere, 45, 347-356, 10.1016/s0045-6535(00)00591-9, 2001.

Mishra, V. K., Kim, K. H., Hong, S., and Lee, K.: Aerosol composition and its sources at the King Sejong Station, Antarctic peninsula, Atmos. Environ., 38, 4069-4084, 10.1016/j.atmosenv.2004.03.052, 2004.

O'Dowd, C. D., Facchini, M. C., Cavalli, F., Ceburnis, D., Mircea, M., Decesari, S., Fuzzi, S., Young, J. Y., and Putaud, J. P.: Biogenically driven organic contribution to marine aerosol, Nature, 431, 676-680, 10.1038/nature02959, 2004.

Pant, V., Siingh, D., and Kamra, A. K.: Size distribution of atmospheric aerosols at Maitri, Antarctica, Atmos. Environ., 45, 5138-5149, 2011.

Paramonov, M., Kerminen, V. M., Gysel, M., Aalto, P. P., Andreae, M. O., Asmi, E., Baltensperger, U., Bougiatioti, A., Brus, D., Frank, G. P., Good, N., Gunthe, S. S., Hao, L., Irwin, M., Jaatinen, A., Jurányi, Z., King, S. M., Kortelainen, A., Kristensson, A., Lihavainen, H., Kulmala, M., Lohmann, U., Martin, S. T., McFiggans, G., Mihalopoulos, N., Nenes, A., O'Dowd, C. D., Ovadnevaite, J., Petäjä, T., Pöschl, U., Roberts, G. C., Rose, D., Svenningsson, B., Swietlicki, E., Weingartner, E., Whitehead, J., Wiedensohler, A., Wittbom, C., and Sierau, B.: A synthesis of cloud condensation nuclei counter (CCNC) measurements within the EUCAARI network, Atmos. Chem. Phys., 15, 12211-12229, 10.5194/acp-15-12211-2015, 2015.

Park, J., Sakurai, H., Vollmers, K., and McMurry, P. H.: Aerosol size distributions measured at the South Pole during ISCAT, Atmos. Environ., 38, 5493-5500, 10.1016/j.atmosenv.2002.12.001, 2004.

Pereira, E. B., Evangelista, H., Pereira, K. C. D., Cavalcanti, I. F. A., and Setzer, A. W.: Apportionment of black carbon in the South Shetland Islands, Antartic Peninsula, J. Geophys. Res.: Atmos., 111, 10.1029/2005jd006086, 2006.

26

Petters, M. D., and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961-1971, 2007.

Pierce, J. R., and Adams, P. J.: Efficiency of cloud condensation nuclei formation from ultrafine particles, Atmos. Chem. Phys., 7, 1367-1379, 2007.

Pritchard, H. D., Ligtenberg, S. R. M., Fricker, H. A., Vaughan, D. G., Van Den Broeke, M. R., and Padman, L.: Antarctic ice-sheet loss driven by basal melting of ice shelves, Nature, 484, 502-505, 10.1038/nature10968, 2012.

Rankin, A. M., and Wolff, E. W.: A year-long record of size-segregated aerosol composition at Halley, Antarctica, J. Geophys. Res. D: Atmos., 108, AAC 9-1 - AAC 9-12, 2003.

Rignot, E., Casassa, G., Gogineni, P., Krabill, W., Rivera, A., and Thomas, R.: Accelerated ice discharge from the Antarctic Peninsula following the collapse of Larsen B ice shelf, Geophys. Res. Lett., 31, L18401 18401-18404, 10.1029/2004gl020697, 2004.

Roscoe, H. K., Jones, A. E., Brough, N., Weller, R., Saiz-Lopez, A., Mahajan, A. S., Schoenhardt, A., Burrows, J. P., and Fleming, Z. L.: Particles and iodine compounds in coastal Antarctica, J. Geophys. Res. D: Atmos., 120, 7144-7156, 10.1002/2015jd023301, 2015.

Schneider, D. P., Deser, C., and Okumura, Y.: An assessment and interpretation of the observed warming of West Antarctica in the austral spring, Clim. Dyn., 38, 323-347, 10.1007/s00382-010-0985-x, 2012.

Sellegri, K., O'Dowd, C. D., Yoon, Y. J., Jennings, S. G., and de Leeuw, G.: Surfactants and submicron sea spray generation, J. Geophys. Res.: Atmos., 111, 10.1029/2005jd006658, 2006.

Shaw, G. E.: Optical, chemical and physical properties of aerosols over the antarctic ice sheet, Atmos. Environ., 14, 911-921, 10.1016/0004-6981(80)90004-9, 1980.

Steig, E. J., Schneider, D. P., Rutherford, S. D., Mann, M. E., Comiso, J. C., and Shindell, D. T.: Warming of the Antarctic ice-sheet surface since the 1957 International Geophysical Year, Nature, 457, 459-462, 10.1038/nature07669, 2009.

Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., and Ngan, F.: Noaa's hysplit atmospheric transport and dispersion modeling system, Bull. Amer. Meteorol. Soc., 96, 2059-2077, 10.1175/bams-d-14-00110.1, 2015.

Tomasi, C., Vitale, V., Lupi, A., Di Carmine, C., Campanelli, M., Herber, A., Treffeisen, R., Stone, R. S., Andrews, E., Sharma, S., Radionov, V., von Hoyningen-Huene, W., Stebel, K., Hansen, G. H., Myhre, C. L., Wehrli, C., Aaltonen, V., Lihavainen, H., Virkkula, A., Hillamo, R., Ström, J., Toledano, C., Cachorro, V. E., Ortiz, P., de Frutos, A. M., Blindheim, S., Frioud, M., Gausa, M., Zielinski, T., Petelski, T., and Yamanouchi, T.: Aerosols in polar regions: A historical overview based on optical depth and in situ observations, J. Geophys. Res.: Atmos., 112, 10.1029/2007jd008432, 2007.

Tunved, P., Ström, J., and Krejci, R.: Arctic aerosol life cycle: Linking aerosol size distributions observed between 2000 and 2010 with air mass transport and precipitation at Zeppelin station, Ny-Ålesund, Svalbard, Atmos. Chem. Phys., 13, 3643-3660, 10.5194/acp-13-3643-2013, 2013.

Twomey, S.: The nuclei of natural cloud formation part II: The supersaturation in natural clouds and the variation of cloud droplet concentration, Geofis. Pura Appl., 43, 243-249, 10.1007/bf01993560, 1959.

Vaughan, D. G., Marshall, G. J., Connolley, W. M., Parkinson, C., Mulvaney, R., Hodgson, D. A., King, J. C., Pudsey, C. J., and Turner, J.: Recent rapid regional climate warming on the Antarctic Peninsula, Clim. Change, 60, 243-274, 10.1023/a:1026021217991, 2003.

Virkkula, A., Teinilä, K., Hillamo, R., Kerminen, V. M., Saarikoski, S., Aurela, M., Koponen, I. K., and Kulmala, M.: Chemical size distributions of boundary layer aerosol over the Atlantic Ocean and at an Antarctic site, J. Geophys. Res.: Atmos., 111, 10.1029/2004jd004958, 2006.

Virkkula, A., Hirsikko, A., Vana, M., Aalto, P. P., Hillamo, R., and Kulmala, M.: Charged particle size distributions and analysis of particle formation events at the Finnish Antarctic research station Aboa, Boreal Environ. Res., 12, 397-408, 2007.

Virkkula, A., Asmi, E., Teinilä, K., Frey, A., Aurela, M., Timonen, H., Mäkelä, T., Samuli, A., Hillamo, R., Aalto, P. P., Kirkwood, S., and Kulmala, M.: Review of aerosol research at the Finnish Antarctic research station Aboa and its surroundings in Queen Maud Land, Antarctica, Geophysica, 45, 163-181, 2009.

5   Weller, R., and Wagenbach, D.: Year-round chemical aerosol records in continental Antarctica obtained by automatic samplings, Tellus Ser. B-Chem. Phys. Meteorol., 59, 755-765, 10.1111/j.1600-0889.2007.00293.x, 2007.

Weller, R., and Lampert, A.: Optical properties and sulfate scattering efficiency of boundary layer aerosol at coastal Neumayer Station, Antarctica, J. Geophys. Res.: Atmos., 113, 10.1029/2008jd009962, 2008.

10

Weller, R., Minikin, A., Wagenbach, D., and Dreiling, V.: Characterization of the inter-annual, seasonal, and diurnal variations of condensation particle concentrations at Neumayer, Antarctica, Atmos. Chem. Phys., 11, 13243-13257, 10.5194/acp-11-13243-2011, 2011.

Weller, R., Minikin, A., Petzold, A., Wagenbach, D., and König-Langlo, G.: Characterization of long-term and seasonal variations of black carbon (BC) concentrations at Neumayer, Antarctica, Atmos.

15   Chem. Phys., 13, 1579-1590, 10.5194/acp-13-1579-2013, 2013.

Weller, R., Schmidt, K., Teinilä, K., and Hillamo, R.: Natural new particle formation at the coastal Antarctic site Neumayer, Atmos. Chem. Phys., 15, 11399-11410, 10.5194/acp-15-11399-2015, 2015.

20   Wolff, E. W., and Cachier, H.: Concentrations and seasonal cycle of black carbon in aerosol at a coastal Antarctic station, J. Geophys. Res. D: Atmos., 103, 11033-11041, 1998.

Yoon, Y. J., Ceburnis, D., Cavalli, F., Jourdan, O., Putaud, J. P., Facchini, M. C., Decesari, S., Fuzzi, S., Sellegri, K., Jennings, S. G., and O'Dowd, C. D.: Seasonal characteristics of the physicochemical properties of North Atlantic marine atmospheric aerosols, J. Geophys. Res.:

25   Atmos., 112, 10.1029/2005jd007044, 2007.

CPC: condensation particle counter
SMPS: scanning mobility particle sizer
CCNC: cloud condensation nuclei counter

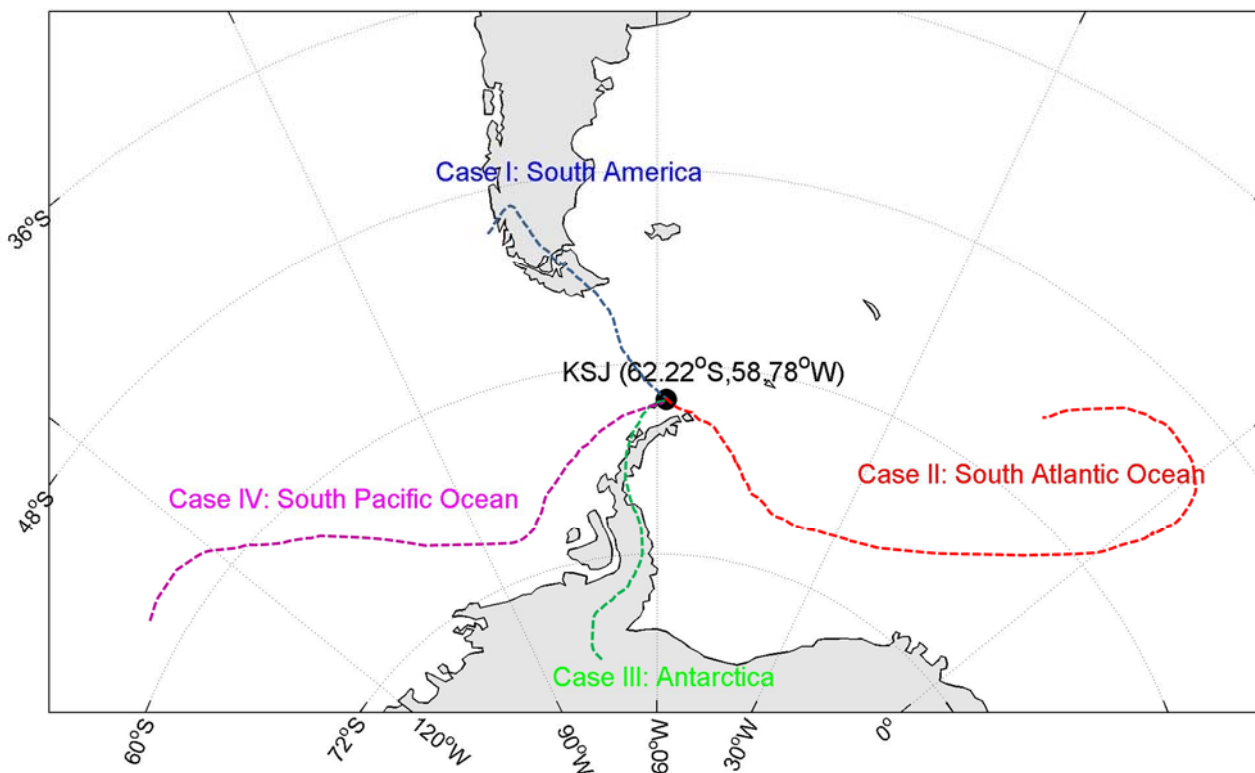5   Figure 1. A schematic diagram for the observation methods used in this study.

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
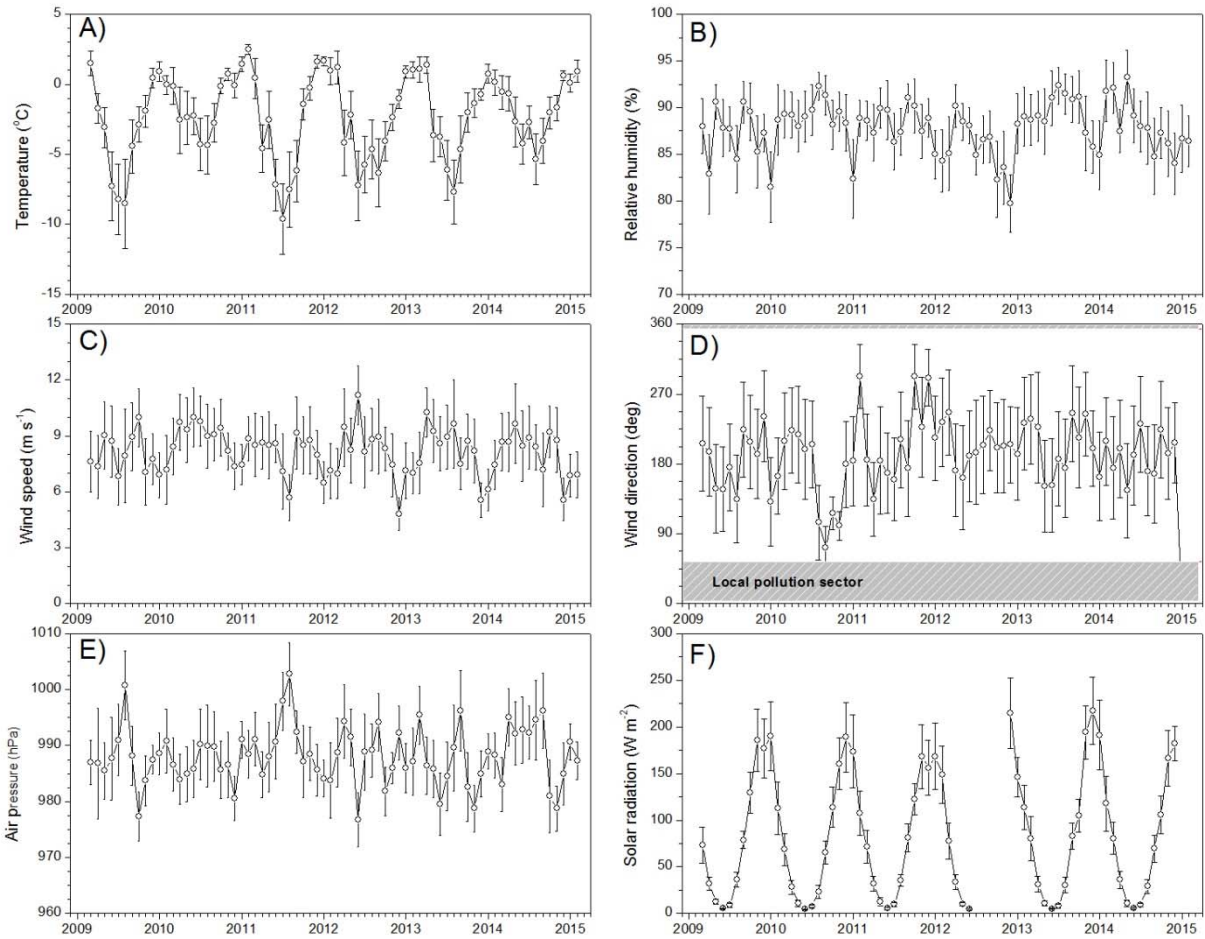
according to cases.

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.
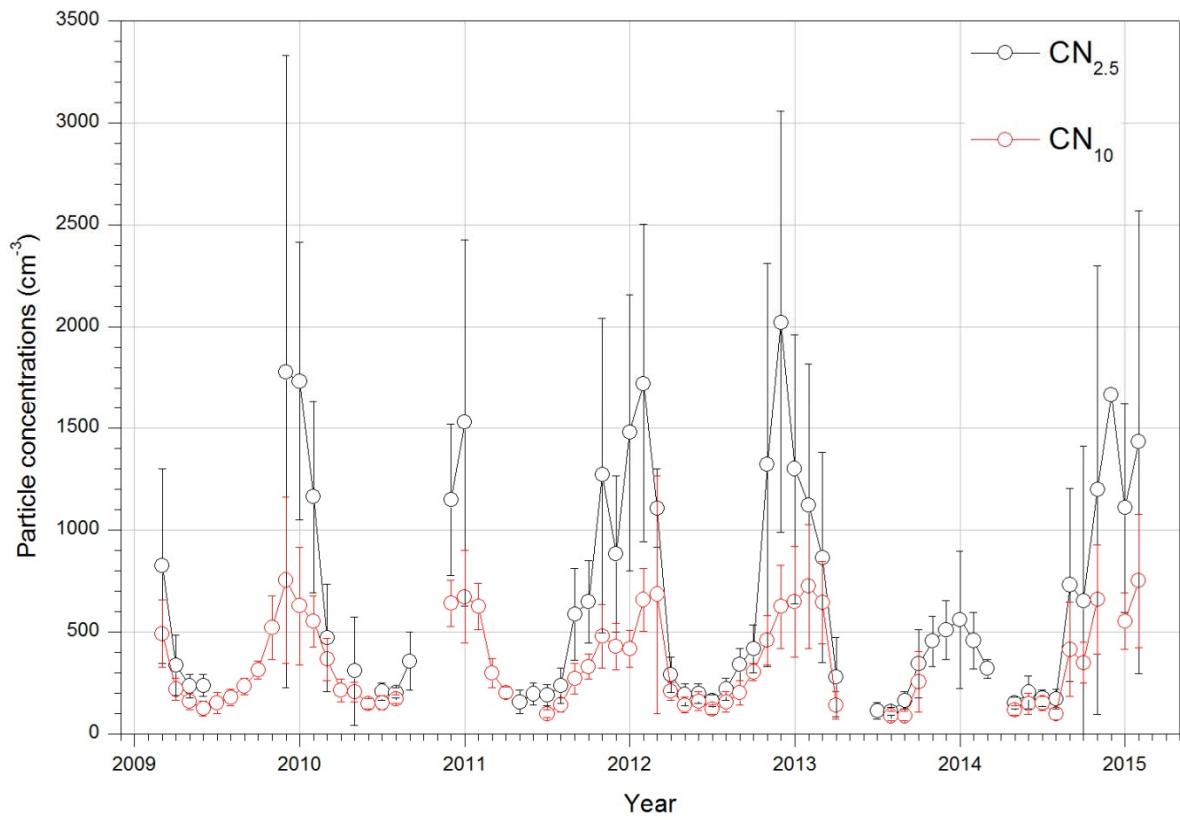
Figure 4. Monthly variations of mean $CN_{2.5}$ (black opened circle) and CN_{10} (red opened circle)

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.
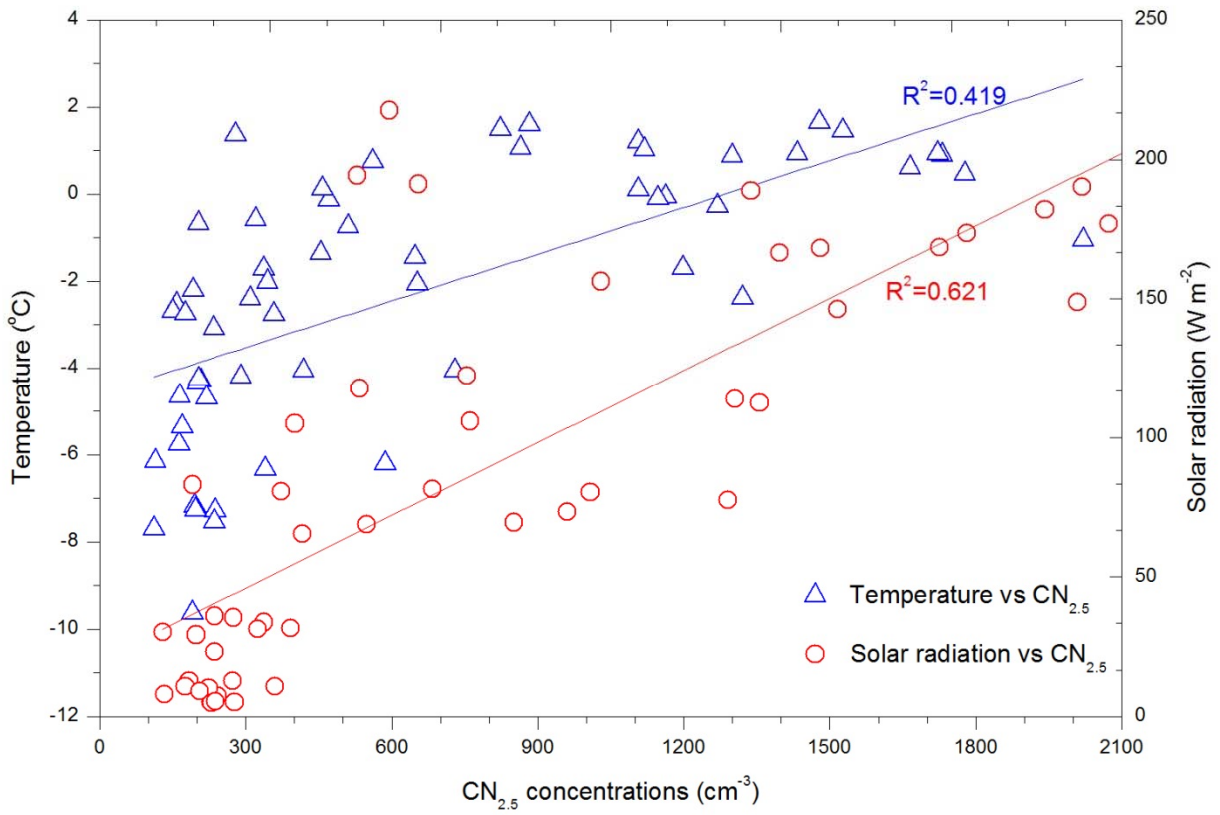
Figure 5. Scatterplot diagram of monthly mean CN$_{2.5}$ concentrations and monthly mean temperature (blue opened triangle) or monthly mean solar radiation intensity (red opened circle). Blue and red solid lines are a regression lines.
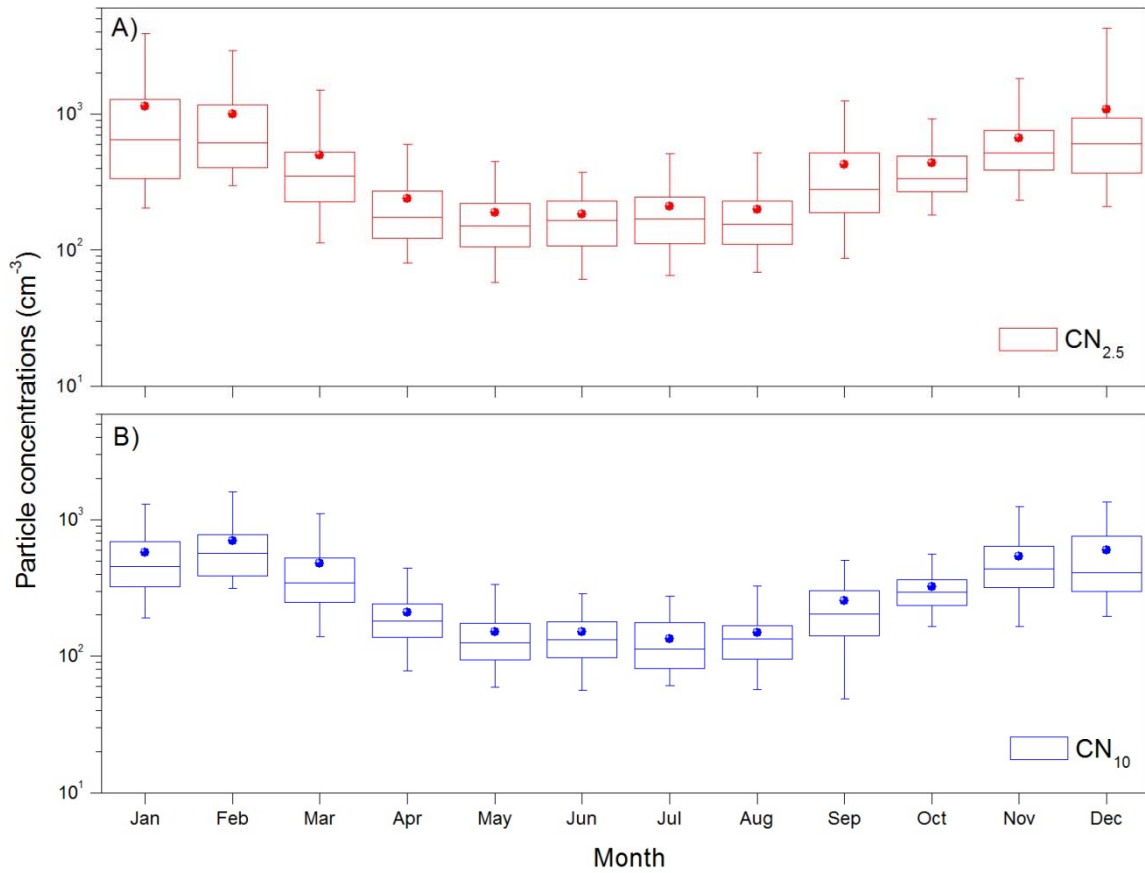
Figure 6. Box plots of seasonality of (a) $CN_{2.5}$ and (b) CN_{10} concentrations. Lines in the middle of the boxes indicate sample medians (mean value is closed circle), lower and upper lines of the boxes are the 25th and 75th percentiles, and whiskers indicate the 5th and 95th percentiles.
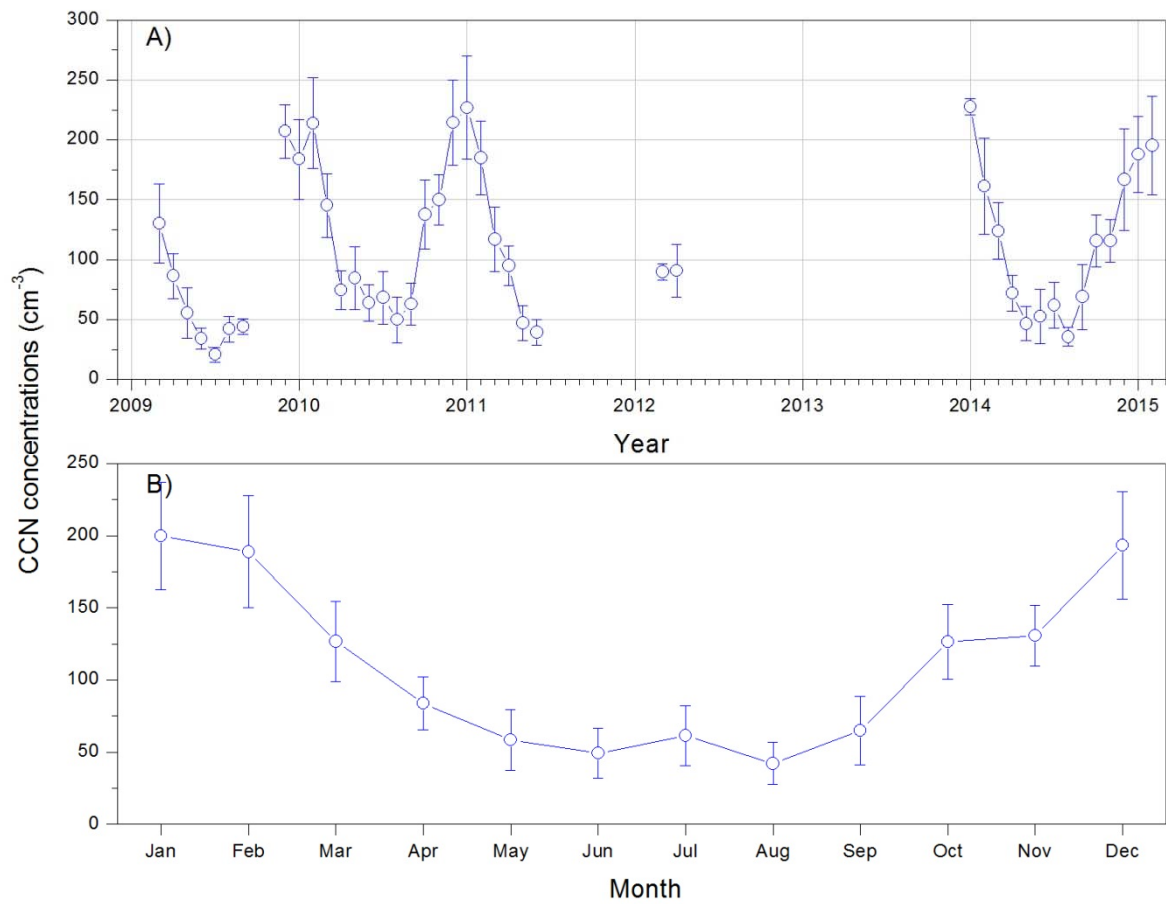
Figure 7. (a) Monthly mean CCN concentrations at the SS of 0.4 % with a standard deviation from March 2009 to February 2015 (b) Seasonal variation of mean CCN concentrations at the SS of 0.4 % with a standard deviation.
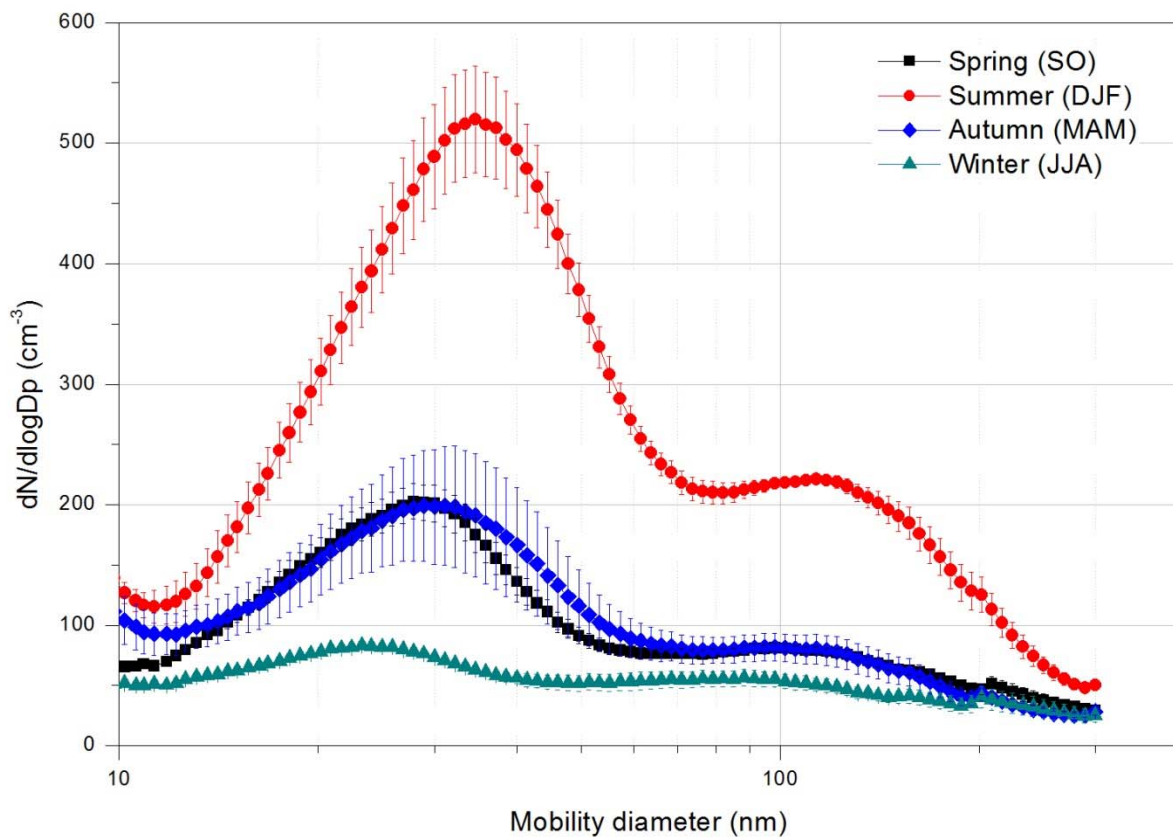
Figure 8. Seasonal mean aerosol size distribution measured by the SMPS at the King Sejong research station over the period from March 2009 to February 2015. The error bars represent the standard deviation of the measurements from the mean value.
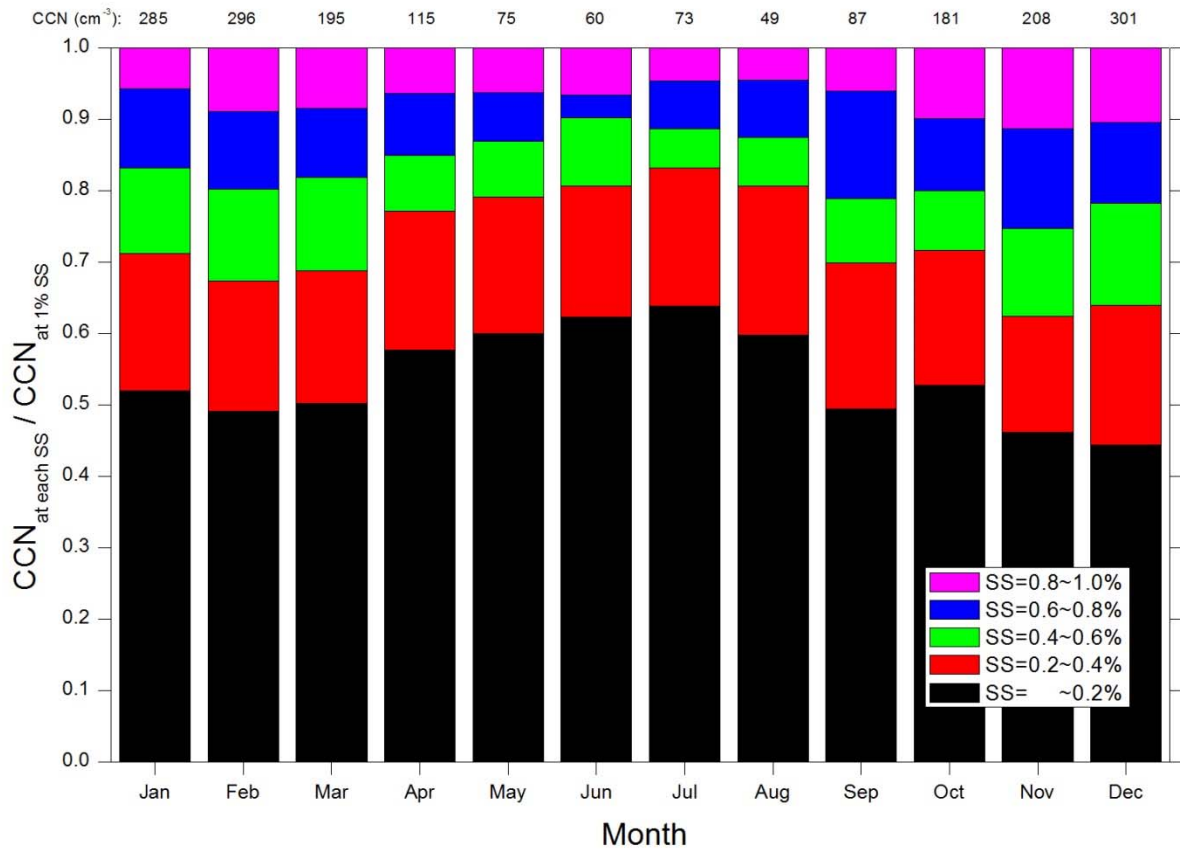
Figure 9. Monthly mean cumulative CCN concentrations shown as fractions of the CCN concentration at the SS of 1.0 %. Colours indicate the SS bins. The number at top of figure represents monthly mean CCN concentrations at the SS values of 1.0 %.
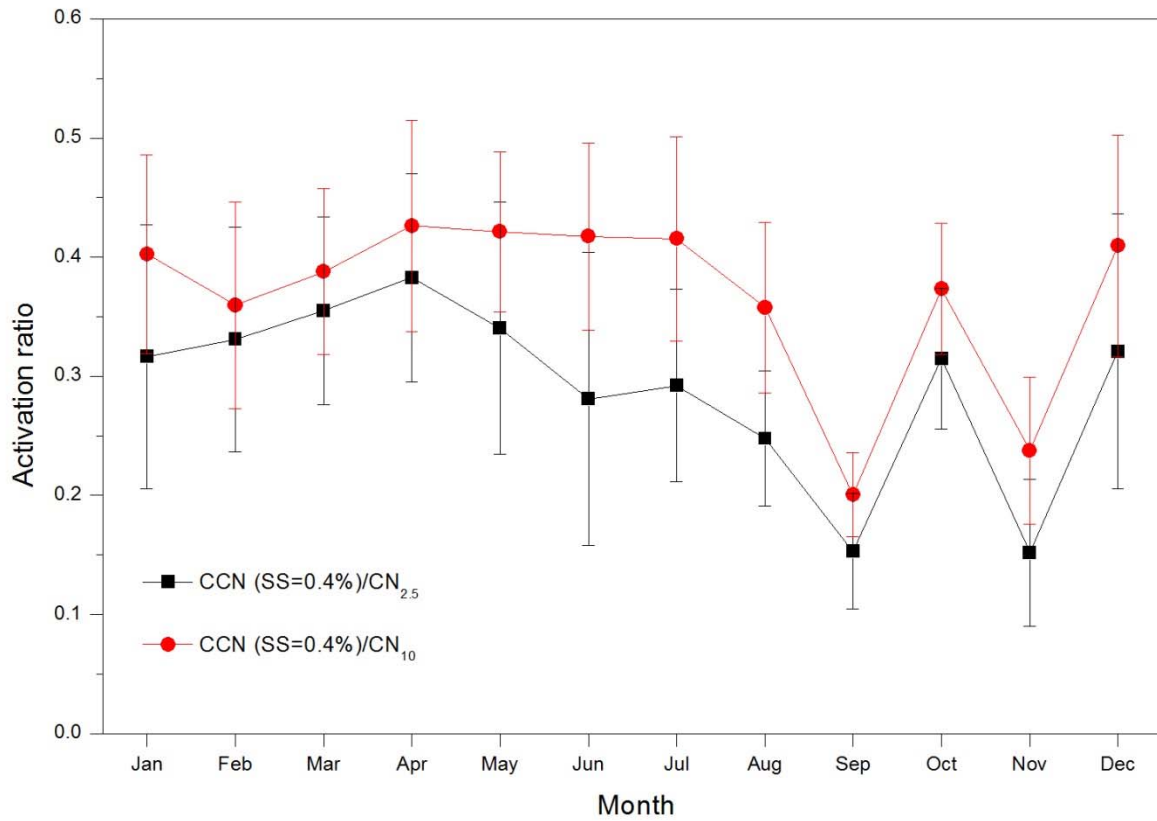
Figure 10. Comparison of the seasonal mean variation of the activation ratio between measurements
(CPC 3776 and CPC 3772) by two CPCs. The error bars represent the standard deviation of the
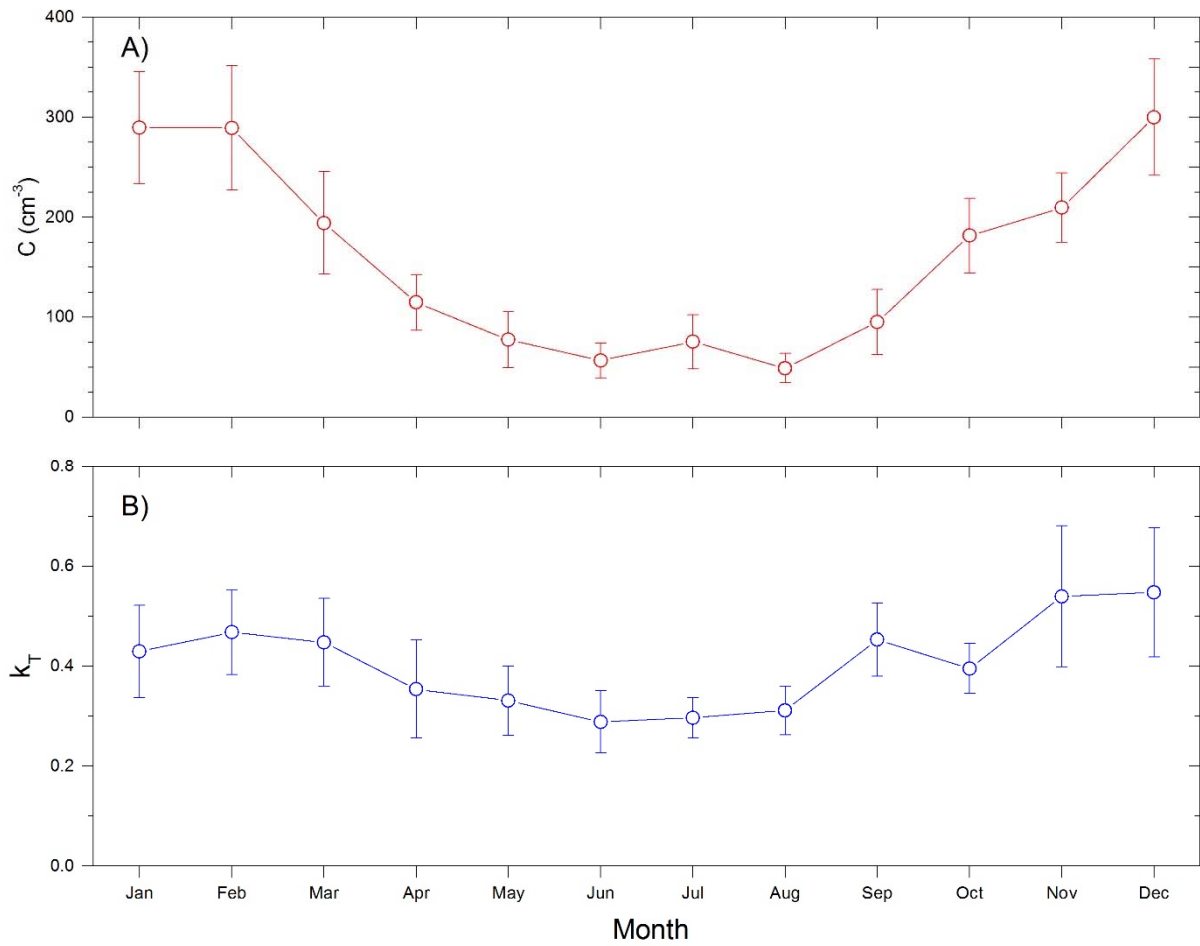measurements from the mean value.

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.
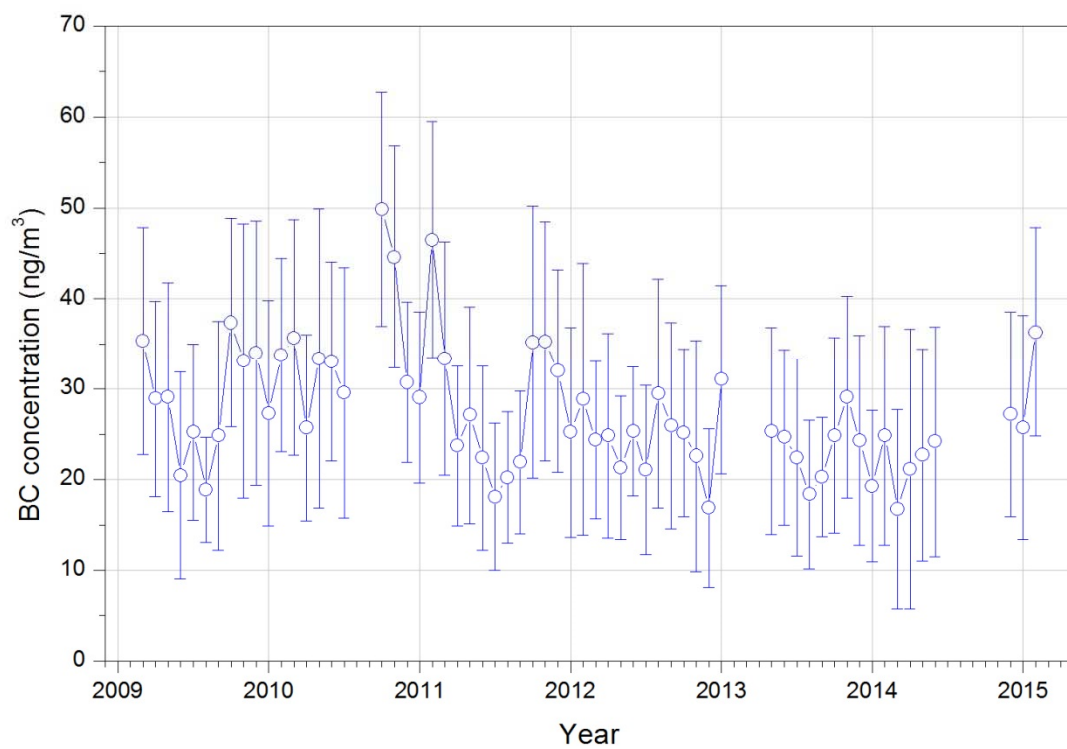
Figure 12. Monthly mean concentrations of black carbon over the period from March 2009 to February 2015. The error bars represent the standard deviation of the measurements from the mean value.
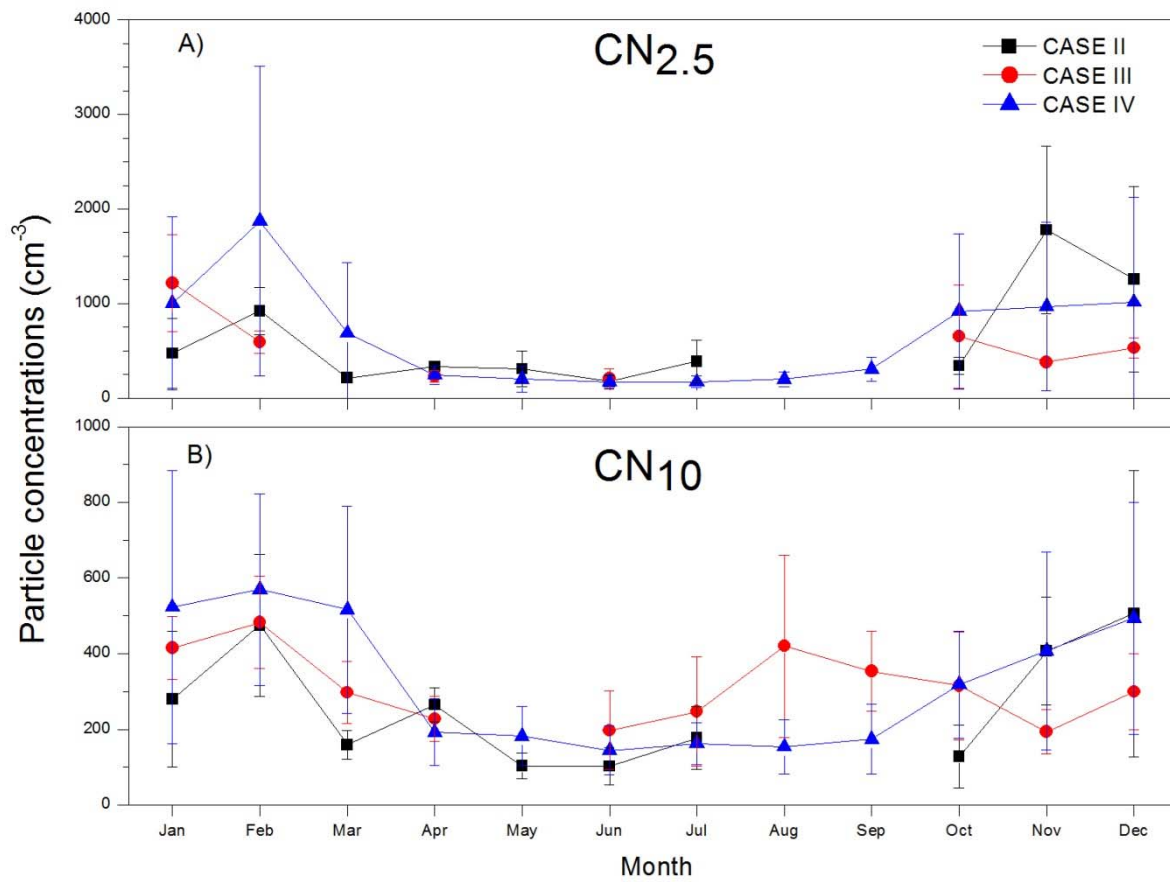
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Figure 13. Seasonal variation of mean (a) $CN_{2.5}$ and (b) CN_{10} concentrations with a standard deviation depending on the air mass origin. The error bars represent the standard deviation of the measurements from the mean value.
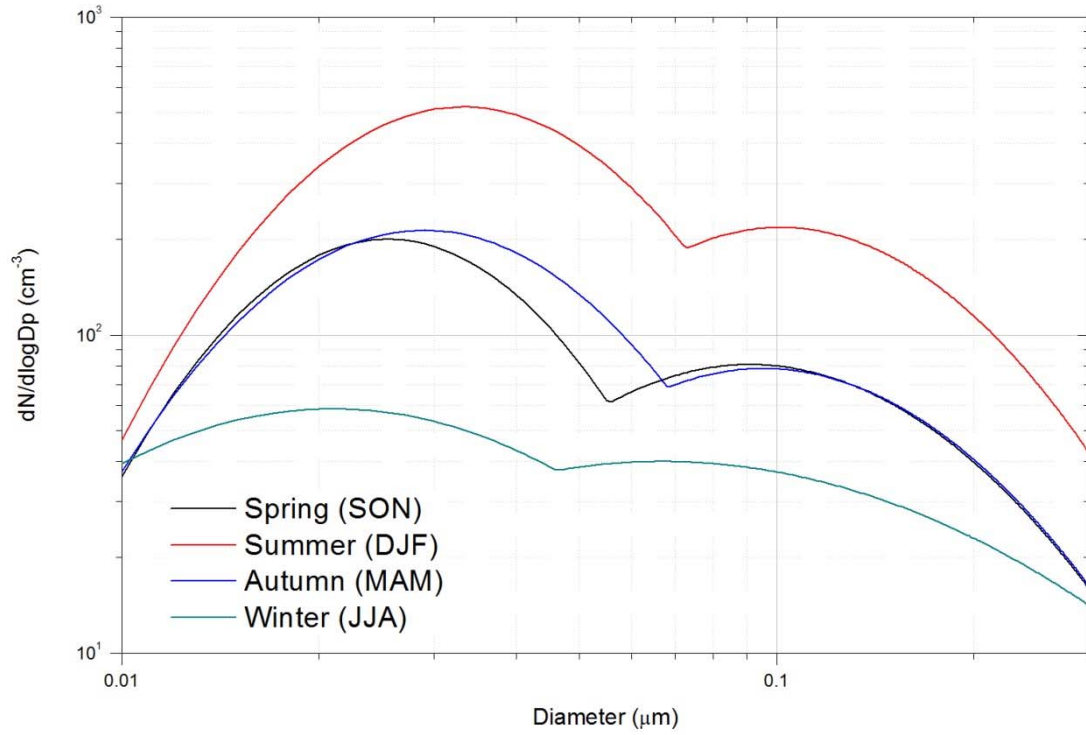
Figure 14. Seasonal lognormally fitted size distribution of aerosol particles originating from the South
Pacific Ocean, ranging from 0.01 to 0.3 µm (Case IV).

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
CN$_{10}$ 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

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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 (μm)	N (cm⁻³)	σ	D_g (μm)
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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