

## Authors' response to Referee #2' comments

Paper No.: ACP-2014-1030

Title: Vertical variation of optical properties of mixed Asian dust/pollution plumes according to pathway of airmass transport over East Asia

We would like to give many thanks to you for the invaluable comments. We found your comments provided significant value to us in preparing the revised manuscript. The criticism and suggestions by you were appropriate and improved the quality of our manuscript. We therefore responded and will revise our original manuscript to address all of the concerns raised.

A point by point response is given below.

Thank you very much for reconsidering this manuscript

### **Anonymous Referee #2**

#### **General comments:**

This manuscript presents a long-term study of multi-wavelength lidar observations of Asian dust and mixtures of Asian dust with anthropogenic pollution over Gwangju, Korea. The results are interesting and valuable. Therefore I recommend publication in ACP after revision of the manuscript.

#### **Major comments:**

**Section 3, Figure 3: The general discussion of the retrieved values should consider the transport way/height. One would expect that the two different cases are also visible in the frequency distribution of the retrieved optical properties. Please comment on this.**

**Response:** We understand referee's concern. The retrieved values of optical properties of Asian dust should explain in consideration of transport pathway and height. In this section we are supposed to give a general description regarding the values of optical properties of Asian dust we observed. The detailed description of the distribution of these values in consideration of the level of pollution emission along the pathway of Asian dust layers or vertical position is presented in section 3.1 and 3.2 after this general description on the distribution of values

However, to clarify the difference in values of optical properties in the frequency distribution, the statement

“The average value of  $\delta_p$  for all observed Asian dust layers is  $0.17 \pm 0.02$ . The average values of  $S$  are  $57 \pm 6$  sr at 355 nm and  $57 \pm 7$  sr at 532 nm. The mean value of  $\mathring{A}_\beta$  is  $0.84 \pm 0.37$ . The optical properties of each individual Asian dust layer vary over a wide range of values. We find values of 0.08-0.33 for  $\delta_p$ , 38-83 sr for  $S$  at 355 nm, 41-73 sr for  $S$  at 532 nm, and 0.38-1.71 for  $\mathring{A}_\beta$ . The maximum value of  $\delta_p$  is 0.33 at 532 nm. The minimum values of  $S$  at 355 nm and 532 nm are 38 sr and 41 sr, respectively. The minimum value of  $\mathring{A}_\beta$  is 0.38. This maximum value of  $\delta_p$  and the minimum values of  $S$  at 355 nm and 532 nm and  $\mathring{A}_\beta$  are similar to the values of optical properties for pure dust particles.

76% of  $\delta_p$  at 532 nm are located in the range between 0.08 and 0.20. 53% of the values of  $S$  at 355 nm are in the range between 60 sr and 85 sr. 47% of the values of  $S$  at 532 nm vary between 60 sr and 75 sr. The Ångström exponents ( $\mathring{A}_\beta$ ) vary between 0.80 and 1.71 and 52% of all cases are in the interval. These values are different from the values of optical properties of pure dust.”

will be added in revised manuscript in order to explain the difference between values in the frequency distribution.

**p. 3389, l. 21 - p. 3390, l. 19: The consideration of all measurements to derive and discuss mean values and to compare these mean values to former findings is misleading as the authors assume differences in the investigated aerosol layers with respect to mixing of dust and pollution. Furthermore the authors assume mixtures of dust and anthropogenic pollution, but the discussion and comparison to former findings is**

**limited to the values of mineral dust. The discussion should be extended to former findings/measurements of pollution aerosols.**

**Response:** We agree with reviewer's comment. The values of optical properties of pollution is needed to explain with respect to changes in optical properties for a mixture of dust with pollution particles.

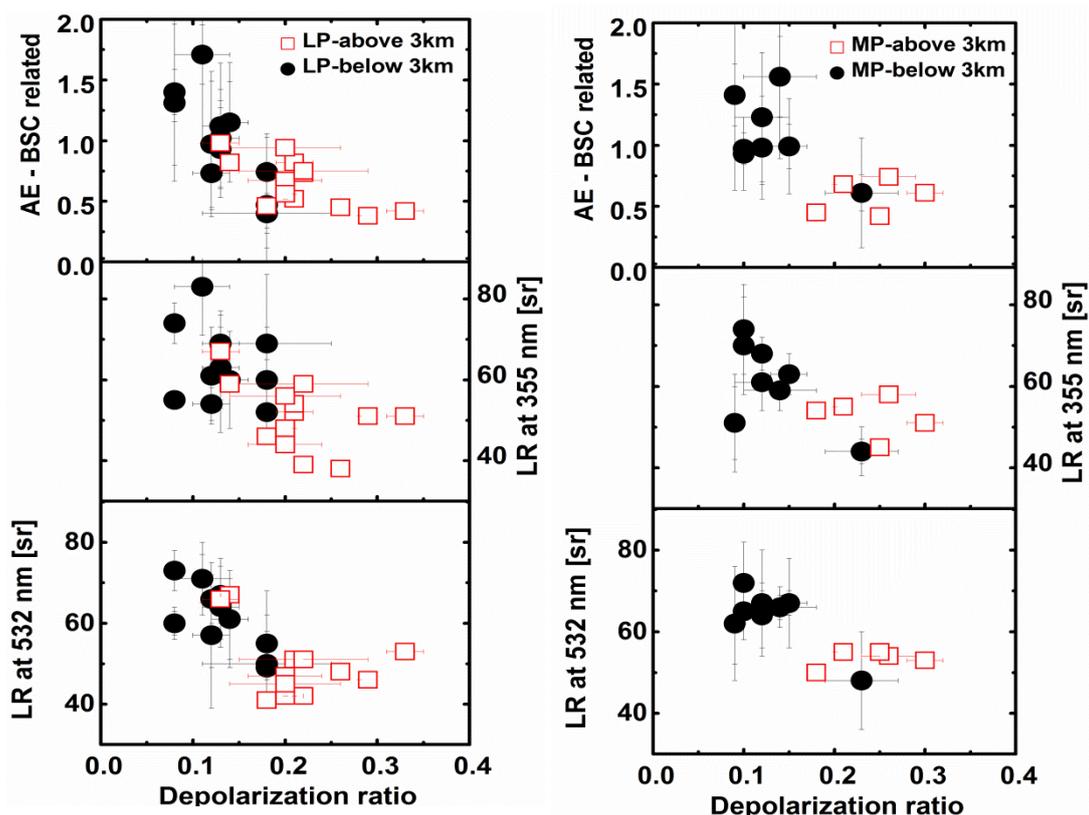
The statement “Ferrare et al. (2002) report a high value of  $68 \pm 12$  sr of the lidar ratio at 355 nm. This high lidar ratio was associated with air masses advected from urban/industrial areas. Omar et al. (2009) finds values of 65-70 sr for the lidar ratio at 532 nm. The numbers describe continental-polluted aerosols and polluted dust.” and “The values of 0.2-0.3 are reported as the values of  $\dot{A}_\beta$  for Saharan dust (Murayama et al., 2002; Tesche et al., 2009). Chen et al. (2007) and Müller et al. (2010) find values of 0.7-1.5 for  $\dot{A}_\beta$  for a mixture of mineral dust with urban haze. Values of 0.8-1.4 for  $\dot{A}_\beta$  were found for heavily polluted continental aerosol layers (Franke et al., 2003).” and “Small values, e.g., values from 0.08 to 0.1 usually are an indicator that dust is mixed with spherical particles (Murayama et al., 2004; Chen et al., 2009; Tesche et al., 2009; Burton et al., 2013). Anthropogenic aerosols normally are spherical with a small depolarization ratio (Pan et al., 2015). The degree of depolarization decreases as the sphericity of particles increases. The depolarization ratio is dependent on the mixing ratio of dust with spherical particles (Somekawa et al., 2008). For instance, Burton et al. (2013) report values of  $\delta_p = 0.13-0.20$  and  $0.03-0.07$  at 532 nm for polluted dust and urban aerosol particles, respectively.” will be added in the revised manuscript.

The references “Omar, A. H., Winker, D. M., Vaughan, M. A., Hu, Y., Trepte, C. R., Ferrare, R. A., Lee, K.-P., Hostetler, C. A., Kittaka, C., and Rogers, R. R.: The CALIPSO automated aerosol classification and lidar ratio selection algorithm, *Journal of Atmospheric and Oceanic Technology*, 26, 1994-2014, 2009.”, “Ferrare, R. A., Turner, D. D., Brasseur, L. H., Feltz, W. F., Dubovik, O., and Tooman, T. P.: Raman lidar measurements of the aerosol extinction-to-backscatter ratio over the Southern Great Plains, *Journal of Geophysical Research: Atmospheres* (1984–2012), 106, 20333-20347, doi:10.1029/2000JD000144, 2001.”, and “Franke, K., Ansmann, A., Müller, D., Althausen, D., Venkataraman, C., Reddy, M. S., Wagner, F., and Scheele, R.: Optical properties of the Indo-Asian haze layer over the tropical

Indian Ocean, Journal of Geophysical Research: Atmospheres (1984–2012), doi: 10.1029/2002JD002473, 108, 2003.” will be added in the revised manuscript.

**Section 3.1: This discussion is misleading as not only ‘the level of pollution emission’, but also the transport height above the polluted area has to be included in the separation. In consequence one would expect different results for the so called LP and MP cases, but the results are very similar for both cases. However, it is obvious, that two different clusters can be seen in the results supporting, that different aerosols/mixtures have been observed. This is also shown in Section 3.2 taking the transport way into account.**

**Response:** We understand the review’s concern. The figure and table as shown below will be added in the revised manuscript to address this issue.



Height of dust layer at pollution regions	Number of observed layers	Depolarization ratio	Lidar ratios (sr)		Ångström exponent
			355nm	532nm	
LP_below 3km	12	0.13±0.03	64±9	62±8	1.00±0.38
LP_above 3km	13	0.21±0.05	51±8	49±9	0.65±0.20
MP_below 3km	8	0.13±0.04	61±10	64±7	1.09±0.30
MP_above 3km	5	0.24±0.05	53±5	53±2	0.58±0.14

The Asian dust layers will be classified in consideration both the vertical position at polluted region in China and the level of pollution emission during transport. The variation of optical properties of Asian dust rather depends on the vertical position of Asian dust at the polluted regions than the level of pollution emission in our study. However, it could be caused by the insufficient number of measurement cases. In order to state this information, the paragraph

“The Asian dust plumes were classified into 4 categories. We considered not only the level of pollution emissions along the transport pathway, i.e., “MP” Asian dust and “LP” Asian dust, but also the vertical position of the layers when they passed over polluted regions of China (“below 3km” and “above 3km”). Figure 8 shows scatter diagrams of  $\hat{A}_\beta$  (wavelength range 355/532 nm), and  $S$  at 355 nm and 532 nm versus  $\delta_p$  at 532 nm in dependence of the level of pollution emission and the vertical position. The frequency distribution of  $\delta_p$  at 532 nm,  $\hat{A}_\beta$  (wavelength range 355/532 nm), and  $S$  at 355 nm and 532 nm for the corresponding clusters are shown in Figure 9. The corresponding mean values of the optical parameters of those clusters are summarized in Table 3. We expect that the optical properties of Asian dust change most if pollution levels (in terms of AOD) are high, (MP Asian case) and when the corresponding air masses passed over industrialized area of China at low altitude (below 3km height above ground). The mean values of  $\delta_p$  at 532 nm and  $\hat{A}_\beta$  are 0.13±0.04 and 1.09±0.30, respectively, for this case which is denoted as “MP\_below 3km”. The mean values of  $S$  are 61±10 sr at 355 nm and 64±7 at 532 nm. However, these values of optical properties of dust for MP\_below 3km are not significantly different from the case of “LP\_below 3km”. In that case the mean values of  $\delta_p$  at 532 nm and  $\hat{A}_\beta$  are 0.13±0.03 and 1.00±0.38, respectively. The mean values of  $S$  are 64±9 sr at 355 nm and 62±8 at 532 nm.

The values of optical properties between “MP” and “LP” at high altitude also do not differ significantly. The mean values of  $\delta_p$  at 532 nm,  $\hat{A}_\beta$ , and  $S$  are 0.24±0.05, 0.58±0.14, and

$53\pm 5$  at 355 nm and  $53\pm 2$  at 532 nm, respectively, for the case "MP\_above 3km". The highest values of  $\delta_p$  and lowest values of  $\mathring{A}_\beta$  are found for this case.

In the case of "LP\_above 3km" the mean values of  $\delta_p$  at 532 nm and  $\mathring{A}_\beta$  are  $0.21\pm 0.05$  and  $0.65\pm 0.20$ , respectively. The mean values of  $S$  are  $51\pm 8$  sr at 355 nm and  $49\pm 9$  sr, respectively. We believe that the changes in the optical properties of Asian dust depend on the vertical position of the dust plume rather than the level of pollution emission during transport."

will be added at the section 3.2 in the revised manuscript.

The caption for the table and the figure will be added in the revised manuscript as

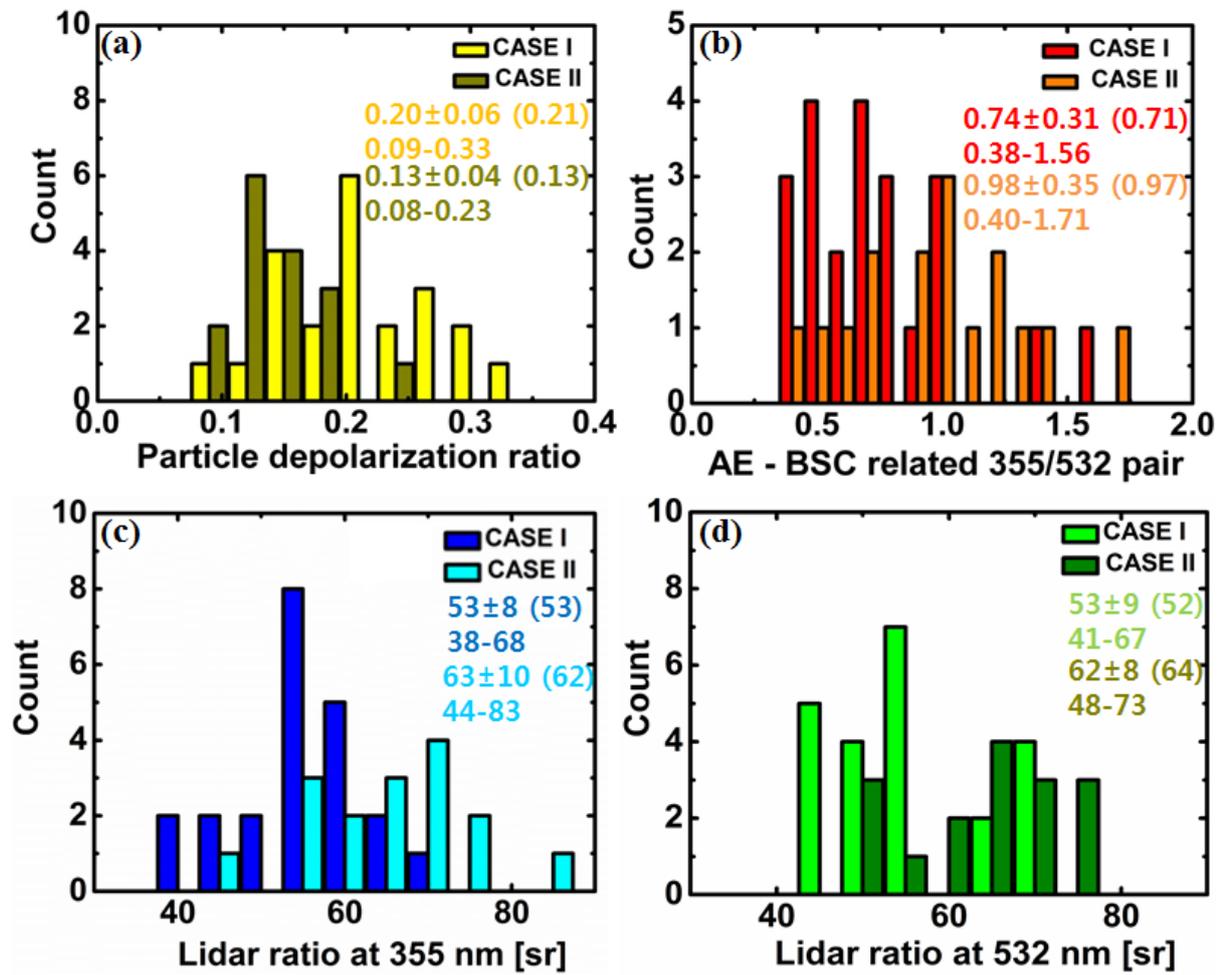
"Table 3. Summary of the linear particle depolarization ratio at 532 nm, the lidar ratios, and the backscatter Ångström exponents of Asian dust layers that passed over China at high altitude (above 3km) and low altitude (below 3km) when the level of pollution emission in China is lower (LP) and higher (MP), respectively."

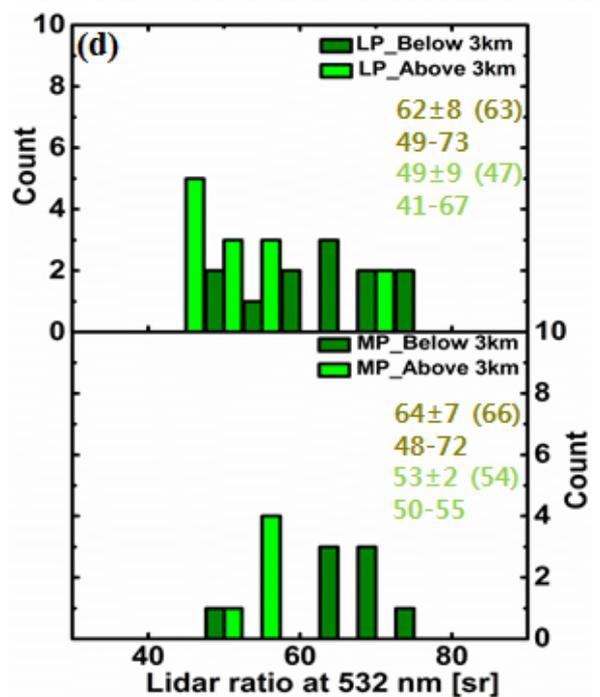
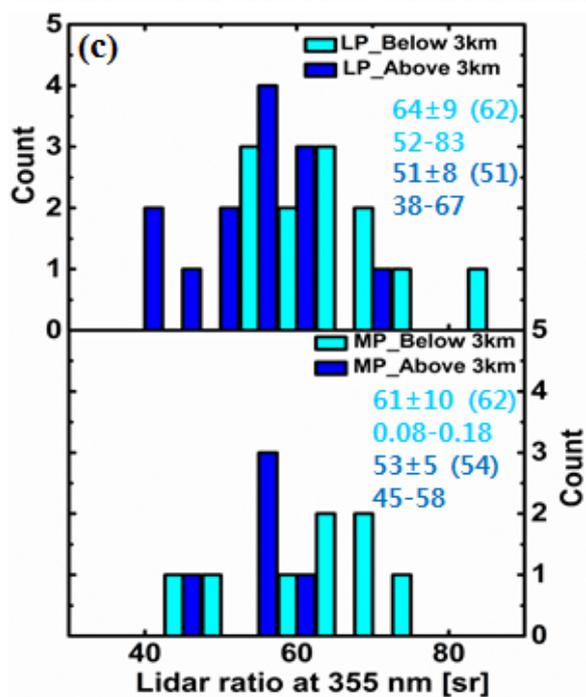
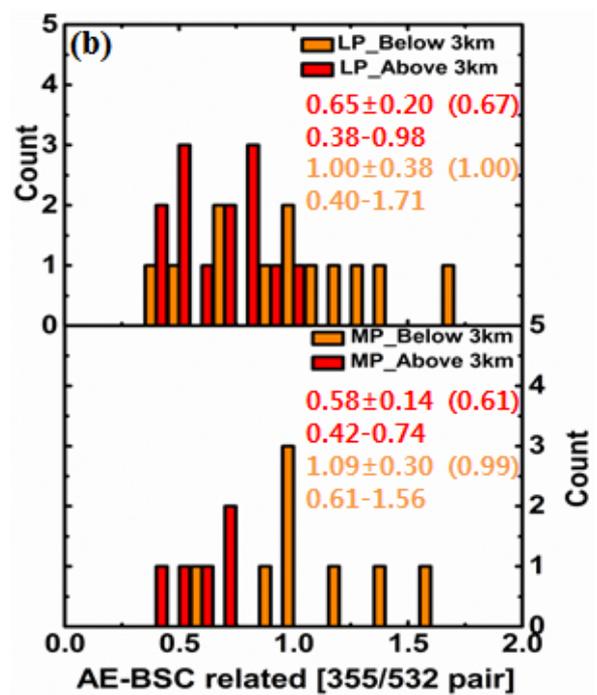
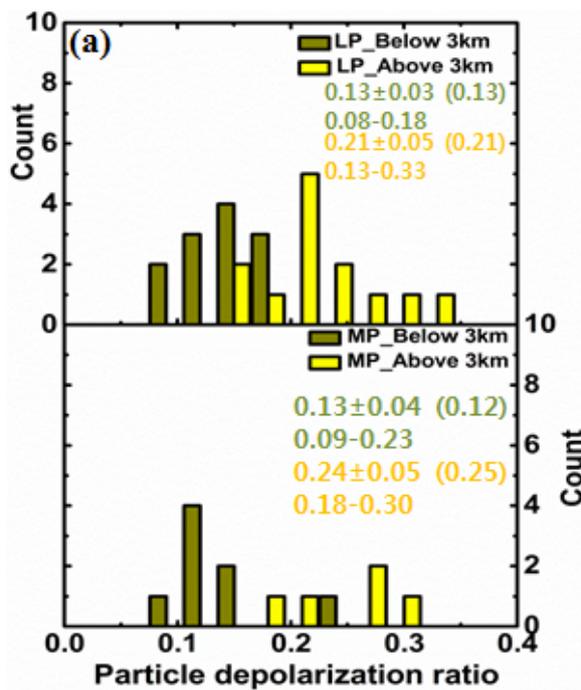
"Figure 8. Scatter diagram of the linear particle depolarization at 532 nm versus (a), (d) the backscatter-related Ångström exponent (355/532 nm wavelength pair), (b), (e) the lidar ratio at 355 nm and (c), (f) the lidar ratio at 532 nm. The left column (a-c) shows the optical properties of Asian dust layers considered as less polluted (LP), the right column (d-f) shows the more polluted cases. The Asian dust layers that passed over polluted regions in China at low altitude are denoted by black circles. The Asian dust layers transported at high altitude are denoted by red squares."

**Section 3.2: It would be valuable to include a frequency distributions of the retrieved optical properties separated for the both cases.**

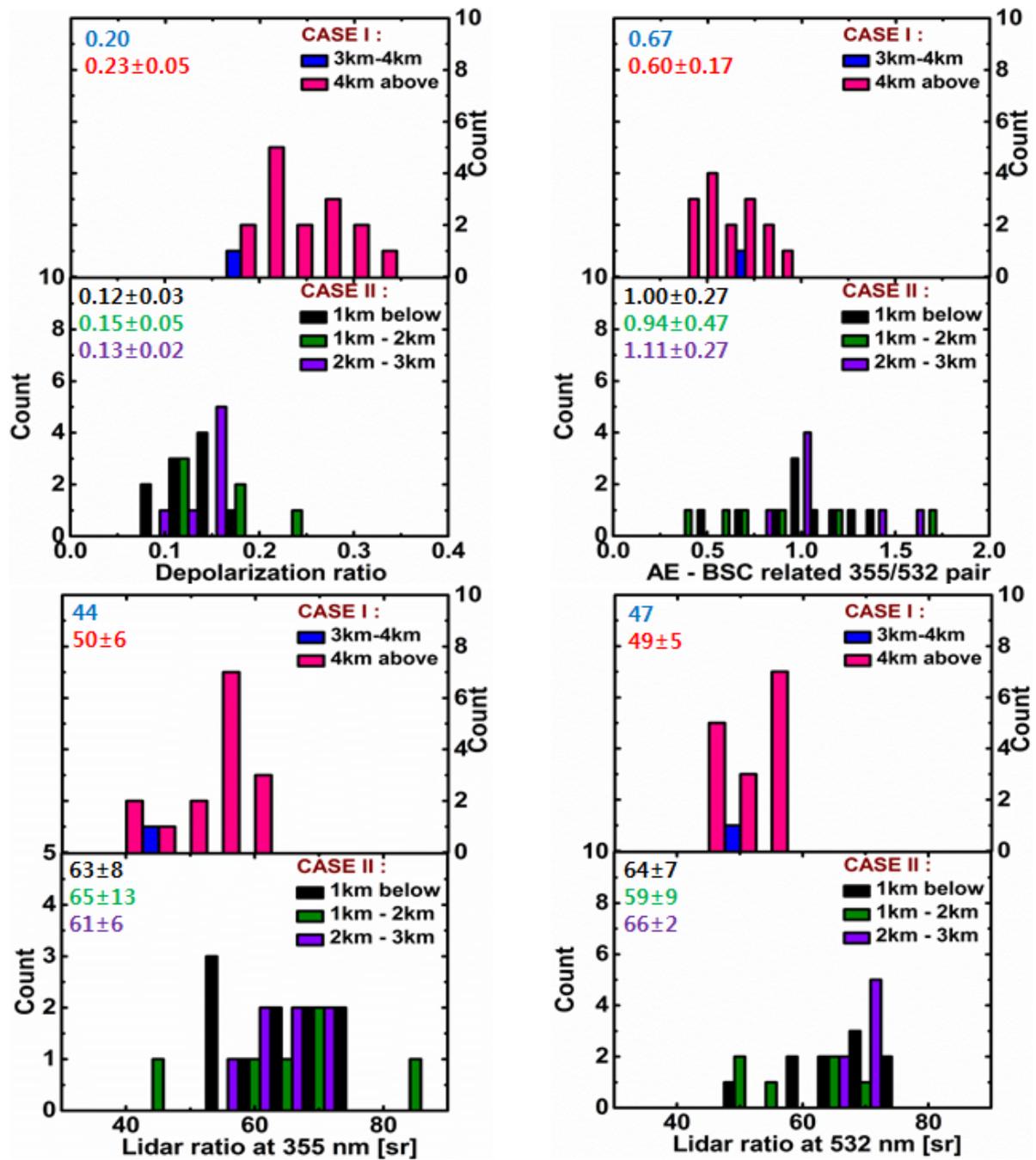
**Response:** We agree with referee. We will include the frequency distributions for all cases in section 3.2 with the figures shown below

This frequency distribution will be added as figure 7 in revised manuscript.





This frequency distribution will be added as figure 9 in revised manuscript.



This frequency distribution will be added as figure 11 in revised manuscript.

### **Minor comments:**

**Within text and figures the authors mostly only use the expression ‘depolarization ratio’. Please make sure whether the volume or the particle depolarization is meant, and change text and figures accordingly.**

**Response:** We analyzed the linear particle depolarization ratio in order to observe Asian dust layer in this study. The expression of “depolarization ratio” will be replaced to “the linear particle depolarization” in all of text and figures as your comment.

**p. 3384, l. 24: change ‘Asian dust plumes’ to ‘aerosol plumes’**

**Response:** It will be changed in revised manuscript.

**p. 3386, l. 6: A short description/summary of the main parameters of the lidar system would be nice.**

**Response:** The statement regarding the description of our lidar system “The light source of the lidar is pulsed Nd:YAG laser that is operated at 355 nm, 532 nm, and 1064 nm. The laser output power is 140, 154, and 640 mJ at the three emission wavelengths. The pulse repetition rate is 10 Hz. We use a beam expander at 532 nm and 1064 nm in order to reduce the divergence of emitted light. The receiver consists of a 14-inch Schmidt-Cassegrain telescope. The collected signals by telescope are according to wavelength with beam splitters and transmitted to photomultiplier tube (PMT). Transient recorders with 12-bit analog to digital converters and 250-MHz photon counter are used for processing the output signals of the PMTs.” will be added in the revised manuscript.

**p. 3386, l. 21: give the height of full overlap for  $\alpha_p$  measurements and thus for Sprofiles.**

**Response:** The statement “In that way we can retrieve vertical profiles of the backscatter coefficient to 400 m above ground. The vertical profiles of the aerosol extinction coefficients ( $\alpha_p$ ) at 355 and 532 nm were derived with the use of the nitrogen vibration Raman signals at 387 and 607 nm (Ansmann et al., 1990), respectively. The aerosol extinction coefficients can

be retrieved above 780 m and 540 m above ground at the measurement wavelengths of 355 nm and 532 nm. We derive particle extinction-to-backscatter ratios (lidar ratios, denoted as  $S$  in this contribution) at 355 and 532 nm from the profiles of  $\beta_p$  and  $\alpha_p$ .” will be added in the revised manuscript in order to describe the full overlap of  $\alpha_p$  and  $S$ .

**p. 3387, l. 20: Do the authors use the volume or the particle depolarization ratio?**

**Response:** We used the linear particle depolarization ratio. The word “depolarization ratio” will be replaced as “the linear particle depolarization ratio” not only in 3387, the whole text and figures in the revised manuscript as well.

**p. 3388, l. 6-7: ‘Asian dust is generally composed of a mixture of pollution: : :’ – this sentence is misleading. Observations were performed for a variety of mixtures of dust and pollution leading to a variety of  $\rho_p$ -values.**

**Response:** We agree with the reviewer’s comments. The statements will be removed.

**p. 3388, l. 11: Measurements of pure ‘aged’ dust plumes (e.g. SAMUM-2) did not show any changes in the retrieved particle depolarization ratio with respect to fresh pure dust. Thus the threshold was used to determine dust in aerosol mixtures.**

**Response:** The reviewer is correct. The threshold values of the particle depolarization ratio which is used in the study of Tesche et al. (2011) are used to determine dust in aerosol mixtures. This value is not suitable to be used as reference values in our study. We removed the reference values from the previous study of Tesche et al. (2011) as

“Shimizu et al. (2004) define 0.1 as threshold value for the determination of polluted dust.”

**p. 3388, l. 19-20: Do the  $\pm$ -values indicate the uncertainty of the mean or the standard deviation of the mean?**

**Response:** It indicated the standard deviation. The statement “The standard deviations were computed for the lidar ratios in each of the layers we could identify.” will be added in the

revised manuscript.

**p. 3389, l. 8-19: How do MACC analyses agree with measurements from satellites and with observations of your lidar measurements?**

**Response:** The re-analysis data from MACC is based on the satellite data (MODIS). The re-analysis assimilates MODIS observation data into a model and this data assimilation system corrects for model departures from observational data (Bellouin et al., 2013\*).

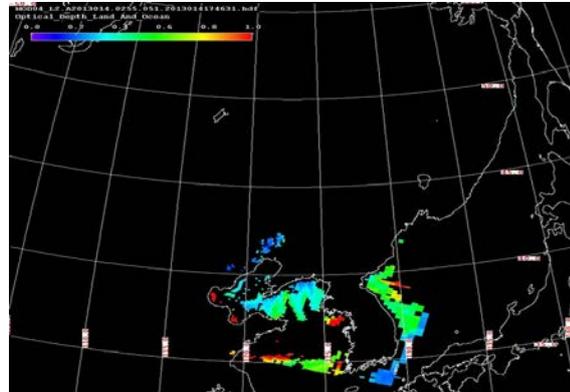
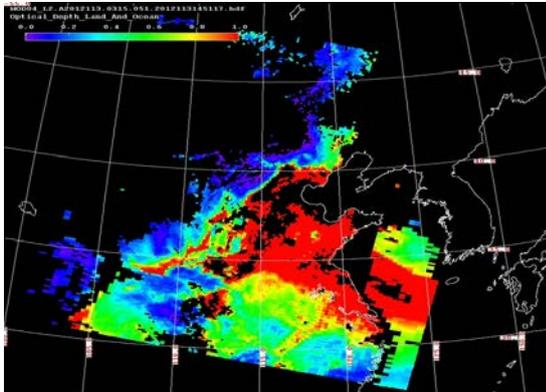
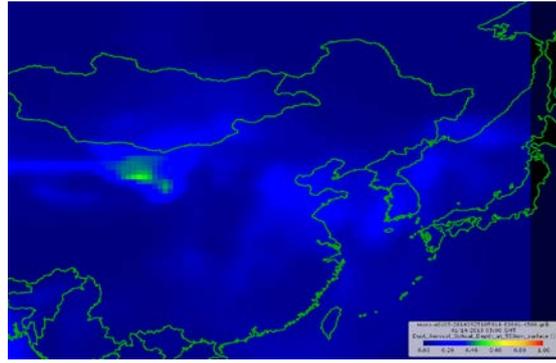
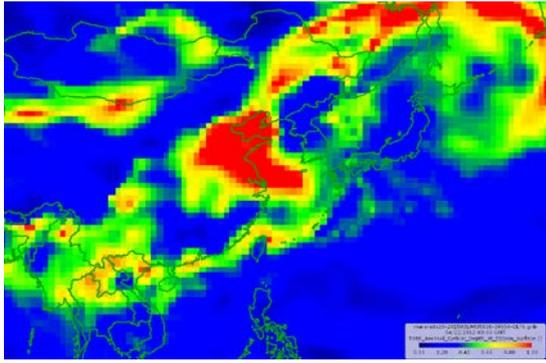
\* Bellouin, N., Quaas, J., Morcrette, J.-J., and Boucher, O.: Estimates of aerosol radiative forcing from the MACC re-analysis, *Atmos. Chem. Phys.*, 13, 2045–2062, doi:10.5194/acp-13-2045-2013, 2013.

The MACC re-analysis data is considered as reliable and widely used to estimate the level of anthropogenic pollution (e.g., AOD for black carbon, organic matter, sulphate).

We are supposed to use the results from MACC in order to evaluate the level of pollution emission in the regions of China that Asian dust plumes passed described as “We used the aerosol AOD from the MACC re-analysis to determine the intensity of pollution (AOD) in densely populated and industrialized regions along the transport path of the dust layers and to investigate the influence of anthropogenic pollution particles on the variation of the optical properties of Asian dust.”

We did not compare results from each measurement.

And the comparisons between AOD retrieved by satellite (MODIS) and MACC are shown as follows



as shown above, the total AOD distribution derived from MACC re-analysis and MODIS are in good agreement for each case.

Moreover, we used AOD for black carbon, organic matter, and sulphated provided from MACC re-analysis data for the identification of level of emission of anthropogenic pollutant which can mix with dust particles. The MODIS provides the total AOD that cannot be distinguished whether it is pollutant aerosols.

**p. 3389, l. 21-22: What is the splitting of the 38 observed dust layers to high and low transport path above China, and to the observed height range above your site?**

**Response:** The vertical distributions of Asian dust layers during observation period were determined with linear particle depolarization ratio measurement as described previously. The statement “These Asian dust layers were identified on the basis of the linear particle depolarization ratio measurements as described in section 2.2. The vertical profiles of the linear particle depolarization ratio allow us to determine the vertical distribution of the Asian dust layers.” will added in the revised manuscript.

**p. 3390, l. 19: What do you mean by ‘moderately aged’? Do you really mean aging (shape/size dependent sedimentation, coating, : : :)?**

**Response:** The most cases of Asian dust layer we observed is considered as external mixture. We will remove the sentences.

**p. 3391, l. 28-29: This sentence is misleading as the lowest S-values in the high \_p-cluster are found for the lowest \_p-values within this cluster. Please change or comment on this.**

**Response:** We agree with the reviewer’s concern. The statement will be changed in the revised manuscript as

“Lower values of  $\delta_p$  are dominantly found in the domain where lidar ratios are above 60-70 sr, except for a few cases.”

**p. 3393, l. 8-10: Please change to ‘mean values of the particle depolarization ratio : : :’ as single measurements agree well with former findings of pure dust layers.**

**Response:** The statement “The depolarization ratios of Asia dust plume” will be changed to “The mean values of the linear particle depolarization ratios of the Asian dust plumes” as the referee’s recommendation.

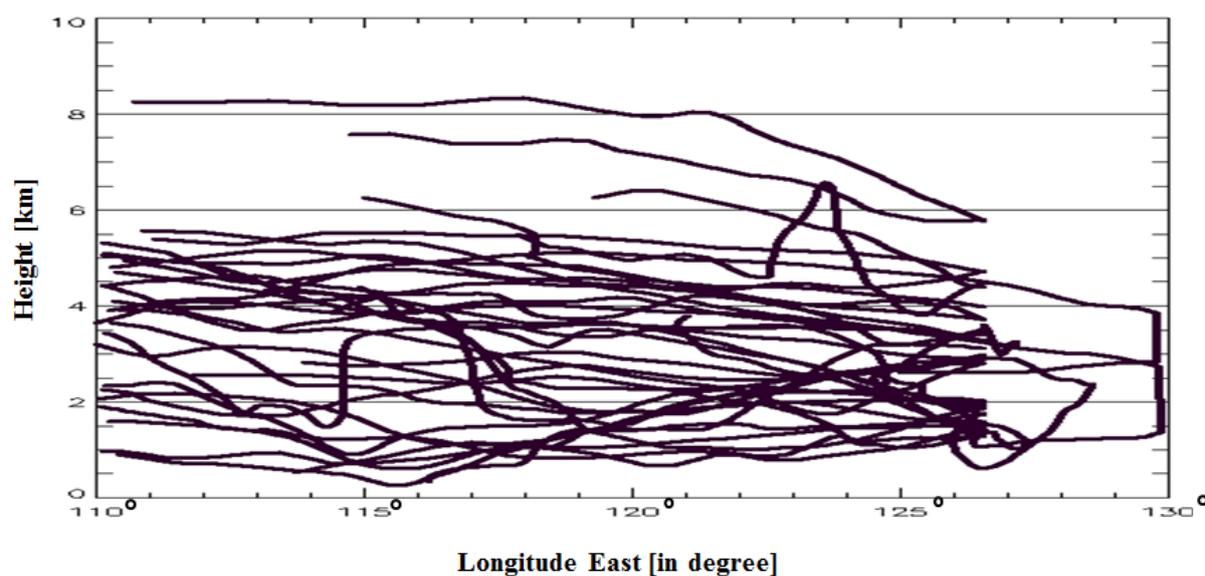
**Figure 3: Why does the shape of Lidar ratio distribution at 355 nm differ from the**

**shape of Lidar ratio distribution at 532 nm?**

**Response:** The larger lidar ratio at 355 nm than the lidar ratio at 532 nm may result from the larger absorption in the UV. However, the accurate description for the wavelength dependence of lidar ratio is not possible because of the dependence on the size distribution, chemical composition, and particle shape.

**Figure 3: An additional plot of the trajectories height would be valuable.**

**Response:** The plot of the trajectories height will be added up in the revised manuscript as follows



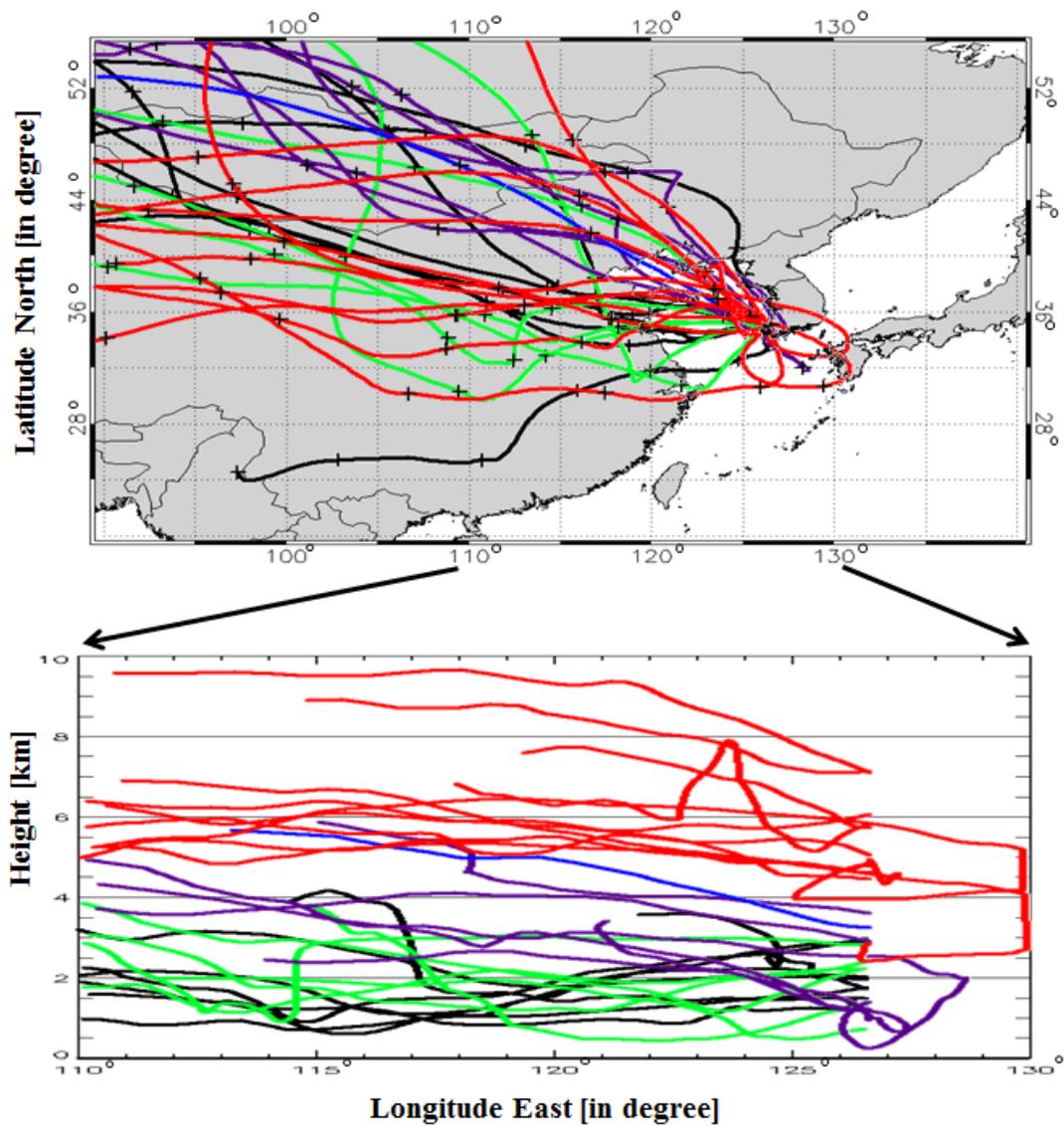
**Figure 6: It should be indicated which colors denote to which case.**

**Response:** The caption for the classification of each cases for figure 6 will be added in the revised manuscript as

“Case I is indicated by red colored circle and Case II is indicated by black colored circle.”

**Figure 7: Color coded trajectories according to their heights above the lidar site would improve this Figure.**

**Response:** The figure will be improved with colored trajectories in the revised manuscript as



**Figure 8: What are the black lines?**

**Response:** The line was supposed to show that we may not be able to find values to the right side and these lines seem to be tilted always in the same way. In any case of misleading this line will be removed in the revised manuscript.