

Dear Editor,

We are submitting a revised manuscript, entitling "Vertical variation of optical properties of mixed Asian dust/pollution plumes according to pathway of air mass transport over East Asia". We would like to give many thanks to editor and reviewers for the invaluable comments. The reviewers pointed out that the idea is interesting and relevant to the ACP, but more scientific explanation/discussion for the results would be needed. We found the comments of the reviewers provided significant value to us in preparing the revised manuscript. We therefore revised our original manuscript to address all of the concerns raised by the reviewers.

A point by point response is given below.

Thank you very much for reconsidering this manuscript.

Authors' response to Referee #1' comments

Paper No.: ACP-2014-1030

Title: Vertical variation of optical properties of mixed Asian dust/pollution plumes according to pathway of air mass 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 #1

General comments:

This present study investigates optical property variations of mixed Asian dust with anthropogenic pollution according to the pathways and vertical distributions during long-range transport. The authors find a decrease of the linear depolarization ratio of the mixed dust/pollution plume in dependence of transport time if the pollution layer travelled over China at low heights, i.e., below approximately 3 km above ground. We can separate aerosol types as anthropogenic pollution particle, smoke particle, sea-salt and dust in the atmospheric aerosol study. However, the mixed aerosol is observed in the

many cases, especially in Asian dust. Asian dust particles, which originate from desert areas in the Asian continent, can be mixed with polluted aerosols that contain black-carbon and/or smoke particles while they are transported over industrial regions. This mixture of dust particles with anthropogenic particles causes changes in optical properties of dust layers. The increased radiative forcing exerted by East Asian dust plumes can be largely attributed to the presence of highly light-absorbing anthropogenic particles that are mixed in dust layers. However, the vertical structure and the degree of vertical mixing between dust and pollution layers during transport are poorly understood, primarily because of the lack of vertically resolved observations of aerosol pollution. In this respect, the results presented in this study are expected to be able to help the understanding of the degree of mixing of dust. In general, this manuscript is well organized and the results are valuable for improving our understanding of the dust particles and the mixing status of dust and pollution particles. The manuscript should be published in "Atmospheric Chemistry and Physics". But it requires some minor revision before to be published.

Comments:

Line 59 : 63 ± 9 sr at 355 nm and 62 ± 8 sr at 532 nm, respectively -> 63 ± 9 and 62 ± 8 sr at 355 and 532 nm, respectively

Response: It will be changed as "63±9 and 62±8 sr at 355 and 532 nm, respectively" at line 61 of the revised manuscript

Line 85, 87 : Change the order of references by year.

Response: The order of references will be changed as order by year as below

- line 88 of revised manuscript : such as ACE-Asia (Huebert et al., 2003) and ADEC (Mikami et al., 2006)

- line 90 of revised manuscript : . Carrico et al. (2003) and Yu et al. (2006)
- line 94 of revised manuscript : (Wang et al., 2007; Sun et al., 2010)

Line 128 : Add “at 532 nm” after “depolarization ratio”

Response: The word “at 532 nm” will be added at line 139 of revised manuscript

Line 137 : Please denote the minimum observation altitude of extinction coefficient.

Response: The sentence “The α_p can be retrieved above 780 m and 540 m at measurement wavelength of 355 nm and 532 nm.” Will be added at line 149 of revised manuscript

Line 142, 146 : Authors used “co-polarized and cross-polarized” in line 142 to explain depolarization ratio. But, “perpendicular and parallel” were also used in line 146. Those expressions are different? If those intend to deliver the same meaning, Please consider using just one of them throughout the manuscript or try to define them together.

Response: The words “co-polarized and cross-polarized” and “perpendicular and parallel” denoted same meaning. The statement will be changed as “Parallel polarized and perpendicular polarized signals are measured at 532 nm.” at line 185 of revised manuscript

Section 2.2 : What is the threshold level of depolarization ratio to identify dust layer?

Response: We used 0.08 as a threshold value to identify dust. The statement "In this study 0.08 was considered as threshold value of δ_p to identify dust." will be added at line 222 of revised manuscript.

Line 182 : as low -> low as

Response: It will be changed from "as low" to "low as".

Line 214-215 : Insert the standard deviation for each mean value.

Response: The standard deviations of each value will be added as "The average value of δ_p for all observed Asian dust layers is 0.17 ± 0.02 . The average values of S are 57 ± 6 sr at 355 nm and 57 ± 7 sr at 532 nm. The mean value of A_p is 0.84 ± 0.37 ." at line 266-267 of revised manuscript.

Section 3.1, line 238-241 : The sentence is hard to understand. How do you classify the dust layers as two episodes? Dose the sentence from line 255 to 261 account for two episodes? If yes, move that sentence to the front of this section and then reorganize the sentence.

Response: The sentences "On the basis of the distribution of AOD of anthropogenic pollution over China, the Asian dust layers were classified as "more polluted", i.e., "MP" Asian dust when the modelled AOD of anthropogenic pollution on that day was higher than the average AOD (modelled) of all 32 observation days considered in this study. In contrast, Asian dust layers that passed over China during episodes of lower AOD, i.e., AOD was below the mean value of modelled AOD of all 32 observation days are denoted as "less polluted", i.e., "LP" Asian dust." will be moved to front of section 3.1 to clarify two episode as your comment.

Line 292 : What kind of the model do you use? You just mentioned "model results".

Response: We used the HYSPLIT mode result to determine the height of the dust layers above ground during transport over China. The statement "The vertical positions of the dust plumes above ground during transport over China were inferred from the model results. We assume that the height of the dust plumes above ground can be distinguished by HYSPLIT model results although the results may have a certain error because of the spatial and temporal complexity of the meteorological fields involved in the computations." will be added at line 345-349 of revised manuscript

Line 307-310 : Remove those sentences. The sentences are same with the sentences in line 216-218.

Response: It has been removed.

Line 437 : Instead of “high altitudes”, display the correct altitude.

Response: The statement “at high altitudes, i.e., above 3 km.” will be added instead of high altitudes at line 436 of revised manuscript

Table 1, 2, 3 : Lidar ratio -> Lidar ratio (sr)

Response: It will be changed as “Lidar ratio (sr)” in all of tables in revised manuscript

Figure 2 : VDPR, PDPR, LR532, LR355, Bsc. Ang. 355/532 -> Use the symbol in the manuscript.

Response: The symbols in figure 2 will be changed as symbol used in text in revised manuscript.

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 δ_p for all observed Asian dust layers is 0.17 ± 0.02 . The average values of S are 57 ± 6 sr at 355 nm and 57 ± 7 sr at 532 nm. The mean value of A_β is 0.84 ± 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 δ_p , 38-83 sr for S at 355 nm, 41-73 sr for S at 532 nm, and 0.38-1.71 for A_β . The maximum value of δ_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 A_β is 0.38.

This maximum value of δ_p and the minimum values of S at 355 nm and 532 nm and A_β are similar to the values of optical properties for pure dust particles.

76% of δ_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 (A_β) 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 ± 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

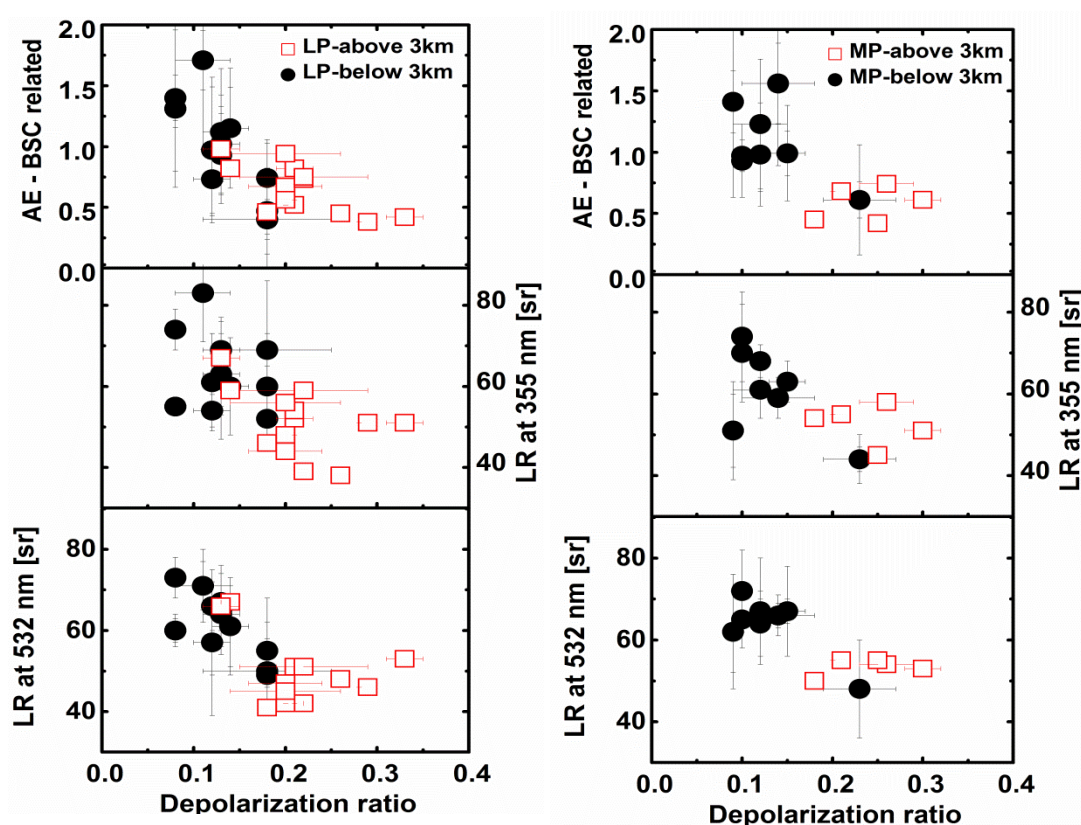
describe continental-polluted aerosols and polluted dust." and "The values of 0.2-0.3 are reported as the values of A_{β} for Saharan dust (Murayama et al., 2002; Tesche et al., 2009). Chen et al. (2007) and Müller et al. (2010) find values of 0.7-1.5 for A_{β} for a mixture of mineral dust with urban haze. Values of 0.8-1.4 for A_{β} 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 A_{β} (wavelength range 355/532 nm), and S at 355 nm and 532 nm versus δ_p at 532 nm in dependence of the level of pollution emission and the vertical position. The frequency distribution of δ_p at 532 nm, A_{β} (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 δ_p at 532 nm and A_{β}

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 δ_p at 532 nm and A_β 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 δ_p at 532 nm, A_β , and S are 0.24 ± 0.05 , 0.58 ± 0.14 , and 53 ± 5 at 355 nm and 53 ± 2 at 532 nm, respectively, for the case "MP_above 3km". The highest values of δ_p and lowest values of A_β are found for this case.

In the case of "LP_above 3km" the mean values of δ_p at 532 nm and A_β are 0.21 ± 0.05 and 0.65 ± 0.20 , respectively. The mean values of S are 51 ± 8 sr at 355 nm and 49 ± 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

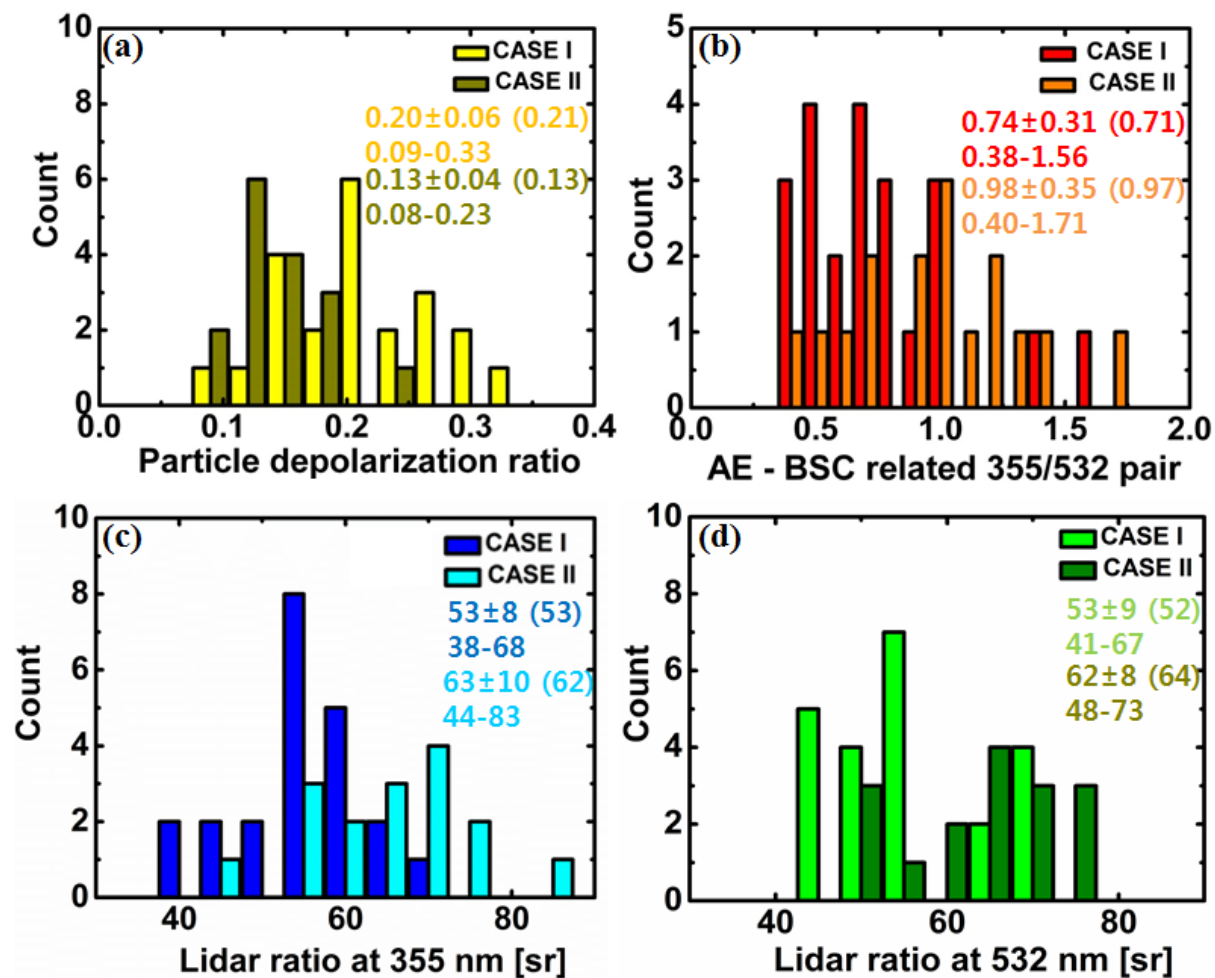
China is lower (LP) and higher (MP), respectively."

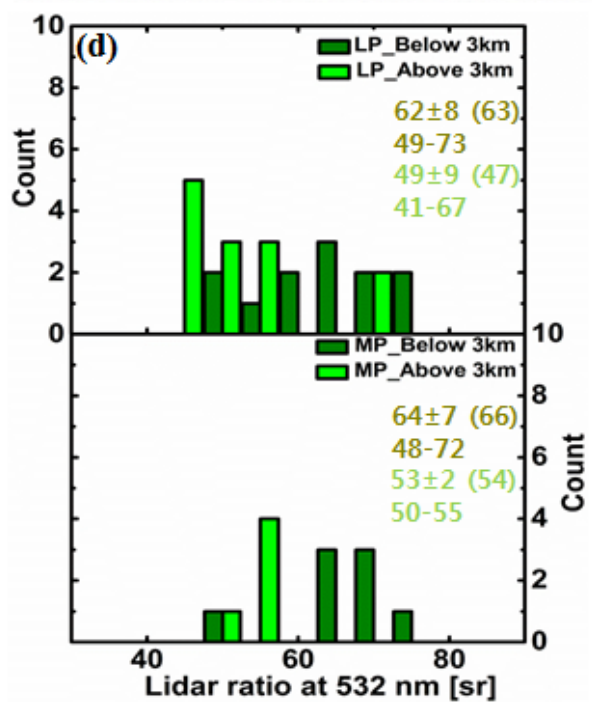
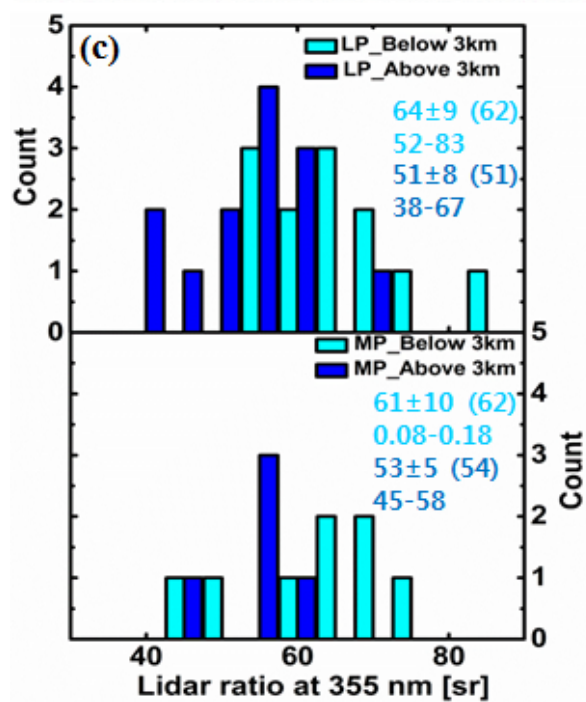
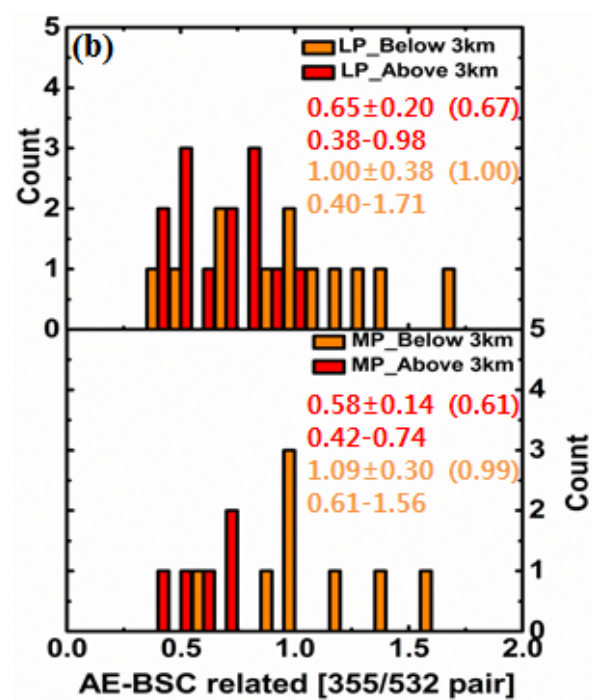
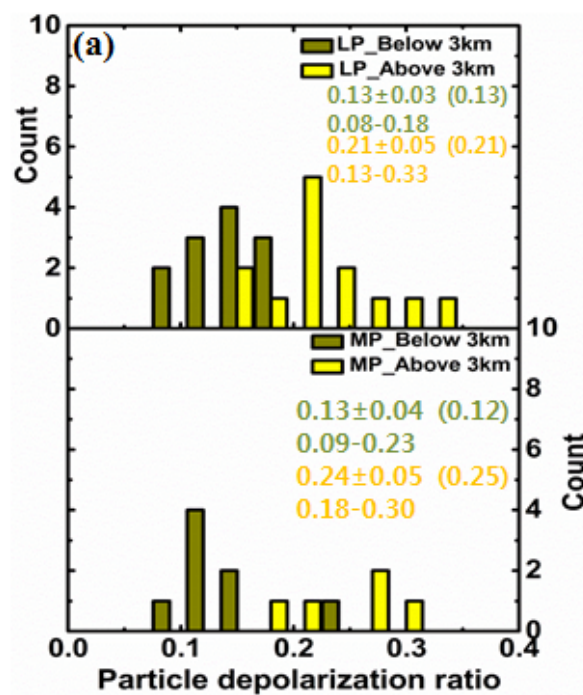
"Figure 8. Scatter diagram of the linear particle depolarization at 532 nm versus (a), (d) the backscatter-related Ångström exponent (355/532 nm wavelength pair), (b), (e) the lidar ratio at 355 nm and (c), (f) the lidar ratio at 532 nm. The left column (a-c) shows the optical properties of Asian dust 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.

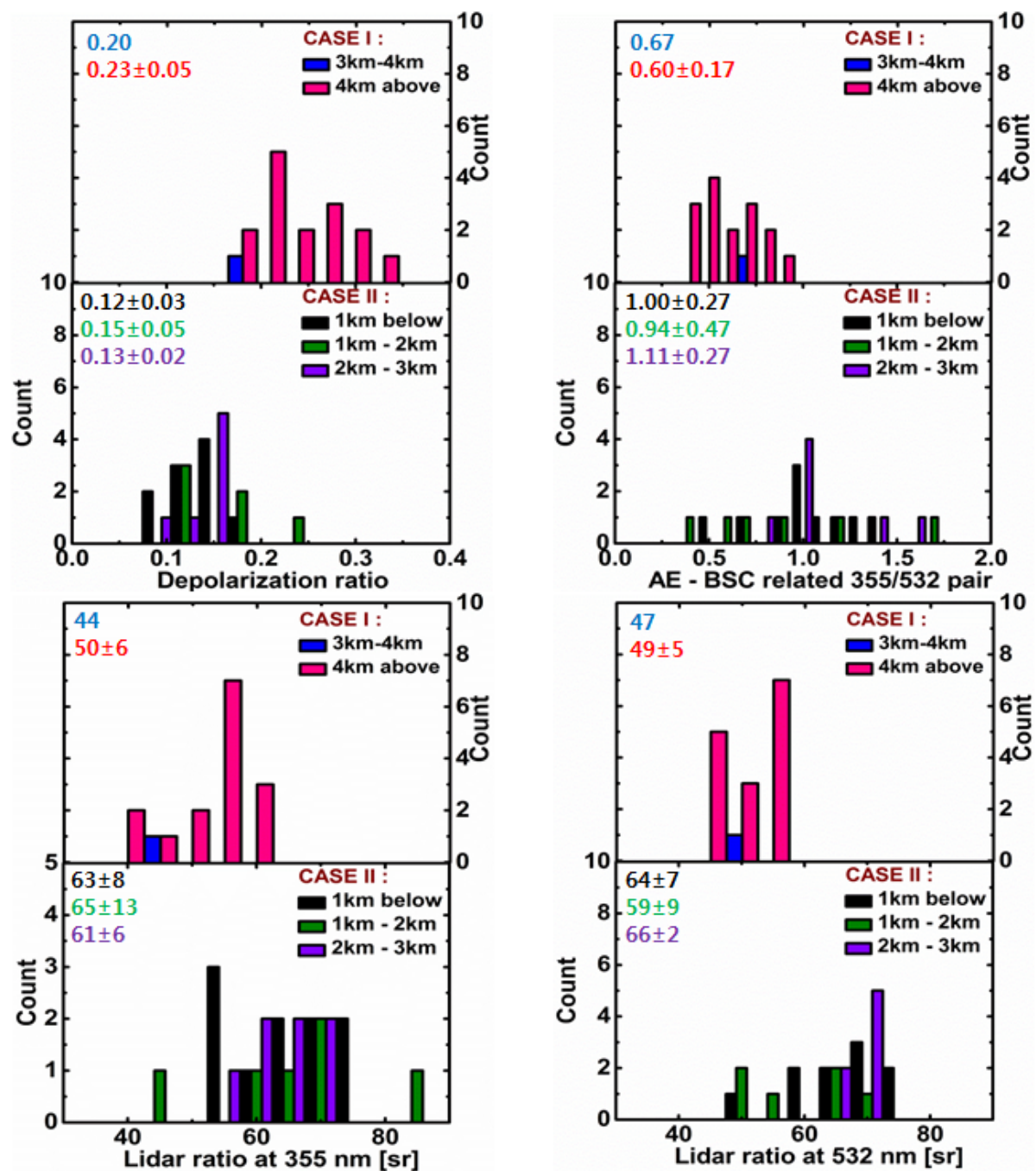
Response: We agree with referee. We will include the frequency distributions for all cases in section 3.2 with the figures shown below

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 β_p measurements and thus for S profiles.

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 (α_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 β_p and α_p ." will be added in the revised manuscript in order to describe the full overlap of α_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 β_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 mode and this data assimilation system correct for model departures from observational data (Bellouin et al., 2013*).

* Bellouin, N., Quaas, J., Morcrette, J.-J., and Boucher, O.: Estimates of aerosol radiative

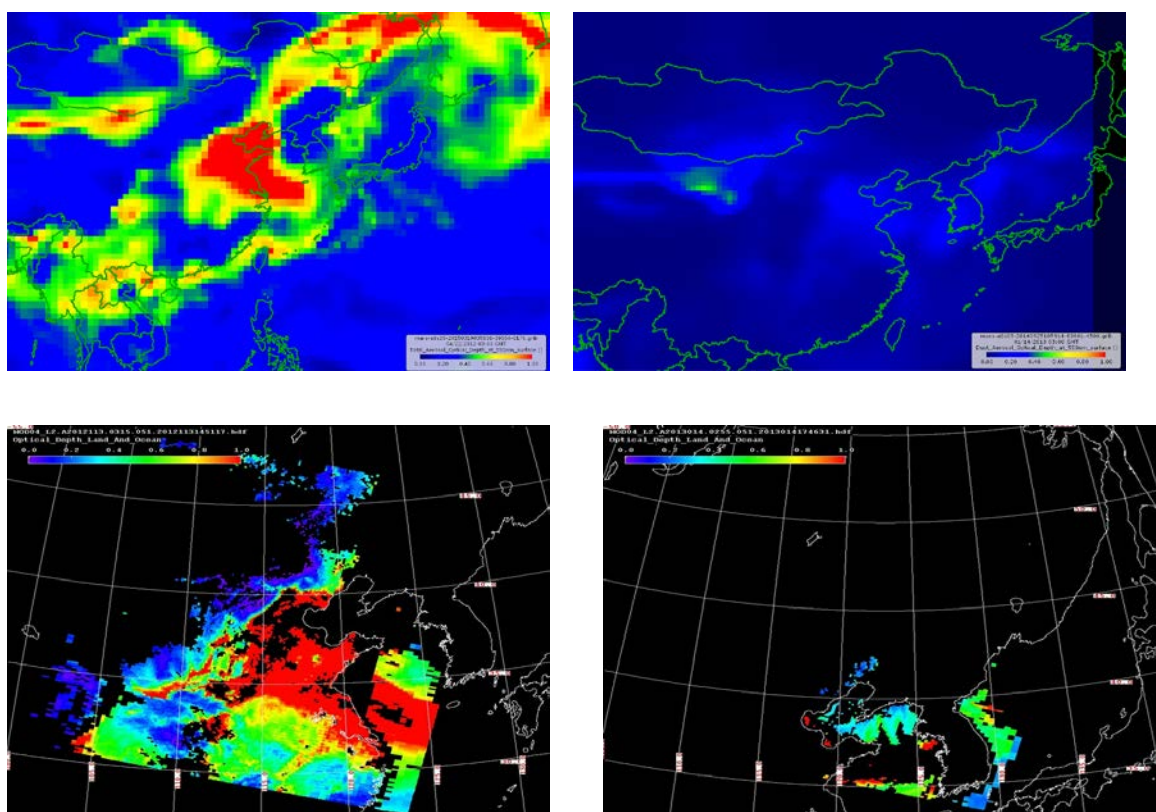
forcing from the MACC re-analysis, *Atmos. Chem. Phys.*, 13, 2045–2062, doi:10.5194/acp-13-2045-2013, 2013.

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 ρ_p -cluster are found for the lowest ρ_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 δ_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

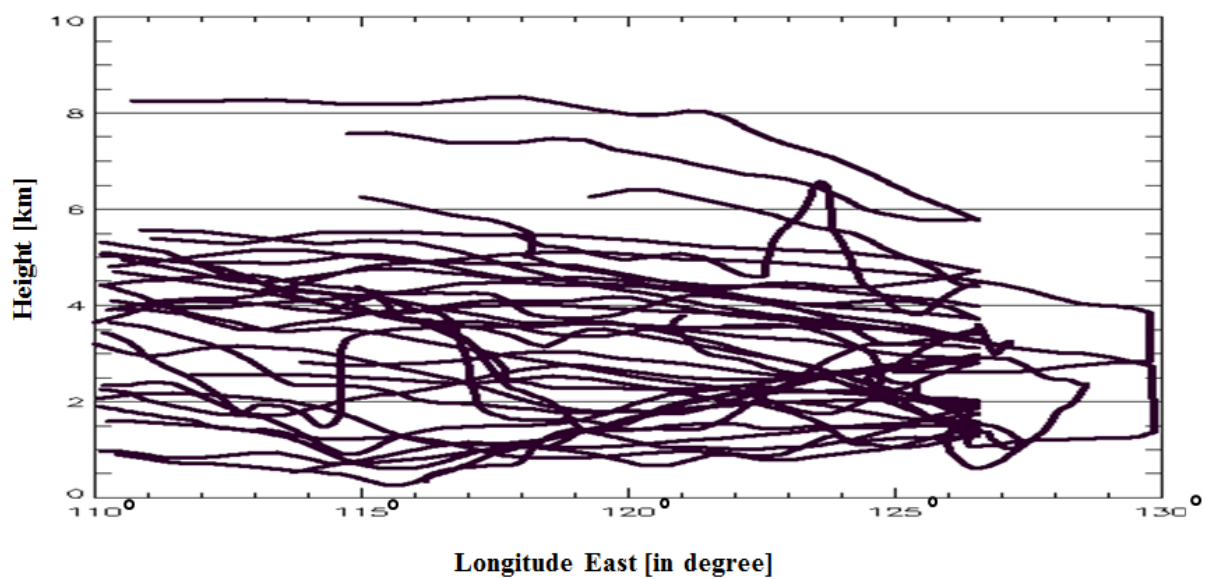


Figure 6: It should be indicated which colors denote to which case.

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

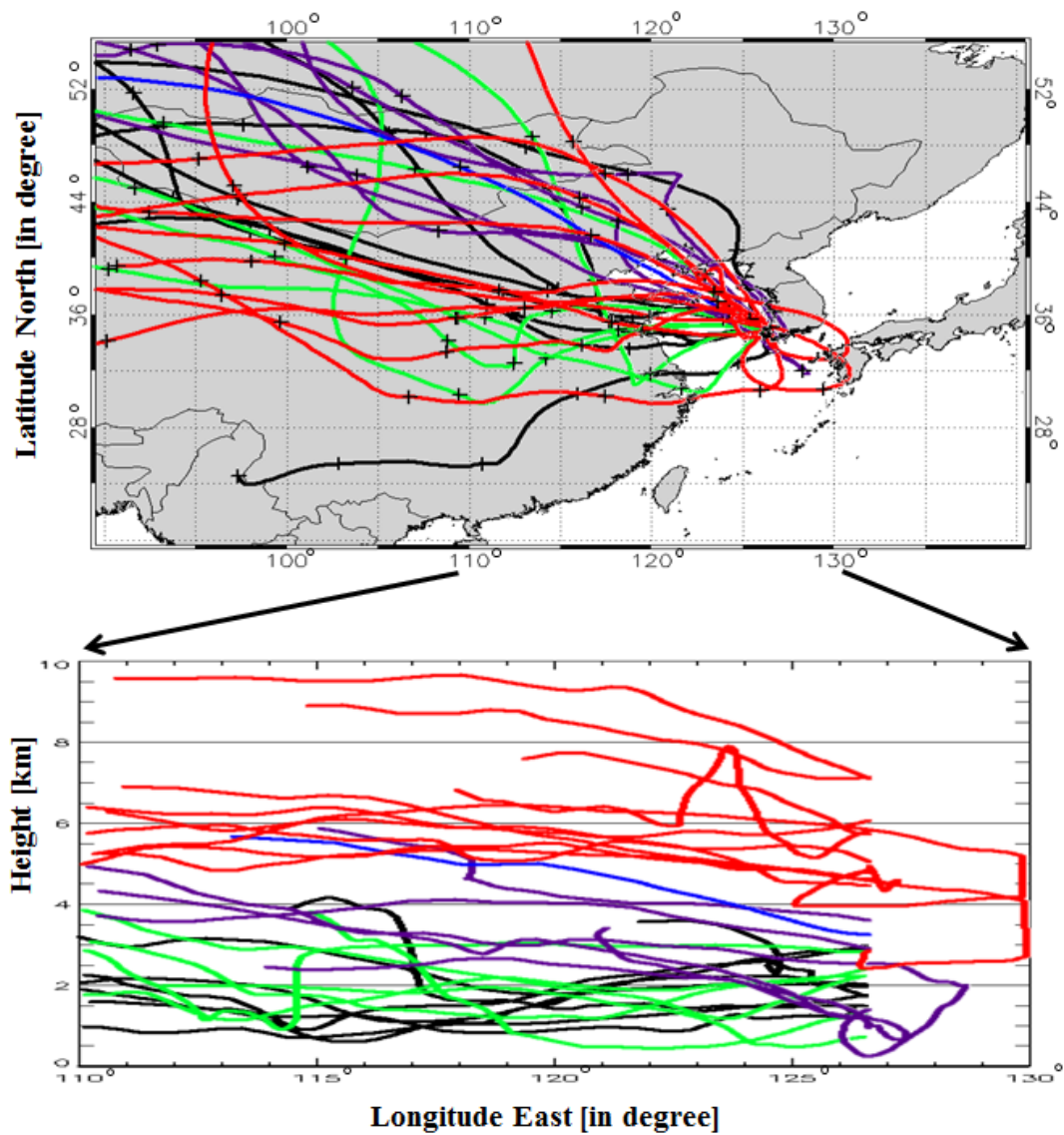


Figure 8: What are the black lines?

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.

Authors' response to Referee #3' comments

Paper No.: ACP-2014-1030

Title: Vertical variation of optical properties of mixed Asian dust/pollution plumes according to pathway of air mass 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 #3

General comments:

The present study investigates dust plumes over East Asia observed by a Multi-Wavelength Raman Lidar at the Gwangju Institute of Science and Technology in the Republic of Korea. The first long-term measurements from this site are presented. A comprehensive analysis of typical LIDAR quantities is used to characterize the observed aerosol from dust plumes which are partially mixed with anthropogenic aerosol from industrialized areas in China.

While these observations are important for the aerosol community and should be published, the discussion could be improved to make the results attractive for a wider audience, e.g. by more interpretation of derived LIDAR quantities. Evidence is given for the pollution of the dust plume with anthropogenic aerosol, a very interesting and currently not-well understood problem, but the method is not convincing. I encourage the authors to improve their method through using satellite observations instead of MACC for the classification of MP and LP events and revising the comparison of pollution magnitudes at different levels (see main comments below). These changes would give a more solid basis for the discussion and add value to the implications of their findings. The overall organization and presentation of the results are good. I recommend publication in ACP after revision of the manuscript.

Main comments:

Section 2.1: State how the linear depolarization ratio and the Angstrom exponent is typically interpreted for aerosol characteristics. Add typical ranges of values for all final quantities that you derive from the measurements so that the reader can better assess the uncertainty of your results later in the manuscript.

Response: We agree with referee's comment. The statement

"The lidar ratios can be used for aerosol typing (Müller et al., 2007). Murayama et al., (2004) find values of $S = 48.6$ sr at 355 nm and $S = 43.1$ sr at 532 in a well-isolated Gobi dust-laden layer observed above 4 km over Tokyo. De Tomasi et al. (2003) report an S value less than 50 sr at 351 nm for a Saharan dust layer. Values of S at 355 nm ranged between 50 sr and 80 sr for dust observed over Leipzig, Germany (Mattis et al., 2002). In contrast, Ferrare et al. (2002) report a high value of 68 ± 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.

The backscatter-related Ångström exponent for the wavelength pair of 355/532 nm (denoted as $\text{\AA}_{\beta}$) is computed, too. The backscatter-related Ångström exponent is a good indicator of the size of particles. High values (>1) are typically observed for accumulation mode particles such as fresh biomass-burning particles. Low values (~ 0) are observed for coarse mode particles such as Saharan dust or Asian dust (Eck et al., 1999; Sakai et al., 2002, Chen et al., 2007). The values of 0.2-0.3 are reported as the values of $\text{\AA}_{\beta}$ 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 $\text{\AA}_{\beta}$ for a mixture of mineral dust with urban haze. Values of 0.8-1.4 for $\text{\AA}_{\beta}$ were found for heavily polluted continental aerosol layers (Franke et al., 2003).

The depolarization ratio is used as indicator of particle shape (Bohren and Huffman, 2008). High values of the depolarization ratio of 0.3 to 0.35 at 532 nm indicate nearly pure dust (Sugimoto and Lee, 2006; Freudenthaler et al., 2009). For example, Freudenthaler et al. (2009) report a value of $\delta_p = 0.31$ at 532 nm for Saharan dust observed during SAMUM 2006. Lidar observations were carried out close to the Taklamakan desert (Iwasaka et al., 2003) and the Gobi desert (Yi et al., 2014). We assume that these dust layers exhibit nearly pure dust conditions as anthropogenic pollution sources in these isolated areas are sparse. Values of δ_p are in the range of 0.3 to 0.35 at 532 nm (Iwasaka et al., 2003; Yi et al., 2014). 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 up in section 2.

Also the paragraph

"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 δ_p , 38-83 sr for S at 355 nm, 41-73 sr for S at 532 nm, and 0.38-1.71 for A_β . The maximum value of δ_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 A_β is 0.38. This maximum value of δ_p and the minimum values of S at 355 nm and 532 nm and A_β are similar to the values of optical properties for pure dust particles.

76% of δ_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 (A_β) 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 up to explain the difference in the values of optical properties in section 3

The reference "Eck, T., Holben, B., Reid, J., Dubovik, O., Smirnov, A., O'Neill, N., Slutsker, I., and Kinne, S.: Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols, Journal of Geophysical Research: Atmospheres (1984–2012), 104, 31333-31349, 1999." will be added.

The references "Anderson, T. L., Masonis, S. J., Covert, D. S., Charlson, R. J., and Rood, M.

J.: In situ measurement of the aerosol extinction-to-backscatter ratio at a polluted continental site, *Journal of Geophysical Research: Atmospheres* (1984–2012), 105, 26907-26915, 2000.", "Ansmann, A., Riebesell, M., Wandinger, U., Weitkamp, C., Voss, E., Lahmann, W., and Michaelis, W.: Combined Raman elastic-backscatter lidar for vertical profiling of moisture, aerosol extinction, backscatter, and lidar ratio, *Applied Physics B*, 55, 18-28, 1992a.", "Liu, Z., Sugimoto, N., and Murayama, T.: Extinction-to-backscatter ratio of Asian dust observed with high-spectral-resolution lidar and Raman lidar, *Applied Optics*, 41, 2760-2767, 2002." will be added.

The references "Sugimoto, N., and Lee, C. H.: Characteristics of dust aerosols inferred from lidar depolarization measurements at two wavelengths, *Applied Optics*, 45, 7468-7474, 2006.", "Somekawa, T., Yamanaka, C., Fujita, M., and Galvez, M. C.: A New Concept to Characterize Nonspherical Particles from Multi-wavelength Depolarization Ratios Based on T-matrix Computation, *Particle & Particle Systems Characterization*, 25, 49-53, 2008.", "Pan, X., Uno, I., Hara, Y., Kuribayashi, M., Kobayashi, H., Sugimoto, N., Yamamoto, S., Shimohara, T., and Wang, Z.: Observation of the simultaneous transport of Asian mineral dust aerosols with anthropogenic pollutants using a POPC during a long-lasting dust event in late spring 2014, *Geophys. Res. Lett.*, 42, doi: 10.1002/2014GL062491, 2015" will be added.

pp. 3392, l. 21-22: It seems you do not trust the model data, but it remains unclear in the text why this is the case. Please specify your criticism.

There are retrieved AODs from satellite products available, e.g. from MODIS. Using those (example below) instead of MACC would eliminate the model uncertainty for your investigation. The model currently determines the classification of your observations in LP and MP events, the differences of which are small (Fig. 5). However,

the model uncertainty prevents to conclude that other factors dominate (pp. 3392, l. 11-13), since you cannot be sure that the classification with the model data is correct.

Response: We are not supposed to deliver that we do not believe the results from model. We attempt to notice that the model results have an uncertainty (as the measurements have a uncertainty) And the pollution particles might be continuously existed on the transport pathway of dust layers although we do not have a results regarding this from direct measurement.

However, the statement "It is clear that this analysis contains significant uncertainty (1) model results are used to determine the height of the dust layers above ground during transport over China and (2) we do not have direct measurement of aerosol optical depth along the trajectories of the particle plumes before arrival over Gwangju" can be understood complicatedly as you pointed out.

This statement "It is clear that this analysis contain significant uncertainty" will be removed.

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 correct for model depatures from observational data (Bellouin, 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.

In the study "MACC Work Package G-AER 3 Monitoring of aerosol direct and indirect forcing" by Nicolas Bellouin and Johannes Quaas, Karsten Peters stated the data from

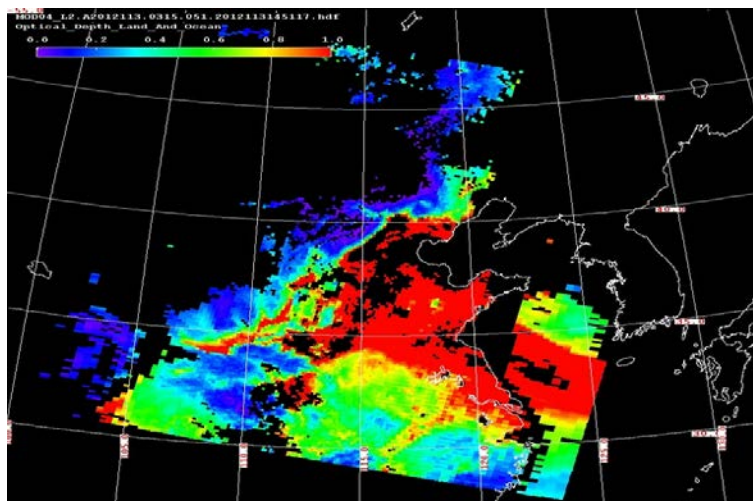
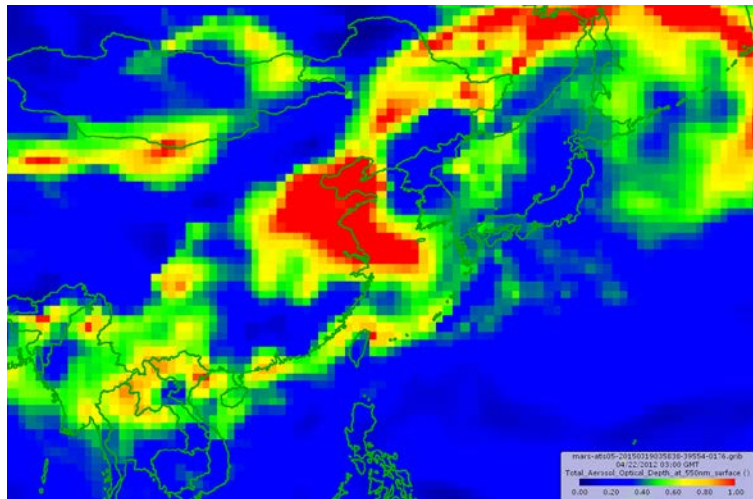
MODIS has tendency to overestimate and the bias is corrected in the MACC re-analysis.

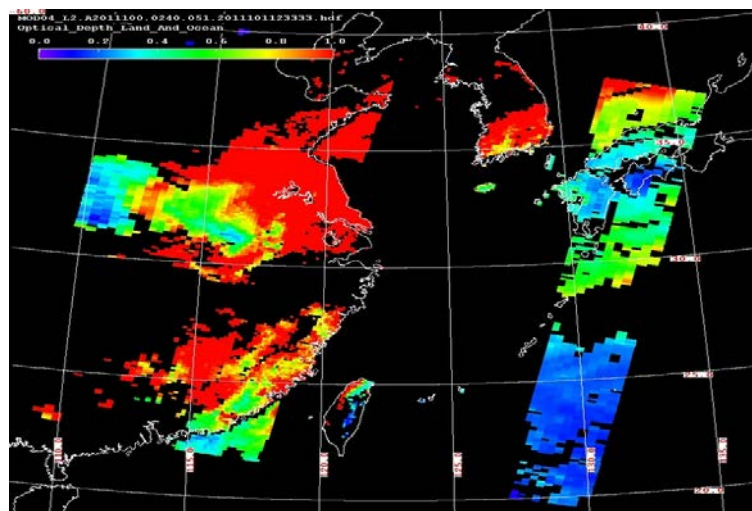
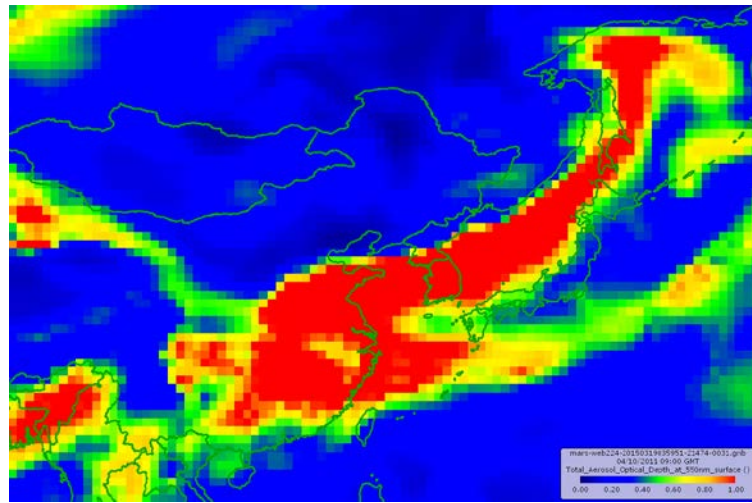
The MACC re-analysis data is considered as reliable and widely used to estimate the anthropogenic pollution or radiative forcing of aerosols.

We compared the AOD observed by MODIS and retrieved by MACC re-analysis data.

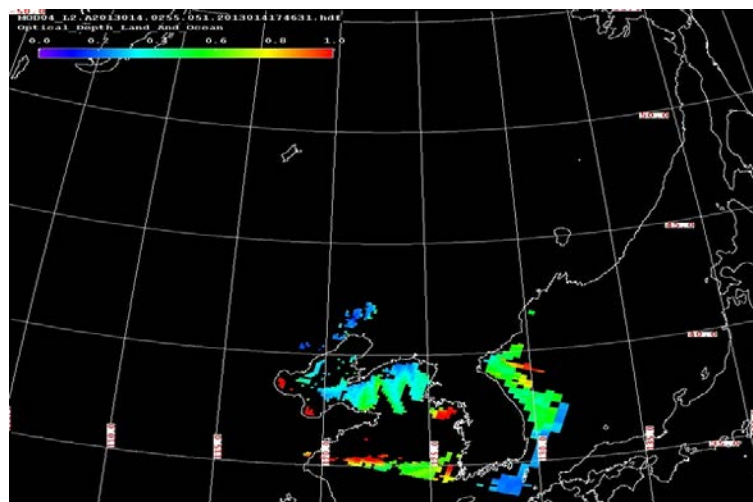
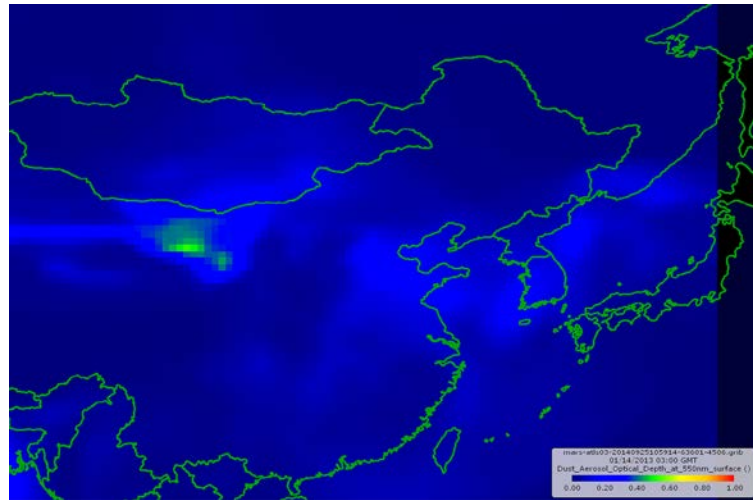
e.g.

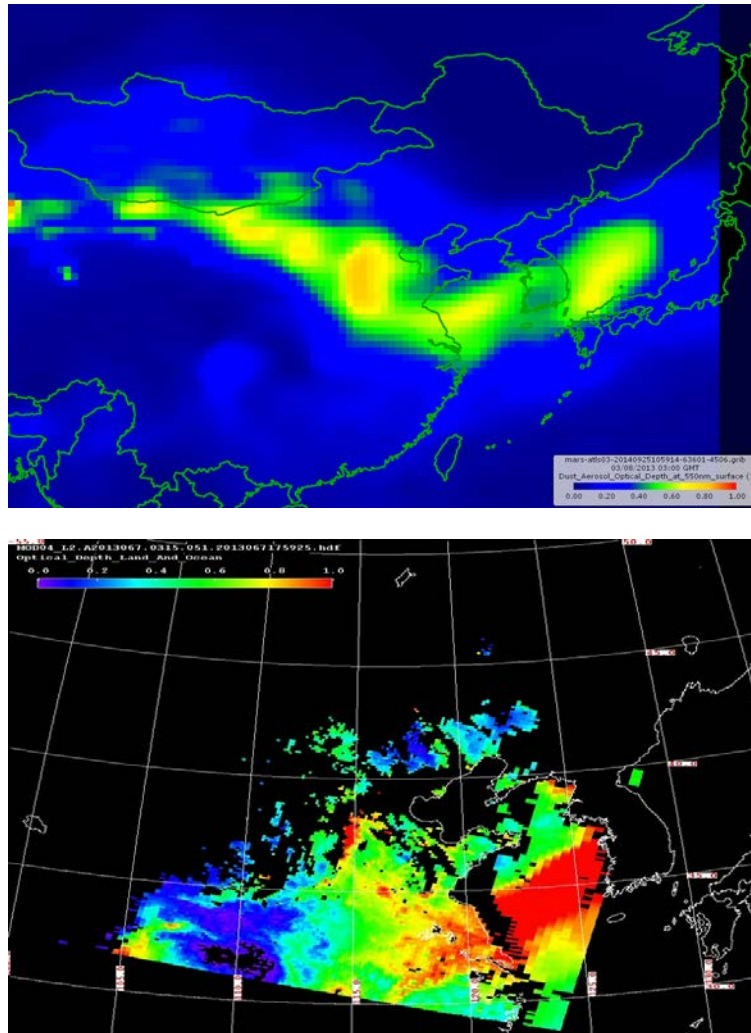
- More Polluted case





- Less Polluted case





as shown above, the total AOD distribution derived from MACC re-analysis and MODIS are in good agreement for each case (Less polluted, More polluted)

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.

In case of any misleading in using MACC re-analysis data as you pointed out, the statement

"The reliability of inferring AOD of pollution from MACC re-analysis is validated by comparing it to results from AERONET sunphotometer measurements. MACC model is widely used to estimate AOD of pollution (Bellouin et al., 2013; Cesnulyte et al., 2014)." will be added before line 1 of 3391 in revised manuscript.

The reference "Cesnulyte, V., Lindfors, A., Pitkänen, M., Lehtinen, K., Morcrette, J.-J., and Arola, A.: Comparing ECMWF AOD with AERONET observations at visible and UV wavelengths, *Atmospheric Chemistry and Physics*, 14, 593-608, 2014." will be added.

pp. 3396, l. 9-21: Why have you chosen the transport time as a critical factor for pollution? Most pollution occurs in the PBL so that predominantly dust plumes at low-levels should be polluted. Here, winds are weak resulting in slow transport compared to upper levels where strong winds result in a quicker transport. Using the transport time as a measure of pollution magnitudes is due to these wind differences misleading.

Response: We agree with referee's comments. Polluted Asian dust that transported slowly over the polluted areas than in pure Asian dust that transported quickly from the dust source region as presented in Sugimoto et al., 2015 (Sugimoto, N., Nishizawa, T., Shimizu, A., Matsui, I., and Kobayashi, H.: Detection of internally mixed Asian dust with air pollution aerosols using a polarization optical particle counter and a polarization-sensitive two-wavelength lidar, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 150, 107-113, 2015) and the wind is moved quickly in the upper level as you mentioned.

As a results, The dust optical properties in upper level (Case I) of course are less influenced by pollution because the dust layer in this height is moved quickly as the scatters of Case I are distributed within 20h. In contrast with this, the spent time is shorter and the dust optical

properties in lower level (Case II) are more influenced by pollution as scatters of Case II are distributed to 50h.

The statement "We find a maximum value of 0.3 for δ_p at 532 nm. On average, the depolarization decreases with increasing residence time over China. However, this dependence differs with respect to the height above ground of dust layers. The change of the depolarization ratio of dust layers travelling above 3 km above ground seems less dependent on the residence time over a given area. We believe that short residence times (fast transport to Korea, 20 hours or less) reduces the chances that pollution may mix with dust, particularly if dust travelled below 3 km above ground. In contrast, longer residence times (slow transport to Korea, >50 hours) of the dust plumes may have increased the chances that pollution mixed with dust if dust travelled below 3km height above ground." will be added.

Besides this common phenomenon, we attempt to show the correlation between changes in optical properties and residence time in polluted regions (Particularly, the changes of dust optical properties in lower altitude because the dust properties in high altitude is less influenced as described)

We first described the differences of optical properties with respect to the height with the results could be commonly explained as the pollution mix with dust commonly occurred in lower altitude than high altitude.

And, we were supposed to emphasize that the correlation between A_β and transport time were differed as we expected and the correlation between S and transport time were differed as we expected.

The possible reasons were described as "With regard to S at 355 nm and 532 nm we

find a maximum value of approximately 75 sr which drops to approximately 40 sr for slow transport. Again, we see that for plumes below 3 km height above ground transport time seems to matter. S drops with increasing transport time. For the case of plumes above 3 km, i.e. dust that likely is not too much affected by mixing with anthropogenic pollution, the lidar ratios do not seem to depend on transport time. This result may however again be caused by the fact that transport times to Korea are comparably short.

We further investigated these results. We initially assumed that A_{β} either should increase with transport time or does not drop significantly for pollution that travels near the ground as there should be a higher share of small anthropogenic pollution particles in the dust plume (large particles). This opposite behaviour may be caused by the state of mixing, i.e., pollution particles attach to the dust particles, thus increasing their mean size. Hygroscopic growth of particles attached to dust may further contribute to the increase of mean size. One point that complicates this interpretation is that A_{β} does not only depend on particle size but also on particle shape and the real and imaginary part (scattering and absorption) of the particles. With regard to S we also expected that S would increase with increasing transport times. If the particles travel at low height above ground more anthropogenic pollution should mix with dust. The decrease of S however suggests an increase of particle size and a decrease of the light-absorption capacity. Hygroscopic particle growth, i.e. increase of mean particle size and decrease of light-absorption by uptake of water might be responsible for this behavior."

After all, the figure 8 and the explanation of correlation between transport time and variation of dust optical properties are shown to deliver this information.

Minor comments:

pp. 3384, l. 6: "comparably strong" Please add what you relate to

Response: The dust has strong light-absorption capacity when it contains Fe_2O_3 , Al_2O_3 , SiO_2 , CaCO_3 , MgCO_3 .

The statement "Its light-absorption capacity is strong in the ultraviolet regions of the solar spectrum (Jacobson 2012). The light-absorption capacity depends on the proportions of Fe_2O_3 , Al_2O_3 , SiO_2 , CaCO_3 , $\text{MgCO}_3(\text{s})$, clays, and other substances."

The reference "Jacobson, M. Z.: Investigating cloud absorption effects: Global absorption properties of black carbon, tar balls, and soil dust in clouds and aerosols, *Journal of Geophysical Research: Atmospheres* (1984–2012), 117, 2012." will be added in revised manuscript.

pp. 3384, l. 10-12: Refer to Fig. 1 for the locations of the deserts.

Response: The reference "(Fig.1)" will be added as

"Central East Asia has large desert regions. Asian dust particles that originate from the Taklamakan desert in west China and the Gobi desert in Mongolia and northwest China (Fig. 1) influence the regional climate over East Asia"

pp. 3384, l. 9: "field" better speak of balance or budget.

Response: It will be changed as "balance" in revised manuscript.

pp. 3386, l. 11: “tracer” better to say proxy

Response: It will be changed in manuscript.

pp. 3388, l. 11: Add that these are lower thresholds for the identification of dust, i.e. dust is identified when the value exceeds the thresholds.

Response: The statement “In this study 0.08 was considered as threshold value of δ_p to identify dust.” will be added in revised manuscript.

pp. 3388, l. 18: Introduce S by name and refer to Fig. 2 d.

Response: The statement “The values of S at 355 and 532nm in layer I are 64_4 sr and 66_4 sr, respectively. The values of 20 S at 355 and 532 nm in layer II are as low 55_4 sr and 55_3 sr, respectively.” will be changed in the revised manuscript as

“The values of the S in layer I are 64 ± 4 sr and 66 ± 4 sr at 355 and 532 nm, respectively. The values of the S in layer II are as low 55 ± 4 sr and 55 ± 3 sr at 355 and 532 nm, respectively, see Fig. 2d”

pp. 3391, l. 6-7: The choice of words is misleading as I assume you did not run MACC yourself. Provide reference.

Response: The statement “The re-analysis data from MACC model are downloadable at the web page of ECMWF (<http://apps.ecmwf.int/datasets/data/macc-reanalysis/>).” will be added

at line 7 of 3391 in revised manuscript.

pp. 3394, l. 13-23: Add arguments/discuss why the values differ for mixed and likely-pure dust (more absorption, more scattering?)?

Response: We agree with referee's comments. In order to state reason of the difference between values for mixed dust and likely-pure dust, the statement

"Lower values of δ_p represent the dominance of spherical particles, i.e. the presence of urban pollution. High values of A_β indicate that small particles dominate in the lower altitude level. The high lidar ratio also indicates the presence of urban pollution which tends to be more light-absorbing (Müller et al., 2007)."

will be added in revised manuscript.

pp. 3394, l. 27: "below or below" replace one with above

Response: It has been changed.

pp. 3395, l. 2-4: The boundary layer height over mid-latitudes does rarely reaches a depth of 3 km. The paper by Basha et al. (2009) investigates a tropical station at 13.5N, which does not apply for conditions in Korea and China.

Response: The Noh et al. (2007) and Xie et al. (2014) have found the range of PBL experimentally with lidar observation as 2.5 km – 3 km in Korea and China. The statement

"This height of 3 km is also in relatively good agreement with the average height of planetary boundary layers." will be replaced with the statement as

"We used 3 km height above ground for the classification. The height of 3 km is reported as the planetary boundary layer. Pollutants emitted at the surface predominantly stay in the planetary boundary layer (Noh et al., 2007; Xie et al., 2015)."

The reference "Xie, C., Zhao, M., Wang, B., Zhong, Z., Wang, L., Liu, D., and Wang, Y.: Study of the scanning lidar on the atmospheric detection, Journal of Quantitative Spectroscopy and Radiative Transfer, 150, 114-120, 2015." will be added.

pp. 3395, l. 14: Spelling of HYSPLIT

Response: Typo, it will be modified in the revised manuscript.

pp. 3395, l. 28: "as" us

Response: Typo, It has been changed.

Figure 1: If there are other loess regions than the Loess Plateau, please name them or speak of a single loess region in the caption.

Response: The loess plateau is most representative loess region in China, however the loess are also distributed around Manchuria.

The caption for figure 1 will be changed as

"Figure 1. Map of the desert regions (Taklimakan desert, Gobi desert, Badain Jaran desert, Ordos Desert, Inner Mongolia plateau, and Manchuria) and loess regions (Loess Plateau and Manchuria). The location of some major cities (Beijing and Shanghai) and industrialized areas of China (Hebei, Shandong, Henan, and Zhejiang province) is also shown, MRS.LEA is located in Gwangju, Korea."

Authors' response to Referee #4' 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 #4

General comments:

This paper describes dependence of optical properties of Asian dust observed with a multi-wavelength Raman lidar in Gwangju, Korea on the dust transport path. The paper presents interesting results on mixing of Asian dust and air pollution particles, and it merits publication in ACP. However, revisions are recommended.

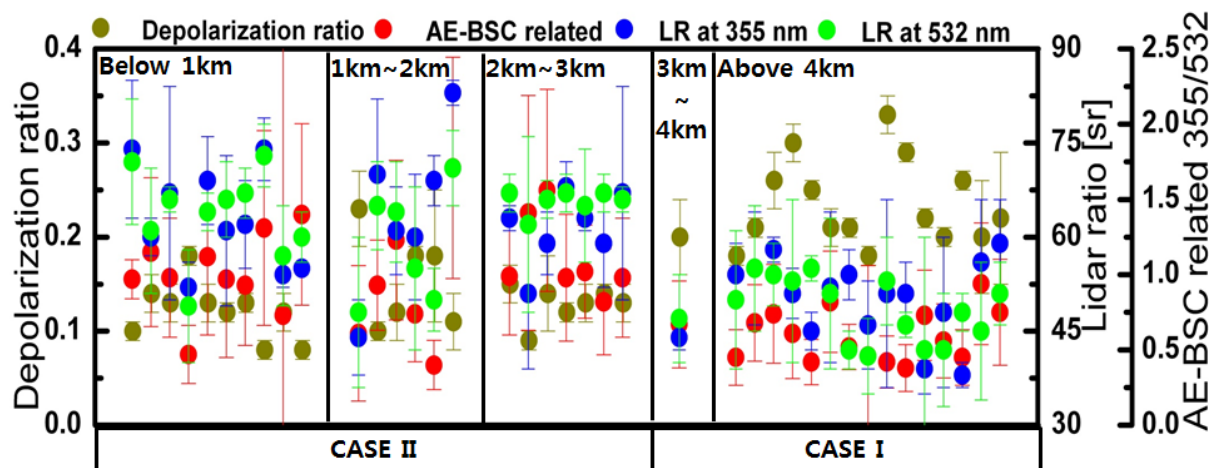
Specific comments:

Fig. 8: There are no explanations of the solid lines in Fig. 8. They look misleading and should be removed.

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.

An analysis in a multi-parameter space would provide more information. In other words, it would be better to give the depolarization ratio, AE-BSC, lidar ratios at 532 nm and 355 nm, at the same time for each data point. It would be useful, for example, to put numbers to typical data points in Fig. 7 (and/or in Fig. 8) for identifying internally mixed cases, though it looks most of the data can be explained by external mixing. Internal mixing of Asian dust and air pollution particles is discussed in a recent JQSRT paper (Sugimoto et al., J. Quantitative Spectroscopy & Radiative Transfer 150 (2015) 107–113, <http://dx.doi.org/10.1016/j.jqsrt.2014.08.003>) They discussed using a depolarization ratio vs. backscattering color ratio plot. This paper might provide further information including the lidar ratios.

Response: The figure 7 will be changed reflecting your comment as below (the optical properties of Asian dust we observed is presented at the same time)



The corresponding values will be given in table 4. The caption for figure 7 has been changed as "Figure 10. (top panel) (a) transport path and classification of East Asian dust layers with respect to (b) their altitude above ground when they passed over industrial regions of China. (bottom panel) transport path and corresponding altitude of Asian dust layers are distinguished by color. (black: 0 km – 1 km; green: 1 km – 2 km; purple: 2 km – 3 km; blue: 3 km – 4 km; red: above 4 km). Scatter plots of the linear particle depolarization at 532 nm (dark yellow), the backscatter-related Ångström exponent (355/532 nm wavelength pair, red), the lidar ratio at 355 nm (blue), the lidar ratio at 532 nm (green) in dependence of the 5 altitude categories (c). The height of the Asian dust layers above ground is separated by vertical lines. Case I included the layers from 3 – 4 km and above 4 km. Case II includes the layers from 0 - 1 km, from 1-2km, and from 2-3km height above ground.

"

To explain the state of mixing (internal or external)

The statement "The optical properties of Asian dust layer observed in our study reflect mixtures between different aerosol types. We notice that these variations of the optical

properties of Asian dust layers may not only result from external mixing. Hygroscopic growth, aging and deposition during transport, and internal mixing might be also affect dust properties (Burton et al., 2014). The interpretation of the mixing state of Asian dust is a challenging task. The mixing state depends on many variables which are poorly known. Sugimoto et al. (2015) tried to identify the mixing state of Asian dust (internal mixing or external mixing) by using analytical relationships inferred from lidar observation. However, we will not go into details here. We assume that most of the Asian dust observed in this study was externally mixed." will be added in revised manuscript

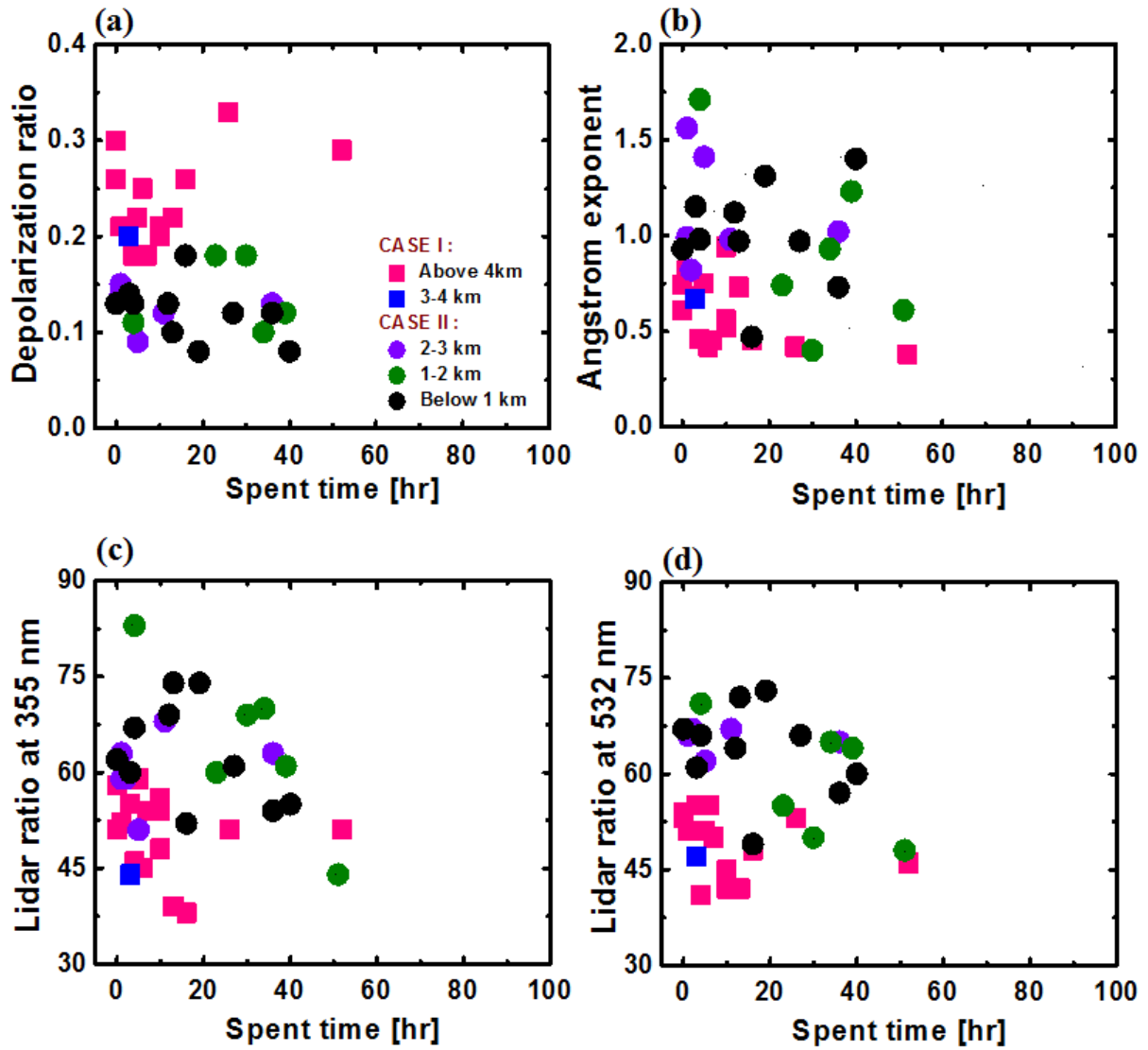
The references to support this statement will be added in the revised manuscript as

Burton, S., Vaughan, M., Ferrare, R., Hostetler, C.: Separating mixtures of aerosol types in airborne High Spectral Resolution Lidar data, *Atmospheric Measurement Techniques*, 7, 419-436, 2014.

Sugimoto, N., Nishizawa, T., Shimizu, A., Matsui, I., and Kobayashi, H.: Detection of internally mixed Asian dust with air pollution aerosols using a polarization optical particle counter and a polarization-sensitive two-wavelength lidar, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 150, 107-113, 2015.

It would be better to plot the data in the lower heights in the foreground in Figs. 7 and 8.

Response: Figure 7 will be changed as shown earlier and figure 8 also will be changed reflecting your comment as below, (the data in lower height has been moved in the foreground)



The depolarization ratio is defined by $(P_{\text{perpendicular}} / (P_{\text{perpendicular}} + P_{\text{parallel}}))$ in this paper. However, some of the authors of the cited papers use the $(P_{\text{perpendicular}} / P_{\text{parallel}})$ definition. It seems they are mixed in the manuscript. More accurate descriptions are required, and the values must be converted if needed.

Response: We agree with the reviewer. We used $P / (P + S)$ as depolarization parameters and some of the author we cited used (P / S) . As shown in Cairo et al., (1999) "Comparison of various linear depolarization parameters measured by lidar" published in optical society of America, These two parameter has the advantage of requiring little processing on the raw

signals. And the values of different depolarization ratio are not that large however, more descriptions are needed .

The statement "The δ can be also defined as $P_{\perp}/P_{\parallel}$ (Cairo et al., 1999). We calculated the δ by using both definitions and compared the difference between the derived values. The results from each individual definition agree within the uncertainty of our depolarization ratio measurements (Tesche et al., 2009; Shin et al., 2013)." will be added from line 148 to 153 of the revised manuscript to prevent any misleading.