Article

Aerosol Properties within and above the Planetary Boundary Layer across the Korean Peninsula during December 2016

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Abstract: During December 2016, airborne aerosol measurements were taken at multiple heights across the Korean Peninsula to examine the vertical properties of aerosols. This study showed that aerosols above the planetary boundary layer (PBL) show similar concentrations and particle size distributions (PSDs), regardless of the relative locations in Korea. On the other hand, aerosols within the PBL differ depending on the geographical location, origin and path of the air mass. The concentrations are the highest in Seoul, followed by Gangneung, East Sea and the Yellow Sea. The known east–west aerosol gradient did not appear and the reasons are discussed in this paper. The study further shows that the aerosols of upwind regions affect the aerosols above the PBL, whereas aerosols in the PBL are affected by local sources and atmospheric conditions in addition to aerosols of upwind areas.

Keywords: vertical structure of aerosols; continental outflow regions; east–west aerosol gradient

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1. Introduction

Aerosols considerably affect human health, air quality and climate systems through the radiative budget by directly scattering and absorbing aerosols and indirectly interacting with clouds [1–7]. Tremendous efforts have been made to monitor surface aerosol concentrations through committed long-term measurements at a location [8], the AERosol Robotic NETwork (AERONET) [9] and dedicated field campaigns [10–12]. However, it is challenging for ground-based point observations to cover the global distribution of aerosols. Satellite measurements provide a global view of aerosols but suffer from retrieval biases [13]. Besides, the vertical distribution of aerosols, an essential component of the aerosol indirect effect, is usually unknown. Lidar measurements provide an excellent tool to illustrate the vertical structure of aerosol concentrations at a location [14]. However, lidars cannot always define the vertical structure of aerosols if clouds and aerosols coexist. Aerosols decrease with heights and are homogeneous horizontally, but the vertical and horizontal distribution can be complicated when the aerosols are advected from remote locations [15,16]. Accordingly, in situ measurements of aerosols, showing the horizontal and vertical distributions, are needed to validate the retrieved aerosol products from remote sensors.

Korea is in the continental outflow regions and is affected by various kinds of aerosols. Aerosol studies in and around the Korean Peninsula have been conducted using ground- and ship-based measurements [10,17–20], satellite-based aerosol optical depth (AOD) [21], lidar retrieval of aerosol mass concentrations [22] and aircraft measurements [19,23,24]. In contrast to the remote sensor, an aircraft can directly probe aerosol properties using in situ instruments. Previously, aerosols were sampled at a constant height of ~3 km over

the inland of the Korean Peninsula and aerosols were obtained from sounding measurements over the ocean [19]. Here, sounding measurements indicate an aircraft probing the atmosphere spiral up or down and a level-leg flight measures aerosols at a constant height. Aerosol data obtained from one constant height hardly represent the overall atmospheric aerosols, especially if the measurements were made higher than the planetary boundary layer (PBL), because sub-cloud layer aerosols are ingested into the clouds, affecting the cloud formation and precipitation processes. Further, sounding measurements have difficulty in representing aerosol values when horizontal variations in aerosols exist. The horizontal variations of aerosols are common when aerosols are transported from remote locations [16]. Therefore, in this study, multiple level-leg flights and sounding measurements were combined to take advantage of both strategies. Airborne aerosol measurements in and around South Korea were recorded [18,19,23]. However, none of them has sampled aerosols at more than a single-level height across the Korean Peninsula. This study provides the first vertical structure of aerosols (concentration and size distribution of aerosols) over the inland of the Korean Peninsula using in situ aircraft measurement.

In situ aerosol airborne measurements were recorded in and around the Korean Peninsula from 1 December to 11 December 2016, to better understand the aerosol properties over the Korean Peninsula, which corresponds to the continental outflow regions. This study provides in situ aerosol measurements over the Korean Peninsula, obtained in and above the PBL, and the results can be used to validate the aerosol retrievals from remote sensors. Although East Asia is a region where a diverse and extensive source of aerosols exists, limited data on aerosol properties are available, especially as a function of height above the surface. As this is the first vertical measurements made over the inland of Korean Peninsula, this study focuses on analyzing the vertical structure of aerosols measured over the Yellow Sea, Seoul, Gangneung and East Sea by the aircraft transects from east to west in the Korean Peninsula. Previous studies [19,21] have shown the east–west aerosol gradients across the Korean Peninsula; thus, this study further verifies the east–west gradients of the total aerosol number concentrations (N_{cn}).

2. Data and Methods

For this study, we used a Cessna206. The aircraft was equipped with a condensation particle counter (CPC, TSI CPC3010) and an aerosol spectrometer called Sky-OPC (GRIMM Aerosol Technik). The CPC measures all aerosols larger than $0.01\ \mu\text{m}$, whereas the Sky-OPC measures aerosols between $0.25\ \mu\text{m}$ and $32\ \mu\text{m}$ at 32 bins. The total aerosol number concentrations (N_{cn}) measured using the CPC during the flights were corrected for pressure using the method described in [18]. The particle size distribution (PSD) and particulate matter (PM) with a diameter of $10\ \mu\text{m}$ or less (PM₁₀) shown in this study were obtained from the Sky-OPC.

The 24 h back trajectory arriving at 500 m and 2000 m above the ground level (AGL) over the Yellow Sea, Seoul, Gangneung and East Sea were calculated using the Hysplit model (http://ready.arl.noaa.gov/HYSPLIT_traj.php, accessed on 4 October 2021) to represent the heights of air mass arriving within and above the PBL, respectively. The PBL height can be referred to by the vertical structure of potential temperature where the upper-air sounding data were downloaded from <http://weather.uwyo.edu/upperair/sounding.html> (accessed on 4 October 2021). PBL defines the interface between a well-mixed layer and the free atmosphere, where the mixed layer is a zone having nearly constant potential temperature and specific humidity with height [25].

The Korean Peninsula is in the continental outflow regions, between China and Japan (Figure 1a) and is heavily affected by aerosols from China. In December 2016, nine research flights (RFs) were made to characterize the background aerosols over the Korean Peninsula, including the Yellow Sea, Seoul, Gangneung and East Sea. Seoul is the capital of South Korea and is a typical large urban area in East Asia. Gangneung is in the east part of the Korean Peninsula and is considered a clean environment, less populated and less polluted than Seoul. Table 1 summarizes the details of the nine individual flights.

Figure 1. (a) Geographical locations of an experimental area with (b) an enlarged area of the Korean Peninsula. A and B in (b) show the Yellow Sea and Gangneung roughly, respectively. In (b), the blue lines indicate the flight patterns for the Yellow Sea and Seoul and magenta lines indicate the flight patterns for the Gangneung area parallel to the coastline. The black line in (b) shows the Han River, which flows across Seoul, the capital of South Korea. Seoul and upper-sounding locations (Osan and Bukgangneung) are overlaid as black dots in (b).

Table 1. Flight Summary.

RF No.	Date	Time (LST) HH:MM	Main Flight Area	Path No.	Note
1	1 December 2016	15:13–16:32	Seoul	3	Two layers along the Han River Flight was made across the coastline
2	2 December 2016	09:36–12:44	Gangneung	1	Flight was made across the coastline
3	2 December 2016	13:51–18:13	Gangneung	1	RF was made along the Han River (the lowest level only), then flew to the Gangneung area (across the coastline)
4	6 December 2016	7:47–11:35	Yellow Sea, Seoul	3	Three layers along the Han River
5	9 December 2016	14:22–16:12	Seoul	3	Three layers along the Han River
6	10 December 2016	7:56–11:37	Yellow Sea, Seoul	3	Three layers along the Han River
7	10 December 2016	13:37–16:21	Gangneung	2	RF was made along the Han River (the lowest level), then flew to the Gangneung area (parallel to the coastline)
8	11 December 2016	7:45–11:22	Gangneung	2	RF was made along the Han River (the lowest level), then flew to the Gangneung area (parallel to the coastline)
9	11 December 2016	12:51–16:39	Gangneung	2	Parallel to the coastline

RF indicates research flight. Path 1: Flew to Gangneung and sampled aerosols across the coastline at three constant heights. Path 2: Flew to Gangneung and sampled aerosols parallel to the coastline at three constant heights over the land and ocean, respectively. Path 3: Flew along the Han River or flew to the Yellow Sea and sampled aerosols along the Han River. Flights sampled aerosols at two constant heights over the Yellow Sea and 2–3 heights along the Han River.

Figure 1b shows two flight patterns in blue and magenta as an example. The blue lines indicate the flight patterns that sampled aerosols over the Yellow Sea and Seoul on 6 December (RF04). Seoul is a densely populated area and, because of the unique military situation on the Korean Peninsula, flying over Seoul at low altitudes is prohibited. Consequently, for Seoul, flying along the Han River was adopted. Here, the Han River flows across Seoul. The measurements over the Yellow Sea were made 80–90 km away from the Gimpo airport, Seoul. The magenta shows the Gangneung area flight pattern, measuring

aerosols over the region along the coast on 11 December (RF09). For the Gangneung area, two flight patterns were adopted to see whether the differences in aerosols exist on landside and oceanside; one was to fly parallel to the coastline (Figure 1b, magenta) and another was to fly across the coast (not shown here). The aircraft measurements over the East Sea were made ~5 km off the coastline and the distance between the level-legs (land-leg and ocean-leg) was ~10 km.

3. Results

3.1. Back Trajectory

The 24 h back trajectories arriving at 500 m and 2000 m (AGL) over the Yellow Sea, Seoul and Gangneung are shown in Figure 2. Here, 500 m and 2000 m are the heights within and above the PBL, respectively. The average location (latitude and longitude) of level-leg flights at a given area was used as the ending points of the back trajectory. Korea is in the continental outflow regions and the prevailing winds during the wintertime are northerly or northwesterly. The result shows that the air mass that arrived at the Korean Peninsula originated from Lake Baikal or northeast China as a whole, regardless of the relative geographical locations in Korea (Yellow Sea, Seoul and Gangneung). Note that the air mass that arrived at Gangneung at 500 m on 11 December (Figure 2f) stayed over the ocean for at least 12 h before being sampled at Gangneung.

Figure 2. A 24 h back trajectory of air mass arriving at 2000 m (a–c) and 500 m (d–f) over the Yellow Sea (a,d), Seoul (b,e) and Gangneung (c,f). The heights of 500 m and 2000 m represent the heights within and above the PBL, respectively. Each symbol indicates 6 h marks. X denotes Beijing.

Vertical profiles of potential temperature at Osan and Bukgangneung are shown in Figure 3 to examine the PBL height. Here, Osan and Bukgangneung are the nearest upper-air sounding locations to Seoul and Gangneung. The locations of Osan and Bukgangneung are overlaid in Figure 1 as dot symbols. Figure 3 shows that the overall PBL heights are at ~1 km and are lower than 2 km, which is used as the height representing the above PBL. The PBL heights for Seoul and Gangneung are ~1 km in [13] and are consistent with the PBL heights in Figure 3. Referring to Figure 3b,c, as seen from the back trajectory in Figure 2, during the observation period, the northwest wind blew mainly in Seoul and Gangneung and the east wind blew 1.5 km below Gangneung on 10 and 11 December.

Figure 3. Vertical profiles of (a) potential temperature and (b) wind direction at Osan 06 UTC and (c) wind direction at Bukgangneung 00 UTC. Each profile was obtained from the day of flight in Seoul and Gangneung.

3.2. Vertical Structure of Aerosols

The vertical profiles of aerosols measured by an aircraft flying across the Korean Peninsula are shown in Figure 4. The figure shows the mean aerosol N_{cn} and PM10 obtained from the level-leg flights over the Yellow Sea along the Han River (Seoul), Gangneung (continental side) and East Sea during the experiment. In Figure 4, the N_{cn} decreases with height, in general. The most distinct feature of the N_{cn} (Figure 4a–d) is that the concentrations measured at Seoul are higher than the aerosols measured in other areas within the PBL, which is expected, because Seoul is one of the megacities in East Asia. The N_{cn} was measured using a CPC, measuring all particles larger than 10 nm. The PBL aerosols at Seoul ranged from 400 to 10,000 cm^{-3} , but mostly appeared around 4000 cm^{-3} (mean and median), except for 1 December (blue triangle), showing a higher N_{cn} than other days. It is not shown here, but a weak yellow-dust passed by the Korean Peninsula on 1 December, confirmed from nearby ground-based lidar and surface PM10 measurements. The air mass on 1 December originated further northeast (east of Lake Baikal; see the blue-filled triangle in Figure 2) than on other days. The N_{cn} at Gangneung (Figure 4c) are higher than those obtained from the East Sea (Figure 4d), although the geographical distance between the two locations is ~10 km. The N_{cn} at Gangneung in the PBL (Figure 4c) ranged from 800 to 6000 cm^{-3} . According to [26], aerosol concentrations for maritime air and continental air are in the order of 10^2 and 10^3 cm^{-3} , respectively. Gangneung is located close to the sea and is less developed and less populated. So, we presumed that Gangneung would have a low aerosol concentration with the characteristics of being close to an oceanic air mass rather than inland. However, the N_{cn} at Gangneung in the PBL was not much different from aerosols measured at Seoul, in which the N_{cn} ranged from 400 to 10,000 cm^{-3} . Aerosols sampled over the East Sea along the coastline near Gangneung (Figure 4d) in the PBL ranged around 1000–3000 cm^{-3} , which are on the same order of the N_{cn} observed over the East Sea in [19,24]. The mean aerosol concentrations (N_{cn} and PM10) shown in Figure 4 are summarized in Table 2.

Table 2. The mean of N_{cn} and PM10 for the level-leg flights shown in Figure 4.

Location	Figure 4 (Colors)	Date	Height (m)	N _{CN} (cm ⁻³)	PM10 (μg m ⁻³)	
Yellow Sea	(a) and (e)	blue dashed	6 December AM	661.2 ± 966.2	1143.1 ± 642.8	6.37 ± 2.34
				1903.0 ± 76.75	85.6 ± 12.12	0.49 ± 0.14
		red solid	10 December AM	431.6 ± 6.08	288.2 ± 103.89	8.51 ± 3.52
				1866.5 ± 8.07	299.88 ± 356.03	0.42 ± 0.08
Seoul	(b) and (f)	sky solid	1 December PM	738.8 ± 19.8	7773.0 ± 770.8	14.6 ± 3.16
				1738.1 ± 14.1	2935.4 ± 1872.6	6.01 ± 3.33
		blue dashed	6 December AM	406.8 ± 465.96	5335.1 ± 752.9	16.27 ± 3.90
				1413.1 ± 29.62	77.2 ± 18.39	0.86 ± 0.79
				2363.5 ± 10.77	96.7 ± 8.71	0.13 ± 0.08
		cooper solid	9 December PM	722.8 ± 10.39	2785.7 ± 693.4	25.69 ± 4.28
				1454.2 ± 13.56	299.6 ± 252.4	2.35 ± 2.69
				2384.4 ± 19.42	76.6 ± 12.10	0.95 ± 0.19
		red solid	10 December AM	708.4 ± 47.28	1231.3 ± 1089.4	7.35 ± 5.37
				1428.5 ± 9.19	457.8 ± 24.3	0.60 ± 0.13
				2359.0 ± 6.72	387.6 ± 22.8	0.11 ± 0.03
		grey	2 December PM	721.8 ± 9.33	5498.3 ± 1507.0	14.83 ± 3.27
		grey	10 December PM	707.5 ± 8.39	4313.3 ± 2410.4	19.5 ± 13.37
		grey	11 December AM	718.6 ± 21.47	464.98 ± 97.0	3.60 ± 0.73
Gangneung	(c) and (g)	blue dashed dot	10 December PM	427.5 ± 7.03	3926.7 ± 545.0	9.9 ± 2.03
				1910.3 ± 8.87	243.7 ± 109.14	0.68 ± 0.55
		cooper	11 December AM	595.8 ± 9.50	862.85 ± 50.66	6.41 ± 1.02
				1909.4 ± 5.41	141.15 ± 377.4	0.07 ± 0.10
		green	11 December PM	580.4 ± 9.73	5084.6 ± 381.64	8.245 ± 1.46
				1937.5 ± 9.98	136.2 ± 21.27	0.16 ± 0.08
East Sea	(d) and (h)	blue dashed dot	10 December PM	426.4 ± 8.18	1581.7 ± 620.72	6.33 ± 1.32
				1906.9 ± 11.53	306.98 ± 150.51	0.77 ± 0.72
		green dot	11 December PM	607.4 ± 430.59	2446.0 ± 430.60	6.45 ± 1.27

In previous studies, the N_{cn} observed over the Yellow Sea within the PBL ranged from 1000 to 30,000 cm^{-3} (Figure 11 in [19]). In this study, aerosols over the Yellow Sea ranged from 200 to 1000 cm^{-3} , which lies on the smaller side of the N_{cn} shown in previous studies [19]. Interestingly, in this study, aerosols over the Yellow Sea showed the lowest N_{cn} among all locations. However, it is not necessarily implied that the N_{cn} over the Yellow Sea are always the smallest among all aerosols over the Korean Peninsula, as many other factors can contribute to the result. For example, aerosol concentrations measured in the afternoon (PM aerosol) were higher than aerosols measured in the morning (AM aerosol). Coincidentally, all level-leg flights over the Yellow Sea were conducted in the morning, whereas the aerosols from other locations were sampled in the mornings and afternoons, indicating that the smallest N_{cn} over the Yellow Sea can be sample bias. However, the N_{cn} over the Yellow Sea was less than the N_{cn} in Seoul on 6 and 10 December, when the aircraft sampled the aerosols at both locations with the same flights, indicating that the lower N_{cn} recorded over the Yellow Sea is not a simple sample bias. The aerosols measured over the Yellow Sea above the PBL are also smaller than or similar to those in Seoul. The smallest N_{cn} over the Yellow Sea within and above the PBL could be related to the air mass origin and path. Figure 2 shows that the air mass observed over the Yellow Sea (Figure 2a,d) originated

near Lake Baikal or northeast China (a relatively clean environment). Furthermore, the air mass stayed over the ocean for at least 6 h before the aerosols were sampled over the Yellow Sea.

Figure 4. The vertical structure of total number concentration (N_{cn} , cm^{-3}) and PM_{10} ($\mu\text{g m}^{-3}$) obtained from the Yellow Sea, Seoul, Gangneung and East Sea. (a–d) indicate N_{cn} and (e–h) represent PM_{10} . Individual dots indicate the mean value of N_{cn} and PM_{10} obtained at constant heights (level-leg flights). The vertical lines connect aerosols obtained from level-leg flights. The horizontal bar represents standard deviation. Gangneung and East Sea flights were made along the coastline, 10 km apart (Figure 1). The colors are the same as those shown in back trajectories in Figure 2. For example, the blue circles with dashed lines in Figure 4a indicate N_{cn} obtained on 6 December AM flight.

Previously, [27] analyzed the air mass origin associated with haze events in Seoul from 2009 to 2010. They showed that the high concentrations of aerosols observed over the Korean Peninsula were linked to the air mass originating from highly populated polluted areas (northern and southern China, such as Beijing and Shanghai) and transported to Korea through the westerly. On none of the days during the experiment, air masses were not originated from near Beijing nor Shanghai.

Figure 4a–d further shows that PM aerosols (filled symbols) within the PBL are higher than morning aerosols (open symbols) as a whole. Specifically, on 11 December, the flight paths were similar in the morning and afternoon flights and the back trajectories were similar too (orange for AM, green for PM in Figures 2f and 4c). On 11 December, the high concentration of aerosols in the afternoon is more evident. The higher N_{cn} in the afternoon (Figure 4a–d) could be related to a photochemical reaction and agrees with previous studies [28], indicating the roles of a local source of aerosols associated with the new particle formation and growth, as indicated in [12,19]. In contrast, the aerosols obtained from above the PBL (above ~1 km) do not show distinct differences according to location (land or ocean) or time of the day (morning or afternoon), which is probably due to the dominant influence of the aerosols from the continental outflows above the PBL.

Airborne PM is a well-known cause of respiratory diseases and lung cancer, among many other diseases or causes of death. PM_{10} , similarly to the N_{cn} , decreases with height (Figure 4e–h). Furthermore, PM_{10} concentrations within the PBL show similar patterns

as the N_{cn} , registering higher concentrations over the land (Figure 4f,g) than those over the ocean (Figure 4e,h). Figure 4e–h shows that afternoon PM10 in Seoul (Figure 4f, filled symbols) was distinctly higher than PM10 in all places and dates. For instance, PM10 in Seoul at ~700 m in the afternoon was $10\text{--}30\ \mu\text{g m}^{-3}$, compared to $3\text{--}10\ \mu\text{g m}^{-3}$ recorded in other places and times. However, the PBL PM10 over the Yellow Sea in the morning (RF06; 10 December; red in Figure 4e) was higher than the PBL PM10 over the East Sea in the afternoon (RF07; 10 December, blue in Figure 4f), indicating higher PM10 over the Yellow Sea than over the East Sea.

The N_{cn} and PM10 decrease rate with heights is more prominent over the land than over the ocean, indicating close links between the surface and the source of PM10 and N_{cn} . The PM10 at Seoul in the PBL (Figure 4f) was $3\text{--}30\ \mu\text{g m}^{-3}$ and $0.1\text{--}1\ \mu\text{g m}^{-3}$ at around 2300 m (above PBL), whereas PM10 over the East Sea in the PBL (Figure 4h) was $\sim 6\ \mu\text{g m}^{-3}$ and $0.7\text{--}0.8\ \mu\text{g m}^{-3}$ at around 2000 m. Besides, PM10 varied widely over the land, compared with that over the ocean (Figure 4f,g versus Figure 4e,h) and in the afternoon (filled versus open symbols).

The higher afternoon aerosol concentrations are more evident in the total number of aerosols (N_{cn} , Figure 4a–d) than PM10 (Figure 4e–h). The N_{cn} measures all particles larger than 10 nm ($D > 0.01\ \mu\text{m}$), whereas PM10 measures particles between 0.25 μm and 10 μm . Therefore, higher afternoon aerosol concentrations indicate that particle growth in the afternoon was in the regime of either $D > 10\ \mu\text{m}$, $0.01\ \mu\text{m} < D < 0.25\ \mu\text{m}$ or in both regimes. A previous study shows the formation and growth of new particles in the regime of $D \sim 0.01\text{--}0.1\ \mu\text{m}$ in the afternoon [19] through photochemical reactions. In summary, aerosol concentrations were higher over the land than those over the ocean (Seoul > Gangneung > East Sea > Yellow Sea), not showing the east–west gradients.

3.3. Particle Size Distribution

The aerosol particle size distributions (PSDs) obtained from the level-leg flights are shown in Figure 5. The PSDs of the same day are shown with the same colors; each corresponds to the PSD obtained within and above the PBL, respectively. The most outstanding feature in Figure 5 is that the PSDs obtained from above the PBL show remarkably similar shapes, regardless of the locations and dates of sampling, monotonically decreasing to $D \sim 1.5\ \mu\text{m}$. PSDs obtained within the PBL also show similar shapes as a whole, monotonically decreasing to $D \sim 1.5\ \mu\text{m}$, then showing a convex upward shape with a maximum value at $D \sim 2\text{--}3\ \mu\text{m}$. However, differences among the PSDs exist depending on the atmospheric conditions and aerosol origin or path. For example, PBL PSDs obtained from the Yellow Sea, Seoul, Gangneung and East Sea show similar patterns with the first peaks at $D < 0.3\ \mu\text{m}$ and the second peaks at $2\text{--}3\ \mu\text{m}$. However, PSD shapes are slightly different from the locations regarding the maximum concentrations and size observed. For instance, PSDs obtained from Seoul (Figure 5b) show slightly higher aerosol concentrations than those obtained from Gangneung (Figure 5c), although both aerosols were obtained over the land. However, the overall shape is similar. The effect of aerosol origin and path on the PSD shape and the maximum size is discussed in Section 4.

PSDs obtained from RF04 (6 December; blue) and RF06 (10 December; red) are compared in Figure 5a,b to show the effects of atmospheric conditions (meteorology, such as clouds and precipitation) on the changes of PSDs. On both days, aerosols were sampled along the Han River (Seoul) and over the Yellow Sea. In Figure 5a, considerable differences among the PSDs exist, especially in the larger particle size regimes ($D > 0.8\ \mu\text{m}$). In the PBL, PSDs on 6 December (blue) show plenty of larger-sized particles with slightly fewer smaller-sized particles than PSDs on 10 December (red). The weather system with precipitation passed by the Korean Peninsula between 6 and 9 December, resulting in no flights between those days. Figure 5a,b shows that larger particles were removed by precipitation (the scavenge effects); thus, fewer larger particles were observed on 10 December (RF06; red) than on 6 December (RF04; blue). The washout of large particles by precipitation was also shown in the PSDs observed at Seoul in Figure 5b (refer to changes from blue to

orange to red). In summary, PSDs obtained from the Korean Peninsula show similar shapes, regardless of the geographical locations of the sampled aerosols. The main differences in PSDs come from whether PSDs are obtained within or above the PBL. Aerosols above the PBL are likely to be affected by continental outflows. By contrast, the characteristics of aerosols within the PBL are influenced by local aerosol sources [19], boundary layer processes [16], atmospheric conditions (Figure 5a,b; e.g., scavenging effect [29]) and the origin of the air mass in addition to continental outflows.

Figure 5. Particle size distributions (PSD) obtained from the (a) Yellow Sea, (b) Seoul, (c) Gangneung and (d) East Sea. Individual lines represent PSDs calculated from level-leg flights within and above the PBL. For example, PSDs on 6 December within and above PBL were obtained from 661 m and 1903 m, respectively.

4. Discussion

In Section 3, the overall PSD shape remains robust, except for the cases associated with the precipitation system. This section further discusses aerosol concentration and PSD differences concerning the origin and path of the air mass (over the ocean or land). Figure 6 shows PSDs on 10 and 11 December (RF07 and RF09 in Table 1). On 10 and 11 December, an aircraft undertook similar flight paths. The aircraft took off at Gimpo airport, then flew out to the Gangneung area. Over the Gangneung area, the aircraft made two sets of level-leg flights along the coastline, one over the land (Gangneung) and another over the ocean (East Sea) at the heights of 600 m and 2000 m for each side of the coastline. The flight patterns on the days are shown as magenta lines in Figure 1. The PSDs in Figure 6 were obtained from the level-leg flights near 600 m and 2000 m in the afternoon.

The 24 h back trajectories show that the air mass observed within the PBL on 11 December (Figure 2f; green solid line) originated from North Korea and stayed over the ocean for more than 12 h before arriving at Gangneung and East Sea. The air mass observed in the PBL on 10 December (Figure 2f; blue dotted) originated further northwest and traveled through the inland before arriving and was sampled at Gangneung and East Sea. Figure 6a,b show that aerosol concentrations are higher over the land (Gangneung, filled) than over the ocean (East Sea, open), regardless of its origin; both air masses originated from northwest Korea (Figure 2f). The aerosol concentrations are related to the air mass path; concentrations are strongly related to whether the air masses stayed over the ocean or land for several hours before being sampled. In Figure 6, aerosol concentrations are lower where the air mass stayed over the ocean before being sampled. Further, larger-sized particles are observed when the air mass stayed over the ocean. Larger-sized particles over the ocean are more evident on 11 December, when the air mass stayed over the ocean in and above the PBL (at both 600 m and 2000 m) than on 10 December, when the air mass stayed over the land before being sampled.

Figure 6. The particle size distributions at 600 m (blue) and 2000 m (red) off Gangneung (land) and East Sea (Ocean) on (a) 11 December and (b) 10 December. The filled and open symbols represent the aerosols obtained from land-legs (Gangneung) and ocean-legs (over the East Sea).

Countries in continental outflow regions are dominantly affected by aerosols of up-wind areas. A previous study has shown the east–west contrast of AOD over the Korean Peninsula with the highest AOD over China, then the Yellow Sea, followed by AOD in the middle of the Korean Peninsula, then the eastern part of Korea [21]. Reference [19] also showed that aerosol concentrations in the PBL were higher over the Yellow Sea than over the East Sea based on airborne measurements. The authors interpreted the east–west contrast of aerosols as being due to the continental outflows. On the other hand, in this study, the total aerosol N_{cn} within the PBL was the highest in Seoul, followed by Gangneung, East Sea and Yellow Sea. According to Figure 6, the aerosol concentrations in the PBL recorded from the ocean-leg (blue, open symbols) were lower than those from the land-leg (blue, filled symbols), even though the origins of the air mass were similar. The lower aerosol concentrations over the East Sea in previous studies [19,21] could not be related to the continental outflows but occurred because the air mass stayed over the ocean a couple of hours before being sampled.

The 3-day back trajectories for the nine flights of [19] are shown in Figure 7 to examine the origin and path of the air mass. The air masses that arrived at 3000 m over the Yellow Sea and East Sea originated from the northern part of China and passed through Beijing to reach the Korean Peninsula (Figure 7a,b). Air masses that arrived at 500 m over the Yellow Sea and East Sea originated from a wider area of China (Figure 7c,d). For the Yellow Sea, air masses originated in northern and southern China and arrived at 500 m over the Yellow Sea via Beijing and Shanghai (Figure 7c). Beijing and Shanghai are among the largest cities in China and they correspond to cities with high air pollution. On the other hand, the air masses that arrived at 500 m over the East Sea originated from further north (Figure 7d). Interestingly, the air mass did not pass over Beijing or Shanghai and stayed a little longer over the sea for several hours before being sampled. Further, the surface winds over the East Sea during most of the experiment periods were easterly (not shown), indicating that

aerosols over the East Sea in their study originated from the open ocean or, at least, stayed over the ocean for several hours before being sampled over the East Sea, even if the aerosols originated from China several days before.

Figure 7. A 3-day back trajectory of air mass arriving at 2000 m (a,b) and 500 m (c,d) over the Yellow Sea (a,c) and East Sea (b,d) for the nine flights in [19]. The figures were created by the author. The 500 m and 2000 m were chosen to represent the heights within and above the PBL, respectively. The same color was used for the same flight.

The satellite-derived AOD is a column-integrated value. Therefore, it is expected that Korea reveals the AOD east–west gradient if the prevailing winds are westerly and the air mass originates in or passes through the polluted area during the observation. As discussed earlier, [27] showed that the air mass that caused high aerosol concentrations in Korea originated from the northern and southern parts of China (Beijing and Shanghai). Here, Beijing and Shanghai are to the west or southwest of Korea, capable of efficiently transporting aerosols to the Korean Peninsula through the westerly winds. However, the air mass originating from northeast China does not transport high aerosols to Seoul. Therefore, the east–west AOD gradients shown in a previous study could be closely related to the high aerosol transport from China to Korea via the westerly wind during the experiments, referred to as back trajectory (Figure 16 in [19]). Note that high AOD does not always guarantee high aerosol concentrations in the PBL, as aerosols originating from remote locations (China) could pass by Korea without transporting them to the ground (Figures 3 and 5 in [30]). Further, if the aerosols above PBL are transported into the PBL, they can be modified by interacting with clouds within the PBL [16]. Figure 2 shows that aerosols sampled in this study originated from northeastern China. Further, they did not pass over either Beijing or Shanghai, indicating that the high aerosol concentrations did not transport from China to the Korean Peninsula during the experiment, resulting in not showing the east–west aerosol contrast in this study.

Although the results on the N_{cn} are in agreement with previous results in the regions where comparable in situ measurements exist, over the Yellow and East Seas, the results of

this study require caution to conclude because the number of flights analyzed here is based on nine individual flights during December 2016. Long-term continuous measurements and comprehensive observations are required for further studies.

5. Summary and Conclusions

Although East Asia is a region where diverse and extensive aerosol sources exist, limited data on aerosol properties are available, especially as a function of height above the surface. This study examined the vertical structure of aerosols in and around the Korean Peninsula from east to west. Nine flights were made in the cloud-free boundary layer between 1 and 11 December 2016. During the experiments, the Korean Peninsula was affected by a northwesterly wind and all the airmasses originated from near Lake Baikal or northeast China. Under these atmospheric conditions, the total aerosol N_{cn} within the PBL was the highest in Seoul, as expected ($1000\text{--}10,000\text{ cm}^{-3}$), followed by Gangneung ($\sim 1000\text{--}5000\text{ cm}^{-3}$), East Sea and the Yellow Sea.

Further, aerosol concentrations obtained in the afternoons were higher than those recorded in the mornings. The higher PBL aerosol concentrations in the afternoon and over the land indicate that local aerosol sources influence the aerosols in the PBL. Aerosols obtained from relatively clean environments (Gangneung and over the oceans) are far more polluted than those obtained from typical maritime environments. PM10 in the PBL was $3\text{--}30\text{ }\mu\text{g m}^{-3}$ in Seoul, $\sim 10\text{ }\mu\text{g m}^{-3}$ in Gangneung and less than $10\text{ }\mu\text{g m}^{-3}$ over the ocean. PM10 near 2000 m was $0.1\text{--}1\text{ }\mu\text{g m}^{-3}$. The PSD in the PBL shows similar shapes, with PSD peaks at $D < 0.25\text{ }\mu\text{m}$ and $D \sim 2\text{--}3\text{ }\mu\text{m}$. However, subtle variations exist because of the cloud–aerosol interactions associated with precipitation and the origin and path of the air mass. Further, PSDs obtained over the ocean showed lower concentrations than aerosols over the land with a few larger-sized particles. However, the N_{cn} and PM10 from above the PBL did not show significant differences with geographical locations (Yellow Sea, Seoul, Gangneung, East Sea) and dates (morning versus afternoon), showing less than $300\text{--}400\text{ cm}^{-3}$. Similarly to aerosol concentrations, PSDs above the PBL did not show significant differences.

This study provides the first vertical structure of aerosols (concentration and size distribution of aerosols) over the inland of the Korean Peninsula using in situ aircraft measurement. The in situ data from this study will be used as a reference to validate the aerosol concentrations derived using lidar, satellite and models. A previous study has shown the east–west gradient of AOD across the Korean Peninsula with the aerosol concentration decreased from west to east, supported by airborne measurements over the East and Yellow Seas. However, the current study shows that the concentration of inland aerosols was the highest, followed by aerosols over the East Sea and Yellow Sea with the lowest aerosol concentrations. It should be noted that the satellite-derived AOD is a column-integrated value; thus, high AOD does not always guarantee high aerosol concentrations in the PBL. Furthermore, concentrations are strongly related to whether the air masses stayed over the ocean or land for several hours before being sampled; the longer the air mass stays over the ocean, the lower the aerosol concentration is. The results also show that aerosols sampled over the Korean Peninsula during the experiments were transported from China to Korea without passing through the polluted areas in China (e.g., Beijing and Shanghai).

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