Replies to "Interactive comment on "Polarization properties of aerosol particles over western Japan: classification, seasonal variation, and implications for air quality" by Anonymous Referee #3

The authors greatly appreciate anonymous reviewer for evaluating our manuscript and providing many insight comments on the manuscript. To response reviewer's major concerns, we did further analysis on (1) the instrumentation of POPC and data analysis procedures, (2) seasonal/diurnal variability of polarization property of aerosol, (3) classification of different aerosol types, and (4) their contributions for local air quality. We concisely made three conclusions that were useful for understanding the polarization property of aerosol in the downstream of dust and pollution source regions, interaction between anthropogenic pollutions and mineral dust in East Asia, and their impacts on air quality in western Japan. Besides, the revised manuscript includes two-year observation period (Jan. 2014 to Dec 2015). To avoid confusion, we used averaged DR value throughout the paper.

As requested by reviewer, aerosol depolarization ratios observed by POPC was compared with ground-based multi-wavelength Mie-Raman lidar (MMRL) observation at the same site. The temporal variation in the Lidar-derived aerosol depolarization ratio (at 532 nm) within the PBL was different from that of POPC measurement (Figure R1), because Lidar observation obtains the volume depolarization properties of all particles in a specific volume of the targeted air parcel, and the large amount of submicron particles dominated the overall polarization of aerosols, particularly under pollution condition. Besides, the aerosol depolarization ratio of Lidar is derived from the backscattering signal at 180º relative to its emitted light direction, and the POPC utilizes the backscattering signal at 120º due to the design limitations of the instrument. Theoretical simulation of DR values for randomly oriented spheroid particles on the basis of T-matrix methodology indicated less than 10% difference between these two configurations. Above all, quantitative comparison between Lidar and POPC is difficult. Therefore, we did not include such discussion in the revised manuscript.
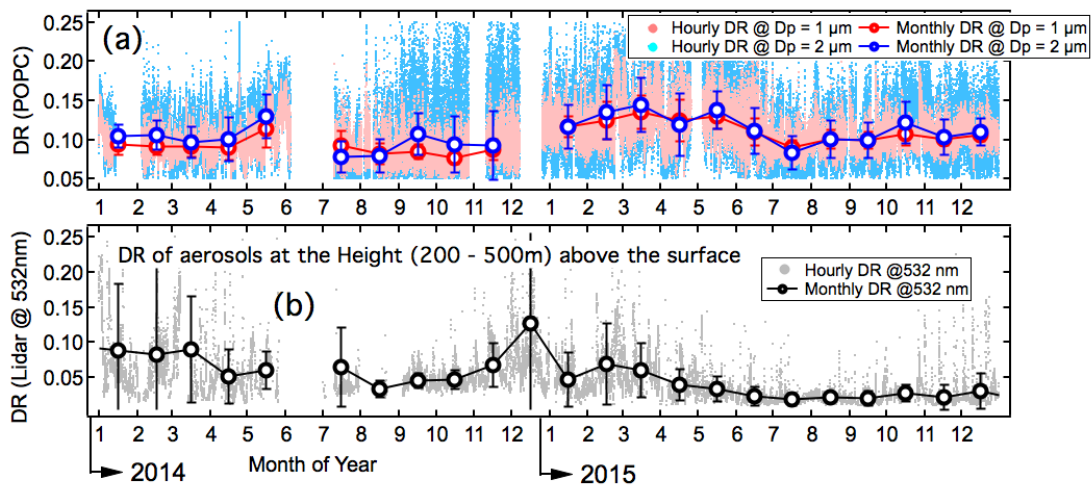
Figure R1 Time series of hourly (color dots) and monthly (colored circles) averaged depolarization ratio for the particles at $D_p = 1$ μm and $D_p = 2$ μm (a), and aerosol volume depolarization ratio determined for the ground-based Lidar observation at Fukuoka (b) in 2014 and 2015.

The specific response to the reviewer's comments are as followings:

Specific comments 1. Page 1, Lines 4 and 5: Correctly arrange affiliation according to increasing number.

Reply: The order of affiliation of coauthors is rearranged.

2. Page 1, Line 1 "By conducting an analysis of online measurements..."–> "By simultaneously conducting an analysis of online measurements..." may be better.

Reply: The expression is revised according to the reviewer's comments.

3. Page 1, Line 19: "..three typical aerosol types (anthropogenic pollutants, dust, and sea salt)."–> This is confusing to me. What type of aerosol is "anthropogenic pollutants"? You may use the word "dominant" somewhere to clarify what you want to say.

Reply: As suggested, the "anthropogenic pollutants" is changed to anthropogenic pollution-dominant.

4. Page 1, Line 20: What is the size for "super micron" particle? State within the parenthesis.

Reply: the definition of "supermicron" is given in the abstract.

5. Page 1, Lines 27-30: It is hard to understand the meaning of this sentence unless one reads the whole manuscript. The abstract section should be understood even without reading the whole manuscript.

Reply: The key points in abstract are reorganized. The sentences in line 27-30 is changed to "Backward trajectory analysis demonstrated that air mass originating from western Pacific contained large amount of spherical particles due to influence of sea salt, in particular in summer; however, for the particles from Asian continent, the dependence of number fraction of spherical particles on air relative humidity was insignificant, indicating of predominance of less-hygroscopic substances (e.g., mineral dust), although mass concentration of anthropogenic pollutants were elevated."

6. Page 2, Line 20: why 532nm within parenthesis?

Reply: Lidar only observe polarization signal at 532 nm, and backscatter signals at 532 nm and 1064 nm. To avoid confusion, the expression is changed to "...ground-based light detection and ranging (Lidar) has also been developed to derive the attenuated backscattering coefficient at 1064 nm and 532 nm and volume depolarization ratio at 532 nm, the latter of which was used to investigate the temporal and spatial characteristics of

dust and pollutant particles"

Reply: The supermicron is particle with Dp larger than 1 μm, it is explained in the abstract.

8. Pages 3-4: Section 2. Measurements:

(i) What is the size range (e.g. Dp > 1um, 2 um etc.) for measurements by POPC?

Reply: The detection range of POPC is 0.5 μm ~ 10 μm. The information is added in the context.

(ii) What is the time resolution of observation by POPC?

Reply: Sampling rate and half-width of full height (WHFH) of the detector's output signal of POPC were 2×10^6 samples/second and ~35 μs, respectively. During the measurements, the pulse signals were sampled in 1 second and processed for 1.2 seconds.

(iii) How do you calibrate the instrument? A brief description may be useful.

Reply: The calibration procedure is added in the revised manuscript.

"POPC were calibrated at the observatory every 6 months. The spherical polystyrene standard (Dynospheres, Dp = 0.5 μm, 1 μm, 3 μm, 5 μm, 10 μm, JSR Life Sciences Corporation) aerosols were generated by a nebulizer at a inject flow rate of 3.5 liter per minute (lpm) and desiccated by passing through a 45 cm Perma casing tube (MD-110-24P, GL Sciences)."

(iv)Have authors validated observation data of this instrument (POPC) by comparing with some standard instruments? If not, is there any literature that did such study in the past? Explain about it in the manuscript.

Reply: Mass concentration of $PM_{2.5}$ was reconstructed on the basis of number concentration of particles measured by POPC and particles density. The particle density was assumed to increase linearly from 1.77 g/cm^3 (0.5 μm) to 2.2 g/cm^3 (10 μm). The result compared well with $PM_{2.5}$ mass determined by ACSA (beta-ray method), with a Pearson coefficient of Pr = 0.7.3 (Figure R2). The POPC results were slightly lower than that of ACSA because the particle with Dp less than 0.5 μm was undetectable for POPC. Till now, numbers of studies using POPC have been published. We will add relevant literatures in the context. The number concentration particles with 1< Dp < 2 μm and 2< Dp < 5 μm was compared well with commercial optical particle counter (KC52, RION, as shown in Figure R3).
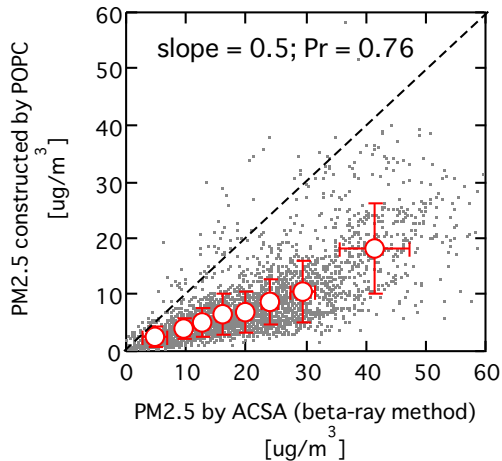
Figure R2 Scatter plot of mass concentration of $PM_{2.5}$ measured by ACSA-12 and constructed by POPC using number concentrations and presumed particle density.
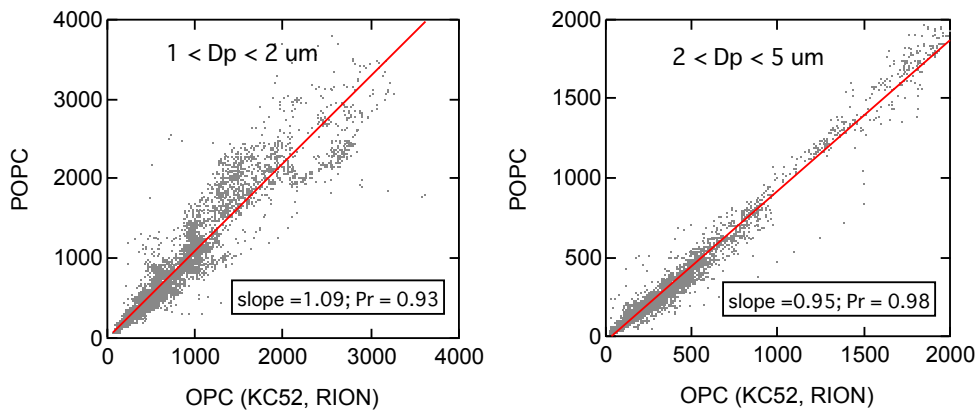
Figure R3. The comparison of number concentrations measured by POPC and OPC (KC52, RION)

(v) Were ACSA-12 and POPC operated simultaneously at the same place during observation? If not, how far were they?

Reply: POPC is working at Chikushi campus of Kyushu university and ACSA-12 is operating at the Fukuoka Institute of Health and Environmental Sciences, within 5 kilometer to the south. Therefore, almost the same air parcels were measured during the observation period.

(vi) ACSA-12 observed at 1 hour interval. How to you integrate POPC data while using ACSA-12 and POPC data together?

Reply: POPC measured particles at the time resolution of 1 second. For combined analysis with ACAS, we took 1 hour average for POPC.

(vii) dV/dlogDp is widely used in this study. How do you determine it? To know it, it is also necessary to describe Dp as indicated in (i).

Reply: dV/dlogDp is the normalized volume concentrations. Aerosol distributions are predominantly lognormal in character. Direct plots of concentration vs. particle diameter can be confusing if the aerosol sizing instruments have different size resolutions. dV/dlogDp is used to avoid this problem. Here, dV is the total volume of particle in the range and dlogDp is the difference in the log of the channel width ($logD_{p,u} - logD_{p,l}$). $D_{p,u}$ and $D_{p,l}$ are the upper and lower channel diameter, respectively. The ratio is useful for intercomparison between different instruments, because a normalized concentration value is independent of the bin width. We will explain the meaning of dV/dlogDp at its first appearance.

(viii) Regarding Figure 2, make the caption clear. For example, MODIS data of which year and month have been used to generate Figure 2? And also what is the source of wind speed data? They should be explained in the caption as well as text.

Reply: The Figure 1 is changed to geographic location of observation site and annual anthropogenic emission $PM_{2.5}$ according to REAS2.0 emission inventory

9. Pages 4-5, Section 3.1. Temporal variation:

(i). Page 4, Line 33: Daily average->Do you mean average of 24 hours? Indicate hours within the parenthesis.

Reply: The starting and ending time of average is shown in the revised manuscript.

(ii). Page 4, Line 33: different aerosol size bins: Figure 1 shows Dp from 1 to 10. Are they mean diameters of size bins?

Reply: The label of y-axis means the size of particle. The mistake is corrected.

(iii). What are the criteria to determine "P", "D", and "M" in Figure 3?

Reply: To avoid confusion, we remove "P", "D", and "M" in Figure 3.

(iv) No unit for dV/dlogDp and MDR in Figure 3.

Reply: In the revised manuscript, we use dV in unit of $\mu m^3/m^3$, and the depolarization ratio (DR) is unitless.

(v) page 5, Line 1: What are the size ranges for submicron and coarse mode ?

Reply: The size range of submicron and coarse mode are explained in the revised manuscript.

10. Pages 5-6, Section 3.2: Size distribution:

(i) The interesting and strange thing for me is that why there is not distinct difference in

dV/dlogDp at different diameters. For summer, it is nearly flat. It is quite interesting to me. Is it natural phenomenon or instrument related issue? Unfortunately, authors did not explain the reason for this type of observation. Authors should give proper reasons to readers for this type of observation. Otherwise, one may suspect in the quality of observation data.

Reply: Great thanks for this good question. I thoroughly checked the POPC data and data analysis procedures, and I found a programmatic mistake that cause the unusual dV/dlogDp result. In the revised manuscript, we corrected all the results. As shown in Figure 4, dV/dlogDp shows evident peak in different season. The discussion in "Size distribution" section is revised as follows:

"The normalized volume size distribution (dV/$d$logDp) of aerosol particles at the site showed prominent seasonal variations, and DR values of particles increased obviously with size, irrespective to season (Figure 4). The monthly averaged DR values at the peak size are shown as read circles in the plot. In spring (MAM: March, April and May), dV/$d$logDp of particles had a broad peak between 4 μm and 6 μm due to the impact of frequent outbreaks and transport of Asian dust (Figure 4a). The typical DR value at the peak size was 21-23%. In summer (JJA: June, July and August) the peak size in dV/$d$logDp was 2 – 3 μm, and the corresponding DR value of particles in fine mode were less than 10%. Such decrease was mostly due to air masses that originated from clean marine regions and consisted of large amounts of sea salt aerosols, which tend to be spherical under high-humidity conditions. In autumn, volume concentrations of particles at site were the lowest because of less impact from Asian continent and favorable meteorological for proliferation of air pollutions. In winter (DJF: December, January and February), dV/$d$logDp have two peaks in both submicron range (D_p = 0.5 μm) and coarse mode (D_p = 4 μm), indicating the combined impact of both anthropogenic pollutions and dust aerosols at the prevailing westerly wind. The DR values at D_p = 0.5 μm and D_p = 4 μm were 9% and 23%, respectively."
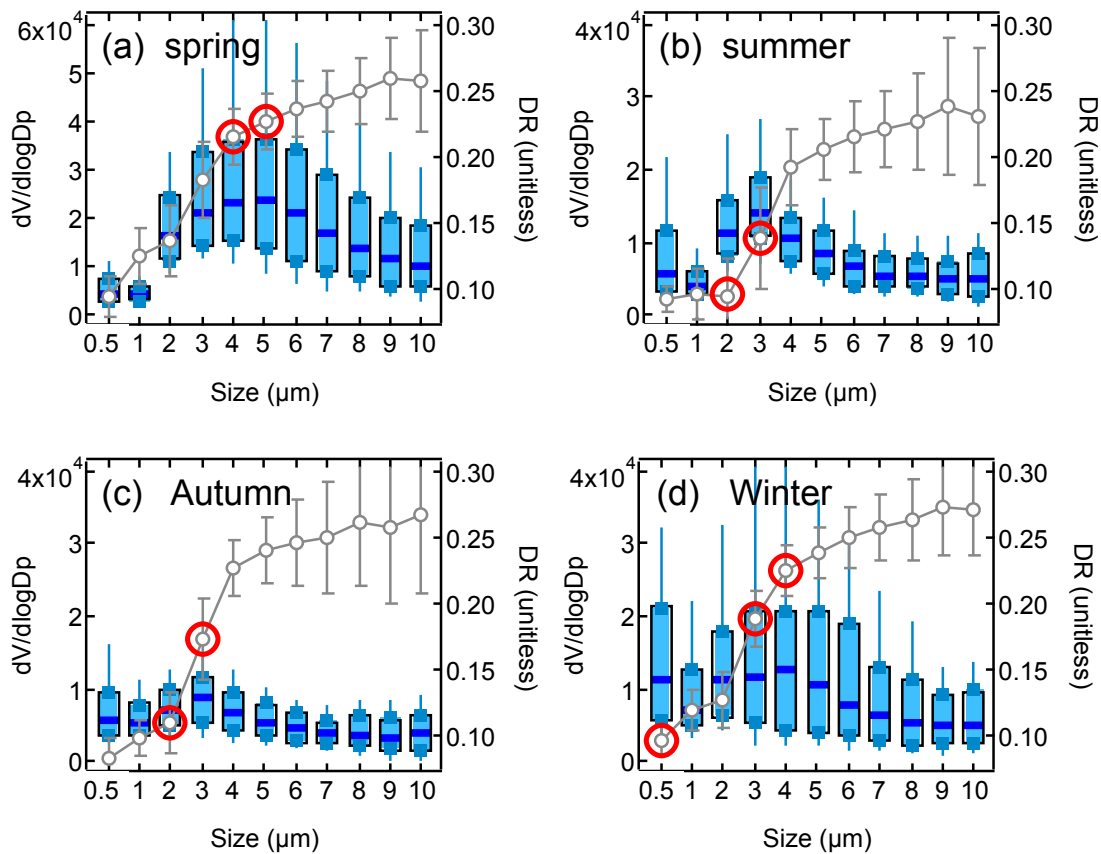
Figure 4 in the manuscript.

(ii). Regardless of the season, we can also see large depolarization ratio (DR) in right hand side of Figure 4 and its distribution as function of aerosol size is nearly same. Though the median value is relatively low in summer, but the mean value is still high According to authors, sea salts are dominant in this season. What types of aerosols are responsible for such large DR in summer? Unfortunately, authors skipped this discussion in the manuscript. Similarly, regardless of all seasons, there is an increase of DR for aerosols of Dp=0.5 micron. Why do you observe this behavior in all seasons? Are they natural phenomenon or instrument related issues? They need to be clarified in the manuscript.

Reply: The discussion in this section was revised as follows:

"It was notable that, observed DR values of particles always increased obviously with size in all season, almost irrelevant to aerosol types. Such characteristic was well predicted by optical model considering particles of Voronoi aggregation (Figure R4). Theoretical simulation also indicated that an increase in the imaginary part of the refractive index could reduce the DR value evidently; however, from the viewpoint of observation, DR value in the coarse mode showed a trend of ascending followed by leveling off, implying that the internal mixing with light-absorbing matter was less important at the site.

Calibration of POPC using polystyrene standard aerosols demonstrated that the DR values were 8%, 9% and 10% for the pure spherical particles at D_p = 5 µm, 7 µm and 10 µm, respectively; and the DR value was almost zero for the fine mode particles. Nevertheless, in this study, the DR values of aerosols in fine and coarse mode were found to be 8% - 10% and 20% - 28%, respectively. It indicated that aerosol particles at the site was still non-spherical generally, even though hygroscopic particles (such as sea salt) may deliquesce and grow at high humid condition."
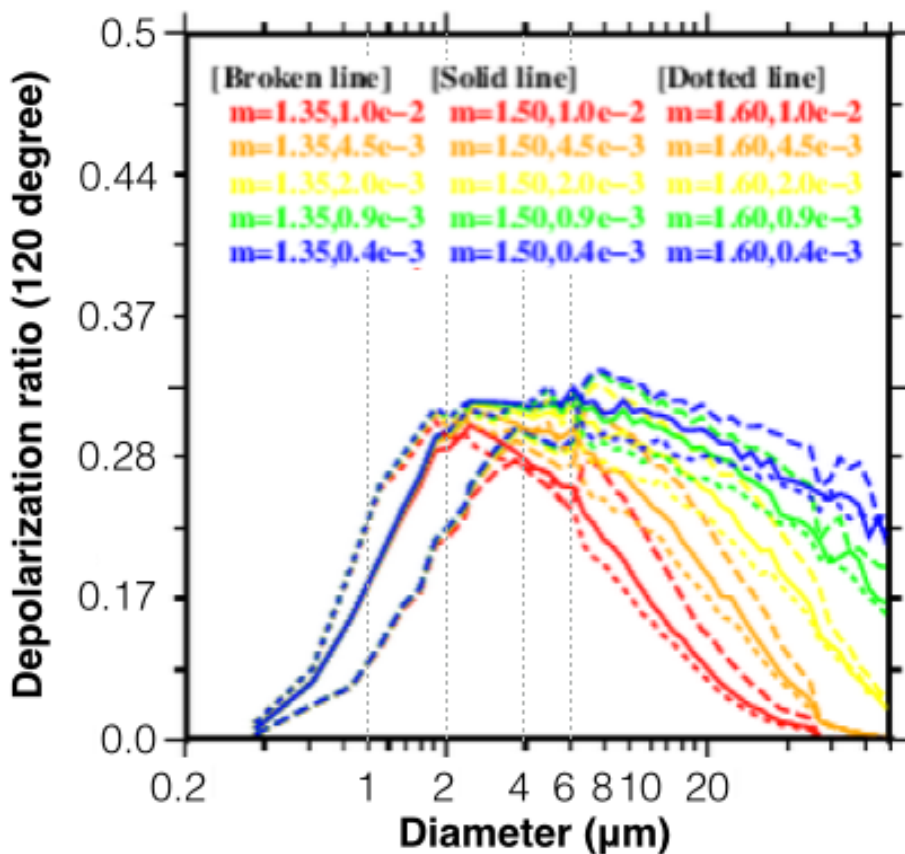
Figure R4 Theoretical calculation of depolarization ratio (at 120 backward direction) as a function of particle size for different refractive index.

11. Pages 6-7, Section 4.1. Size polarization properties of aerosol particles:

(i) It is not reasonable to assume that only a single type of aerosol (Dust or sea salt) can exist in the atmosphere; it is better to use word "dominant" somewhere while classifying group in this section.

Reply: To avoid misleading, we will use "dominant-" for classification of aerosol types.

(ii) Total number of hours is given in Table 1, but date and time are not given. Specify date and time for each event shown in Table 1.

Reply: We will add detailed information in the Table 1

(iii) I do not understand how you make Figure 5. Are they the average values of whole event period? If so, write clearly in the text as well as Figure. I further do not understand why median value is used for only DR.

Reply: Figure 5 depicts the statistics of DR as a function of particles size, and the color represents the normalized volume concentration (maximum = 1) during specific periods. In the context, such plot was referred to as DR-D_p-Volume plot. As concerned, the volume concentration is averaged values of whole event period and Y-axis actually is the DR.

(iv) DR value of 0.1 is said to be a threshold value to distinguish spherical and non-spherical particles. It is also one of the important conclusions of the paper; however, mode DR of specific period, rather than DR of instantaneous observation time, is used in Figure 5. In fact, how do you determine this mode DR and why is it important to discuss your result, taking into account mean DR, median DR can be also calculated? Probably, this value may change depending on the number of samples; I doubt this threshold value is applicable for all situations. More data analysis of different scenarios is required before recommending such threshold value.

Reply: Sorry for misleading. Figure 5 describes the total volume concentration of all the particles at specific particle size (X-axis) and DR value (Y-axis, not mode DR) during the specific periods. Actually the DR-Dp-Volume patterns were consistent at different period (Figure R5). For confirmation, the number size distribution at different DR level was also plotted in the Figure R5. It clearly demonstrated that the threshold of 0.1 was applicable for all the scenarios. We will give more explanations in the revised manuscript.
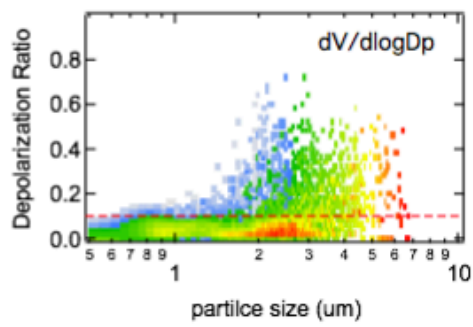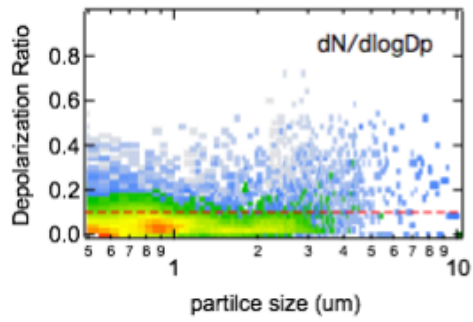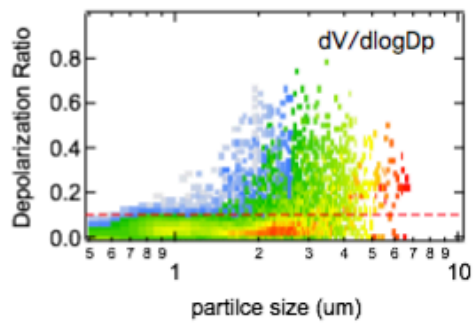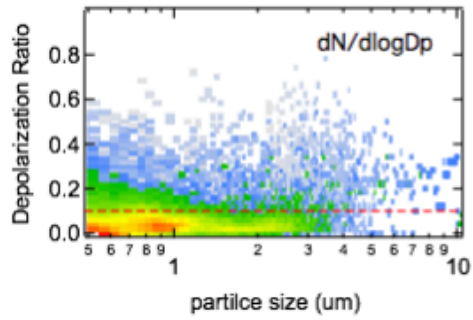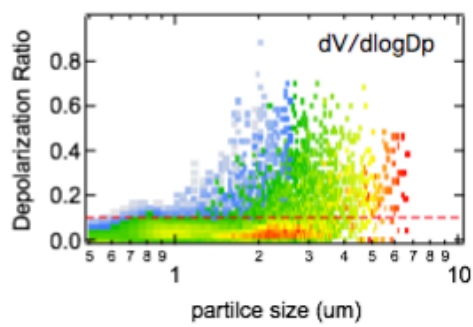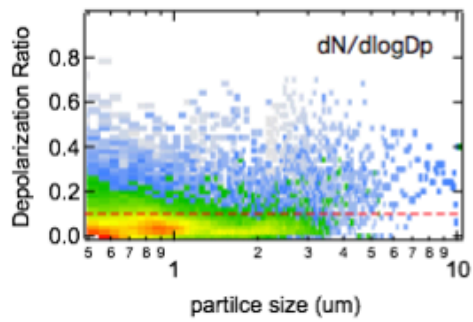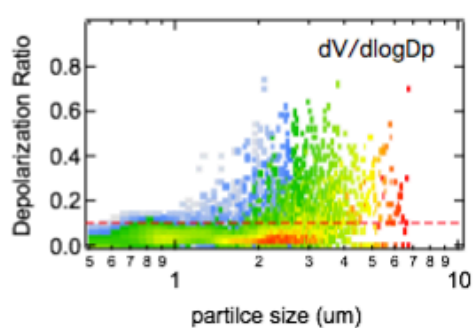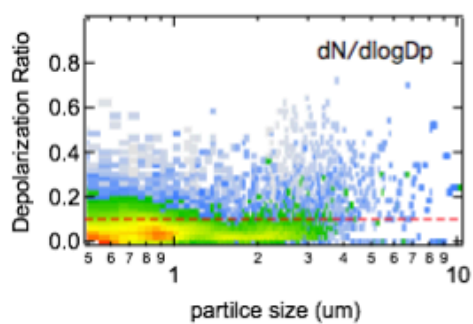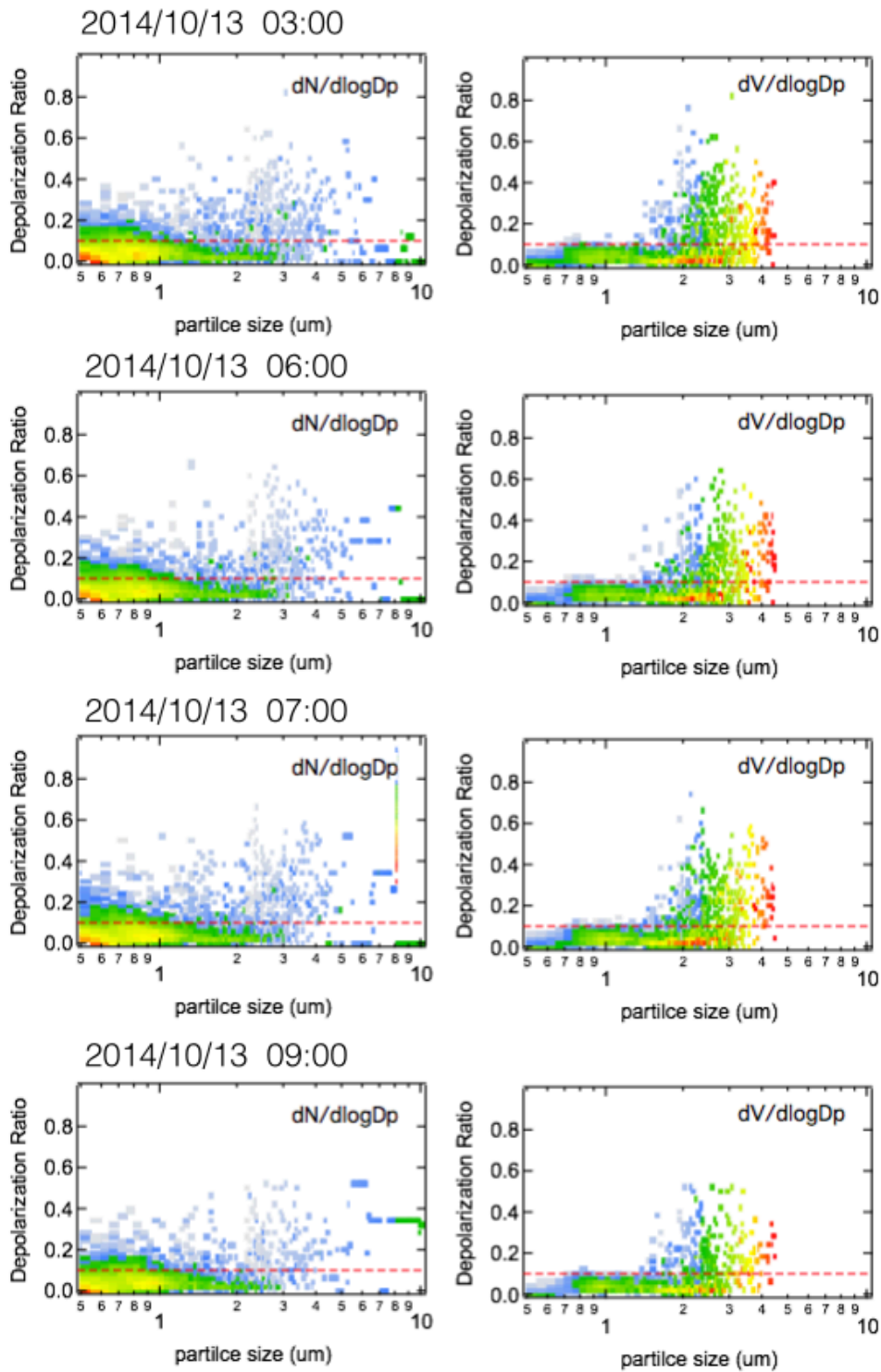
Figure R5 dN/dlogDp (left panel) and dV/dlogDp (right panel) of aerosols as a function of DR value and particle size during typhoon period.

(v) RH value is discussed in this section. What is the source of RH data?

Reply: The RH is from the local meteorological observation at Fukuoka. The source information will be added in the revised manuscript.

12. Pages 7-8, section 4.2. Contributions of different aerosol types to local air quality

(i) What criteria do you use to classify on different groups, namely "Dust", "Sea salt" , and so on? Specify those criteria specifically and quantitatively (e.g., DR range).

Reply: As suggested, classification of sea salt or dust dominant episode was on the basis of both volume mode and DR values; we revised the manuscript as follows:

"Using the criteria suggested in section 4.2, the impacts of different aerosol types on local air quality in western Japan were investigated. The period when DR-D_p-volume plot has only one prominent volume mode in A1 with mode DR value > 0.1, was regarded as being anthropogenic pollution-dominant. Dust dominant period was characterized by a distinct volume mode in A2. Periods when three distinct modes occurred simultaneously in A1, A2 and A3 were considered to represent a "mixed type"(Figure 5d). Determination of sea salt dominant period was based on both two distinct volume mode in fine mode, and small DR values (< 0.1). The circumstance that DR-D_p-volume plot has only two volume modes in both A2 and A3 was also observed during a long-lasting dust event at the end of May 2015 (Pan *et al.,* 2015), we regarded such case as "dust dominant"."

(ii) As I wrote in 11 (iv), DR of Figure 5, which is the foundation for classifications in this section, is a mode value of specific period, which is subject to change depending on the situation. Do authors perform a type of cross-check for classification results discussed in this section?

Reply: As replied in 11(iv).

(iii) Classifications are made based on plot of each day starting from 00:00-24:00 local time. However, it is hard to say that air mass of same origin exists for the whole day. It also raises question on classification discussed in this section.

Reply: As reviewer suggested, the origin of air mass at observation site may changed in a day, resulting in variations of the mass concentration of PM2.5. Therefore discussion on the contribution of different aerosol types to local air quality was much meaningful if hourly DR-D_p–volume pattern was considered. However, we are mainly interested in the days those failed to meet the Ambient Air Quality Standard in Japan (daily averaged PM2.5 > 3.5 ug/m^3). The compromising solution is to use DR-D_p–volume plot in a daily averaged basis. In this study such data treatment can be accepted because we just focused on the seasonal variability of the main contributors that caused the degradation of local quality.

(iv) Only "mixed" is shown in Figure 6(c). Does it represent pollution and dust mixed or what ?

Reply: We will use "mixed type" in the legend of Figure 6c. It represent the pollution

type with three distinct mode at A1, A2 and A3 (as explained in the section 4.1)

13. Pages 8-9, Section 4.3. Polarization properties of aerosols from different origin

(i) Similar to Figure 5, I do not understand how you plot Figure 7 (right). Is dV/dlogDp the mean value or median value or what ? Unit is also missed for dV/dlogDp in Figure 7(right). Make it clear both in text as well as figure caption.

Reply: To avoid confusion, the Figure 7 (right) represents the normalized (maximum = 1, unitless) total volume concentration of particles at specific size and DR values for the air mass came from different regions. We will make it clear in the caption and text.

(ii) Page 8, line 22: "..spherical particles (e.g., sea salt) in fine mode..."–> Sea salt may be in fine mode, but the contribution of anthropogenic pollutants may be significant that sea salt. Give logical discussion with proper evidence.

Reply: In the revised manuscript, we removed the dubious conjecture. We just say, "The small polarization degree of aerosol particles in the fine mode indicated that the aerosol particles tend to be spherical."

14. Pages 9-10, section 4.4. Fraction of spherical particles as a function of RH

(i) page 9, line 18: It is said that RH was calculated from back ward trajectory. How do you calculate RH from backward trajectory?

Reply: The Hysplit mode can provide meteorological data along the trajectory, and we extracted the RH value for each trajectory point, and we made the average.

(ii) Describe about background points of Figure 8 in both text and figure.

Reply: The background points are the origin data that were used for the statistics (data points with error bar), we will describe it in the caption of Figure 8

(iii) Chemical composition data shown in Figure 9 are for fine mode aerosols (according to caption of Figure 9); however Figure 8 discusses results for relatively large particles , including diameter larger than 3 micron. Is it reasonable to use such different data sets to discuss your results?

Reply: We agreed with reviewers that it is difficult to explain the less-hygroscopicity of coarse mode particles by chemical composition in fine mode. In the revised manuscript, we will use the particles' Acidity ($c\Delta H^+$, defined as the change of molar concentration of hydrogen ion after sampled particles dissolved into the extracting solution) to indicate the presence of crustal matter in the coarse mode. The discussion is modified as follows:

"The chemical analysis indicated that the mass fraction of water-soluble matter in coarse mode particles ($WSM = cSO_4^{2-} + cNO_3^- + cWSOC$) from the Region II was responsible for only 15% of the mass concentration of $PM_{2.5-10}$, almost the same with air mass from other origins. The missing part in mass balance was likely to be related to crustal species.

(iv) Probably, the lines of Figure 8 represent the average value. It is important to note that aerosol concentration as well as their chemical composition cannot be considered to be unique for air masses coming from any region. For example, Region II can have aerosols of different origins depending on the season as well as meteorology. I do not think that Figure 8 represent aerosol characteristics of each region. For this type of study, authors should divide data depending on season by carefully taking into account backward trajectory and chemical composition. For example, how confidently can you say that dust aerosols are always present when air mass comes from Region II? Authors are suggested to gather more evidence and then discuss the results more logically based on gathered information.

Reply: We agree with reviewer's comments that Figure 8 indicates the average condition for the aerosol particles coming from different region, and dust aerosols may not always present in the air mass that came from Region II. We will clarify these points in the revised manuscript. As shown in Table 2, air mass coming from different regions had clear seasonality (Table 2) and acidity of particles in coarse mode ($c\Delta H^+$) were obviously different. The air mass from Region I was comparatively concentrated in summer and autumn ($c\Delta H^+ = 3.6 \pm 1.4$ nmol/m^3). The air mass from Region II was intensive in winter ($c\Delta H^+ = -4.7 \pm 1.0$ nmol/m^3) and spring ($c\Delta H^+ = -2.8 \pm 1.0$ nmol/m^3), and the negative $c\Delta H^+$ indicated that the air mass consisted of alkaline substances (Ca^{2+}, Mg^{2+} etc.) It was only in summer that the air mass came form Region III ($c\Delta H^+ = 2.0 \pm 3.0$ nmol/m^3) and Region IV ($c\Delta H^+ = 7.5 \pm 4.9$ nmol/m^3). Overall, DR-D$_p$-volume plots in Figure 8 represented well polarization characteristics of particles from different regions.

Table 2 Number count of trajectories used in Figure 7 and corresponding monthly averaged acidity ($c\Delta H^+$) of particles in coarse mode for different regions. The hyphen mark represents that number count of trajectories are less than 10 in this month.

Month in 2014	Region I		Region II		Region III		Region IV	
	Num.	$c\Delta H^+$ (nmol/m^3)	Num.	$c\Delta H^+$ (nmol/m^3)	Num.	$c\Delta H^+$ (nmol/m^3)	Num.	$c\Delta H^+$ (nmol/m^3)
Jan.	-	-	203	-5.2 ± 4.5	-	-	-	-
Feb.	-	-	108	-5. 3 ± 2.4	-	-	-	-
Mar.	-	-	114	-1.8 ± 2.6	-	-	-	-
Apr.	-	-	175	-3.0 ± 3.4	18	-1.4 ± 2.4	-	-
May	-	-	276	-3.5 ± 6.7	-	-	-	-
Jun.	102	3.7 ± 2.5	69	3.4 ± 2.5	-	-	14	13.2 ± 16.7
Jul.	34	2.5 ± 1.3	33	4.7 ± 3.1	-	-	85	4.2 ± 3.8
Aug.	132	4.8 ± 2.9	-	-	136	1.5 ± 3.1	71	5.1 ± 2.7
Sep.	22	2.4 ± 3.6	-	-	104	5.9 ± 2.7	-	-
Oct.	26	4.7 ± 3.7	107	1.1 ± 4.3	-	-	-	-
Nov.	-	-	629	-1.7 ± 4.4	-	-	-	-
Dec.	12	"-5.9 ± 1.3"	527	-3.7 ± 1.8	-	-	-	-