

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 at the King Sejong Station in the Antarctic Peninsula was investigated over the period of March 2009 to February 2015. Clear seasonal cycles of 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 the 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 of the cloud condensation nuclei (CCN) concentrations coincided with the CN concentrations, where the concentrations were minimum in the winter and maximum in the summer. ~~Based on measured CCN data at each supersaturation ratio (SS), empirical parameterization were also conducted using formula expressed by power-law function ($N_{\text{cen}} = C \times (\text{SS})^k$), where N_{cen} is the CCN concentrations at a given SS, and C and k are the fitting parameters~~ The measured CCN spectra were approximated by a power law fitting function relating the number of CCN at a given supersaturation (SS) to SS with fitting coefficient C and k . 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 ranged between 0.07 and 2.19, with a mean of 0.41 ± 0.10 . In particular, the k 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 SS of 0.4%. ~~Furthermore,~~ The effects of the origin and the pathway travelled by the air mass on the physical characteristics of aerosol particles were determined. The modal diameter of aerosol particles that originated from the South Pacific Ocean showed seasonal variations; being equal to $0.023 \text{ }\mu\text{m}$ in the winter and $0.034 \text{ }\mu\text{m}$ in the summer for the Aitken mode and $0.086 \text{ }\mu\text{m}$ in the winter and $0.109 \text{ }\mu\text{m}$ in the summer for the accumulation mode.

1. Introduction

Aerosol particles in the atmosphere may be emitted 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 influence 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, the direct and indirect climate effects are still ~~unknown~~ highly uncertain (IPCC, 2013). Moreover, in order to understand the sources and the processes of the atmospheric aerosol particles, there should be a need to have long-term observations at different regions because aerosol particles vary temporally and spatially.

The Antarctic region is highly sensitive to climate changes due to complex interconnected environmental systems (e.g. snow cover, land ice, sea-ice, and ocean circulation) (Chen et al., 2009). Previous studies show that the Antarctic Continent and the Antarctic Peninsula have experienced noticeable 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 warming rate of more than 5 times that of the other regions on earth (Vaughan et al., 2003; IPCC, 2013). The Antarctic climate system can be linked with aerosol particles by complex feedback processes that involve aerosol-cloud interactions. In addition, 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 ~~the~~ their physical properties in Antarctica, e.g. total particle concentrations, size distributions and concentrations of black carbon and activated CCN, is necessary.

Over the years, measurements of aerosol particles have been widely conducted at various 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 their size
5 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 studies 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).
10 Although various studies have been performed, the measurements taken at the Antarctic Peninsula and the long-term observations of aerosol particles are still insufficient.

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

2 Methods

2.1 Sampling site and instrumentation

Continuous observations of the physical properties of aerosol particles have been carried out since
25 March 2009 at the King Sejong Station (62.22°S, 58.78°W) in the Antarctic Peninsula. Detailed information of the sampling site is given by Choi et al. (2008). In brief, the King Sejong Station is located on the Barton Peninsula of King George Island (KGI). The population density of KGI is

higher than that of other regions in Antarctica due to the various research activities carried out from eight permanent on-site stations. The observatory is located approximately 400 m southwest of the main buildings, which include the power generator and crematory of King Sejong Station. Thus, the northeastern direction (355° - 55°) was designated as the local pollution sector because of the emissions from the power generator and crematory at the station. We therefore discarded data from the local pollution sector to improve data quality, and data where BC concentrations were higher than 100 ng m^{-3} were also discarded. In order to ensure the reliability of measurements, only dataset, of which acquisition rate higher than 50%, were used during all analysis procedures. In this study, we present the analysed results of the physical characteristics of aerosol particles obtained during March 2009 to February 2015.

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

Based on Global Atmosphere Watch (GAW) aerosol measurements guidelines and recommendations, we installed cylindrical stainless common inlet. 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, 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 flow rate of total sample air was maintained as 150 lpm. The Reynolds number in the common inlet was 2388. It represents that the flow in the common inlet is transition regime ($2000 < \text{Re} < 4000$). For sampling, short L-bend tube made of stainless steel was placed at center of the common inlet. Instruments were connected with the common inlet using conductive tubing to minimize the particle loss. 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, any drying system was not used during sampling. The total counting efficiency of the measurement system was estimated. 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 expect for the common inlet was conductive tubing to minimize the known particle loss.

Total particle number concentrations were examined with two CPCs: a TSI model 3776 that measured particles > 2.5 nm in diameter and a TSI model 3772 that measured particles > 10 nm. Sample aerosol flow rates of CPC 3776 and CPC 3772 were 1.5 lpm and 1.0 lpm, respectively.

The aethalometer (Magee Scientific, AE16) was used to measure the concentration of light absorption particles at 880 nm wavelengths. The flow rate of the sample was constant at 5.0 lpm. The main purposes of measuring BC concentrations were to investigate long-range transport aerosol particles and to assess the influence exerted by local pollution.

To measure the CCN concentrations, a CCNC (DMT CCN-100) was used at five different supersaturation ratios (SS) (0.2, 0.4, 0.6, 0.8, and 1.0 %) and total flow rate of 0.5 lpm. The CCN concentrations were 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 0.2 % SS) before it was changed to the next SS value. For a 0.2 % SS, CCN concentrations were measured for 10 min because it required additional time to achieve stability after completing measurements at a 1 % SS.

Aerosol size distributions were continuously measured with the SMPS, which consisted of a differential mobility analyser (DMA; HCT 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 scanning time was set to 120 s for mobility particle diameters from 0.01 to 0.30 μm . A closed sheath-

air loop with a diaphragm pump in the control unit was used to maintain the sheath flow of DMA. The flow rate of sheath air of DMA was 10 lpm. The ratio of aerosol flow to sheath flow of DMA was 1:10.

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

2.2 Back-trajectory analysis

In order to associate the physical properties of aerosol particles to their source areas for the sampling periods, the air mass back trajectory analysis was conducted using the 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 the sampling site. The results where the origin and pathway of the air masses for at least 12 h were similar at three different heights were used for the analysis in this study. Based on this analysis, we have classified the air mass into four groups according to their origin and pathway: two continental regions (South America and Antarctica) and two oceanic areas (South Atlantic and South Pacific Ocean), as are shown in Fig. 2.

3 Results and Discussion

3.1 Meteorological conditions

Fig. 3 depicts monthly variations of the meteorological parameters measured from ~~and-an~~ automatic weather system (AWS) during the whole 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 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 ~~condition~~ 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 temperature are observed during a six-year period due to a relatively short observation period. In this manuscript, we focused on correlation analysis between temperature (or solar radiation) and CN concentration.

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3.2 Seasonality in the physical characteristics of aerosol particles

3.2.1 Total particle number concentrations

Fig. 4 shows the monthly mean particle number (CN) condensations measured with two types of instruments (TSI CPC 3776 and 3772) over the period from March 2009 to February 2015. All the seasons mentioned in this study are austral seasons. As can be seen in Fig. 4, there is an evident seasonal cycle of CN concentrations, which are the maximum in the summer (from December to February, DJF) and minimum 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 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 approximately 110 cm⁻³ and 90 cm⁻³ in August 2013, respectively. Our results were 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 long-term daily average CN concentrations over the period from November 2003 to January 2007 from observations at Aboa, the Finnish Antarctic research station at a coastal region in Antarctica. The maximum monthly average CN concentrations were observed 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 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 oxidation of dimethyl sulphide (DMS) produced by phytoplankton (Fattori et al., 2005; Weller et al., 2011). The DMS concentrations increase sharply

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when biological activity is enhanced due to increasing temperatures and solar radiation (Virkkula et al., 2009). Since our sampling site was in the Antarctic Peninsula, ocean biological activity was considered to be an important factor in the particle formation and growth of aerosol particles. *Since the higher summer than winter differences between $CN_{2.5}$ and CN_{10} concentrations correlate with higher summer than winter biological activity, and solar radiation and temperature, our hypothesis*

~~The difference between $CN_{2.5}$ and CN_{10} concentrations typically increased in the summer season (DJF) due to high biological activity, whereas those in the winter season (JJA) decreased when biological activity is low. Our hypothesis~~ is that the trends of the difference should be related to secondary aerosol formation caused by biological activity. To better understand the effect of temperature and solar radiation intensity on $CN_{2.5}$ concentrations, we compared the relationship between monthly mean $CN_{2.5}$ concentrations and solar radiation intensity, and monthly mean $CN_{2.5}$ concentrations and temperature. The correlation coefficient between $CN_{2.5}$ and the solar radiation intensity (opened circle; $R^2=0.621$) was slightly higher than that between $CN_{2.5}$ and temperature (opened triangle; $R^2=0.419$), as shown in Fig. 5. Our results suggest that the $CN_{2.5}$ concentrations may be more closely coupled with solar radiation intensity than with temperature.

Unique results of $CN_{2.5}$ 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 not be explained by solar radiation intensity and temperature because the solar radiation and the temperature did not show any distinctive variation compared with other years. The possible reason is type of air masses reached to the sampling site. Although air mass originated from the South Pacific Ocean (Case IV: 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 Sec 2.2, frequency of air mass originated from the South Atlantic Ocean (Case II) in the summer of 2013-2014 was higher than other years and frequency of air mass originated from Case IV was lower than other years. ~~In case of Case II, peak $CN_{2.5}$ concentrations were in November, while maximum $CN_{2.5}$ concentrations of Case IV were in February.~~ Unfortunately, neither CN_{10} nor SMPS data are available for the austral summer season of 2013-2014 because of mechanical failures, it is not possible to directly explain the low concentrations of $CN_{2.5}$ for this season in terms of the potential effects of air mass characteristics on the concentration of 2.5-10 nm size particles.

Nevertheless, it is likely that the ~~Therefore, it is that increasing-increased~~ frequency of air mass ~~originated originating~~ from the South Atlantic Ocean (Case II) ~~would explain~~ might have resulted in ~~this the~~ lower CN_{2.5} concentration of the austral summer season of 2013-2014.

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

A more detailed comparison of the monthly trends in the CN_{2.5} and CN₁₀ 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 CN_{2.5} concentrations during the summer period increased sharply compared to the CN₁₀ concentrations, the 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 SS value of 0.4 % over the period from March 2009 to February 2015. There is a long 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 five SS values (0.2, 0.4, 0.6, 0.8, and 1.0%) during the third Palls Cloud Experiment (PaCE-3). They showed correlation between CDNC and CCN concentrations at each supersaturation. The relationship between CDNC and CCN concentrations at the SS value of 0.4% was approximately linear, while CCN concentrations were lower than CDNC when the SS value was lower than 0.4% and CCN concentrations at upper 0.4% higher than CDNC. Based on this result, in this study, the supersaturation of 0.4% was chosen to

investigate seasonal variations of CCN. We found monthly variations in the CCN concentrations with the maximum values being observed during the summer periods (DJF) and the minimum concentrations were observed during the winter periods (JJA). The monthly mean CCN concentrations were in the range of 20.63 cm^{-3} in July 2009 and 227.52 cm^{-3} in January 2014, with a mean of $112.80 \pm 39.05 \text{ cm}^{-3}$. Fig. 7(b) also shows seasonality in CCN concentrations at an 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 $199.89 \pm 37.07 \text{ cm}^{-3}$ and the minimum CCN concentration in August was $42.13 \pm 14.51 \text{ cm}^{-3}$. This clear seasonality of CCN concentrations follows the seasonal trend-march of CN concentrations. As shown in Fig. 6, CN_{10} concentrations as well as $\text{CN}_{2.5}$ concentrations increased during the summer. In addition, the aerosol size distributions measured by SMPS showed that concentrations of 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 CCN concentrations at each SS value in 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 SS of 1.0 % was carried out, and 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 SS of 1.0 %. Although a clear seasonal trend of 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 SS value of 0.2 % in 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 mean CCN concentrations at the SS

values of 1.0 %. The fraction of particles activated to CCN at the 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 SS value of 0.2 % during the winter (JJA) was similar to those (~ 0.52) measured in Mace Head, which is a representative site of a marine environment (Paramonov et al., 2015). Although CCN concentrations were low in the winter, our observations suggest that aerosol particles that are activated to CCN during the winter season should be more hygroscopic than those during the summer period.

3.2.3 Activation ratio and Fitting parameter of CCN

The seasonal variations in the mean activation ratio of CCN concentrations at a SS of 0.4 % to the CN concentrations measured from two CPCs (TSI 3776 and 3772), as shown in Fig. 10. The mean values of activation ratios of CCN/CN_{2.5} and CCN/CN₁₀ were about 0.33 ± 0.10 and 0.40 ± 0.08 , respectively. Our results suggest that hygroscopic compounds were less dominant in the aerosol particles at our sampling site compared to levels in aerosol particles in the Arctic regions (Latham et al., 2013). Although clear changes were observed in the monthly variation in the CN and CCN concentrations as shown in Fig. 6 and Fig. 7(b), it was seen that the activation ratio (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 the concentrations of accumulation mode particles, as can be seen in Fig. 8. The lower activation ratios in September and November are mainly because of the size and chemical properties of aerosol particles. Both, the size and chemical components of aerosol particles may have 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 concentrations in August. Thus, the activation ratio decreased dramatically. Unfortunately, we did not confirm aerosol size distribution because our aerosol size distribution data in November was unreliable due to malfunctioning instruments.

The CCN concentrations at SS values can be represented by a power-law function, defined by

Twomey (1959):

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

where N_{CCN} is the concentration of CCN at given a supersaturation values (SS), C and k are coefficient constants estimated from CCN spectra. The correlation coefficient between the measured CCN concentrations at the SS of 0.4% and the power-law fit values, r , was 0.978. ~~The average correlation coefficient, r , was 0.978.~~ The values of C varied from 6.35 cm^{-3} to 837.24 cm^{-3} , with a mean of $171.48 \pm 62.00 \text{ cm}^{-3}$. The daily mean κ values ~~of k~~ range between 0.07 and 2.19, with a mean of 0.41 ± 0.10 . The monthly variations of k and C values are also summarized in Fig. 11. A comparison with CCN concentrations indicated that the values of k 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 during the winter.

It is useful to infer hygroscopic properties of aerosol particles with a hygroscopicity parameter, κ . The κ values varies from 0 for insoluble particles to larger than 1 for water-soluble salt particles (Petters and Kreidenweis, 2007). The κ value 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 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 where SMPS scan starts. The CCN/CN ratio indicates the fraction of CCN-active aerosols among total particle concentrations.

In this study, the kappa values were estimated using the monthly mean CCN concentrations at the SS of 0.4%, the monthly mean CN concentrations measured by CPC and the monthly mean size number distribution results obtained from SMPS data. The annual mean kappa value was calculated to be 0.15 ± 0.05 . This value is comparable to the previous studies from Arctic and subarctic areas. For example, Latham et al. (2013) who measured the CCN activity at the Arctic by using aircraft measurements reported the kappa value of 0.32 ± 0.21 . Martin et al. (2011) inferred total kappa of 0.33 ± 0.13 during cruise observation in Longyearbyen, Svalbard. Kammermann et al. (2010) reported the kappa values varied between 0.07-0.21 in the period of 18 days within the Arctic Circle in Sweden. Jaatinen et al. (2014) also showed the kappa value of 0.13 ± 0.07 using 13-day set of data at subarctic area in Finland (Pallas-Sodankylä station).

3.2.4.3 Black carbon (BC) concentrations

Fig. 12 shows variations of monthly mean BC concentrations over the whole sampling period. To eliminate effect of local pollution on observations, in 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 were 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 the Baton Peninsula of King George Island. In particular, six stations are located within a 10 km radius from the King Sejong Station. ~~This should be extra bias of data from BC concentrations due to effect of other stations.~~ The presence of these stations should affect the measured BC concentrations at the King Sejong Station, causing a positive bias.

Additionally, no clear seasonal patterns were observed in our study throughout the entire observation period. However, clear seasonal patterns 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 Hally station and 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 to Pereira et al. (2006), although BC concentration during the summer increased 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 air mass on the physical characteristic of aerosol particles is presented. As mentioned earlier in Sec. 2.2, we classified air masses into four groups based on air mass back trajectory analysis. The wind data and aerosol characteristics with the four types of air masses during the entire observation period are listed in Table 1. The very few cases of air masses ~~originated~~originating from the continent of South America (Case I) showed the highest BC and CCN concentrations (Table 1). This might be due to anthropogenic influences at the source and the aging of aerosol particles. The CN₁₀ concentrations were 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 the CN concentrations based on the 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 mean values. This is probably because of new particle formation, causing a bias, new particle formation mainly contribute a sudden

increase of aerosol number concentrations (Kulmala et al, 2004; Pierce and Adams, 2009). Minimum concentrations of aerosol particles ($CN_{2.5}$ and CN_{10}) originating from the ocean (Case II and IV) were observed from April to September, whereas concentrations of aerosol particles ($CN_{2.5}$) originating from the South Atlantic (Case II) and the South Pacific (Case IV) Oceans were the highest in November and February, respectively. Here we found that the peak month of the $CN_{2.5}$ concentrations had discrepancies in accordance with the air mass history. This is probably due to difference in chemical compounds that contributed to aerosol formation processes and/or in variations of biogenic activity according to the origin and transport pathway of air masses. To verify this, further studies on chemical compositions of aerosol particles need to be carried out in the future.

When air masses were transported from the South Pacific Ocean to the King Sejong Station (Case IV), the seasonality of aerosol size distribution was also investigated. Aerosol size distribution parameters fitted to log-normal distribution were derived for each season. The seasonally averaged log-normal aerosol size distributions are shown in Fig. 14. The lognormal-fitted aerosol size distribution ranged from 0.01 to 0.3 μm is presented in Fig. 14. The computed modal diameters along with standard deviation and number concentrations are summarized in Table 2. It is obvious 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): 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. The number concentrations for the summer are also higher than the value those for the winter for the (304.36 \pm 20.10 against 49.16 \pm 3.88 cm^{-3} , respectively) and accumulation (140.25 \pm 10.64 against 44.78 \pm 14.24 cm^{-3} , respectively) modes. Aitken and accumulation modes, 49.16 \pm 3.88 cm^{-3} during the winter and 304.36 \pm 20.10 cm^{-3} during the summer for the Aitken mode and 44.78 \pm 14.24 cm^{-3} in the winter and 140.25 \pm 10.64 cm^{-3} in the summer for the accumulation mode. The enhancement of number concentrations for the Aitken mode during the summer should be linked to new particle formation over oceans as a product

of biological activity. The spring and autumn seasons show intermediate values. Our 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 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 March 2009 to February 2015. An obvious seasonal variation of particle number concentrations (CN) exists, with the maximum concentrations in the austral summer (DJF) and the minimum concentrations in the winter (JJA). The maximum CN concentrations of particles larger than 2.5 nm (CN_{2.5}) and 10 nm (CN₁₀) were approximately 2000 cm⁻³ in December 2012 and about 800 cm⁻³ in December 2009 and February 2015, respectively. In particular, CN_{2.5} concentrations increased sharply during the summer compared to CN₁₀ concentrations, suggesting that the particle formation processes were probably driven by the high biological activity during the season.

In addition, we presented the clear seasonal trends of CCN concentrations at the supersaturation (SS) of 0.4%. 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). ~~It suggests that aerosol particles in Antarctic Peninsula should be less hygroscopic than those in Arctic.~~ We also estimated C and k values from measured CCN results at each SS value. The measured CCN spectra were approximated by a power law fitting function relating the number of CCN at a given SS to SS with fitting coefficient C and k ~~The C and k are constants were estimated using approximate formula~~

~~expressed by a power-law function ($N_{CCN}=C \times (SS)^k$) (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 of k ranged between 0.07 and 2.19, with a mean of 0.41 ± 0.10 . The k values during austral the 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 SS of 0.4%.

Based on the backward trajectory analysis, we classified the air mass into four groups according to their origin and pathway: two continental regions (South America and Antarctica) and two oceanic areas (South Atlantic and South Pacific Ocean). We found that most air masses originated from the oceanic areas. ~~The very few cases of air masses originated-originating form-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), in particular, the $CN_{2.5}$ concentrations show clear seasonal variations; 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 Case IV, an analysis of aerosol size distributions in the $0.01\text{-}0.3 \text{ }\mu\text{m}$ range was performed. The modal diameters also showed seasonal variations, $0.023 \text{ }\mu\text{m}$ in the winter and $0.034 \text{ }\mu\text{m}$ in the summer for the Aitken mode and $0.086 \text{ }\mu\text{m}$ in the winter and $0.109 \text{ }\mu\text{m}$ in the summer for the accumulation mode.

Overall, this study is the first of its kind to analyze seasonal variations in the physical characteristics of 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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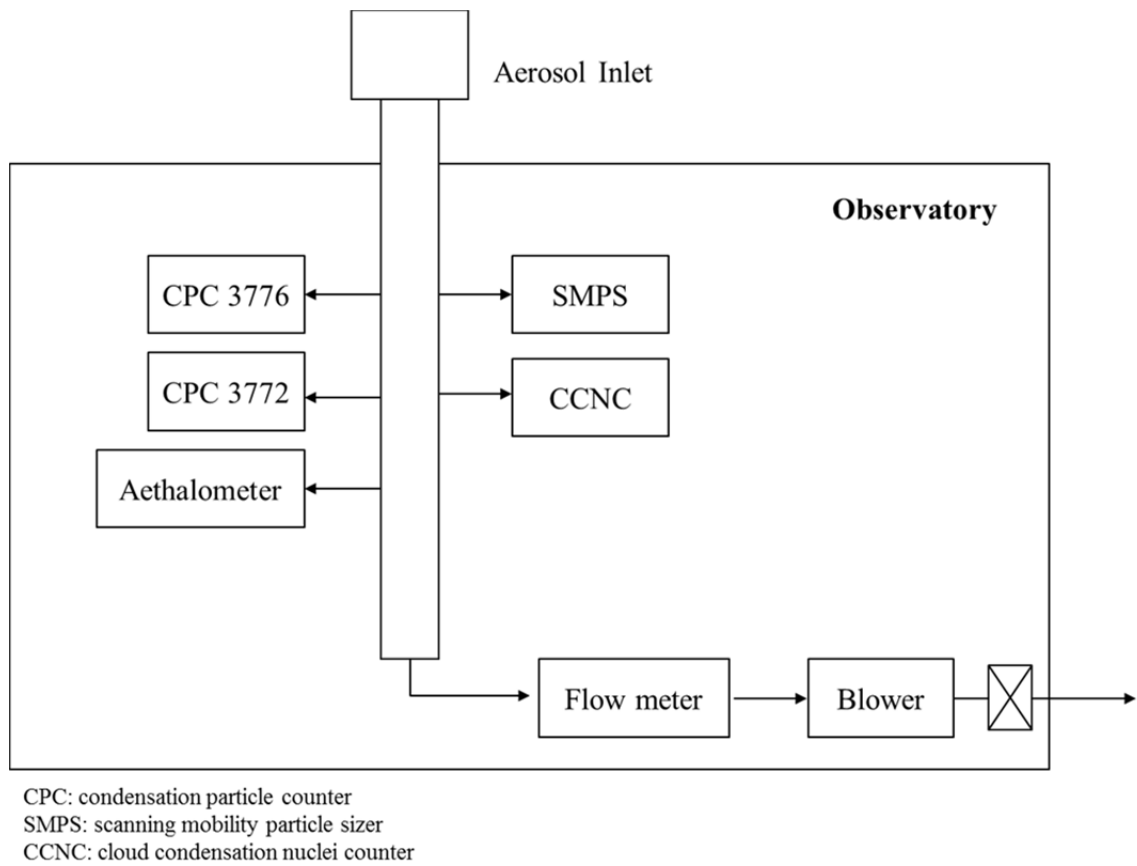
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5 Figure 1. A schematic diagram for the observation methods used in this study.

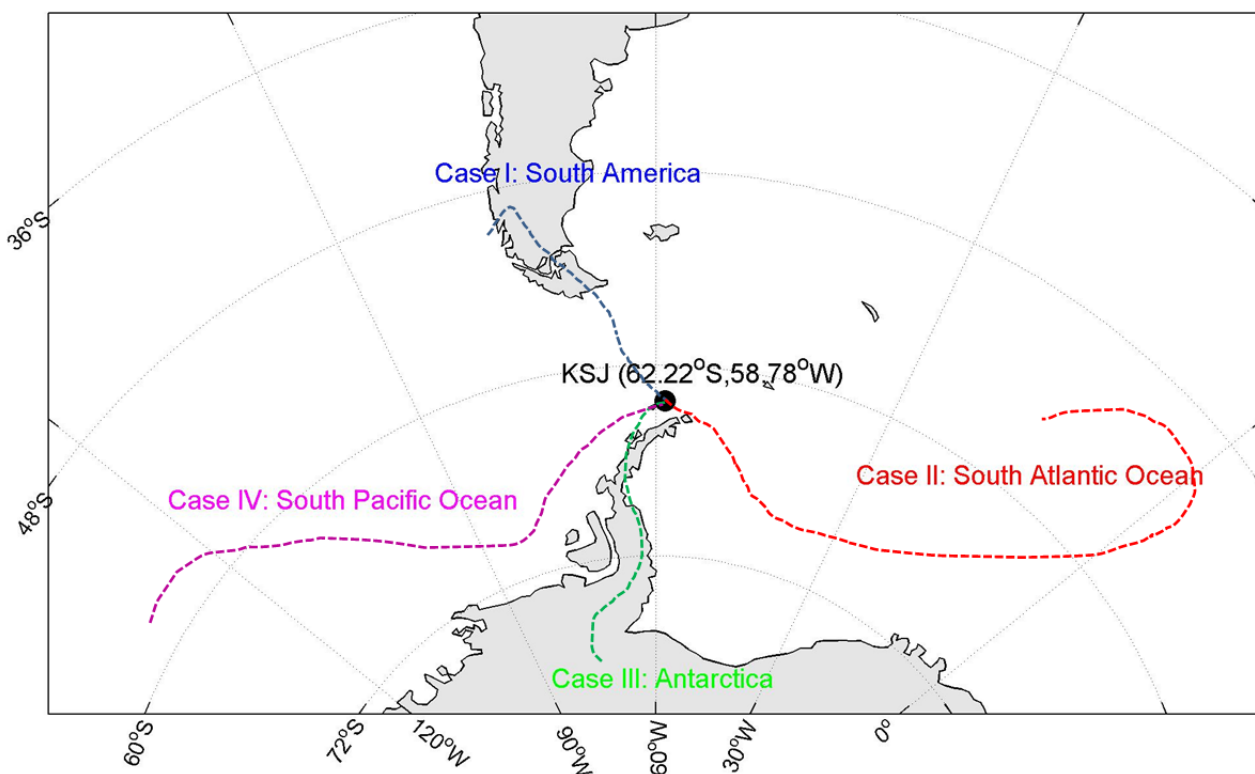


Figure 2. Map of the sampling site (62.22°S, 58.78°W; black circle) and classification of the four cases according to the origin and pathway of the air masses. Dot lines represent example of back trajectories according to cases.

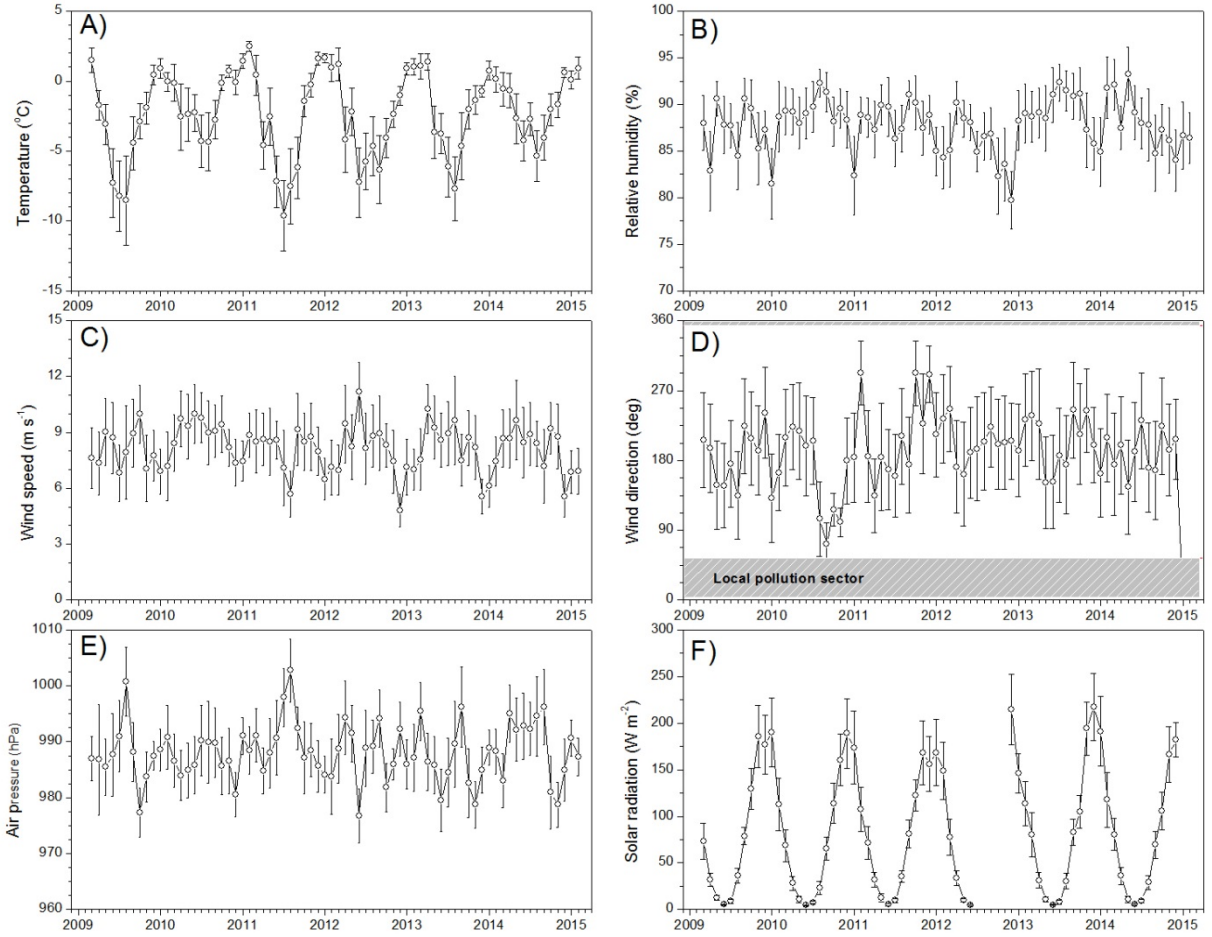


Figure 3. Monthly mean variation of (a) temperature, (b) relative humidity, (c) wind speed, (d) wind direction, (e) air pressure, and (f) solar radiation over the period from March 2009 to February 2015. The shaded area in Figure 3(d) represents the wind direction for the local pollution sector.

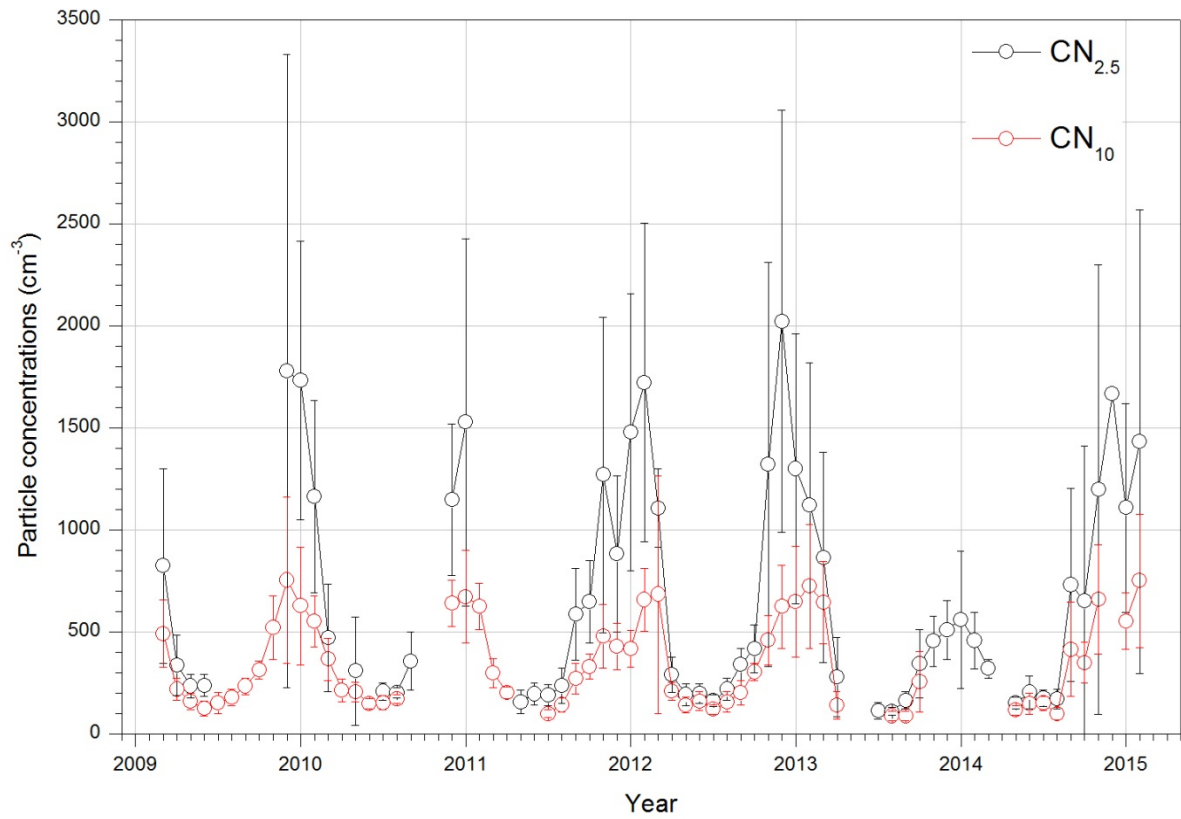


Figure 4. Monthly variations of mean $CN_{2.5}$ (black opened circle) and CN_{10} (red opened circle) concentrations with a standard deviation from March 2009 to February 2015. ~~Here the~~ The error bars represents the standard deviation of the measurements from the mean value.

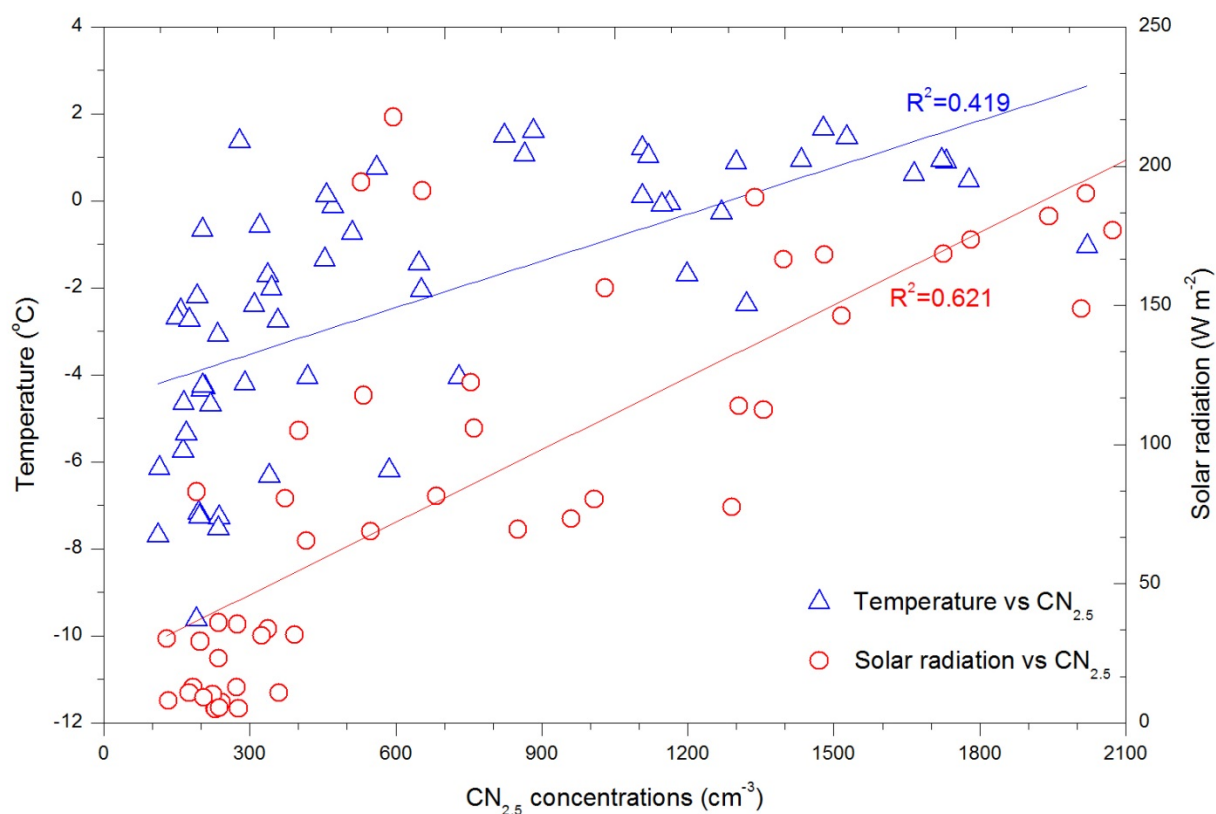


Figure 5. Scatterplot diagram of monthly mean $\text{CN}_{2.5}$ concentrations and monthly mean temperature (blue opened triangle) or monthly mean solar radiation intensity (red opened circle). Blue and red solid lines are a regression lines.

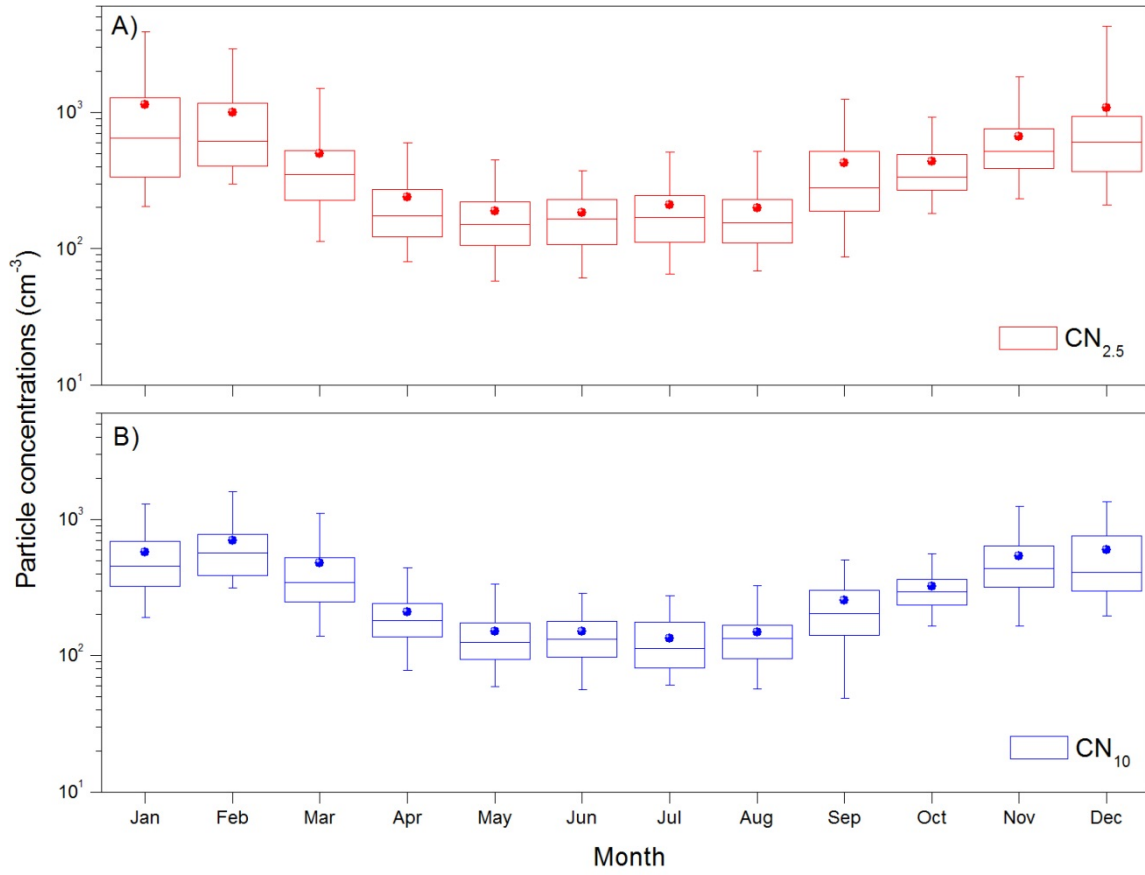


Figure 6. Box plots of seasonality of (a) CN_{2.5} and (b) CN₁₀ concentrations. Lines in the middle of the boxes indicate sample medians (mean:— value is closed circle), lower and upper lines of the boxes are the 25th and 75th percentiles, and whiskers indicate the 5th and 95th percentiles.

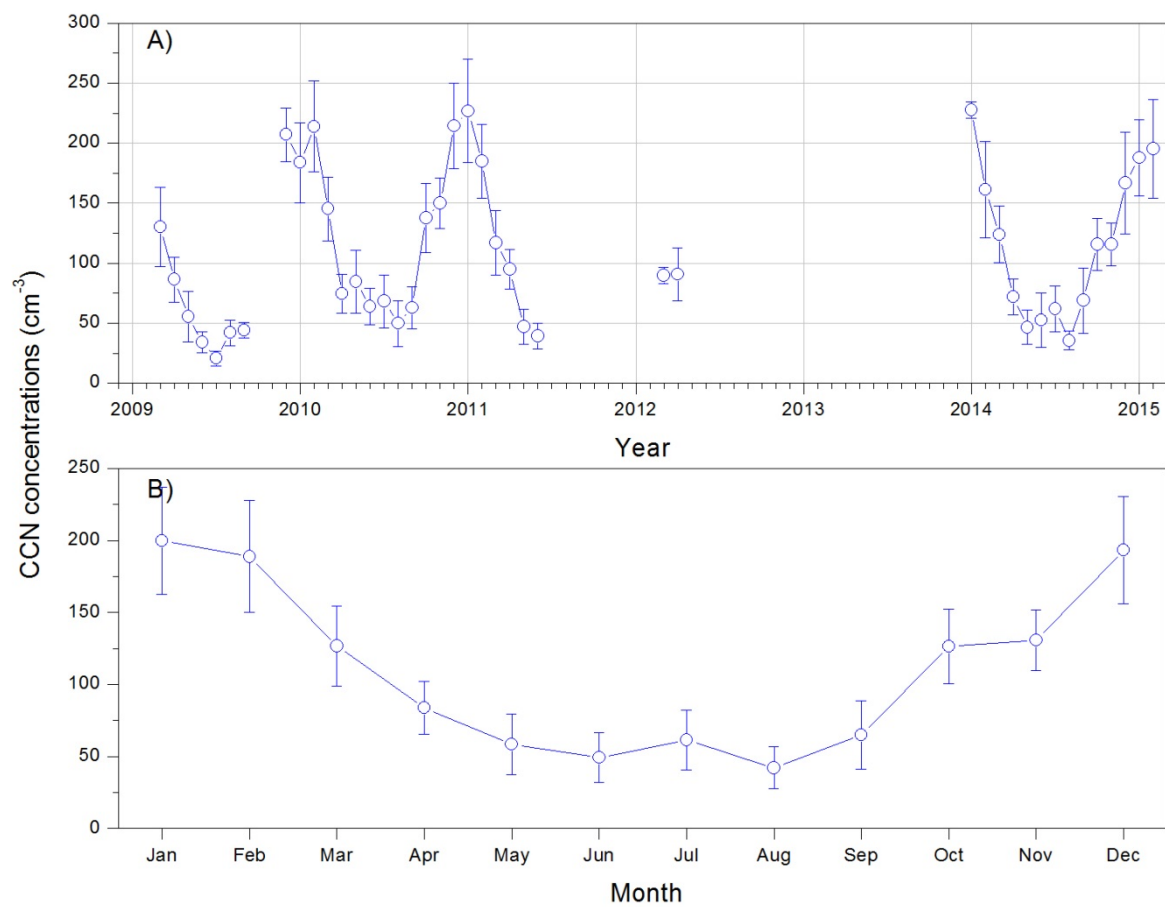


Figure 7. (a) Monthly mean CCN concentrations at the SS of 0.4 % with a standard deviation from March 2009 to February 2015 (b) Seasonal variation of mean CCN concentrations at the SS of 0.4 % with a standard deviation.

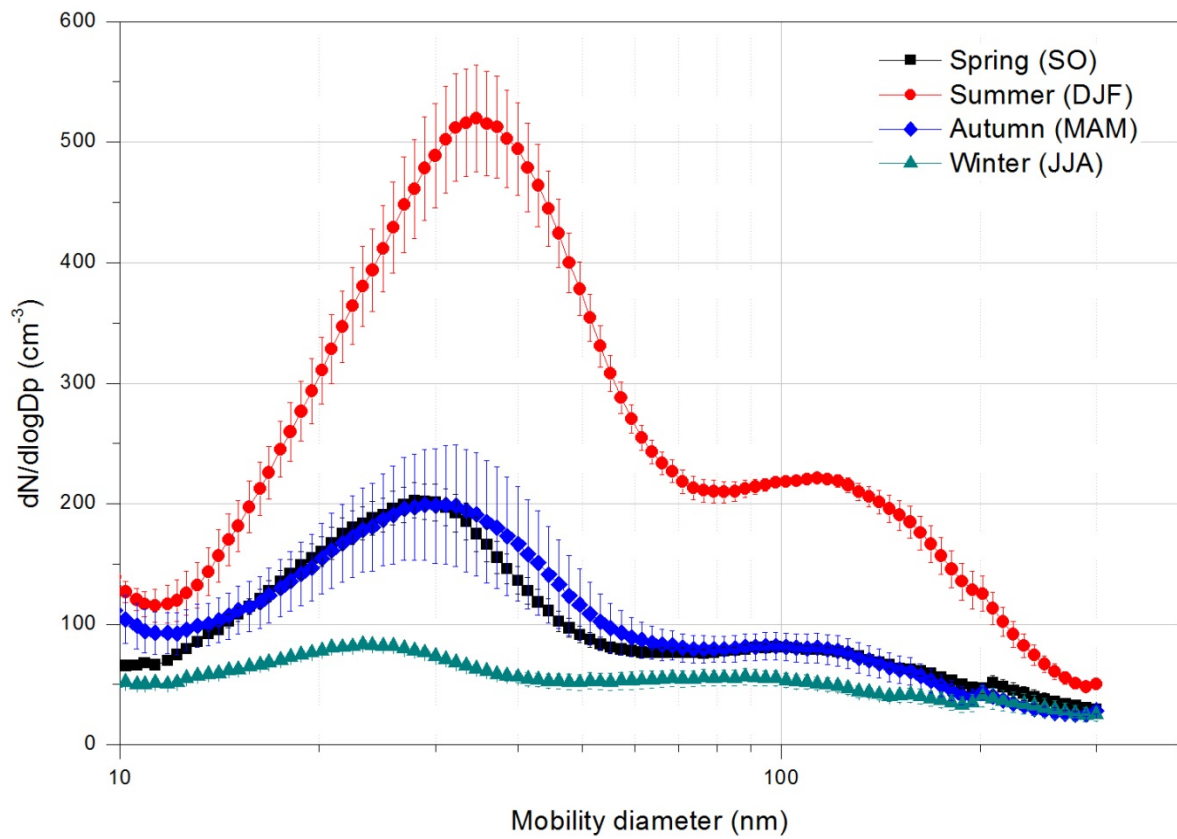


Figure 8. Seasonal mean aerosol size distribution measured by the SMPS at the King Sejong research station over the period from March 2009 to February 2015. ~~Here the~~ The error bars represents the standard deviation of the measurements from the mean value.

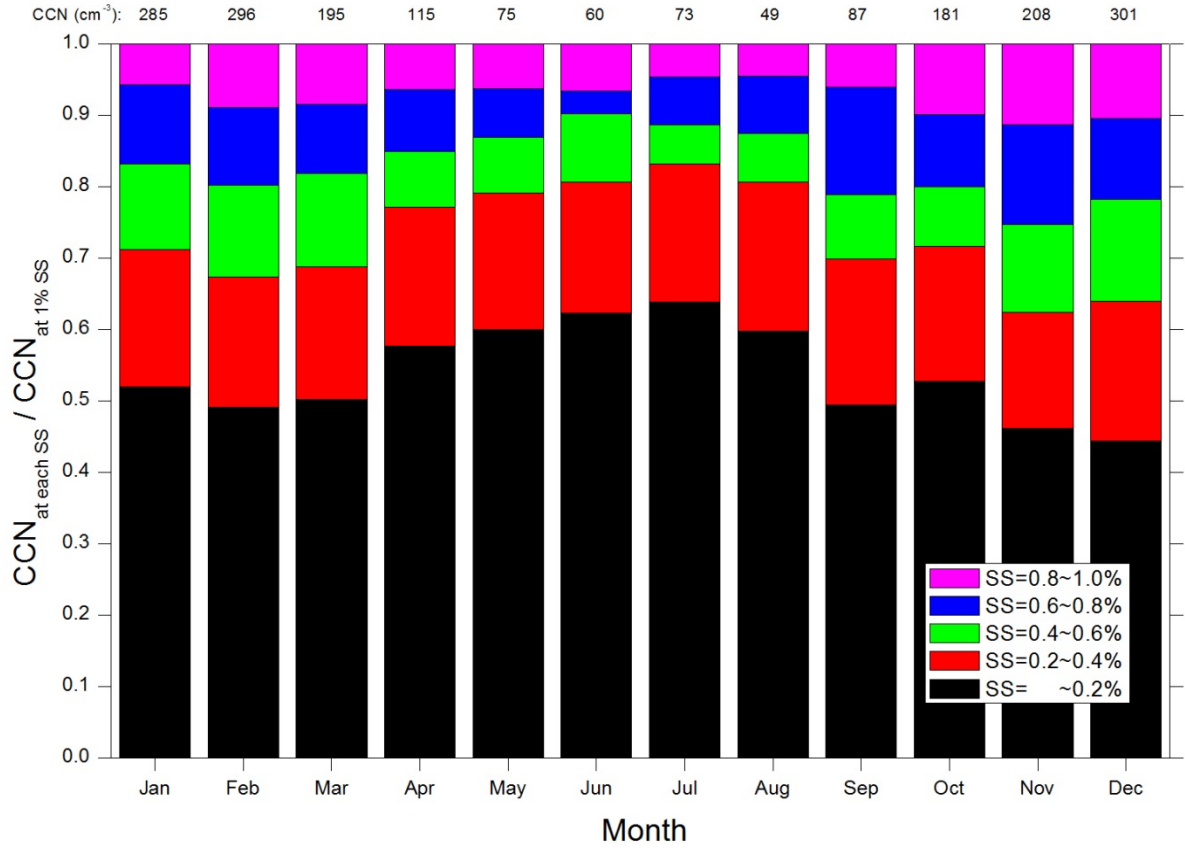


Figure 9. Monthly mean cumulative CCN concentrations shown as fractions of the CCN concentration at the SS of 1.0 %. Colours indicate the SS bins. The number at top of figure represents monthly mean CCN concentrations at the SS values of 1.0 %.

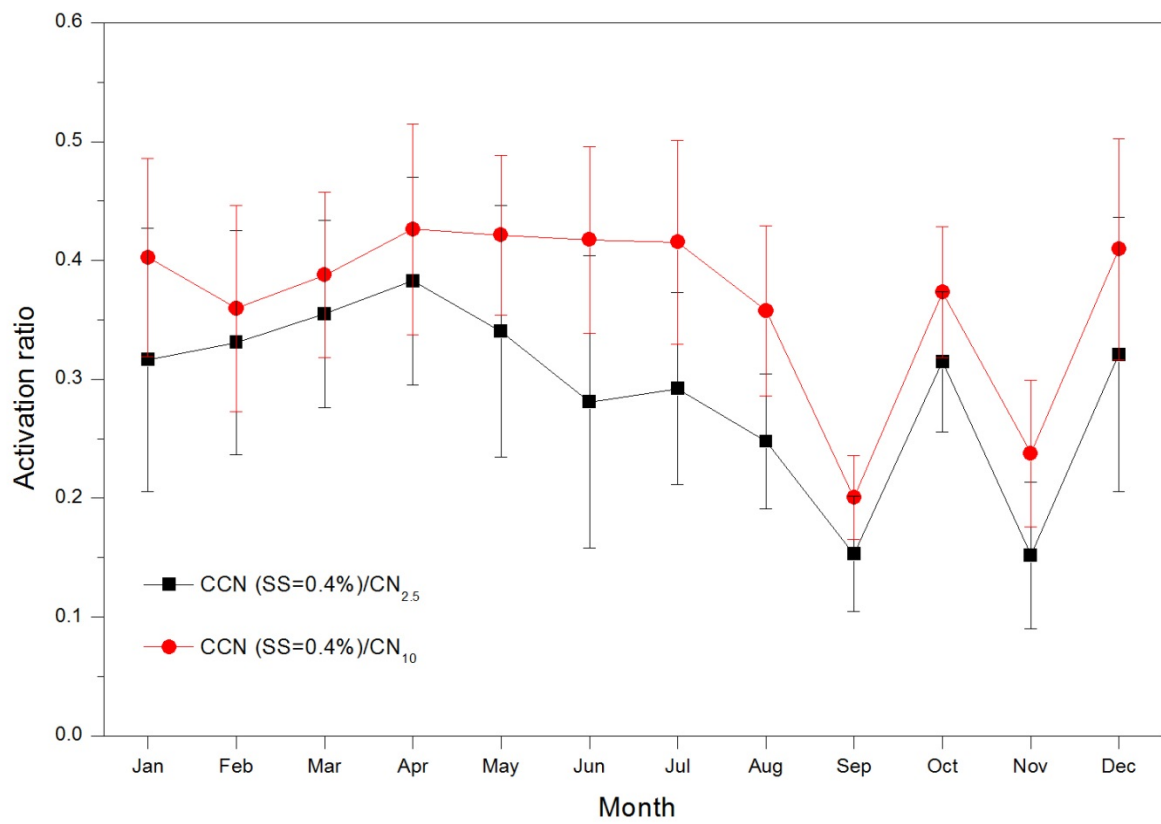


Figure 10. Comparison of the seasonal mean variation of the activation ratio between measurements
 5 | (CPC 3776 and CPC 3772) by two CPCs. ~~Here the~~ The error bars represents the standard deviation
 of the measurements from the mean value.

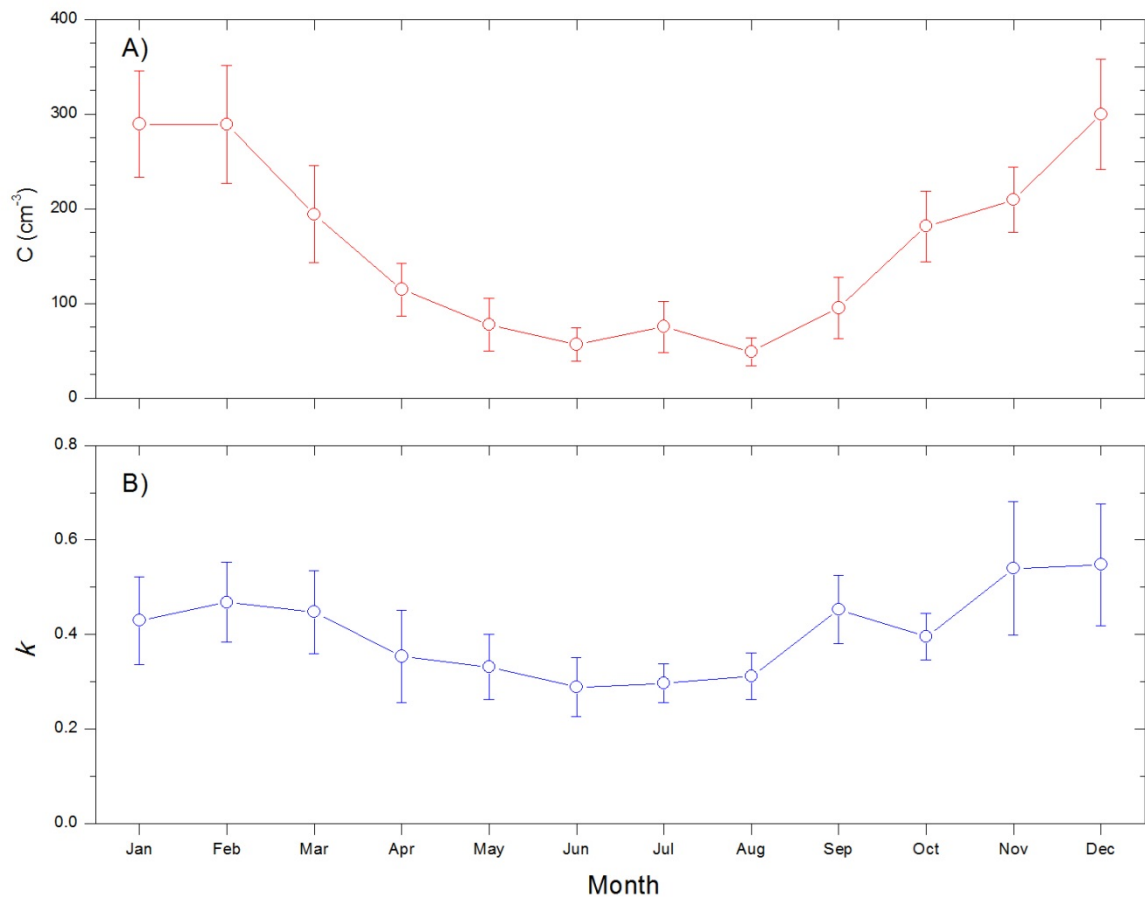


Figure 11. Seasonality of monthly mean values of (a) C and (b) k over the whole observation periods.

5 | ~~Here the~~ The error bars represents the standard deviation of the measurements from the mean value.

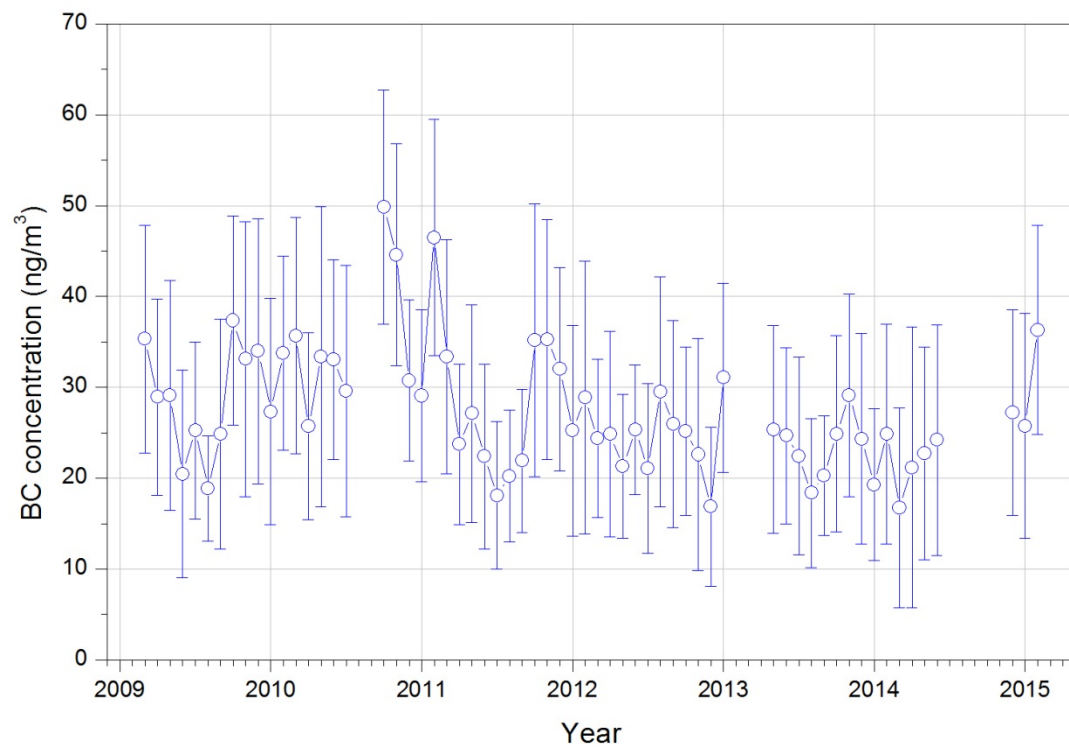


Figure 12. Monthly mean concentrations of black carbon over the period from March 2009 to February 2015. ~~Here the~~The error bars represents the standard deviation of the measurements from the mean value.

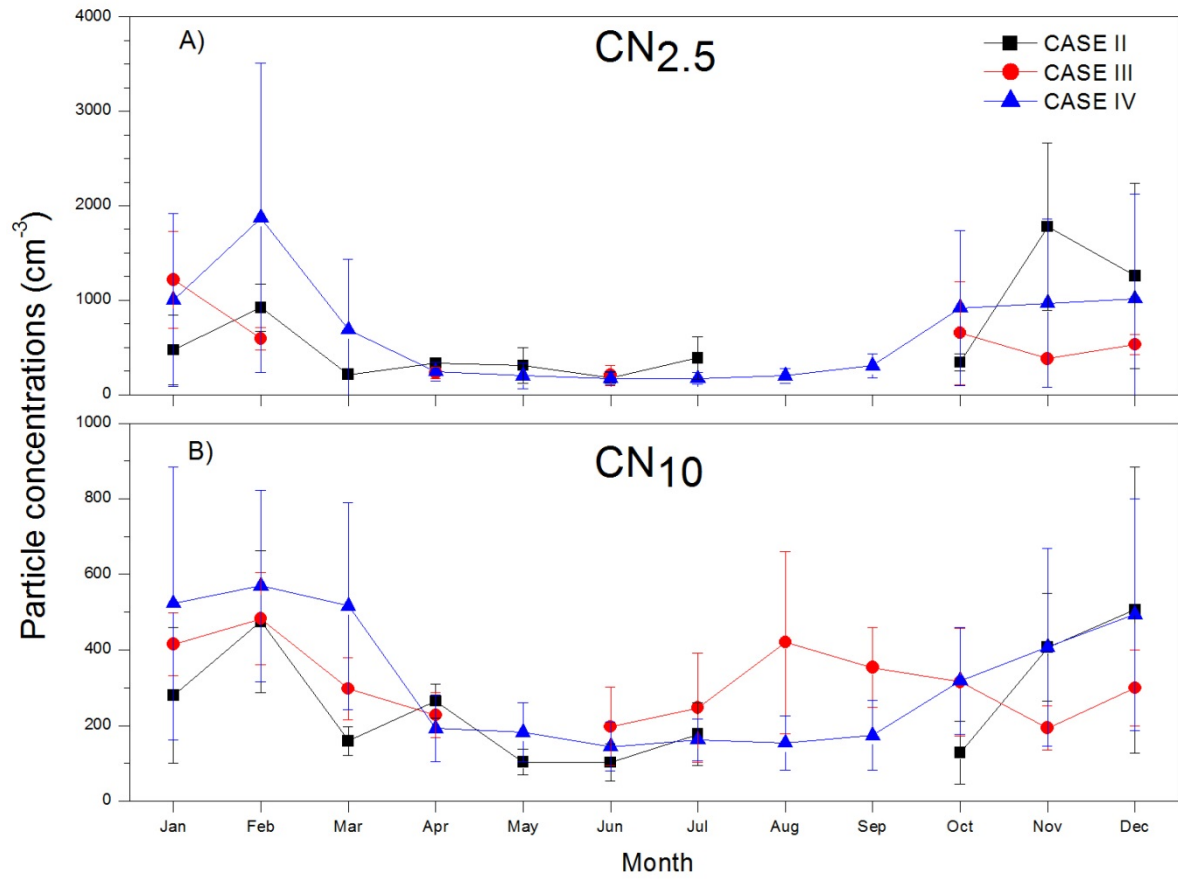


Figure 13. Seasonal variation of mean (a) CN_{2.5} and (b) CN₁₀ concentrations with a standard deviation depending on the air mass origin. ~~Here the~~ The error bars represents the standard deviation of the measurements from the mean value.

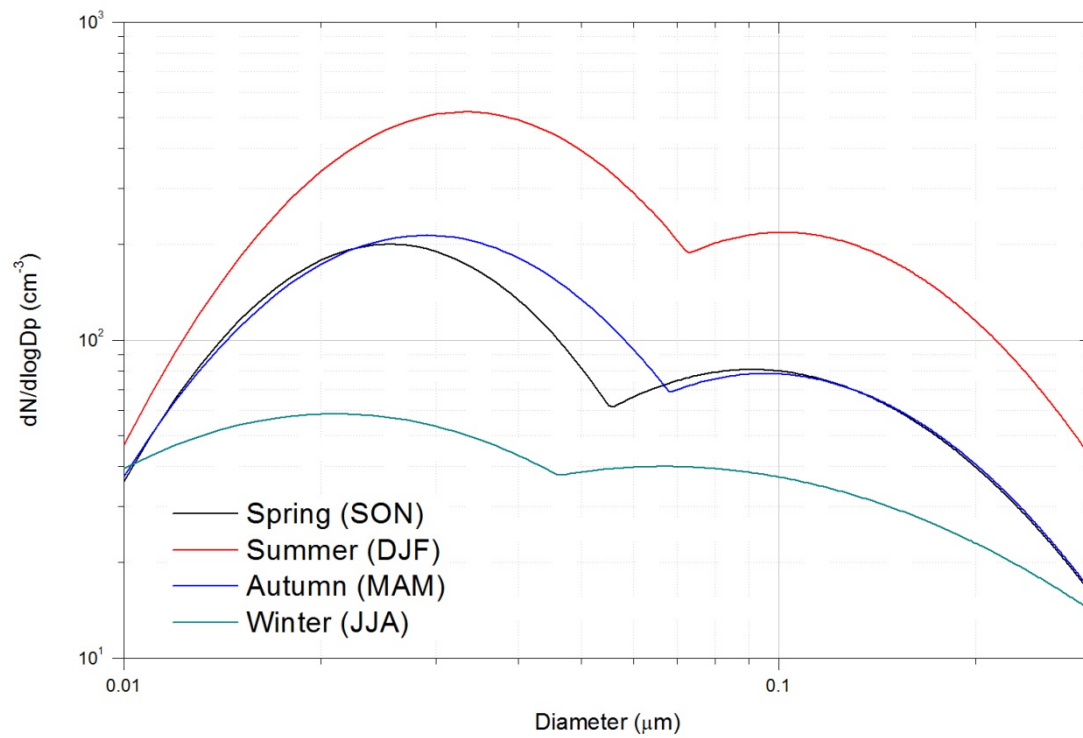


Figure 14. Seasonal lognormally fitted size distribution of aerosol particles originating from the South Pacific Ocean, ranging from 0.01 to 0.3 μm (Case IV).

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
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

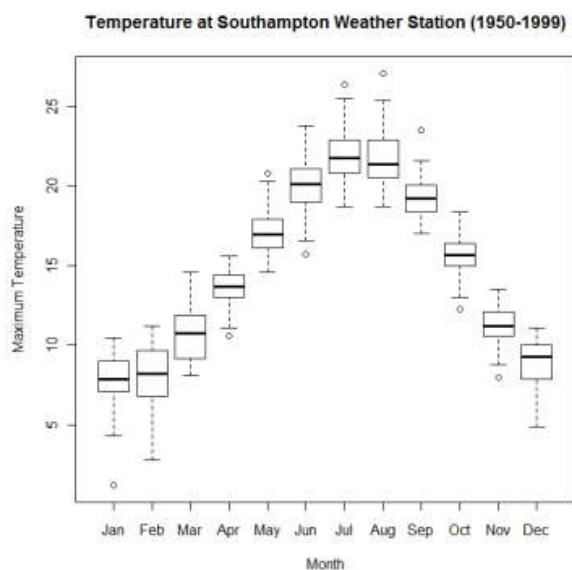
Table 2. Seasonal size distribution lognormal fitting parameters for the Aitken and Accumulation mode of aerosol particles originating from a Case IV scenario. N , σ , and D_g refer to the number concentrations, a standard deviation, and the geometric mean diameter, respectively.

	Aitken mode			Accumulation mode		
	$N \text{ (cm}^{-3}\text{)}$	σ	$D_g \text{ (}\mu\text{m)}$	$N \text{ (cm}^{-3}\text{)}$	σ	$D_g \text{ (}\mu\text{m)}$
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

Anonymous Referee #2

The authors have addressed most of my concerns. I still have one minor and one major concern.

1) Minor concern: Figure S1, as presented to me in the authors' response, is unreadable. I have not been able to check the supplemental document but if it looks the same the authors need to make it clearer. Just to be perfectly clear, this is what I think a box-whisker plot should look like (I took this random plot from the web):



Authors' response: For the reviewer's and editor's reference, we have redrawn box-whisker plots of particle size distribution as shown in following figures (to show supplementary information of the Figure S1(a)-(d) of the 'author's reply to the reviewers') as:

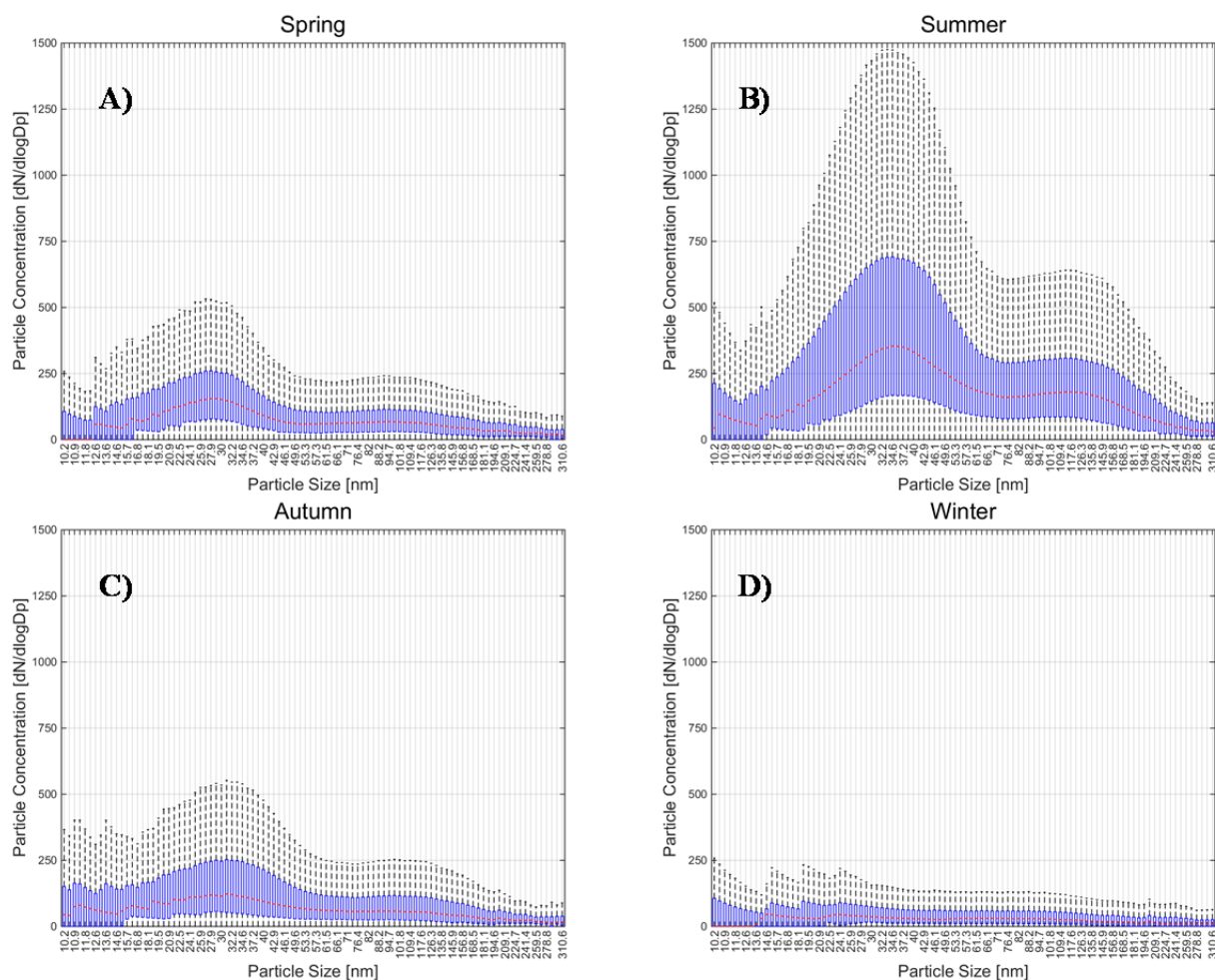


Figure S1. Box-whisker plot of particle size distribution during (a) Spring, (b) Summer, (C) Autumn, and (d) Winter season.

These new plots used same data-set as the figure 8 of the manuscript, do not show clear increase of recently formed particles. The pattern that the concentration of Aitken mode particles show clear sudden increase during summer season can be seen from the both formats (figure 8 of the manuscript and figure S1 of the reply to the reviewers), authors have chosen to show the seasonal size distribution in the way of mean and standard deviation, as the figure 8 of the manuscript.

2) Major concern: The authors' response to my comment on the CCN measurements does not give me great comfort that the authors have made a careful analysis of their data. While I am happy that the authors took the time to calculate kappa, I would argue two issues:

(a) It is, in fact, quite common to estimate kappa based on bulk measurements of CCN concentrations and SMPS-derived size distributions. As the authors point out, an internal mixture must be assumed. While it's true that this may introduce uncertainties, it is still an important and in fact is an often utilized, method for estimating kappa. Just a quick literature search uncovered three

studies where this was done [1-3], and I am sure there are dozens more.

(b) If the authors feel that they have performed a correct calculation of kappa then they should clearly state their methods and result in their manuscript. It is OK if the value of kappa is unreasonable (and 1.18 is an unreasonable value!!). But it is not OK (in my opinion) to perform a good calculation that provides an unsatisfactory answer and then ignore the result.

Authors' response: We re-calculated the kappa value using monthly mean CCN concentrations at the SS of 0.4%, monthly mean CN concentrations measured by CPC and monthly mean size number distribution results obtained from SMPS data. The critical diameter was estimated using equation introduced by Furutani et al. (2008), as follows:

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

where N_{tot} is total number concentrations of aerosol particles measured by SMPS. D is the electric mobility diameter observed by SMPS. D_0 means smallest size measured by SMPS and D_{act} represents the critical diameter. The CCN/CN ratio indicates the fraction of CCN-active aerosols. Subsequently, the kappa value with the critical diameter and SS value can be estimated using the following equation (Petters and Kreidenweis, 2007);

$$\kappa = \frac{4A^3}{27D_{act}^3 \ln^2 SS}$$

$$A = \frac{4\sigma_w M_w}{RT\rho_w}$$

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. Re-calculated kappa value was found as 0.15 ± 0.05 , a reasonable value compared to the previous studies (Kammermann et al., 2010; Martin et al., 2011; Lathem et al., 2013; Jaatinen et al., 2014).

The possible reason for difference of the kappa value between the previous calculation and the present calculation should be attributed to the difference in the calculation of the critical diameter. In the previous calculation, we decided the critical diameter by comparing CCN concentrations with the

integrated SMPS results (SMPS data integrated from larger to smaller particles: 300 nm to 10 nm). In the previous calculation, the critical diameter was decided when the integrated number concentrations equal to the total CCN concentrations. Because SMPS adopted in this study can scan particle number distribution only up to 300 nm, this might mislead information of the critical diameter in the previous calculation.

In the revised version, we re-calculated the critical diameter and kappa value by comparing fraction of non-CCN-active aerosol particle with the integrated SMPS data (using equation 3 of revised manuscript). The re-calculated kappa value ranged from 0.044 to 0.343, showing an annual mean value of 0.153. These new calculation of the kappa and discussion were added in section 3.2.2.

In the revised manuscript, we added following paragraph for estimation and discussion of hygroscopicity parameter, kappa, on Page 13 Lien 15-Page 14 Line 15:

“It is useful to infer hygroscopic properties of aerosol particles with a hygroscopicity parameter, kappa. The kappa values varies from 0 for insoluble particles to larger than 1 for water-soluble salt particles (Petters and Kreidenweis, 2007). The kappa value 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 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 where SMPS scan starts. The CCN/CN ratio indicates the fraction of CCN-active aerosols among total particle concentrations.

In this study, the kappa values were estimated using the monthly mean CCN concentrations at

the SS of 0.4%, the monthly mean CN concentrations measured by CPC and the monthly mean size number distribution results obtained from SMPS data. The annual mean kappa value was calculated to be 0.15 ± 0.05 . This value is comparable to the previous studies from Arctic and subarctic areas. For example, Latham et al. (2013) who measured the CCN activity at the Arctic by using aircraft measurements reported the kappa value of 0.32 ± 0.21 . Martin et al. (2011) inferred total kappa of 0.33 ± 0.13 during cruise observation in Longyearbyen, Svalbard. Kammermann et al. (2010) reported the kappa values varied between 0.07-0.21 in the period of 18 days within the Arctic Circle in Sweden. Jaatinen et al. (2014) also showed the kappa value of 0.13 ± 0.07 using 13-day set of data at subarctic area in Finland (Pallas-Sodankylä station)”

In the revised manuscript, we also added the following sentence on Page 2 Line 17-18 in the abstract section and on Page 18 Line 4-5 in the summary and conclusions section.

“Furthermore, the annual mean hygroscopicity parameter, kappa, was estimated as 0.15 ± 0.05 , for SS of 0.4%.”

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3. Jaatinen, A., et al., The third Pallas Cloud Experiment: Consistency between the aerosol hygroscopic growth and CCN activity. *Boreal Environment Research*, 2014. 19: p. 368-382.

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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.

Anonymous Referee #4

Review of the manuscript entitled “Seasonal variations in physical characteristics of aerosol particles at the King Sejong Station, Antarctic Peninsula” by J. Kim, Y. J. Yoon, Y. Gim, H. J. Kang, J. H. Choi, and B. Y. Lee, with reference no.: acp-2016-795.

This manuscript presents and analyses measurements of aerosol properties (number concentrations, size distributions), cloud condensation nuclei (CCN) and black carbon concentrations for a six-year period (March 2009 to February 2015) at the King Sejong research station in Antarctica.

The Cryosphere and especially Arctic and Antarctica are key components of the Earth’s system, and are inherently sensitive to a changing climate serving as the most stunning indicators of climate change. On the other hand, among climate change drivers, aerosols still contribute the largest uncertainty to the total climate forcing estimate especially through the aerosol – cloud interactions. This is due to the great variety of aerosol types, both natural and anthropogenic, their short atmospheric lifetimes and to the subsequent high spatiotemporal variability of their physical and optical properties. The Antarctic continent being the most remote area on the planet from other continents and thus from anthropogenic activities and emissions, it is an ideal place for studying natural aerosol processes in order to understand them and to correctly distinguish between natural and anthropogenic factors influencing the climate. Apart from some long-range transported pollution aerosols, primary aerosol sources like mineral dust, vegetation, soot or secondary aerosols from gas to particle conversion are virtually absent on this almost completely ice-covered continent. Hence, marine air masses advected from the Southern Ocean surrounding the continent, remains the dominant source to the Antarctic aerosol load. Therefore, any dataset of original and accurate measurements that helps to elucidate physical processes taking place in a such climatically sensitive region is important.

In this framework the submitted manuscript is interesting and relevant to the topics of ACP. Moreover it is well written and organised and thus it could be published in the ACP Journal after taking into account the following comments.

The manuscript presents interesting results on Antarctic aerosols based on continuous relatively long term (six years) observations at King Sejong station. The dataset is unique and the analysis of measurements is quite adequate. Core of the manuscript is data analysis on a seasonal basis and at a

next level according to the origin of air masses though timeseries of monthly mean data of some variables are presented (e.g. Fig. 3, Fig. 4, Fig. 7a and Fig. 12). Authors analyse the intra-annual variation of examined parameters and they discuss the main features of their seasonal behavior making an effort to provide possible explanations to interpret their findings. In some cases, they compare their results with that of other research works conducted in Antarctica. However, the whole analysis doesn't go deeper to gain an important insight into the factors determining the aerosol properties seasonality and the factors affecting the CCN activation. For instance, is the new particle formation the only or the main factor that induces seasonal variation in particle concentration (total and CN2.5)?

My main concern is about the gain of the new knowledge that this paper brings. Authors cite in the text, especially in the introduction, several works on Antarctic aerosols and their properties. So, what is the contribution of this manuscript to this knowledge? In the introduction authors state "Although various studies have been performed, the measurements taken at the Antarctic Peninsula and the long-term observations of aerosol particles are still insufficient" but they do not discuss any inter-annual variability or trend (except for the exceptional year of 2014). They focus on the seasonal variation. In order to support their work, authors should clearly state what are the new approach, analysis and/or findings compared to literature and this should be clearly presented in the concluding section as well. Otherwise, they can discuss their results compared to other works, examining whether they are in agreement strengthening thus the existing knowledge since current results are issued from multi-year observations. Actually they do it sometimes. For instance, authors state that the revealed seasonal pattern of CN2.5 and CN10 is in agreement with the results of previous studies (page 8, lines 8-10). I am wondering if the consideration of the seasonal variation of CN2.5 and CN10 separately, is additional and further information compared to previous works. On the other hand, throughout the discussion regarding timeseries, seasonal behavior of CCN concentrations, particles size distribution and CCN activation ratios, there are no references to other relative studies in Antarctica. If this analysis and its findings give new or additional information should be stated by authors adding thus value to their work. The same is valid for the analysis regarding the effects of air masses origin on the aerosols physical properties.

I should however state that the lack of new knowledge doesn't reduce the value of a dataset of original measurements of aerosol properties with a relatively long temporal coverage, in a remote, not easily accessible and very interesting from climate change point of view, area of the planet.

Authors' response:

We appreciate positive feedback from the reviewer. We think the value of this manuscript lies on the fact that although many studies have been performed in Antarctica, research on seasonal variations of CN, CCN, and size number distribution was less conducted in Antarctic Peninsula. In addition, published papers obtained by long-term observations were rare. Based on the multi-year observation in Antarctica, in particular, analysis on characteristics of CCN has been carried out for the first time, to authors' knowledge.

Some minor remarks

- Authors give enough information about instrumentation but they do not discuss any quality control assessment that they apply to their raw records.

Authors' response: To minimize the effect of local source on CN, CCN, and SMPS data, all data were eliminated when wind direction was within 355-55° and the BC concentrations were higher than 100 ng m⁻³ (see Page 5 Line 5-7). When daily and monthly mean concentrations of CN and CCN were estimated with remaining data, subsequently, daily data that the rate of daily data acquisition was higher than 50% were only used to secure the quality of raw data.

In the revised manuscript, we added following sentence on Page 5 Line 7-8.

"In order to ensure the reliability of measurements, only dataset, of which acquisition rate higher than 50%, were used during all analysis procedures."

- Authors trying to interpret the exceptional CN concentration levels during the period 2013-2014, found that air masses origin was differentiating this period compared to previous years. Air masses from south Atlantic were more frequent than other years. A comparison of CN concentration levels with analogous measurements (if there are published) at stations which are affected mostly by south Atlantic air masses could support this argument.

Authors' response: Virkkula et al. (2009) showed CN concentrations measured at Aboa Station from December 2003 to January 2007. Aboa Station is located in coastal area in inland Antarctica and is affected by south Atlantic air masses. Although there is variation of the CN concentrations year by year, the daily CN concentrations during astral summer were ~ 600 cm⁻³. It is quite similar value comparing with our results during the period of 2013-2014.

In the revised manuscript, we added the following sentence on Page 10 Line 4-8:

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

- Analyzing the CCN concentration, it was found that its seasonal variation follows the seasonal cycle of particles concentration which is logical. I have however point out that the CCN concentrations during the period 2013-2014 seem to be unaffected by the low particles concentration in that period as they remain similar to other years.

Authors' response:

Authors appreciate the issue raised by the reviewer pointing out the fact that CCN concentration in the summer season of 2013-2014 shows normal value. Unfortunately, we do not have CN10 or SMPS data during the period 2013-2014 (only CN2.5 data are available). Authors think the normal CCN concentrations for 2013-2014 summer season may imply the hypothesis that the nucleation of new particles ($2.5\text{nm} < D_p < 10\text{nm}$) was less frequent or weaker for 2013-2014 summer season, judging from the fact that (i) the lower concentration of CN2.5 compared with other summer seasons, and (ii) the fairly normal concentration of CCN (see figure 7 (a)).

For reviewer's reference, in the section 3.2.1, we have been modified to relate frequent air mass origin with the lower CN2.5 concentrations in the summer season of 2013-2014, by adding the following paragraph in revised manuscript on Page 9 Line 26-Page 10 Line 3:

“Unfortunately, neither CN10 nor SMPS data are available for the austral summer season of 2013-2014 because of mechanical failures, it is not possible to directly explain the low concentrations of CN2.5 for this season in terms of the potential effects of air mass characteristics on the concentration of 2.5-10 nm size particles. Nevertheless, it is likely that the increased frequency of air mass originating from the South Atlantic Ocean (Case II) might have resulted in the lower $\text{CN}_{2.5}$ concentration of the austral summer season of 2013-2014.”

- Page 3, lines 8-9. In the sentence “.... the direct and indirect climate effects are still unknown (IPCC, 2013).”, I think the word “unknown” is not appropriate. Actually, according to IPCC report, aerosol effects contribute the largest uncertainty in the total radiative forcing. Thus you can replace the word ‘unknown’ by ‘highly uncertain’.

Authors' response: In the revised manuscript, text was changed accordingly, see Page 3 Line 9.

- Page 3, line 21. In the sentence “For these reasons, the observation of the physical properties in Antarctica, ...”, replace the word ‘the’ by ‘their’

Authors' response: In the revised manuscript, text was changed accordingly, see Page 3 Line 21.

- Page 7, line 14. In the sentence “Fig. 3 depicts monthly variations of the meteorological parameters measured from and automatic weather system (AWS) ...” replace the word ‘and’ by ‘an’

Authors' response: In the revised manuscript, text was changed accordingly, see Page 7 Line 22.

- Page 7, line 18. In the sentence “the observation site was relatively humid and warm condition compared to inland Antarctic stations”, remove the word ‘condition’

Authors' response: In the revised manuscript, we removed “condition” accordingly, see Page 7 Line 26.

- Page 15, line 6. In the sentence “Our results are similar those of previous laboratory and field experiments (Sellegrì et al., 2006; Yoon et al., 2007).”, add the word ‘to’ after the word ‘similar’.

Authors' response: In the revised manuscript, text was changed accordingly, see Page 17 Line 2.

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