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 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 #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 $\boldsymbol{S}$ = 48.6 sr at 355 nm and $\boldsymbol{S}$ = 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 $\boldsymbol{S}$ value less than 50 sr at 351 nm for a Saharan dust layer. Values of $\boldsymbol{S}$ 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 $\boldsymbol{Å}_{\beta}$) 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 $\boldsymbol{Å}_{\beta}$ 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 $\boldsymbol{Å}_{\beta}$ for a mixture of mineral dust with urban haze. Values of 0.8-1.4 for $\boldsymbol{Å}_{\beta}$ 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 $\boldsymbol{\delta}_{\mathbf{p}}$ = 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 $\boldsymbol{\delta}_{\mathbf{p}}$ 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 $\boldsymbol{\delta}_{\mathbf{p}}$ = 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 $\boldsymbol{\delta}_{\mathbf{p}}$, 38-83 sr for $\boldsymbol{S}$ at 355 nm, 41-73 sr for $\boldsymbol{S}$ at 532 nm, and 0.38-1.71 for $\boldsymbol{\mathring{A}}_{\boldsymbol{\beta}}$. The maximum value of $\boldsymbol{\delta}_{\mathbf{p}}$ is 0.33 at 532 nm. The minimum values of $\boldsymbol{S}$ at 355 nm and 532 nm are 38 sr and 41 sr, respectively. The minimum value of $\boldsymbol{\mathring{A}}_{\boldsymbol{\beta}}$ is 0.38. This maximum value of $\boldsymbol{\delta}_{\mathbf{p}}$ and the minimum values of $\boldsymbol{S}$ at 355 nm and 532 nm and $\boldsymbol{\mathring{A}}_{\boldsymbol{\beta}}$ are similar to the values of optical properties for pure dust particles.

76% of $\boldsymbol{\delta}_{\mathbf{p}}$ at 532 nm are located in the range between 0.08 and 0.20. 53% of the values of $\boldsymbol{S}$ at 355 nm are in the range between 60 sr and 85 sr. 47% of the values of $\boldsymbol{S}$ at 532 nm vary between 60 sr and 75 sr. The Ångstöm exponents ($\boldsymbol{\mathring{A}}_{\boldsymbol{\beta}}$) 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 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 can not 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, 5 J., Morcrette, J.-J., and Boucher, O.: Estimates of aerosol radiative forcingfrom the MACC re-analysis, Atmos. Chem. Phys., 13, 2045–2062, doi:10.5194/acp-13-2045-2013, 2013.

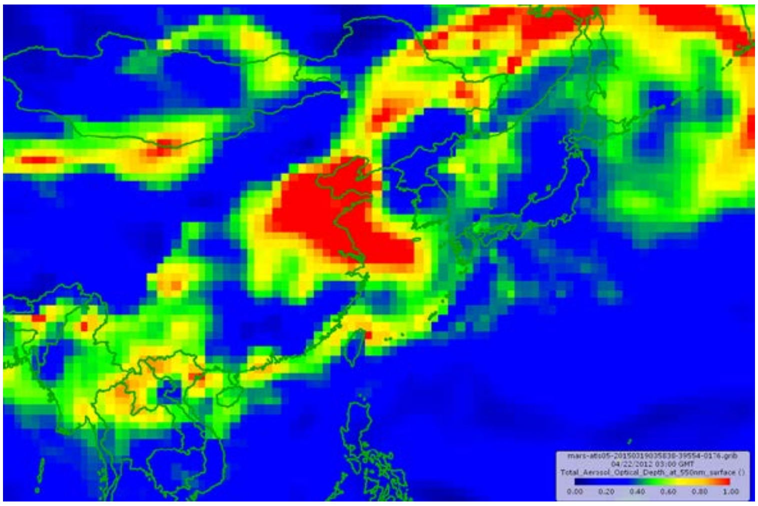
In the study “MACC Work Package G-AER 3 Monitoring of aerosol direct and indirect forcing” by Nicolas Bellouin and Johannes Quaas, Karsten Peters stated the data from MODIS has tendency to overestimate and the bias is corrected in the MACC re-analysis.

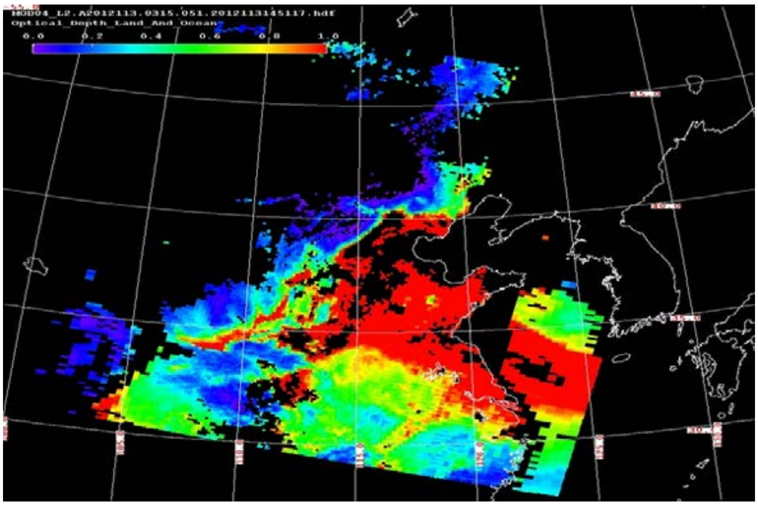
The MACC re-analysis data is considered as reliable and widely used to estimate the anthropogenic pollution or radiative forcing of aerosols.

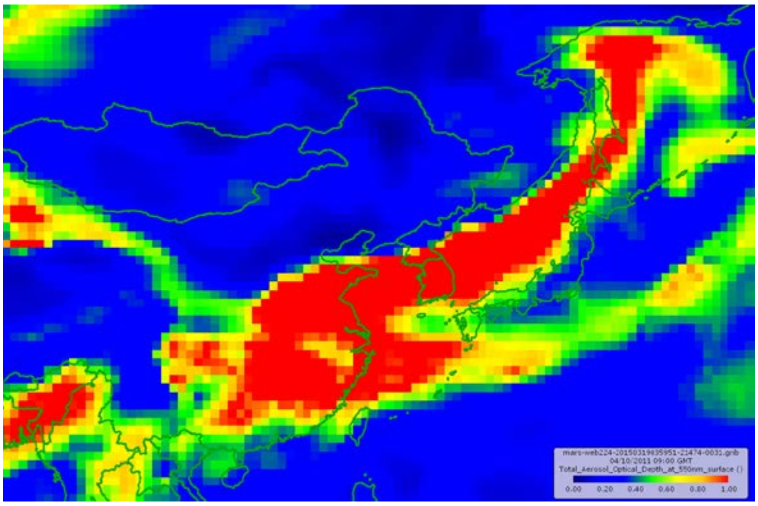
We compared the AOD observed by MODIS and retrieved by MACC re-analysis data.

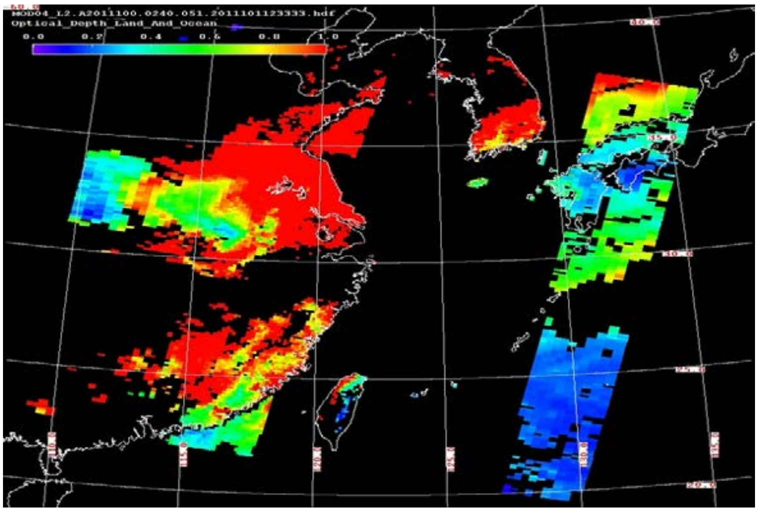
e.g.

- More Polluted case

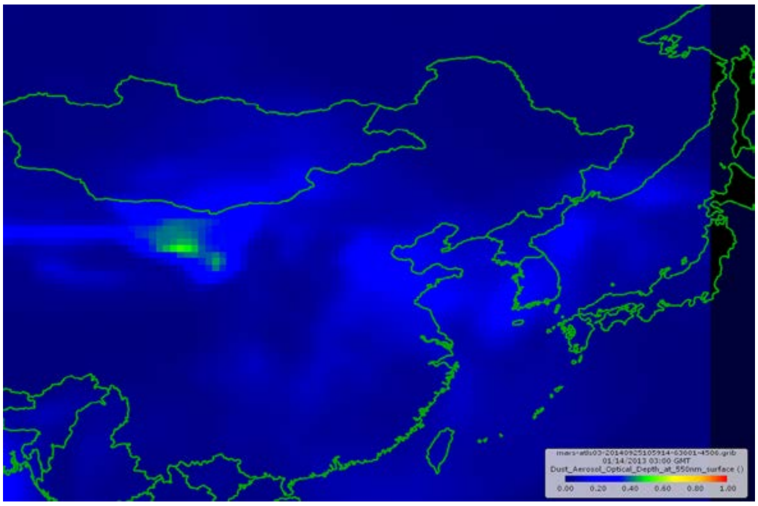


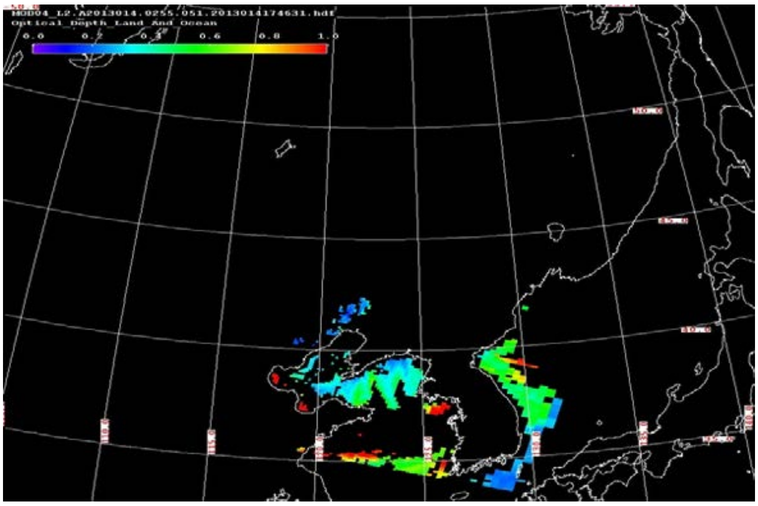


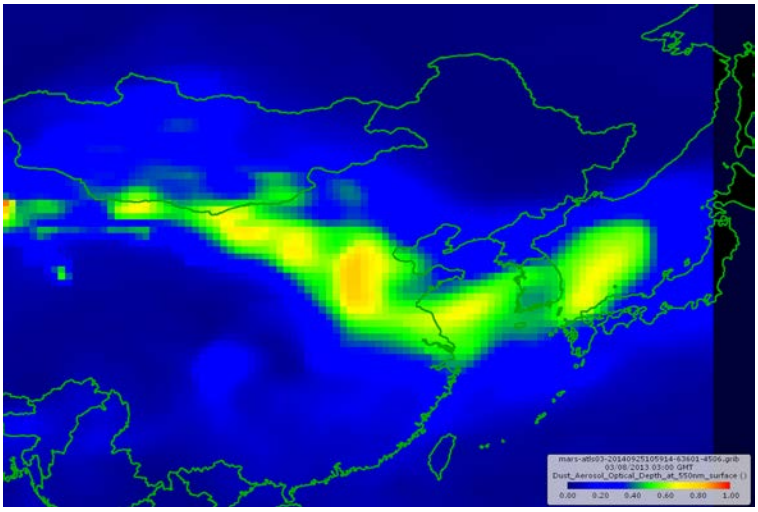

04/10/2011 09:00 GMT
Total_Aerosol_Optical_Depth_at_550nm_surface ()
0.00 0.20 0.40 0.60 0.80 1.00

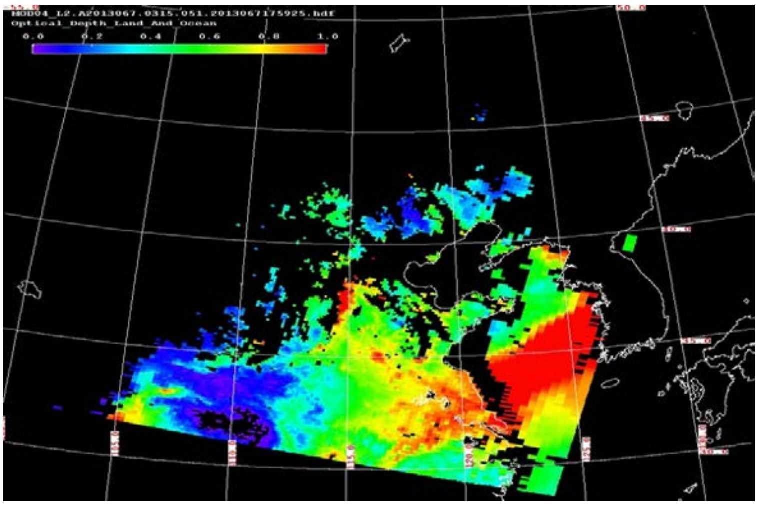

MOD04_L2.A2011100.0240.051.2011101123333.hdf
Optical_Depth_Land_And_Ocean
0.0 0.2 0.4 0.6 0.8 1.0

- 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 suphated 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 choosen 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 lever 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 $\boldsymbol{\delta}_{\mathbf{p}}$ 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 $\mathring{A}_{\beta}$ 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 $\mathring{A}_{\beta}$ 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 $\mathring{A}_{\beta}$ 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, $CaCO_3$, $MgCO_3$(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 of20 *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 $\boldsymbol{\delta_p}$ represent the dominance of spherical particles, i.e. the presence of urban pollution. High values of $\boldsymbol{\AA_\beta}$ 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."