Reply to the comments of anonymous reviewer #1 on manuscript entitled "Variability of depolarization of aerosol particles in Beijing mega city: implication in interaction between anthropogenic pollutants and mineral dust particles"

The authors appreciate very much for reviewing our manuscript and your insight comments. As suggested, we carefully revised the manuscript thoroughly according to the valuable advices, as well as proof-read the manuscript to minimize typographical, grammatical, and bibliographical errors.

General Comments:

Tian et al. discuss aerosol particle polarization properties from November 29, 2015 to July 29, 2016 at Beijing. The dataset presented is interesting, particularly in showing particle polarization in anthropogenic pollution, mixed type pollution and pure Asian dust cases. Using an optical particle counter equipped with a polarization detection module is a new method to study mixing processes. However, the discussion in the current draft is insufficient for these data and results: a number of results/conclusions are not obtained from lab experiment or data prove, just derivate from published paper. I believe the article therefore should add data prove or lab experiment, and require major revisions.

Reply: We fully agree with the reviewer's suggestions that simultaneous measurement of chemical compositions of particles and lab experiment evaluations will benefit the understanding of the aerosol's physical changes during mixed pollution. The change in aerosol's depolarization ratio can only represent that overall characteristics of the surface of dust aerosols, not the chemical reactions on it. As the reviewer suggested, we add more observational and experimental results in laboratory to clarify the chemical process happened during mix pollution, and the depolarization ratio of particles with meteorological factors, $PM_{2.5}$ and PM_{10} concentrations, and aerosol chemical compositions are carefully discussed. To respond to the reviewer's major concerns, we made thoroughly revisions and corrections according to all the insight comments of the reviewers. Besides, more crucial/supporting information and analysis will be added in the revised manuscript, as follows:

(1) A detailed description and specification of calibration system of POPC and calibration results in this study will be added in the revised manuscript.

(2) Comparison between observation and the simulation result with T-matrix method on randomly-oriented Voronoi aggregation and elongated ellipsoid particles will be added.

(3) A comparison result of measurement of size-resolved number concentration of particles between POPC and KC52 optical particle counter will be added in the revised manuscript.

(4) Comparison of observation results between POPC measurement and LIDAR observation during several dust events in Beijing will be discussed. The cross validation on the dust events provide more direct evidence of variation in the depolarization properties of dust particles.

Besides, to make the conclusions more pertinent to this study, we revised the expression and concise the discussion about specific chemical composition and reaction. Our conclusions were useful for understanding the long-term depolarization property of aerosols in East Asia, interaction between anthropogenic pollutions and mineral dust in East Asia and their impacts on local air quality, and the sensitive relation between particle morphology and meteorological parameter. This long-term study in Beijing motivate revisit on decades of Lidar data in dust-pollution interactions, and it indicates the necessity of a reliable optical model of internally mixed polluted dust for a detailed analysis of polarization remote sensing observations.

The initial purpose of this observational study is to investigate the long-term morphology properties of particles in Beijing because the North China region has long experienced seasonal haze and dust events due to the special geographical location and industrial development stage. As far as we know, due to limit of observation techniques, there was no studies reported on long-term variability of morphological characteristics of aerosols in north China till now. Alternatively, depolarization ratio of single particle is an applicable parameter indicating of general irregularity of aerosol particles. For this reason, an eight-month field measurement was performed at an urban site in Beijing megacity, we detected single particle depolarization ratio

properties using a new developed beach-top instrument. Take advantage of meteorology/pollution data and trajectory analysis during the same period, the relationship between morphology change of particles and meteorological factors, especially water content in the atmosphere, was investigate. As pointed out, electro-microscopy inspection and composition results would provide direct evidence of chemo-physical changes of the particles, we will add more supporting information in the revised manuscript. The knotty point is that, single particle-based laboratory inspections need passive samples with variable time scales of 1 min to several days, it is difficult to have enough samplings to represent the long-term variability of aerosols statistically. We will follow reviewer's advice to conduct single-particle based electro-microscopic study in our next study. Chemical compositions of particles is helpful for understanding the possible chemical reaction during observation period. Unfortunately, we didn't have concurrent measurement during the experiment.

According to long-term real-time measurements of aerosol particle composition in Beijing in previous study, the seasonal variations of secondary inorganic aerosol (SIA, i.e., sulfate C nitrate C ammonium) concentrations were not significant, contributions of SIA were observed in summer (57 – 61 %) and in winter (43 – 46 %) in fine mode aerosols (Sun et al., 2015). Although we did not have *in situ* measurement in this study in Beijing, a high $PM_{2.5}/PM_{10}$ ratio still indicates that the components of soluble inorganic salts was greatly possible to be relatively high, and in the case of high humidity. Therefore, morphology and corresponding depolarization ratio of particles should be affected. As expected, I did find a similar correlation between the ratio of $PM_{2.5}/PM_{10}$ and the depolarization ratio of coarse-mode aerosols, proving the great possibility that the inorganic salts might be trapped by large particles, or the occurrence of reaction between reactive gas and alkaline dust particles. At present, depolarization ratio is deemed as the most applicable method to rapidly or simultaneously distinguish types or mixing state of pollutions. Such characteristic are widely used by satellite-borne remote sensing and ground-based Lidar observations (Burton et al., 2012; Shimizu et al., 2004a; Shimizu et al., 2004b). In this study, combining analysis with LIDAR measurement, we hope to find an alternative and relatively simple indicator, such as the ratio of $PM_{2.5}/PM_{10}$, to estimate the possibility of morphology change of particles in the coarse-mode. In the manuscript, we revised the expression and weaken the discussion about specific chemical composition and reaction. In our next research, we would like to follow reviewer's suggestions to perform comprehensive measurements on the dependence of both morphology properties and chemical information of particles in different mixing state.

Sun, Y. L., Wang, Z. F., Du, W., Zhang, Q., Wang, Q. Q., Fu, P. Q., Pan, X. L., Li, J., Jayne, J., and Worsnop, D. R.: Long-term real-time measurements of aerosol particle composition in Beijing, China: seasonal variations, meteorological effects, and source analysis, Atmos Chem Phys, 15, 10149-10165, 10.5194/acp-15-10149-2015, 2015.

Burton, S. P., Ferrare, R. A., Hostetler, C. A., Hair, J. W., Rogers, R. R., Obland, M. D., Butler, C. F., Cook, A. L., Harper, D. B., and Froyd, K. D.: Aerosol classification using airborne High Spectral Resolution Lidar measurements – methodology and examples, Atmospheric Measurement Techniques, 5, 73-98, 10.5194/amt-5-73-2012, 2012.

Shimizu, A., Sugimoto, N., Matsui, I., Arao, K., Uno, I., Murayama, T., Kagawa, N., Aoki, K., Uchiyama, A., and Yamazaki, A.: Continuous observations of Asian dust and other aerosols by polarization lidars in China and Japan during ACE - Asia, Journal of Geophysical Research: Atmospheres, 109, 2004a.

Shimizu, A., Sugimoto, N., Matsui, I., Arao, K., Uno, I., Murayama, T., Kagawa, N., Aoki, K., Uchiyama, A., and Yamazaki, A.: Continuous observations of Asian dust and other aerosols by polarization lidars in China and Japan during ACE-Asia, Journal of Geophysical Research-Atmospheres, 109, 10.1029/2002jd003253, 2004b.

Specific Comments.

1. *Page 2, Line 2-3, only consumption of fossil fuel lead serious air pollution in East Asia? How about vehicle exhaust?*

   Reply: We agree to the reviewer's comments. On-road vehicle exhaust plays an important role in formation of urban air pollution. We revised the expression in the manuscript as following, "Rapid economic growth and urbanization processes in East Asia has caused serious air pollution in the past decades owing to the substantial consumption of fossil fuel. Anthropogenic emissions (industrial, traffic, residential etc.) emits substantial amount of pollutant gases (SO_2, NO_2, NH_3,

VOCs etc.) and primary aerosols (Akimoto, 2003;Akimoto et al., 2006;Kurokawa et al., 2013;Li et al., 2017), resulting in formation of $PM_{2.5}$."

Akimoto, H.: Global air quality and pollution, Science, 302, 1716-1719, 10.1126/science.1092666, 2003.

Akimoto, H., Ohara, T., Kurokawa, J.-i., and Horii, N.: Verification of energy consumption in China during 1996–2003 by using satellite observational data, Atmospheric Environment, 40, 7663-7667, 10.1016/j.atmosenv.2006.07.052, 2006.

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., and Akimoto, H.: Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2, Atmos Chem Phys, 13, 11019-11058, 10.5194/acp-13-11019-2013, 2013.

Li, M., Zhang, Q., Kurokawa, J.-i., Woo, J.-H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H., and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP, Atmos Chem Phys, 17, 935-963, 10.5194/acp-17-935-2017, 2017.

2. *Page 2, Line 6, mineral dust particles only originated from natural sources? In the manuscript 3.4 part, the authors said emissions in famous mining areas in China can result in a huge amount of dust.*

Reply: The statement in section 3.4 made a misleading. What we want to express is that mining areas produce huge amount of pollutant gases SO_2, CO, as well as NO_x related to vehicle exhaust in North China. In case C, there was a convergent air mass from natural dust sources (desert and Gobi) and mining areas in Beijing, according to FLEXPART trajectory analysis. It lead to a huge amount of dust mixed with pollutant-rich gaseous atmosphere in the Beijing area. We will revise the expression in the revised manuscript in section 3.5.1. As the reviewer pointed out, mineral dust particles in city environment might also be emitted from on-road vehicle and construction dust. These fugitive dust usually have a large diameter and a short lifetime due to rapid gravity settlement velocity. The sentence has been revised as "Mineral dust particles in the atmosphere also have a detrimental impact on air quality and human health such as reducing visibility, increase respiratory morbidity."

3. *Page 3, Line 3, the authors should add more descriptions about depolarization of aerosol particles.*

Reply: We will add more descriptions about depolarization of particles. Volume depolarization ratio of particle groups is widely used in LIDAR detection to distinguish the types of aerosol particles. The volume depolarization information of target air parcel just represents an overall depolarization. In this study, we use a new instrument to measure real-time variations in single particle depolarization ratio. This measurement can avoid the influences from the presence of substantial amount of small spherical particles. As mentioned, direction of polarization of scattering light was identical with the incident light for spherical particle; however, for the non-spherical particles, the direction of polarization of light will be deviated. According to such phenomenon, mineral dust and anthropogenic pollution dominant particles can be distinguished. Sugimoto et al. (2015) found that backscattering depolarization ratio in polluted dust was smaller compared to pure Asia dust, Pan et al. (2017) reported the vital role in observed depolarization ratio decrease that calcium nitrate played via coating process and hygroscopic growth on the surface of dust particles. In a word, single particle-based depolarization ratio is applicable for quantitatively investigating the evolution of the mixing of dust particles during their transport. As suggested by the reviewer, we will add and clarify the relevant explanation about depolarization ratio in the revised manuscript.

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 & Radiative Transfer, 150, 107-113, 10.1016/j.jqsrt.2014.08.003, 2015.

Pan, X., Uno, I., Wang, Z., Nishizawa, T., Sugimoto, N., Yamamoto, S., Kobayashi, H., Sun, Y., Fu, P., Tang, X., and

Wang, Z.: Real-time observational evidence of changing Asian dust morphology with the mixing of heavy anthropogenic pollution, Sci Rep, 7, 335, 10.1038/s41598-017-00444-w, 2017.

4. *Page 3, Line 4-5, the authors said studies on the polarization characteristics are necessary, how about depolarization characteristics?*

Reply: To avoid misleading, we will use depolarization ratio of particle in the whole context. Actually, identical laser hit particles and producing different polarized light signals. The different is caused by particle morphology and best described by the ratio of "depolarization". I will make corresponding corrections in revised manuscript.

5. *Page 3, Line 10-14, why key pollutants and meteorological data were obtained from two different sites? The key pollutants and meteorological data are the same in the two different sites?*

Reply: We select the nearest state-controlled environmental monitoring station and weather station to the observation experiment site to support this study. As known, the horizontal scale of an air mass is 10 kilometers to hundreds of kilometers, the synoptic condition of the air mass (pollutant concentration, humidity and temperature) in city scale are normally the similar (as shown in Figure 1), though differences existed due to possible local sources.

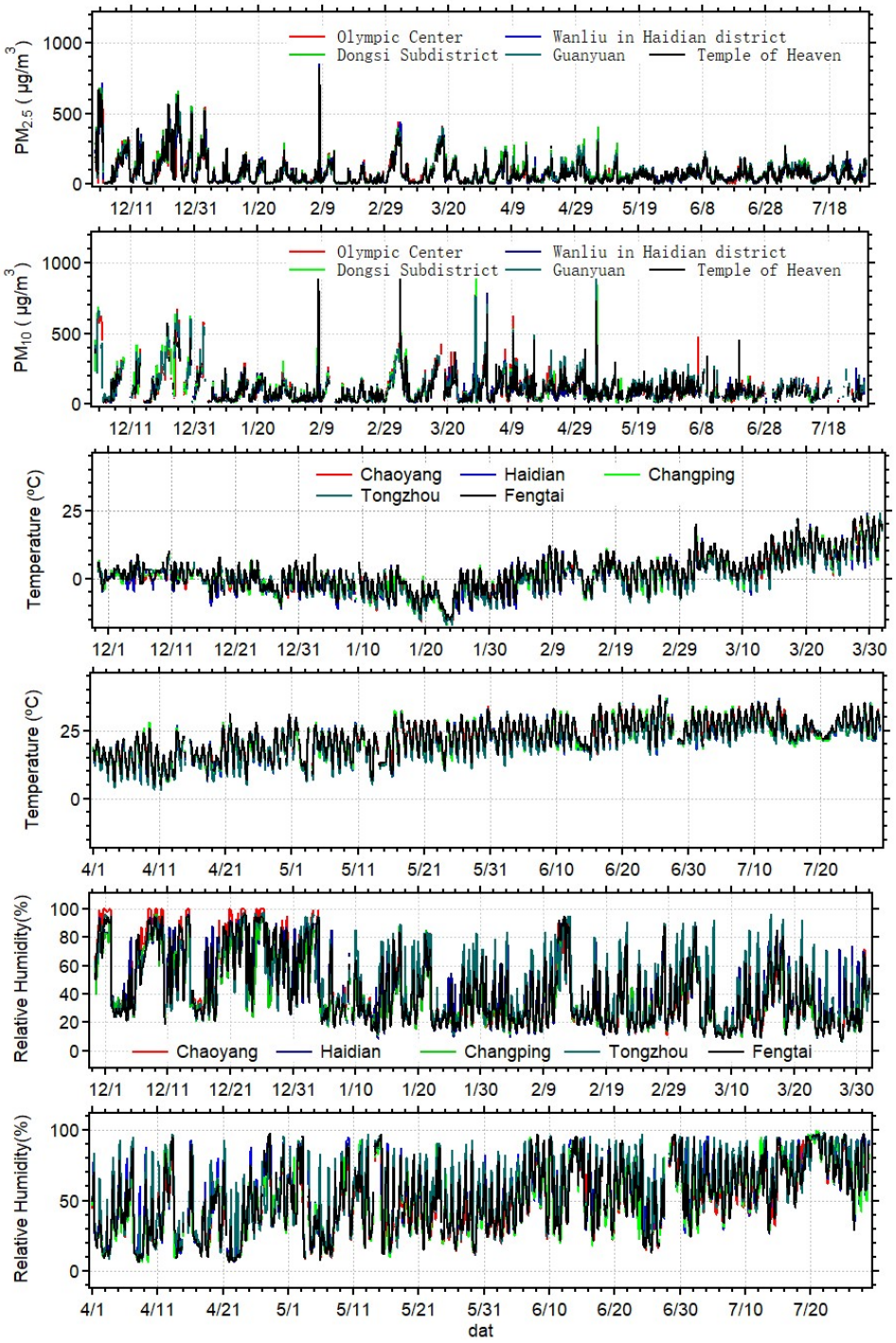

Figure 1. Time serious of particle mass concentration of $PM_{2.5}$ and PM_{10}, and meteorology factors (relative humidity and temperature) in different site in Beijing area.

6. *Page 4, Line 28-29, why 150 m above ground level was chose? Why not is 100m or 200m? Why the simulation time of the backward trajectory is 5 days, not 3 days or 2 days?*

Reply: We performed backward trajectory simulation starting from height of 150 m, because our observation is on a top of two-story building (~10 meter high above the ground), and urban canopy of Beijing city was estimated to be less than 200 m. Considering that we use trajectories to indicate synoptic transport of dust aerosols, a starting height of 150 m is chosen. As a matter of fact, the trajectories starting from 150 m and 200 m are similar, and did not change our major verdict. A weather process is generally last for 3-7 days. We choose 5 days simulation because it can better reflect the long-distance transmission of particles. 2 or 3 days simulation may only reflect the local impact, but the typically continuous dust process has a great probability to be influenced by long-distance transmission in North China.

7. *Page 5, Line 9-11, What about the sources of particle size in 1 μm – 4 μm? A number of published papers said that $PM_{2.5}$ also mostly derived from anthropogenic sources. The reference of Kurosaki and Mikami, (2003) is a bit old.*

Reply: The particle with aerodynamic diameter less than 2.5 μm is defined as fine mode, and the particle size between 2.5 – 10 μm is defined as coarse mode. As mentioned, the particles between 1 – 4 μm consisted of both anthropogenic pollutants such as sulfate, nitrate and organic matters and mineral dust particle from both nature and anthropogenic sources. Numbers of previous studies indicate that the mode diameter of dust particle in North China is normally about 5 μm. It was consistent with our measurement of size distribution which showed a broad peak between 4 – 8 μm. We would like to cite Lue et al. (2010) to support our conclusion in the revised manuscript.

Lue, Y. L., Liu, L. Y., Hu, X., Wang, L., Guo, L. L., Gao, S. Y., Zhang, X. X., Tang, Y., Qu, Z. Q., Cao, H. W., Jia, Z. J., Xu, H. Y., and Yang, Y. Y.: Characteristics and provenance of dustfall during an unusual floating dust event, Atmospheric Environment, 44, 3477-3484, 10.1016/j.atmosenv.2010.06.027, 2010.

8. *Page 5, Line 24-26, the depolarization ratios of fine mode particles in summer are not lower than any other season? Why the authors emphasized depolarization ratios of particles with an optical diameter of 2 μm are lower than any other season?*

Reply: Sorry for misleading here. The depolarization ratios of all mode particles in summer were lower than other seasons, as shown in Figure 2b-c and Figure 5 in the manuscript. This seasonal variability was more obvious for fine mode aerosols. It was because in summer, high relative humidity could lead to absorption and deliquesce process happened on soluble component in aerosols. Depolarization ratio of 2 μm particles was separately mentioned as an example. We will clarify this point in the revised manuscript as "In general, the δ value of particles in urban Beijing had prominent seasonal variability with a summer-low and spring-high pattern due to the different composition, origins of aerosols and atmospheric meteorology at the site. The averaged δ values of particles in both fine and coarse mode was highest in March 2016 (0.26) and lowest in July 2016 (0.19). This seasonal variability was much obvious in fine mode particles……"

9. *Page 6, Line 4-7, wind speed, biomass burning or other factors have no impact on high depolarization ratio? Only transport lead high depolarization ratio?*

Reply: We consent to the reviewer's comment that depolarization properties of different aerosol types are different. For instance, the soot particles from biomass burning normally have complex chain-like morphology which was distinct from spherical particles. In this study, the depolarization ratio actually reflect the type of particle: mineral dust, and anthropogenic pollutant or sea salt aerosols (Kobayashi et al., 2014). We will revise the interpretation in the manuscript as "For mineral dust-dominant episodes, the air mass mainly originated from large areas in western Mongolia and Gobi Desert, and the footprint pattern represented comparatively large dust loading in the atmosphere in spring. While for anthropogenic pollution-dominant period, air mass past through Beijing-Tianjin-Hebei region where anthropogenic emission was significantly strong. To note that, the RH along the trajectories was 13.9 % averagely during dust-dominant dominate cases (mostly in spring-time) and 87.6 % in anthropogenic pollution-dominant cases (mostly in winter-time). It means that origin of aerosol particles and their interaction with water vapor as well as consecutively heterogeneous reactions can lead to pronounced morphological changes of particles."

Kobayashi, H., Hayashi, M., Shiraishi, K., Nakura, Y., Enomoto, T., Miura, K., Takahashi, H., Igarashi, Y., Naoe, H., Kaneyasu, N., Nishizawa, T., and Sugimoto, N.: Development of a polarization optical particle counter capable of aerosol type classification, Atmospheric Environment, 97, 486-492, 10.1016/j.atmosenv.2014.05.006, 2014.

10. *Page 6, Line 7-10, have the authors ruled out other factors (such as wind speed, biomass burning)? These factors also can lead high $PM_{2.5}$.*

Reply: Yes, as pointed out by the reviewer, factors including wind speed and biomass burning also can lead high $PM_{2.5}$. Previous studies of Wang et al. (2013) and Chang and Zhan (2017) have demonstrated the necessary effect of weak wind

speed on haze formation. Chen et al. (2017) and Cheng et al. (2014) have also demonstrated the importance of biomass burning high $PM_{2.5}$ levels by observing facts. It reflected that the biomass combustion and specific synoptic situation was of great importance in variation of $PM_{2.5}$. In this study, we forces on the depolarization properties in ambient air in Beijing area, and the trajectory analysis was to explain the seasonal change of depolarization ratio. In our next study, we would like to quantify the relationship between biomass burning and $PM_{2.5}$ levels, and investigate the change and effect on depolarization ratio of ambient air.

Wang, Y., Yao, L., Wang, L., Liu, Z., Ji, D., Tang, G., Zhang, J., Sun, Y., Hu, B., and Xin, J.: Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China, Science China Earth Sciences, 57, 14-25, 10.1007/s11430-013-4773-4, 2013.

Chang, W., and Zhan, J.: The association of weather patterns with haze episodes: Recognition by PM2.5 oriented circulation classification applied in Xiamen, Southeastern China, Atmospheric Research, 197, 425-436, 10.1016/j.atmosres.2017.07.024, 2017b.

Chen, J., Li, C., Ristovski, Z., Milic, A., Gu, Y., Islam, M. S., Wang, S., Hao, J., Zhang, H., He, C., Guo, H., Fu, H., Miljevic, B., Morawska, L., Thai, P., Lam, Y. F., Pereira, G., Ding, A., Huang, X., and Dumka, U. C.: A review of biomass burning: Emissions and impacts on air quality, health and climate in China, Sci Total Environ, 579, 1000-1034, 10.1016/j.scitotenv.2016.11.025, 2017.

Cheng, Z., Wang, S., Fu, X., Watson, J. G., Jiang, J., Fu, Q., Chen, C., Xu, B., Yu, J., Chow, J. C., and Hao, J.: Impact of biomass burning on haze pollution in the Yangtze River delta, China: a case study in summer 2011, Atmos Chem Phys, 14, 4573-4585, 10.5194/acp-14-4573-2014, 2014.

11. *Page 6, Line 30-31, the authors said "Mixed type pollution was also necessary for substandard days in Jan. (44%), Feb. (50%), and Apr. (35%)". But in Page 5, Line 17-18, the authors said mixed type pollution is "B" (March 3rd to 4th, 2016). And please clearly describe how to decide mixed type pollution.*

Reply: In this study, mixed type pollution means coexistence of anthropogenic pollutants and dust particles in the atmosphere. We will make it clear in the revised manuscript. To better reflect the influence of dust-related heterogeneous processes on pollution formation, we changed this part into the comparison of δ value in substandard days and clean days as shown in Figure 5 and section 3.4 in the revised manuscript. We accept suggestions from the reviewer and will further explain and discuss the reasons and vital influence of mixed pollution in the revised manuscript.

12. *Page 7, Line 7-9, the authors obtained the conclusion is based on the report in published papers, not obtained from the authors' observation or lab experiment. So possibly the conclusion didn't confirm.*

Reply: We agree the suggestion of reviewer and will carefully make discussion and conclusion in revised manuscript as "This implies that pollution days in North China was great possible to induce internal mixing of dust and pollutant, especially in high humid atmospheric environment, and dust-related heterogeneous processes on the dust surface can aggravate the deterioration of air quality as a feedback."

13. *Page 7, Line 25-26, why the authors confirm that increasing $PM_{2.5}$ level in case A was caused by weak diffusion conditions and high relative humidity? How about other aspects, such as emissions?*

Reply: The heating season in north China is from mid-November to mid-March of the following year. The pollutant emission in Beijing area in this period remained at a high level compared with any other seasons (Wang et al., 2015). Haze pollution can essentially be attributed to two aspects: pollutant emissions into the lower atmosphere and favorable meteorological conditions. In particular, meteorological conditions control the occurrence of haze pollution (Zhang et al., 2016; Wang et al., 2013; Jacob and Winner, 2009; Ilten and Selici, 2008; Chang and Zhan, 2017). High emission of primary pollutant precursors in winter time combine with stable weather conditions (weak diffusion conditions) and favorable conditions for liquid phase reaction of pollutant (high humid), resulting in the formation of secondary pollutants $PM_{2.5}$.

And the clarity of the pollution process is usually accompanied by a strong cold air which from Siberia spills south into East Asia. We will make the reasons for high $PM_{2.5}$ clearer in the manuscript.

Wang, Q., Sun, Y., Jiang, Q., Du, W., Sun, C., Fu, P., and Wang, Z.: Chemical composition of aerosol particles and light extinction apportionment before and during the heating season in Beijing, China, Journal of Geophysical Research: Atmospheres, 120, 12708-12722, 10.1002/2015jd023871, 2015.

Zhang, Z., Zhang, X., Gong, D., Kim, S. J., Mao, R., and Zhao, X.: Possible influence of atmospheric circulations on winter haze pollution in the Beijing–Tianjin–Hebei region, northern China, Atmos Chem Phys, 16, 561-571, 10.5194/acp-16-561-2016, 2016.

Wang, Y., Yao, L., Wang, L., Liu, Z., Ji, D., Tang, G., Zhang, J., Sun, Y., Hu, B., and Xin, J.: Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China, Science China Earth Sciences, 57, 14-25, 10.1007/s11430-013-4773-4, 2013.

Jacob, D. J., and Winner, D. A.: Effect of climate change on air quality, Atmospheric Environment, 43, 51-63, 10.1016/j.atmosenv.2008.09.051, 2009.

Ilten, N., and Selici, A. T.: Investigating the impacts of some meteorological parameters on air pollution in Balikesir, Turkey, Environ Monit Assess, 140, 267-277, 10.1007/s10661-007-9865-1, 2008.

Chang, W., and Zhan, J.: The association of weather patterns with haze episodes: Recognition by PM2.5 oriented circulation classification applied in Xiamen, Southeastern China, Atmospheric Research, 197, 425-436, 10.1016/j.atmosres.2017.07.024, 2017.

14. *Page 7, Line 29-33, Fig, 7b didn't present height of air parcel, and in the paper, the authors also didn't present descriptions about height of air parcel in the draft. Why the authors can get conclusion that emissions in mining areas can lead a huge amount of dust? And in the paper, the descriptions about emissions in mining areas in March 3 to 4 are also not given.*

Reply: As suggested, we will add detailed simulation information in section 2.2 and section 3.5. In the case C, the air mass was a convergence of deviating northwest and south streams. The eastward-conveying air mass originated from the Taklimakan Desert in Sinkiang and channeled the Tengger Desert in Inner Mongolia, which were major source of dust in east Asia (Fu et al., 2012; Chen et al., 2013; Chen et al., 2017; Zhang et al., 2018); the south branch of the air mass past central Henan Province and southern Hebei Province where emission of gaseous pollutants from industries, residential and transport sources were significant (Huang et al., 2012;Zhao et al., 2012;Wang et al., 2014). What we want to express here was the eastward transport air mass may lead to a huge amount of dust mixed with pollutant-rich gaseous atmosphere in the Beijing area. We will make a more clear and accurate revision in the section 3.5.3 in the manuscript.

Fu, P., Zhong, S., Huang, J., and Song, G.: An observational study of aerosol and turbulence properties during dust storms in northwest China, Journal of Geophysical Research: Atmospheres, 117, n/a-n/a, 10.1029/2011jd016696, 2012.

Chen, S., Huang, J., Zhao, C., Qian, Y., Leung, L. R., and Yang, B.: Modeling the transport and radiative forcing of Taklimakan dust over the Tibetan Plateau: A case study in the summer of 2006, Journal of Geophysical Research: Atmospheres, 118, 797-812, 10.1002/jgrd.50122, 2013.

Chen, S., Huang, J., Kang, L., Wang, H., Ma, X., He, Y., Yuan, T., Yang, B., Huang, Z., and Zhang, G.: Emission, transport, and radiative effects of mineral dust from the Taklimakan and Gobi deserts: comparison of measurements and model results, Atmos Chem Phys, 17, 2401-2421, 10.5194/acp-17-2401-2017, 2017.

Zhang, X.-X., Sharratt, B., Liu, L.-Y., Wang, Z.-F., Pan, X.-L., Lei, J.-Q., Wu, S.-X., Huang, S.-Y., Guo, Y.-H., Li, J., Tang, X., Yang, T., Tian, Y., Chen, X.-S., Hao, J.-Q., Zheng, H.-T., Yang, Y.-Y., and Lyu, Y.-L.: East Asian dust storm in May 2017: observations, modelling and its influence on Asia-Pacific region, Atmospheric Chemistry and Physics Discussions, 1-31, 10.5194/acp-2018-205, 2018.

Huang, X., Song, Y., Li, M., Li, J., Huo, Q., Cai, X., Zhu, T., Hu, M., and Zhang, H.: A high-resolution ammonia emission inventory in China, Global Biogeochemical Cycles, 26, n/a-n/a, 10.1029/2011gb004161, 2012.

Zhao, B., Wang, P., Ma, J. Z., Zhu, S., Pozzer, A., and Li, W.: A high-resolution emission inventory of primary pollutants for the Huabei region, China, Atmos Chem Phys, 12, 481-501, 10.5194/acp-12-481-2012, 2012.

Wang, L. T., Wei, Z., Yang, J., Zhang, Y., Zhang, F. F., Su, J., Meng, C. C., and Zhang, Q.: The 2013 severe haze over southern Hebei, China: model evaluation, source apportionment, and policy implications, Atmos Chem Phys, 14, 3151-3173, 10.5194/acp-14-3151-2014, 2014.

15. *Page 8, Line 6-9, please connected the cited part of Itahashi et al. (2010) with your study.*

Reply: In the original manuscript version, we cite the work of Itahashi to explain the descending of dust plume on March 5 in case C. Considering that the discussion of meteorological factors is not the focus of this study, we removed this part in the revised manuscript. For better discuss about the internal/external mixing state of dust particle, and compare the two depolarization ratio detecting instruments which is Lidar and POPC, we will add more observation result for Lidar at the same period in section 3.52 and section 3.5.3.

16. *Page 8, Line 15, please give data prove or corresponding references.*

Reply: Considering the heating season was now over for a month, $PM_{2.5}$ that associated with high burning of fossil fuels for heating purpose was relatively weak, we would like to cite Wang et al., (2015) in the manuscript,

Wang, Q., Sun, Y., Jiang, Q., Du, W., Sun, C., Fu, P., and Wang, Z.: Chemical composition of aerosol particles and light extinction apportionment before and during the heating season in Beijing, China, Journal of Geophysical Research: Atmospheres, 120, 12708-12722, 10.1002/2015jd023871, 2015.

17. *Page 8, Line 16-17, how does the author judge no anthropogenic pollution emissions in case C, and obtain a pure dust process? In the paper, there are no data prove.*

Reply: For easier and better understanding of the manuscript, we redefined the time of the two dust case, that was case B on April 9-10 and case C on March 4-6, 2016. We agree with the reviewer that we cannot describe case B as completely free of anthropogenic pollution due to lacking of chemical composition information. In the period, the daily PM_{10} was 273.6 μg/m^3 and $PM_{2.5}$ account for 33.4% in PM_{10} on April 9. POPC analysis (Figure 6e in the manuscript) showed that particles in this process was concentrated in the particle size range larger than 3 μm, in which that contribution secondary formed aerosol particles was less important. During case B, air mass originated from Gobi desert in Mongolia and transport rapidly by a strong wind. Recent filter-based observation (Pan et al., 2017) showed that the water-soluble compounds in coarse mode was insignificant, compared to crustal matters, during dust event. To avoid misleading, we will defined case B as a typical dust-dominant period. We will make a more comprehensive interpretation of this conclusion in the revised manuscript according to the comments of reviewers.

Pan, X., Uno, I., Wang, Z., Nishizawa, T., Sugimoto, N., Yamamoto, S., Kobayashi, H., Sun, Y., Fu, P., Tang, X., and Wang, Z.: Real-time observational evidence of changing Asian dust morphology with the mixing of heavy anthropogenic pollution, Sci Rep, 7, 335, 10.1038/s41598-017-00444-w, 2017.

18. *Page 9, Line 21-22, why the authors use $PM_{2.5}/PM_{10}$ ratio? In 3.1 part, the authors said the fine mode (<1 μm), the coarse mode (4 μm – 8 μm). Why the authors didn't use $PM_{1.0}$, $PM_{4.0-8.0}$?*

Reply: Sorry for the misleading. The particles with aerodynamic diameter less than 2.5 μm is defined as fine mode, and the particles with diameter between 2.5 and 10 μm is defined as coarse mode. The widely adopted commercial online instruments normally measure the mass concentrations of $PM_{2.5}$ and PM_{10}. For POPC measurement, the mass concentrations of particles of $PM_{1.0}$ and $PM_{4.0-8.0}$ could be reasonably calculated on the basis of size distribution of particles.

However, this measurement at the state-control observatories are very limited in China, which make it difficult to estimate the variability of aerosol types. Alternatively, we adopted the ratio $PM_{2.5}/PM_{10}$ to identify the sources of primary pollutant. A higher ratio was generally ascribed to anthropogenic related secondary particles (sulfate, nitrate etc.), and a lower ratio indicate significant contributions mainly re-suspended or fugitive mineral dust particles due to some mechanical processes (Chan et al., 2005; Pérez et al., 2008; Akyuz and Cabuk, 2009; Chan and Yao, 2008; Xu et al., 2017). In a typical dust period, the higher the ratio was, the more possibility that anthropogenic pollution and dust particles were mixing, and consequentially morphological changes. We agree with the reviewer's concerns and will add the explanations for using the $PM_{2.5}/PM_{10}$ ratio in the manuscript.

Chan, C. Y., Xu, X. D., Li, Y. S., Wong, K. H., Ding, G. A., Chan, L. Y., and Cheng, X. H.: Characteristics of vertical profiles and sources of PM2.5, PM10 and carbonaceous species in Beijing, Atmospheric Environment, 39, 5113-5124, 10.1016/j.atmosenv.2005.05.009, 2005.

Pérez, N., Pey, J., Querol, X., Alastuey, A., López, J. M., and Viana, M.: Partitioning of major and trace components in PM10–PM2.5–PM1 at an urban site in Southern Europe, Atmospheric Environment, 42, 1677-1691, 10.1016/j.atmosenv.2007.11.034, 2008.

Akyuz, M., and Cabuk, H.: Meteorological variations of PM2.5/PM10 concentrations and particle-associated polycyclic aromatic hydrocarbons in the atmospheric environment of Zonguldak, Turkey, J Hazard Mater, 170, 13-21, 10.1016/j.jhazmat.2009.05.029, 2009.

Chan, C. K., and Yao, X.: Air pollution in mega cities in China, Atmospheric Environment, 42, 1-42, 10.1016/j.atmosenv.2007.09.003, 2008.

Xu, G., Jiao, L., Zhang, B., Zhao, S., Yuan, M., Gu, Y., Liu, J., and Tang, X.: Spatial and Temporal Variability of the PM2.5/PM10 Ratio in Wuhan, Central China, Aerosol and Air Quality Research, 17, 741-751, 10.4209/aaqr.2016.09.0406, 2017.

19. *Page 9, Line 25-26, generally we firstly presented some evidences, such as data prove or results in published paper, then derivate possible conclusion.*

Reply: We will cite more relevant reference to support our conclusion in the revised manuscript as “This negative relationship reflected the spheroidization of the dust particles as a result of hygroscopic properties of mineral dust aerosols. Polluted air generally contains abundant HNO_3 (Liang et al., 2007; Shi et al., 2014; Bytnerowicz et al., 2016). Li and Shao (2009) reported that mineral particles are mainly covered with coating including $Ca(NO_3)_2$, $Mg(NO_3)_2$, and $NaNO_3$ in North China. Deeper interaction between alkaline mineral dust and reactive acidic gases and the trapping process of atmospheric secondary inorganic salt modified the hydrophilic state of dust aerosols.”

Liang, Q., Jaeglé, L., Hudman, R. C., Turquety, S., Jacob, D. J., Avery, M. A., Browell, E. V., Sachse, G. W., Blake, D. R., Brune, W., Ren, X., Cohen, R. C., Dibb, J. E., Fried, A., Fuelberg, H., Porter, M., Heikes, B. G., Huey, G., Singh, H. B., and Wennberg, P. O.: Summertime influence of Asian pollution in the free troposphere over North America, Journal of Geophysical Research, 112, 10.1029/2006jd007919, 2007.

Shi, Y., Chen, J., Hu, D., Wang, L., Yang, X., and Wang, X.: Airborne submicron particulate (PM1) pollution in Shanghai, China: chemical variability, formation/dissociation of associated semi-volatile components and the impacts on visibility, Sci Total Environ, 473-474, 199-206, 10.1016/j.scitotenv.2013.12.024, 2014.

Bytnerowicz, A., Hsu, Y. M., Percy, K., Legge, A., Fenn, M. E., Schilling, S., Fraczek, W., and Alexander, D.: Ground-level air pollution changes during a boreal wildland mega-fire, Sci Total Environ, 572, 755-769, 10.1016/j.scitotenv.2016.07.052, 2016.

Li, W. J., and Shao, L. Y.: Observation of nitrate coatings on atmospheric mineral dust particles, Atmos Chem Phys, 9,

1863-1871, DOI 10.5194/acp-9-1863-2009, 2009.

20. *Page 9, Line 29-31, the result was obtained by the authors' lab experiment? If no, please add data prove or references.*

Reply: We consented to the reviewer's comments. The over discussion about chemical component here was removed to avoid misleading.

21. *Page 10, Line 3-20, please give corresponding data prove or lab experiment, then obtain conclusions.*

Reply: We agree with the reviewer's suggestions, corresponding lab experiment or composition observation data would enhance the credibility of the conclusions. We will revise the expression and concise the discussion about specific chemical reactions in section 3.6.

Minor Comments:

1. *Page 5, Line 11-13, "The growth of fine mode particles mainly stems from high anthropogenic emissions during heating seasons." confused with "The distinct growth period of fine mode particles was usually induced by an increase in relative humidity (i.e., RH)". Please afresh write the two sentences.*

Reply: The sentences will be rewrite in the revised manuscript, as "It can be seen in Figure 2a that volume size distribution generally had two size modes during the whole observation periods. The occurrence of fine mode (peak at ~1 μm) was accompanied with increase of RH.

2. *Page 6, Line 31, what is JJ? Please explain it.*

Reply: The "JJ" has been explained as June and July in the revised manuscript.

3. *Page 7, Line 3,4, NOx should be NO_x.*

Reply: Thanks for pointing out the mistake, and the "NOx" has been revised as "NO_x" in the manuscript.

4. *Page 8, Line 6, please clearly point out "dominant mechanisms". What were dominant mechanisms?*

Reply: I will delete the "dominant mechanisms" since it is not discussed in the context.

5. *Page 8, Line 10-11, the authors said the first processes (22st -23rd, Dec. 2015) and the third process (9th, Apr. 2016). But in Page 5, Line 17-18, the authors said "A" (December 22nd to 23rd, 2015), "B" (March 3rd to 4th, 2016) and "C" (April 9th, 2016), respectively. Please unite.*

Reply: We will change the date in the manuscript to a uniform format.

6. *Page 8, Line 15, please added data descriptions or references for "primary anthropogenic pollution emissions in this period were also lower compared to the winter time".*

Reply: We revised the sentence as suggested, as follows: " Considering the heating season was now over for a month, $PM_{2.5}$ that associated with high burning of fossil fuels for heating purpose was relatively weak (Wang et al., 2015). "

Wang, Q., Sun, Y., Jiang, Q., Du, W., Sun, C., Fu, P., and Wang, Z.: Chemical composition of aerosol particles and light extinction apportionment before and during the heating season in Beijing, China, Journal of Geophysical Research: Atmospheres, 120, 12708-12722, 10.1002/2015jd023871, 2015.

7. *Page 9, Line 23-25, please added references.*

Reply: We will cite more previous studies to support relevant content in the revised manuscript.

8. *Figure 7f, the date is 04/10/2016, not 04/09/2016?*

Reply: The date on Figure 7f will be changed to "04/09/2016".

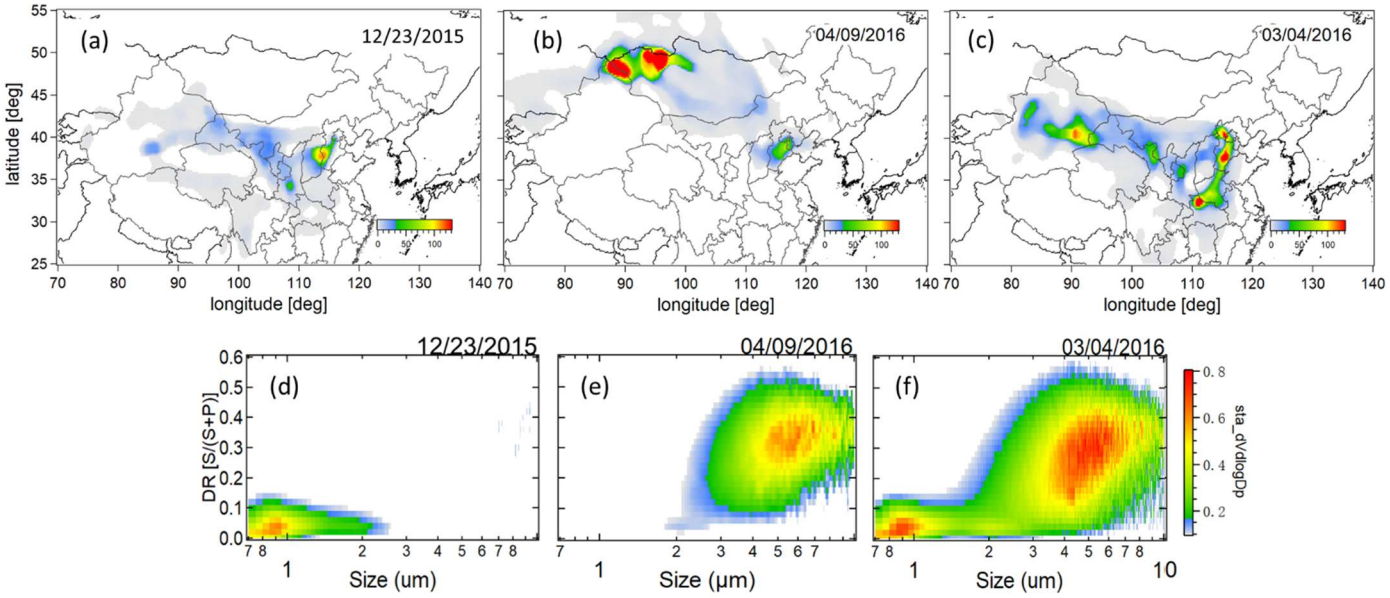

Figure 6. Backward trajectories from Beijing calculated by the FLEXPART dispersion model for (a) the anthropogenic pollution case, (b) the dust-dominant case, and (c) the mixed pollution period. Variation in the standard δ value as a function of particle size for corresponding episodes: (d), (e), and (f).

9. *Page 9, Line 24-25, please added references.*

Reply: We will cite Liang et al. (2007), Shi et al. (2014) and Bytnerowicz et al. (2016) to support the content here.

Liang, Q., Jaeglé, L., Hudman, R. C., Turquety, S., Jacob, D. J., Avery, M. A., Browell, E. V., Sachse, G. W., Blake, D. R., Brune, W., Ren, X., Cohen, R. C., Dibb, J. E., Fried, A., Fuelberg, H., Porter, M., Heikes, B. G., Huey, G., Singh, H. B., and Wennberg, P. O.: Summertime influence of Asian pollution in the free troposphere over North America, Journal of Geophysical Research, 112, 10.1029/2006jd007919, 2007.

Shi, Y., Chen, J., Hu, D., Wang, L., Yang, X., and Wang, X.: Airborne submicron particulate (PM1) pollution in Shanghai, China: chemical variability, formation/dissociation of associated semi-volatile components and the impacts on visibility, Sci Total Environ, 473-474, 199-206, 10.1016/j.scitotenv.2013.12.024, 2014.

Bytnerowicz, A., Hsu, Y. M., Percy, K., Legge, A., Fenn, M. E., Schilling, S., Fraczek, W., and Alexander, D.: Ground-level air pollution changes during a boreal wildland mega-fire, Sci Total Environ, 572, 755-769, 10.1016/j.scitotenv.2016.07.052, 2016.

Reply to the comments of anonymous reviewer #2 on manuscript entitled "Variability of depolarization of aerosol particles in Beijing mega city: implication in interaction between anthropogenic pollutants and mineral dust particles"

The authors appreciate very much for reviewing our manuscript and your insight comments. As suggested, we carefully revised the manuscript thoroughly according to the valuable advices. We will response to all the comments as follows:

The authors present a study of aerosol particles in Beijing, China, with a polarization optical particle counter (POPC). They present 8 months of statistics and 3 case studies of polluted conditions, dust conditions and a dust-pollution mixture. Especially the results presented in Fig. 7 are of interest, because they give insight in the mixing process of dust and pollution and help in the discussion about external and internal mixture. While reading the manuscript, I was missing the main new points. In the revision process, I would strongly recommend to strengthen the impact of the study: the separation of dust and pollution with a POPC and the discussion about internal or external mixture. Then, this can be applied to the statistics collected over an East Asian megacity, which lies in a global hot spot of dust and pollution mixtures.

The main point, why I have to reject the paper is that the retrieved depolarization values are questionable. I further checked the publication by Kobayashi et al., AE 2014, and I could not find any information on the calibration process of the depolarization ratio measured with the POPC. Especially the depolarization ratio can be influenced by multiple factors. I am sure, that the authors developed a calibration process to check the trustworthiness of the measured data, but it is missing in the publication. Also there are no comparisons of the retrieved depolarization ratios at 120◦ scattering angle to existing measurements or simulations of spherical or non-spherical particles. The uncertainties of the depolarization ratio values are not given. These points are essential to present trustworthy results. And as the depolarization ratio is one of the main quantities measured in this study, I have to insist on a proper calibration. Therefore, I recommend resubmitting the paper after clarifying the uncertainties of the depolarization ratio.

Reply: We greatly appreciate the reviewer for insight comments on the manuscript. To respond to the reviewer's major concerns, we made thoroughly revisions and corrections according to all the insight comments of the reviewers. Besides, more crucial information and analysis will be added in the revised manuscript, as follows:

(1) A detailed description and specification of calibration system of POPC and calibration results in this study will be added in the revised manuscript.

(2) Comparison between observation and the simulation result with T-matrix method on randomly-oriented Voronoi aggregation and elongated ellipsoid particles will be added.

(3) A comparison result of measurement of size-resolved number concentration of particles between POPC and KC52 optical particle counter will be added in the revised manuscript.

(4) Comparison of observation results between POPC measurement and LIDAR observation during several dust events in Beijing will be discussed. The cross validation on the dust events provide more direct evidence of variation in the depolarization properties of dust particles.

Besides, to make the conclusions more pertinent to this study, we revised the expression and concise the discussion about specific chemical composition and reaction. Our conclusions were useful for understanding the long-term depolarization property of aerosols in East Asia, interaction between anthropogenic pollutions and mineral dust in East Asia and their impacts on local air quality, and the sensitive relation between particle morphology and meteorological parameter. This long-term study in Beijing motivate revisit on decades of Lidar data in dust-pollution interactions, and it indicates the necessity of a reliable optical model of internally mixed polluted dust for a detailed analysis of polarization remote sensing observations.

As fas as we known, this was the first study that long-term observation on the single particle polarization properties performed in China, the aim of this study is to investigate the long-term morphology properties of particles in North China because this region has long experienced serious air pollution and dust events due to its special geographical location and industrial

activities. As suggested, in the revised manuscript, we will strengthen discussion about the separation of dust and pollution and their internal or external mixture. Comprehensive study with previous studies using the polarization optical particle counter will be performed, for instance the Lidar measurement. Single particle depolarization ratio measure is able to quantitatively investigate the evolution of the mixing of dust particles during their transport. According to the depolarization ratio of their scattering signals, every mineral dust aerosols and anthropogenic pollutants can be distinguished because the direction of polarization of scattering light was identical with the incident light for spherical particle; for the non-spherical particles, a vertical polarization signal will be produced (Pan et al., 2015). Based on this characteristic, a number of studies in East Asia have been conducted that focued on the depolarization ratio of single particles. Sugimoto et al. (2015) found that backscattering depolarization ratio in polluted dust was smaller compared to pure Asia dust for the measurement at Seoul. Pan et al. (2017) reported coating processes such as heteorogeneous reaction and hygroscopic growth on the surface of dust particles play a vital role in decreasing of depolarization ratio (DR) of particles in coarse mode. There were also lots of direct evidences of internally mixed dust particle on the basis of electronic microscopy (Li and Shao, 2009; Li et al., 2011). Note that, these studies were just a case study, and long-term measurement of their mixing states and seasonal varibility in China was still lacking. Therefore we performed an eight-month observe experiment in urban Beijing. At the present stage of this manuscript, the description about instrument calibration and uncertainty assessment was not shown, we will add relavent important information and make an detailed explanation in the revised manuscript.

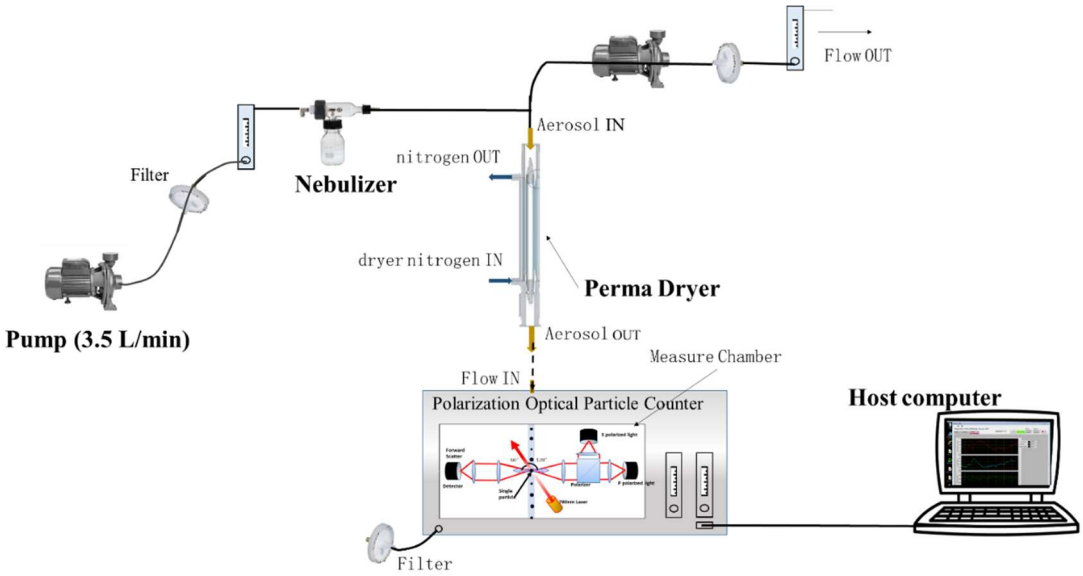

Figure 1. A schematic diagram of the laboratory calibration process

Figure 1 shows the schematic diagram of the laboratory calibration process of POPC. The spherical polystyrene standard aerosols were Dp = 0.048 μm, 1.005 μm, 3.210 μm, 5.125 μm, 7.008 μm, 10.14 μm (JSR Life Sciences Corporation). Aerosols were generated by a nebulizer at an flow rate of 3.5 L/min, desiccated by passing through a vrtically placed 45 cm Perma casing tube (MD–110–24P, GLSciences). Depolarization ratio of typical spherical particles at Dp = 5.125 μm, 7.008 μm and 10.14 μm were found to be 0.075, 0.085 and 0.102 with an pervasive uncertainty of ± 0.01,and was almost zero for the fine particles (Dp = 0.048 μm, 1.005 μm) (Figure 2). In this study, the DR values of aerosols in coarse mode were found to be centered on ~ 0.3, much larger than calibration results, indicate that coarse mode aerosol particles at the site was non-spherical generally.

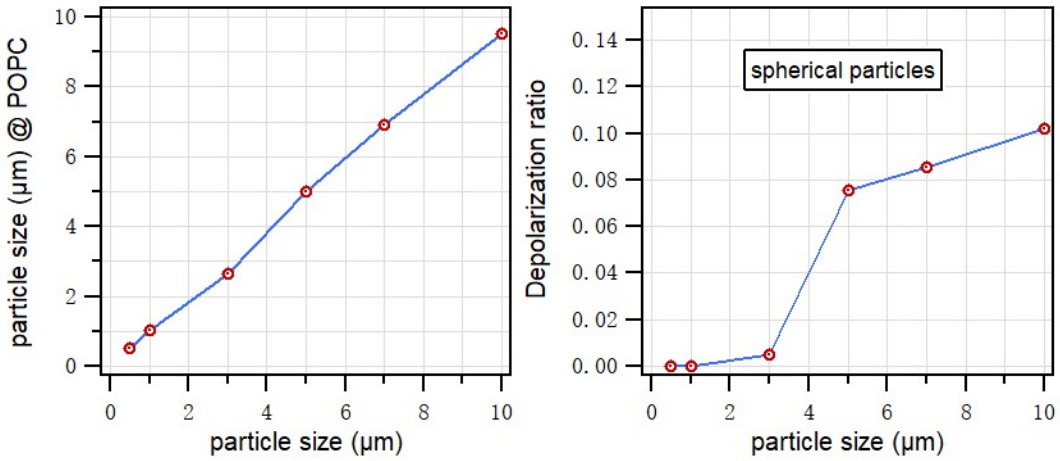

Figure 2. The calibration results of POPC. Optical size of the particle converted from a forward scattering signal at 60 degree (left); Depolarization of the standard particles detected from a backward scattering signal at 120 degree (right).

Due to the limitation of standard aerosols, we only did the calibration of the spherical particles. For non-spherical aerosols, we did some simulation to show the depolarization ratio at a scattering angle of 120 °. The polarization property of randomly oriented elongated ellipsoid particles was simulated on the basis of the T-matrix methodology (Dubovik et al., 2006). The non-sphericity is indicated by the aspect ratio (the ratio of long axis to its orthogonal short axis). As indicated in Figure 3, the observed depolarization ratio (range from 0.1-0.5, and centered on 0.3) in this study corresponded to an aspect ratio of 1.20 - 1.55 for coarse mode dust particles, with mode value 1.50. During the dust-dominant period on April 10, the aspect ratios of the dust particles were estimated to be 1.51 as the depolarization ratio of the dust particles was 0.34. While on polluted dust period on March 4, the aspect ratios of the dust particles were estimated to be 1.48 as the depolarization ratio of the dust particles was 0.30, providing that the dust particles has a great possibility to underwent partial deliquescent and hygroscopic growth on the surface in mixed pollution period.

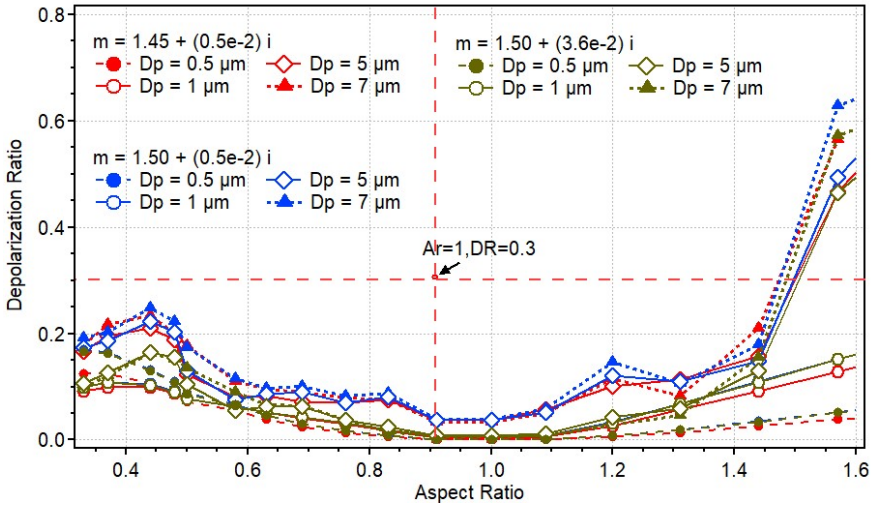

Figure 3. Theoretical simulation of the depolarization ratio of randomly oriented elongated ellipsoid particles as a function of the aspect ratio at fine mode: Dp = 0.5 μm,1 μm, and Dp = 5 μm, 7μm and on the basis of the T-matrix methodology.

Such characteristic was also well predicted by optical model considering particles of Voronoi aggregation (Ishimoto et al., 2010) (Figure 4). Theoretical simulation showed that an change in refractive index could affect the DR value evidently, for the coarse modal particles observed in this observation experiment, the simulation results showed that the depolarization ratio showed a leveling off tendency at 0.31 ± 0.02, confirmed with the depolarization ratio observed in our study.

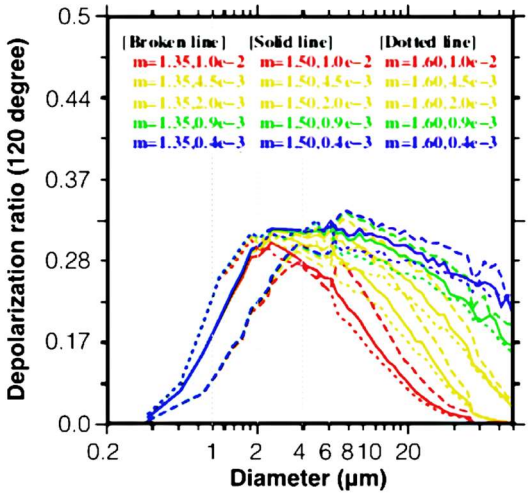

Figure 4. Theoretical calculation of depolarization ratio (at 120 backward direction) as a function of particle size for different refractive index.

As the reviewer suggested, providing the comparison that between the retrieved depolarization ratios at 120◦ scattering angle to existing measurements would better enhance the credibility of our data. The reality is that we are the first study that focused on the single particle polarized optical spectrometer in China. As far as I know, Lidar measurement and satellite on-board remote sensing (CALIPSO) (Glen et al., 2013) could provide tempo-spatial profile of volumn integrated depolarizaiton ratio, however the defination of Lidar's depolarizaiton ratio (S/P, backward at angle of 180°) is different from POPC and the two methods all persuming externally mixed of spherical particles and dust aerosols. Beyond that, there is no relevant comercial instrument that can make such measurement. Mass concentration of PM was reconstructed on the basis of number concentration of particles measured by POPC and particles density. The particle density was assumed to increase linearly from 1.77 g/cm^3 (0.5 μm) to 2.2 g/cm^3 (10 μm). The result compared well with commercial optical particle counter (KC52, RION, as shown in Figure 5), especially in the coarse mode size range observation. We will add relevant literatures and additional remarks in the context.

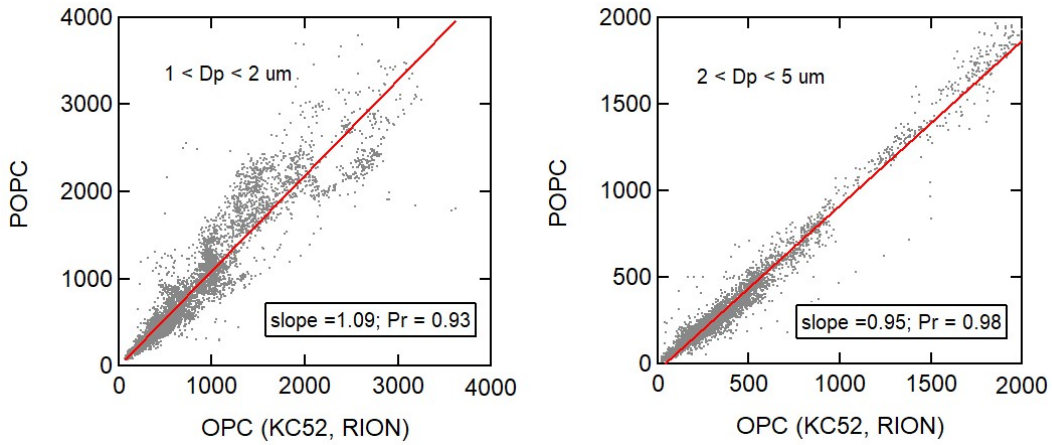

Figure 5. The comparison of number concentrations measured by POPC and OPC (KC52, RION).

The uncertainties of the depolarization ratio values was affected by various factors, including voltage variance of power supply (σ_{vol}^2), emvironment water content (σ_{WC}^2) and complex refraction index (σ_{nf}^2) of the aerosol. Light scattering signals are stored in the form of electrical impulses, the voltage instability will affect the pulse signals. The power supply voltage of POPC was set 15 V, and have a fluctuation by 5% on test. The residence time of diluted air mass in POPC was estimated to be 0.7 s, which was generally sufcient for aerosol particles to achieve equilibrium before measurement in the detecting chamber. It suggested that the wet particles tend to shrink due to loss of water. Zhang et al. (2015) reported aerosol backscattering coefficient increased 25% as the RH increased from 40% to 85 %. The measurement uncertainty in DR was estimated to be 10% caused

by humidy change. Theoretical simulation also indicated that an increase in the imaginary part of the refractive index could reduce the DR value evidently (Ishimoto et al., 2010), according to the simulation experiments variations in the particle's refractive index (the real and imaginary part) can explain 6% depolarization variability (Figure 4). Under the calculation method ($\sigma_{DR} = \sqrt{\sigma_{vol}^2 + \sigma_{nf}^2 + \sigma_{WC}^2}$), we estimate the uncertainty of depolarization ratio was < 13%. As indicate in Figure 14 in the manuscript, the depolarization ratio has a decrease of 50% and 43% for 3 μm and 5 μm aerosols, we conclude that dust particles in the atmosphere does contain hygroscopic component.

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, Geophysical Research Letters, 42, 1593-1598, 10.1002/2014gl062491, 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 & Radiative Transfer, 150, 107-113, 10.1016/j.jqsrt.2014.08.003, 2015.

Pan, X., Uno, I., Wang, Z., Nishizawa, T., Sugimoto, N., Yamamoto, S., Kobayashi, H., Sun, Y., Fu, P., Tang, X., and Wang, Z.: Real-time observational evidence of changing Asian dust morphology with the mixing of heavy anthropogenic pollution, Sci Rep, 7, 335, 10.1038/s41598-017-00444-w, 2017.

Li, W. J., and Shao, L. Y.: Observation of nitrate coatings on atmospheric mineral dust particles, Atmos Chem Phys, 9, 1863-1871, DOI 10.5194/acp-9-1863-2009, 2009.

Li, W. J., Zhang, D. Z., Shao, L. Y., Zhou, S. Z., and Wang, W. X.: Individual particle analysis of aerosols collected under haze and non-haze conditions at a high-elevation mountain site in the North China plain, Atmos Chem Phys, 11, 11733-11744, 10.5194/acp-11-11733-2011, 2011.

Dubovik, O., Sinyuk, A., Lapyonok, T., Holben, B. N., Mishchenko, M., Yang, P., Eck, T. F., Volten, H., Munoz, O., Veihelmann, B., van der Zande, W. J., Leon, J. F., Sorokin, M., and Slutsker, I.: Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust, Journal of Geophysical Research-Atmospheres, 111, 10.1029/2005jd006619, 2006.

Ishimoto, H., Zaizen, Y., Uchiyama, A., Masuda, K., and Mano, Y.: Shape modeling of mineral dust particles for light-scattering calculations using the spatial Poisson–Voronoi tessellation, Journal of Quantitative Spectroscopy and Radiative Transfer, 111, 2434-2443, 10.1016/j.jqsrt.2010.06.018, 2010.

Glen, A., and Brooks, S. D.: A new method for measuring optical scattering properties of atmospherically relevant dusts using the Cloud and Aerosol Spectrometer with Polarization (CASPOL), Atmos Chem Phys, 13, 1345-1356, 10.5194/acp-13-1345-2013, 2013.

Zhang, L., Sun, J. Y., Shen, X. J., Zhang, Y. M., Che, H., Ma, Q. L., Zhang, Y. W., Zhang, X. Y., and Ogren, J. A.: Observations of relative humidity effects on aerosol light scattering in the Yangtze River Delta of China, Atmos Chem Phys, 15, 8439-8454, 10.5194/acp-15-8439-2015, 2015.

Further comments for improving the manuscript:

1. *Add an outline of the paper at the end of the introduction*

Reply: As the reviewer suggested, we will add an outline of the paper at the last paragraph of the introduction as follows:

"In this study, a comprehensive ground-based measurement of depolarization properties of aerosol particles was performed at an urban site in Beijing, at the State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC, Longitude: 116.37E; Latitude: 39.97N), Institute of Atmospheric Physics/Chinese Academy of Sciences. Morphological variability of ambient aerosol particles were investigated from November 2015 to July 2016

using a Polarization Optical Particle Counter (POPC), and the seasonal characteristics of depolarization ratio (δ) of atmospheric aerosols were explained. Three pollution events including anthropogenic pollutants, mineral dust-dominant and polluted dust were classified according to its size distribution and δ value, as well as trajectory analysis, to investigate the interactions between dust particles and pollutants. The objective of this study focuses on the variation of the δ value of aerosol particles and its relationship with secondary pollutants. This study represents the first time such long-term observation of ambient aerosol morphology performed in the megacity of Beijing, and provides more applicable data for evaluating the mixing processes of atmospheric aerosols and their climate impact."

2. *Mention, which size range is covered with your optical particle counter.*

Reply: The effective size range of the optical particle counter was 0.5-10 μm, we will add it into the manuscript.

3. *The term "fine mode particles" normally refers to particles with a diameter smaller than 1 μm. PM1 data would be helpful to assess the fine mode particles.*

Reply: Thanks for the reviewer's suggestion, we carefully checked relevant studies on atmospheric aerosols, and find that fine mode particles usually refers to particles with an aerodynamic diameter below 2.5 microns (Adams et al., 2001; Rathnayake et al., 2017a; Rathnayake et al., 2017b) and PM_1 (Bari et al., 2015; Shi et al., 2014) was often referred to as the sub-micron scale.

Adams, H. S., Nieuwenhuijsen, M. J., and Colvile, R. N.: Determinants of fine particle (PM2.5) personal exposure levels in transport microenvironments, London, UK, Atmospheric Environment, 35, 4557-4566, 10.1016/s1352-2310(01)00194-7, 2001.

Rathnayake, C. M., Metwali, N., Jayarathne, T., Kettler, J., Huang, Y., Thorne, P. S., amp, apos, Shaughnessy, P. T., and Stone, E. A.: Influence of rain on the abundance of bioaerosols in fine and coarse particles, Atmos Chem Phys, 17, 2459-2475, 10.5194/acp-17-2459-2017, 2017a.

Rathnayake, C. M., Metwali, N., Jayarathne, T., Kettler, J., Huang, Y. F., Thorne, P. S., O'Shaughnessy, P. T., and Stone, E. A.: Influence of rain on the abundance of bioaerosols in fine and coarse particles, Atmos Chem Phys, 17, 2459-2475, 10.5194/acp-17-2459-2017, 2017b.

Bari, M. A., Kindzierski, W. B., Wallace, L. A., Wheeler, A. J., MacNeill, M., and Heroux, M. E.: Indoor and Outdoor Levels and Sources of Submicron Particles (PM1) at Homes in Edmonton, Canada, Environmental Science & Technology, 49, 6419-6429, 10.1021/acs.est.5b01173, 2015.

Shi, Y., Chen, J., Hu, D., Wang, L., Yang, X., and Wang, X.: Airborne submicron particulate (PM1) pollution in Shanghai, China: chemical variability, formation/dissociation of associated semi-volatile components and the impacts on visibility, Sci Total Environ, 473-474, 199-206, 10.1016/j.scitotenv.2013.12.024, 2014.

4. *State clearer the difference between the depolarization ratio used in lidar studies (s-polarized to p-polarized ratio at 180◦ backscatter) and the depolarization ratio used in the POPC study (s-polarized to s+p polarized ratio at 120◦ backscatter). To add value to the lidar studies by using single particle analyses, it would be better to use the same depolarization ratio or at least to present a method to convert the POPC depolarization ratio to a lidar retrieved depolarization ratio. But I dare, that this would be rather complicated for non-spherical particles.*

Reply: The main difference between the depolarization ratio (δa) used in LIDAR and the POPC was: Lidar measured was all the particles' depolarization ratio in backward scattering angle 180° in a beam volume. The overall depolarization ratio of dust particles might be underestimated due to substantially presence of small spherical particles in the dust plume. It means external mixing of a large amount of fine particles with mineral dust aerosols also results in a lower δa; POPC measured all the single particle's depolarization ratio in the backward scattering without interference. According to theoretical simulation and optical lens design, POPC only measure depolarization ratio of scatter light at angle of 120°.

Considering that the scattering phase function of particles was not linearly varying at different angles, especially for non-spherical particles, depolarization ratio at backscatter 120 ° was difficult to switch to the results of 180 °. Now we are working on a project of remould the light road of POPC, in the new version, we design to detect the scattered light information at around 180 °. In our next research, we would like to follow the review's suggestion to try provide a comparison for LIDAR result.

5. *Add the distance and the direction (north, east,…) to the description of the two additional measurements sites (Olympic Sport Centre state control site and meteorological station) and not just the coordinates. It makes it easier for the reader.*

Reply: Thanks to the reviewers' suggestions, we will add information about the distance and the direction to the description of the two additional measurements sites.

Variability of depolarization of aerosol particles in Beijing mega city: implication in interaction between anthropogenic pollutants and mineral dust particles

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Abstract. East Asia is suffering from both severe air pollution problem due to intensive anthropogenic emissions and natural mineral dust aerosols. During transport, the aerosol particles undergo complex mixing processes, resulting in great impacts on regional air quality, human health and climate. In this study, we conducted a long-term observation using an Optical Particle Counter equipped with a Polarization detection module (POPC) at an urban site in Beijing. Mass concentrations of both $PM_{2.5}$ and PM_{10} estimated from POPC compared well with ground-based measurements. The results revealed that the observed depolarization ratio (δ, termed as the ratio of the intensity of the s-polarized signal to the intensity of the 120 degree backward scattering signal [s/(s + p)]) for aerosol particles in the fine mode was generally much lower in summer than that in spring as a result of predominance of different aerosol types. Mineral dust particles in the coarse mode normally had a large δ value (0.3 ± 0.05) owing to their non-spherical shape; however, particles in the fine mode consisted of large proportion water-soluble compositions, which lead to apparent decrease of their δ values in particular high relative humidity (RH) condition. Because the observation site was subject to frequent impact of dust event in spring, δ value of particle at 1 μm was almost twice as high as that (0.07 ± 0.01) in summer. Based on size-resolved δ values, anthropogenic pollutants, mineral dust and polluted mineral dust particles, and their contribution to local air quality were well distinguished. About 26.7% of substandard days (daily averaged $PM_{2.5}$ concentration larger than 75 $\mu g/m^3$) in Beijing was featured by high atmospheric loading of coarse-mode particles in winter-spring time. In particular during severe pollution episode in winter, the δ values of coarse mode particles decreased by 13%, implies the high possibility of dust-related heterogeneous processes on pollution formation. During dust event, δ values of particle at Dp = 5 μm decreased evidently with increase of $PM_{2.5}/PM_{10}$ ratio as well as RH, indicating of the morphological changes of mineral dust. This study confirmed that high RH tends to promote water absorption processes on the dust surface as well as coating of soluble compounds, and suggested that complex mixing of dust and anthropogenic particles may lead to underestimate impact of dust particles in urban area based on remote sensing technique, and interaction between dust particle and pollutants should be well considered by the optical model.

1 Introduction

Rapid economic growth and urbanization processes in East Asia has caused serious air pollution in the past decades owing to the substantial consumption of fossil fuel. Anthropogenic emissions (industrial, traffic, residential etc.) emits substantial amount of pollutant gases (SO_2, NO_2, NH_3, VOCs etc.) and primary aerosols (Akimoto, 2003;Akimoto et al., 2006;Kurokawa et al., 2013;Li et al., 2017), resulting in formation of $PM_{2.5}$. Mineral dust particles in the atmosphere also have a detrimental impact on air quality and human health such as reducing visibility, increase respiratory morbidity. Beijing is located in vicinity of dust source region. In dry seasons, mineral dust particles emitted from Taklimakan, Gobi, Mongolia plateau and Loess Plateau may transport eastwardly across the northern part of China (Takemura et al., 2002;Jickells et al., 2005;Uno et al., 2009), and undergo complex mixing with anthropogenic pollutants.

The environmental and climate effects of these mixing processes are notable because of dramatic changes in the physical, chemical and optical properties of mixed particles (Pan et al., 2009). In polluted urban area, soluble salts coated on dust aerosols reduce the critical supersaturation, and there is a stronger tendency for polluted aerosols to serve as CCN (cloud condensation nuclei), influencing the formation of cloud (Sullivan et al., 2009;Tang et al., 2016). The soluble salts are derived from directly trapping inorganic salt, as well as the heterogeneous reactions between reactive gases (mostly HNO_3, HCI, SO_2 and NO_2) and alkaline mineral dust. Continuous coating and hygroscopic growth processes on the surface modify the shapes of dust particles (Li et al., 2011). A recent study pointed out that coexistence of NO_x and mineral dust may lead to a gas-particle conversion process and promote the conversion of SO_2 to sulfate (He et al., 2014). There is also observational evidence that heavy dust mixed with anthropogenic pollution may trigger new particle formation(Dupart et al., 2012;Nie et al., 2014), which exaggerates the degradation of regional air quality. The above scientific discoveries all indicate the importance of study on the mixing states of dust and pollution aerosols.

Widely used technologies to distinguish aerosol types include high-precision ground-based Lidar (Light Detection And Ranging) systems and satellite-borne observations (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation; CALIPSO)(Winker et al., 2009;Cesana et al., 2016;Venkata and Reagan, 2016) that measures an targeted air parcel's volume depolarization ratio from backward scattering signals. Aerosol types can be distinguished by the distinct parts formed by data points on a figure of their volume depolarization ratio(δ_a = s/p, at 532 nm) of aerosols versus backscattering color ratio (1064 nm/532 nm); however, bias in the classification of internal mixed dust (normally has a large color ratio and small δ_a) is sometimes unavoidable since external mixing of a substantial amount of fine particles (δ_a< 0.1) with mineral dust aerosols (δ_a > 0.35) can also results in decrease of of δ_a value. To respond to this need, an optical particle counter with a depolarization module was developed. Single particle δ value measure is able to quantitatively investigate the evolution of

the mixing of dust particles during their transport (Pan et al., 2015). According to the size-resolved δ value of scattering signals, particles in spherical shape can be distinguished because the direction of polarization of scattering light was identical with the incident light; for the non-spherical particles, direction of polarization will be deviated significantly. Generally, secondary formatted particles tend to be spherical with small δ value, while natural mineral dust has a larger δ value for its irregular shape (Kobayashi et al., 2014). Real-time measurement of δ value on a single-particle-based help to avoid the misclassification of aerosol types. Sugimoto et al. (2015) found that backscattering δ value in polluted dust was smaller compared to pure Asia dust for the measurement at Seoul. Previous study (Pan et al., 2017) in Beijing indicated that coating processes such as heteorogeneous reaction and hygroscopic growth on the surface of dust particles play a vital role in decreasing of depolarization ratio (δ) of particles in coarse mode. As far as we known, long-term measurements of interaction of both anthropogenic pollution and mineral dust and their effect on dust morphological changes in North China are still lacking.

In this study, a comprehensive ground-based measurement of depolarization properties of aerosol particles was performed at an urban site in Beijing, at the State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC, Longitude: 116.37E; Latitude: 39.97N), Institute of Atmospheric Physics/Chinese Academy of Sciences. Morphological variability of ambient aerosol particles were investigated from November 2015 to July 2016 using a Polarization Optical Particle Counter (POPC), and the seasonal characteristics of depolarization ratio (δ) of atmospheric aerosols were explained. Three pollution events including anthropogenic pollution case, typical dust-dominant case and mixed type pollution period were classified according to its size distribution and δ value, as well as trajectory analysis, to investigate the interactions between dust particles and pollutants. The objective of this study focuses on the variation of the δ value of aerosol particles and its relationship with secondary pollutants. This study represents the first time such long-term observation of ambient aerosol morphology performed in the megacity of Beijing, and provides more applicable data for evaluating the mixing processes of atmospheric aerosols and their climate impact.

2 Observation

2.1 Iinstrument overview

The observation of the depolarization properties of single particles in Beijing was performed using a Polarization Optical Particle Counter (POPC) (Kobayashi et al., 2014). The instrument was installed on the second floor of an air-conditioned two-story building. The inlet was ~50 cm above the roof of the building, and ambient air was drawn into room through ½ inch stainless tube with rainproof cap. The total flow rate of inlet sampling air was set to 13 lpm (litres per minute) with a supporting pump. For POPC, the detecting size range was 0.5-10 μm. A polarized laser beam at a wavelength of 780 nm illuminated the particles. The POPC uses a forward scattering light at a scattering angle of 60° to determine the size of particles; backward scattering intensity at 120° is divided into two components with polarizer: p-polarized light is in the plane of the incident and

reflected beams, known as the horizontal polarized light, while s-polarized light is perpendicular to the plane. Normally, the ratio of the s-polarized signal to that of the total backward light scattering signal is defined as the depolarization ratio (δ), which can provide morphological information about the particle (Muñoz and Hovenier, 2011). The acceptance angle (angle of the backscattered light received by the polarizer) for the polarization detector is 45°. To avoid coincidence error (several particles passing through the laser beam simultaneously), sampling flow of the POPC is set to 80 ccm (cubic centimetre per minute) with a dilution flow of 920 ccm.

During the observation, POPC was calibrated using standard known-size particles (JSR Life Sciences Corp.) at Dp = 0.048 μm (SC-0050-D), 1.005 μm (SC-103-S) and DYNOSHERE polystyrene standard aerosols at 3.210 μm (SS-032-P), 5.125 μm (SS-052-P), 7.008 μm (SS-074-P) and 10.14 μm (SS-104-P). Aerosols were generated by a nebulizer at a flow rate of 3.5 L/min, desiccated by passing through a vertically placed 45 cm Perma Casing tube (MD–110–24P, GL Sciences), as the laboratory calibration process Figure S1 showed. δ value of typical spherical particles at Dp = 5.125 μm, 7.008 μm and 10.14 μm were found to be 0.075, 0.085 and 0.102, and the δ value was almost zero for the fine mode particles (Dp = 0.048 μm, 1.005 μm) (Figure S2). The uncertainties of the δ value was affected by various factors, including voltage variance of power supply (σ_{vol}^2), emvironment water content (σ_{WC}^2) and complex refraction index (σ_{nf}^2) of the aerosols, we estimate the uncertainty of δ value was < 13%. For comparison, mass concentrations of pollutants in Beijing was obtained from a ground-based state control site (Longitude: 116.40E; Latitude: 39.98N; 2.7 km northeast of LAPC) and corresponding meteorology data from the climatological station (Longitude: 116.48E; Latitude: 39.95N; 9.5 km southeast of LAPC), we analysed the artificial pollution process and special mineral dust cases that occurred in Beijing.

Vertical profile of extinction coefficient, depolarization ratio of aerosol particles was concurrently measured using a NIES Lidar system from National Institute for Environmental Studies (http://www-lidarnies.go.jp/AD-Net) (Singh et al., 2008;Shimizu et al., 2016). The polarization Mie-Lidar is a powerful instrument for identify the change of optical properties of mineral dust (Shimizu et al., 2004). The data from the Lidar was processed in 15 min resolution to derive the volume depolarization ratio at 532 nm, attenuated backscattering coefficient at 1064 nm and 532 nm and the extinction coefficient estimated for spherical aerosols (mainly air pollutant) and non-spherical particles (mainly natural dust). The energy is 20 mJ /pulse and 30 mJ/pulse for 1064 nm and 532nm laser, and the light is emitted vertically with a pulse repetition of 10 Hz (Shimizu et al., 2016). Telescopes with a diameter of 20 cm (Lidar in Beijing) are used to collect the scattered light from the sky at an observation wavelength of 532 nm.

2.2 Dispersion and trajectory analysis

Long-range and mesoscale dispersion of air parcels over the Asia region was simulated using the FLEXPART (FLEXible PARTicle Dispersion Model) Dispersion Model. FLEXPART is a Lagrangian transport and dispersion model (https://www.flexpart.eu) developed by Norwegian Institute for Air Research. This model is suitable for the simulation of a

large range of atmospheric transport processes (Stohl et al., 2005), which can do with forward simulating to trace particles from source areas and backward simulating to track particles from given receptors. The meteorological fields for FLEXPART are taken from NCEP's (National Centers for Environmental Prediction) reanalysis GDAS (Global Data Assimilation System) dataset on a 1° lat × 1° lon grid, which provides global observation meteorological data at 0000, 0600, 1200 and 1800 UTC and forecast data at 0300, 0900, 1500, and 2100 UTC (http://nomads.ncep.noaa.gov/pub/data/nccf/com/gfs/prod/). During the simulation, 1 unit mass of particles considered as an air sample was released from the observation site at 150 m above ground level. The spatial distribution of the footprint region of the air samples was calculated on the 5 days of backward simulation considering flow meteorology, turbulent motions, the sub-grid terrain effect and Earth's water cycle. Besides, a footprint region of air parcels of interest were simulated used the HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory Model) Trajectory Model developed by NECP (National Centers for Environmental Prediction) and NCAR (National Center for Atmospheric Research), which is based on the Lagrangian transport model (https://ready.arl.noaa.gov/HYSPLIT_traj.php) (Stein et al., 2015). The dataset provided for HYSPLIT is the global reanalysis data in GDAS format that was stored for 5 weeks per month (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1). It produces meteorological data four times a day, namely, at 0000, 0600, 1200, 1800 UTC, and the horizontal resolution is 2.5° lat × 2.5° lon. The vertical direction is 17 floors, ranging from the ground surface to 10 hPa. Elements, including wind, temperature, humidity, potential height and ground precipitation, are provided. During the simulation, the trajectory ensemble option starts multiple trajectories from the first selected starting location. Each member of the trajectory ensemble is calculated by offsetting the meteorological data by a fixed grid factor (one grid meteorological grid point in the horizontal and 0.01 sigma units in the vertical). Air samples were released at 150 m above ground level from LAPC, and the simulation time of the backward trajectory is 5 days.

3 Results and discussion

3.1 Size distribution of ambient aerosols

Hourly-averaged volume size distribution of aerosol from 29 November 2015 to 29 July 2016 is shown in Figure 2a. The particle size was derived according to the calibration curve between the forward scattering intensity and standard spherical particles (Figure S3). Mass concentrations of particle matters were reconstructed on the basis of number concentration of particles measured by POPC and particles density. The particle density was assumed to increase linearly from 1.77 g/cm^3 (0.5 μm) to 2.2 g/cm^3 (10 μm). To test the accuracy of the POPC detection, the $PM_{2.5}$ and PM_{10} inverted by POPC were compared with the observed data from the Olympic Sport Centre state control station (Figure S4). The correlation coefficients are 0.91 and 0.89 (significance level: 0.001) for $PM_{2.5}$ and PM_{10}, respectively. The result also compared well with commercial optical particle counter (KC52, RION, as shown in Figure S5) especially in the coarse mode size range observation.

It can be seen in Figure 2a that volume size distribution generally had two size modes during the whole observation periods. The occurrence of fine mode (peak at ~1 μm) was accompanied with increase of RH. The coarse mode (4~8 μm) mainly

occurred in the spring when eastward transport of dust events were significant (Lue et al., 2010). According to the POPC observations, totally five main dust episodes (March 3-5; March 16-22; March 30-31; April 9-10; April 28-May 1) happened at the site during observation period. For the cases on March 3-5 and March 16-18, volume size distribution of ambient particles showed two peaks in both fine and coarse mode (Figure S6), suggesting the interaction of anthropogenic pollutant and dust particles, and high possibility of existence of internally mixed dust particles (discussed in section 3.5). In winter 2015, POPC observed anthropogenic-dominant pollution cases for five times. All of them was related to high emissions for residential heating purpose and unfavourable air diffusion circumstance (Wang et al., 2015). In summer, the volume concentration of aerosols in all size mode were significantly low because of relative moderate anthropogenic emission, better diffusing boundary layer condition and frequent precipitation. Besides, relative strong turbulence in the Planetary Boundary Layer also increase the dry deposition processes of particles.

3.2 Seasonal patterns of d δ value of ambient aerosols

Figure 2b illustrates the size-resolved δ value as a function of time during observation period. The δ value of particles increased significantly with the size increases. Episodes influenced by mineral dust could be easily discerned due to the increase in both the volume concentration of coarse mode aerosols and the δ value of fine mode aerosols. In general, the δ value of particles in urban Beijing had prominent seasonal variability with a summer-low and spring-high pattern due to the different composition, origins of aerosols and atmospheric meteorology at the site. The averaged δ values of particles in both fine and coarse mode was highest in March 2016 (0.26) and lowest in July 2016 (0.19). This seasonal variability was much obvious in fine mode particles. Figure 2c shows temporal variations of hourly and monthly averaged δ values for the typical particle size at 1 μm and 5 μm, the error bar depicts the monthly averaged deviation. For fine particles at Dp ＝1 μm, their δ values in winter and summer were 0.09 ± 0.01 and 0.07 ± 0.01 respectively, however they could increase up to 0.2 as they were subject to dust events in spring. The phenomena of coexistence of fine mineral dust with anthropogenic pollutants has been reported by electro-microscopic studies in literatures (Li and Shao, 2009;Li et al., 2011). The daily averaged δ at Dp ＝ 5 μm varied significantly between 0.12 ~ 0.4 with a monthly mean value of 0.3 ± 0.05. We could discern that their δ values for 5μm particles in winter could decrease dramatically down to 0.15, however they had a much small deviation in spring. It was because water-soluble anthropogenic pollutant in winter and summer was substantial. Although dV/dlogDp of coarse mode particles was comparatively low, heterogeneous processes on the surface of particles in the coarse mode at high RH condition was inevitable, which may result in decrease in δ value.

3.3 δ value of aerosols from different origins

The dependence of the hourly-averaged δ value of particles at Dp = 1 μm and Dp = 5 μm on the wind speed and directions in different seasons are plotted in Figure 3. It can be seen that, the δ values at the both size increased when the observation site

was prevailing northwest wind, almost irrelevant to the season. For the particles at Dp = 5 μm, δ values in all direction in spring were generally higher than those in winter and summer because of impact of mineral dust aerosols, and the δ values also generally increased with wind speed (Figure 3b), implying of impact from re-suspended road dust or floating dust at strong wind condition. For particles at Dp = 1 μm, it was only during northwest wind period that the δ values were 40~50% higher than other direction (Figure 3d-f). It demonstrated that the morphology of particles in the fine mode were altered significantly only during dust event. We noted that particles with low δ values in all size mode was observed when the site was prevailing southeast wind in summer, indicating of presence of large fraction of spherical particles, and high ambient water content greatly contributed to deliquescence process of soluble component in the atmosphere (Figure 3c and 3f).

In order to understand the region source of air masses in different pollution types and further explain the seasonal characteristics of δ values of atmospheric aerosols, the footprint of the air mass in typical anthropogenic pollution cases and mineral dust dominant cases were analysed. Generally speaking, $PM_{2.5}/PM_{10}$ ratios have been used for identifying the sources of primary pollutant (Chan et al., 2005;Pérez et al., 2008). Higher ratio was generally ascribed to anthropogenic related secondary particles (sulfate, nitrate etc.), and a lower ratio indicate significant contributions mainly re-suspended or fugitive mineral dust particles due to some mechanical processes (Chan and Yao, 2008;Akyuz and Cabuk, 2009;Xu et al., 2017). Here, the specific pollution incidents were identified based on the size-resolved volume distribution and δ values. The criterion of $PM_{2.5} > 250$ μg/m^3 and $PM_{2.5}/PM_{10} \geq 0.8$, and $PM_{10} > 150$ μg/m^3 and $PM_{2.5}/PM_{10} \leq 0.4$ were chosen for heavy anthropogenic pollution-dominant and dust-dominant cases, as shown in Table 1.

The 5 days backward trajectories were calculated from HYSPLIT ensemble calculations, resulting in 27 members for all-possible offsets around the release point. The different directions from which the air mass originated to Beijing are shown in Figure 4. For mineral dust-dominant episodes, the air mass mainly originated from large areas in western Mongolia and Gobi Desert, and the footprint pattern represented comparatively large dust loading in the atmosphere in spring. While for anthropogenic pollution-dominant period, air mass passed through Beijing-Tianjin-Hebei region where anthropogenic emission was significantly strong. To note that, the RH along the trajectories was 13.9 % averagely during dust-dominant dominate cases (mostly in spring-time) and 87.6 % in anthropogenic pollution-dominant cases (mostly in winter-time). It means that origin of aerosol particles and their interaction with water vapor as well as consecutively heterogeneous reactions can lead to pronounced morphological changes of particles.

3.4 δ variability of atmospheric aerosols in clean and substandard days

Figure 5a shows the number of substandard days that daily-averaged $PM_{2.5}$ exceed 75 μg/m^3, the secondary standard of Chinese Ambient Air Quality Standard. Figure 5b shows the mean mass concentrations of $PM_{2.5}$, PM_{10} and $PM_{2.5}/PM_{10}$ ratio in substandard days. About 26.7% of substandard days was featured by high atmospheric loading of coarse-mode particles ($PM_{2.5}/PM_{10} < 0.6$) in winter-spring time. There were 18 substandard days on December 2015 with mean values of 199.6 μg/m^3,

240.1 μg/m^3 for $PM_{2.5}$ and PM_{10}. A high $PM_{2.5}/PM_{10}$ ratio (0.83) suggested that anthropogenic pollutants were dominant. We found that, median δ value of the particles at Dp = 5 μm (0.27) during substandard days in winter of 2015 was 12.9% lower than that (0.31) on clean days, as shown in Figure5c. It indicated that during the substandard days the dust particles were more likely to be modified in shape due to the coexistence of huge amount of pollutants. The second most pollution days occurred in March (15 days) and April (14 days), and the $PM_{2.5}/PM_{10}$ was 0.67 and 0.65 respectively. The δ value of particles at Dp =5 μm particles were almost the same for substandard and clean days. This demonstrated that almost all of the mineral dust particles were in irregular shape.

The peak of δ value of the particles at Dp = 1 μm particles occurred in April, 0.15 ± 0.03 (0.13 ± 0.02) in substandard (clean) days (Figure 5d). The high 90th percentile value indicated the observation site was influenced by several intensive dust events in spring. Anthropogenic pollution was dominant in substandard days in summer that $PM_{2.5}$ account for 0.76 and 0.87 in PM_{10} in June and July. δ value of aerosols at Dp = 1 μm and 5 μm in summer were 0.07 ± 0.01 and 0.27 ± 0.03, and there were no apparent different for substandard and clean days. This was because under the humid and high oxidizing environment in summer-time, catalysing process (Nie et al., 2012;Dupart et al., 2012) on the surface of mineral dust aerosols affect the hygroscopicity of dust and affect evolution of particle morphology. Previous studies found that mineral dust coexisting with NO_x can promote the conversion of SO_2 to sulfate (He et al., 2014). Nie et al. (2014) also found that mixed plumes provide abundant reactive species, and dust-induced photocatalytic reactions accelerate oxidisation in SO_2 and volatile organic compounds (VOC_s). This implies that pollution days in North China was great possible to induce internal mixing of dust and pollutant, especially in high humid atmospheric environment, and dust-related heterogeneous processes on the dust surface can aggravate the deterioration of air quality as a feedback.

3.5 δ value of different types of aerosols

Li et al. (2011) showed that surface of mineral dust aerosols provide a suitable space for heterogeneous reactions with gaseous pollutants, leading to the changes in size, shape and chemical components. On the basis of depolarization properties of single particles, the evolution of mixing state of dust particles could be estimated properly. Here, three pollution cases were carefully chosen considering the variation of both the volume size distribution and δ value anthropogenic pollution, typical dust-dominant case and mixed type pollution period (coexistence of anthropogenic pollutants and dust particles in the atmosphere that caused severe air pollutions). Note as “case A” on December 22-23, 2015, “case B” on April 9-10, 2016 and “case C” on March 4-6, 2016 respectively. In this study, we simulated 5-day footprint region of air mass based on FLEXPART model. The inert particles were released at 0900 UTC each day in 23 December 2015, 9 April 2016 and 4 March 2016. The releasing point was at LAPC in Beijing at 150 m above ground level. The footprint regions for the three cases are shown in Figure 6a-c. The variation of volume concentration of particles as a function of both δ value and particle sizes are depicted in Figure 6d-f. For

better comparison, the standard volume concentration was normalized to maximum value = 1 using the formula: normalized value = (truth value – minimum) / (maximum - minimum).

3.5.1 Anthropogenic pollution-dominant period

In case A, daily-averaged $PM_{2.5}$ was 281.3 μg/m^3 with a $PM_{2.5}/PM_{10}$ ratio of 0.72. The volume concentration of particles in this period had a peak in the submicron range, with a δ value of < 0.1 (Figure 6d), reflecting of predominance of secondary formation pollutants. As shown in Figure 7, δ value of particles with Dp less than 2 μm were normally less than 0.12 and increased gradually to ~ 0.27 (Dp > 4 μm), implying of influence from dust aerosol, even in typical anthropogenic pollution-dominant period. Footprint of air mass covered west of North China Plains, which are characterized by heavy industrialization and high emissions. (Zhao et al., 2012). As suggested by many previous studies (Ilten and Selici, 2008;Wang et al., 2013;Zhang et al., 2016;Chang and Zhan, 2017), unfavourable meteorological conditions played an important role in the occurrence of severe pollution. In case A, the RH was ranging 80% ~ 90% from 0000 LST on December 22 to 1200 LST on December 23 and the diffusion condition was weak (wind speed < 1.5 m/s) (Figure S7). Air mass was mostly stagnant within a high emission region, resulted in substantially formation of secondary pollutants from primary pollutant precursors in the atmosphere (Wang et al., 2014).

3.5.2 Dust dominant case

In the typical dust-dominant case occurred on April 9-10, 2016. The main body of dust plume was arrived at the site on April 9 with a daily-averaged PM_{10} was 273.6 μg/m^3, and $PM_{2.5}/PM_{10}$ ratio of 0.33. Footprint analysis shows that air mass originated from western Mongolia and transport rapidly by a strong wind (~ 5 m/s). The vertical structure of the dust extinction coefficient determined by ground-based Lidar measurement indicated that the dust plume present a layered structure while arrive in the observation site (Figure 8 a-b). The lowest dust layer that in the altitude of < 700 m first arrived at Beijing at 0600 LST on April 9 containing huge amount of coarse mode particles with hourly averaged $PM_{2.5-10}$ reached 395 μg/m^3 at noon. This layer was lifted up and become mixed with the dust layer at height 1 km on afternoon April 9. Impact of anthropogenic pollutants on this dust event was weak for less pollutant. POPC analysis shows no feature of internally mixed dust particle. The dVdlogDp has a peak at 5 μm with a mean δ value of 0.34, consistent with the result in western Japan (Pan et al., 2015). The δ values of aerosols in coarse mode were found to be about 3 times larger than calibration result (0.07 - 0.1) for standard spherical aerosols (Figure S2), which suggest the coarse mode particles at the site was great non-spherical. To note that, the δ values (0.18 ± 0.02) of particles in fine mode was twice higher than that during anthropogenic pollutant-dominant case, indicating thepresence of irregular dust particles.

3.5.3 Mixed pollution case

The vertical profile of extinction coefficient and depolarization ratio during March 4-6, 2016 are shown in (Figure 8), and evolution of depolarization properties of dust particles were simultaneously observed by POPC. During the occurrence of the

dust event on March 4, daily-averaged PM_{10} was 376.3 μg/m^3 and $PM_{2.5}/PM_{10}$ was 0.19, and the dust plume existed at an altitude up to 3.5 km on March 5. At 1200 UTC March 5, the main body of dust plume descended to an altitude of less than 1.5 km. The $PM_{2.5}/PM_{10}$ concurrently increased to 49 % because of mixing with higher concentration of pollutant and rapid gravitational settlement of large particles. δ value (0.28) for particles at 5 μm decreased 17.6% on midnight March 5 compared to 0.34 on March 4 (Figure S8). The 5 day back trajectory implied that the air masses were from a convergence air flow of deviating northwest and south streams, being affected by both mineral dust in Midwest China and emissions of anthropogenic pollution in East China. The dV/dlogDp in case C has two peaks in both submicron range (0.9 μm) and coarse mode range (4.5 μm) (**Figure 7**a) corresponding to δ values of < 0.1 and 0.3 ± 0.2 on Figure 6f. To be noted, the averaged δ value for particles at Dp larger than 4 μm was about 0.3, that was 11.8% lower compared to case B (Figure 7 b). T-matrix simulations indicated that the aspect ratios of the dust particles were estimated to be 1.48 ($\delta = 0.30$) (Figure S9), presuming that the dust particles was in spheroid shape.

Figure 9 shows the scatter diagram of the averaged particle depolarization ratio (PRD) at 532 nm versus ratio of backscattering coefficient at 1064 nm/532 nm on the two dust cases and an anthropogenic-dominant cases according to Lidar measurement. For comparison, the results from an observation study of mixed type pollution in Seoul (Sugimoto et al., 2015) was also plotted in the figure. As shown, the daily averaged δa value for dust aerosols in case C was 0.26 ± 0.1 with a backscattering averaged color ratio of 1.21 ± 0.39. It was ~ 36.6% lower than the δa value in case B (0.41 ± 0.14), but the color ratio (1.32 ± 0.14) was relatively consistent with case C. The air pollution aerosols had a color ratio of 0.32 ± 0.25 and δa value 0.1 ± 0.02. The results of δa value in pure dust and polluted dust plume was similar to the study in Seoul, even though the coarse mode aerosols observed in Seoul had a smaller size range due to gravitational settlement of large particles during longer range transport. One possible reason for the decrease in δa value was that the mineral dust did be involved in internal mixing process through trapping or heterogeneous reactions. However, we cannot eliminate the possibility that the decrease of δa was just caused by the external mixing of huge amount of dust and anthropogenic pollutants. Because Lidar observations only provide an averaged δ value of all the particles in the detecting volume, the external mixing of dust particles with substantial amounts of spherical secondary anthropogenic particles could also result in a lower δa value. Which means the environmental impact of transported Asian dust in polluted areas in East Asia may be underestimated since shadow area of Lidar in properly recognize polluted dust particles.

3.6 Implication on heterogeneous processes on dust particles

Coarse mode particles observed in North China was reported to contain large amount of Ca (Yuan et al., 2006;Geng et al., 2014), generally existing in the form of $CaCO_3$ (component of calcite), which is the most widely investigated component of mineral dust particles. Previous studies indicate that the interaction between water vapor and $CaCO_3$ particles was significant. Hatch et al. (2008) showed that the mass of adsorbed water on $CaCO_3$ is equal to ~8% of the mass of dry $CaCO_3$ particles at

78% RH. Studies also found that 1-9 monolayers of adsorbed water are formed on $CaCO_3$ particles at 50% - 95% RH (Gustafsson et al., 2005;Ma et al., 2012). Actually, the composition of dust particles was complex, may also contain SiO_2 (component of quartz, illite, feldspar), Al_2O_3 (illite) and CaO (feldspar), aluminosilicate (kaolinite) etc. These substances are comparatively insoluble and not sensitive to water vapor in the air (Tang et al., 2016). This means the heterogeneous reactions and trapping process on the dust surface was closely related to the dust sources and residential time in the atmosphere. For example, Wang et al. (2011) found that the ratio of Ca component in mineral dust in Beijing was high when air mass was from Loess Plateau, while chemical components of dust from desert/Gobi areas contained more crustal element oxides such as SiO_2, Al_2O_3, Fe_2O_3 (Ta et al., 2003). Dust with inert component require longer transport or residential times before morphology changing.

Here, we investigate the vital roles that ambient air humidity and air pollution content in the air played in the morphological changes of dust particles in Beijing area. The high $PM_{2.5}/PM_{10}$ ratio indicate that the components of soluble inorganic salts or reactive gases loading in the atmosphere was greatly possible to be relatively high, and in the case of high humidity. It also indicate a high collision probability between pollutant and dust. Therefore, morphology and corresponding δ value of particles should be affected. According to our 8-month observation in Beijing, the δ value had a general trend of increasing with particle size but decreasing with the $PM_{2.5}/PM_{10}$. The decrease degree was not equal for different particle size: for particles at Dp = 5 μm, δ value was ~ 0.3 while $PM_{2.5}/PM_{10}$ was less than 0.6 and decreased ~ 20 % when $PM_{2.5}/PM_{10}$ increased to 1; for particles at Dp = 1 μm, δ value decreased by 42.9% from 0.14 to 0.08 (Figure S10). In fact, atmospheric humidity played a vital role in the observed δ value decrease. Figure 10 shows the relationship between the δ value of dust particles (Dp = 5 μm), vaper content (RH) and $PM_{2.5}/PM_{10}$ in the air. It can be seen, δ value of particles in coarse mode decreased as $PM_{2.5}/PM_{10}$ ratio increased, especially under high RH conditions. For particles at Dp = 5 μm, their δ value was generally 0.28 - 0.35 when the ambient air was dry (RH < 10%), and it decreased by ~ 28.6% when the ambient RH increase to > 90%. δ value of for particles at Dp = 3 μm decreased by ~ 36.4% (Figure S11). For small dust particles, the interaction of water vapor and pollutant on the surface was more obvious in the change of morphology and reduction in δ value. This negative relationship reflected the spheroidization of the dust particles as a result of hygroscopic properties of mineral dust aerosols. Polluted air generally contains abundant HNO_3 (Liang et al., 2007; Shi et al., 2014; Bytnerowicz et al., 2016). Li and Shao (2009) reported that mineral particles are mainly covered with coating including $Ca(NO_3)_2$, $Mg(NO_3)_2$, and $NaNO_3$ in North China. Deeper interaction between alkaline mineral dust and reactive acidic gases and the trapping process of atmospheric secondary inorganic salt modified the hydrophilic state of dust aerosols. According to Sullivan et al. (2009), pure $CaCO_3$ with diameter of ~ 2 μm need a supersaturation values of 0.6 to 0.9 to be activated, while the soluble salts of $Ca(NO_3)_2$ and $CaCl_2$, are more likely to be activated with supersaturation values ranging from 0.07 to 0.4. This means that the more dust particles become involved in chemical mixing or coagulation process, the more easily they become hydrophilic and are incorporated into cloud processes and effect regional/global climate (Shi et al., 2008;Koehler et al., 2009). Besides, the theoretical simulation of

Ishimoto et al. (2010) indicated that a change in refractive index could also affect the δ value, the simulation results depicted that the δ value showed a levelling off tendency at 0.31 ± 0.02 for the coarse modal particles (Figure S12), which means variations in the particle's refractive index can only explain limited depolarization variability (6%). At present, observation studies for single particle δ value combined with aerosol chemical composition analysis are still scant. To clarify the chemical process happened during mix pollution, more observational and experimental results in lab are needed.

4 Conclusions

Interactions between mineral dust and anthropogenic pollutants has great impact on physical properties and climate effect of dust particles in East Asia. In this study, a long-term observation of atmospheric aerosols in the megacity of Beijing from 29 November 2015 to 29 July 2016 was performed using an Optical Particle Counter equipped with a Polarization detection module (POPC). Combined with trajectory and dispersion model analysis, the temporal variability of depolarization characterization of aerosols, mixing state of dust particles, and their effect on local air quality were investigated. The major conclusions are as follows: (1) The depolarization ratio (δ) of particles in urban Beijing has significant seasonal variabilities due to different components and origins of aerosols. The averaged δ value of particles in both fine and coarse mode was highest in March 2016 (0.26) and lowest in July 2016 (0.19). The seasonal variation of the δ value in fine mode particles (< 2 μm) showed a more pronounced winter-spring high and summer low pattern owing to emissions as well as weather conditions in different seasons. During the substandard days in Beijing, the decrease of δ value of particles in coarse mode implied of internal mixing of mineral dust and pollutant, especially under humid atmospheric conditions. (2) Although both Lidar and POPC could identify impact of dust aerosols, single-particle based depolarization measurement could provide more accurate information about the sphericity of particles, which could indicate the possible modification of morphology of particles as a result of mixing processes. Case studies on the evolution of a typical Asian dust event, we found that the δ of dust particles obtained by POPC could decrease by 17.6% with mixing effect of polluted. Volume δ value measured by Lidar also showed an obvious decreasing trend, however the interference of substantial amount of small spherical anthropogenic pollutant make it difficult to evaluate their interactions. During a typical polluted dust event, the shape of mineral dust can be modified as a result of heteorogeneous processes. In this study, δ value of particles at Dp = 5μm decreased by 28.6% when RH increase up to 90%. (3) These findings bring attention to comprehensive study on the complex mixing processes and possible mechanisms of morphological change of Asian dust. In addition to Lidar and satellites, single-particle based depolarization measurement could provide a reliable detection method for separating aerosol types and investigating mixing processes of aerosol along the transport. This study also indicate an urgent need for a realistic optical model for predicting the physical changed of Asian dust particles during transport.

Acknowledgement

This work was supported by the National Natural Science Foundation of China (Grant No. 41675128 and 41620104008). Air quality data was provided by ministry of environmental protection, and meteorological data was from the Chinese meteorological department. The author also thank the NCEP for provision of GDAS dataset, NILU for develop FLEXPART Transport and Dispersion model, and NCAR for exploiting HYSPLIT Trajectory Model collaborating with NCEP.

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Table

Table 1: Crucial air quality indices for selected pollution cases in Beijing.

Selected pollution cases	Year/Month/Day	$PM_{2.5}$ ($\mu g/m^3$)	PM_{10} ($\mu g/m^3$)	$PM_{2.5}$/ PM_{10}	AQI
Anthropogenic-dominant cases	2015/12/01	490	578	0.85	476
	2015/12/23	255	298	0.86	305
	2015/12/25	477	510	0.94	485
	2015/12/29	279	338	0.83	329
	2016/01/02	266	299	0.89	316
Dust-dominated cases	2016/03/05	58	290	0.20	170
	2016/03/06	73	182	0.40	116
	2016/03/28	70	195	0.36	123
	2016/04/09	54	245	0.22	148
	2016/04/10	41	192	0.21	121
	2016/05/05	62	153	0.40	102
	2016/05/06	57	182	0.31	116

Figures

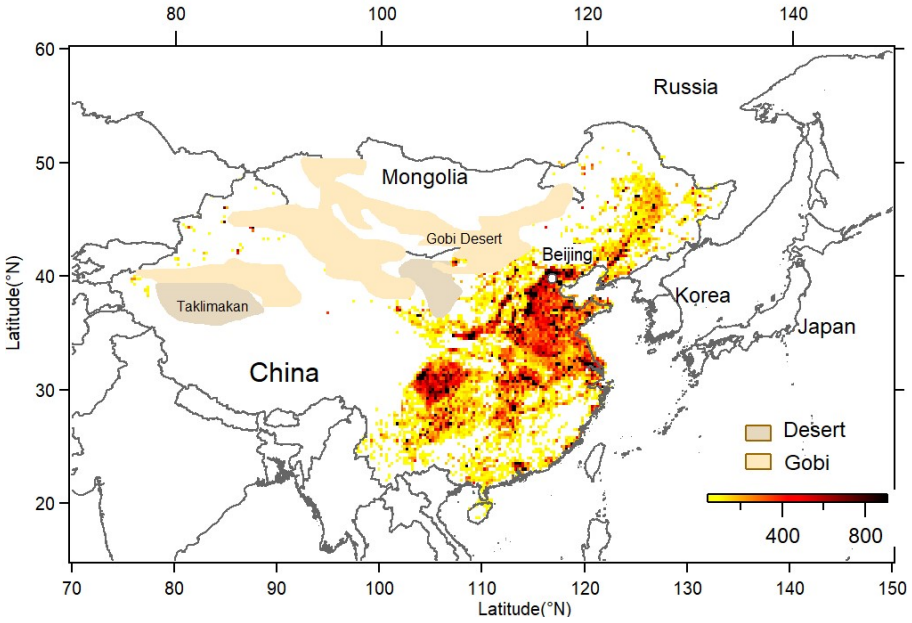

Figure 1: Geographical location of the observation site in Beijing, $PM_{2.5}$ emissions in China and the location of major deserts and the Gobi in East Asia.

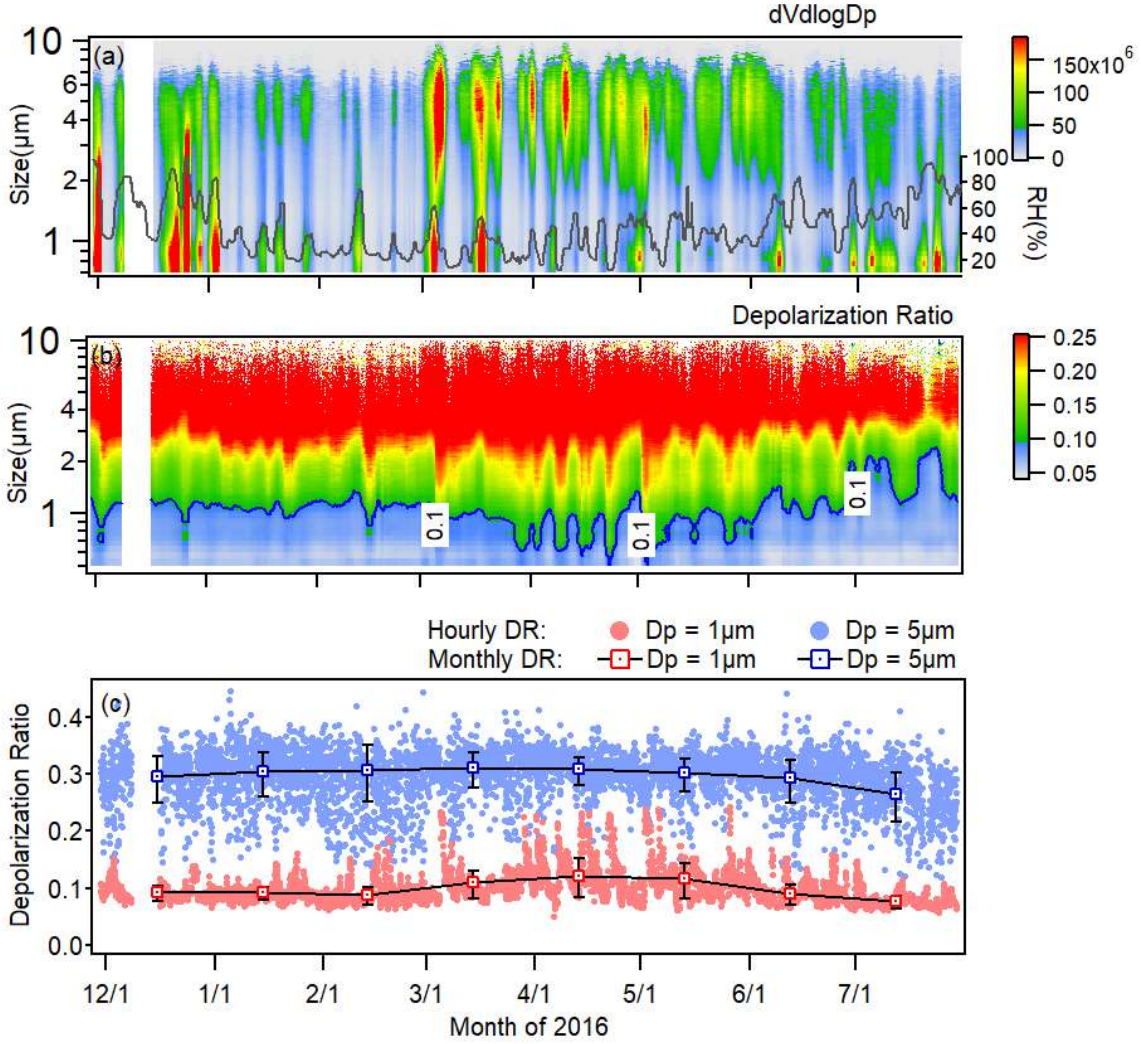

Figure 2: Time series of (a) volume size distributions and RH (gray solid line), (b) size-resolved δ value(Green solid line: δ = 0.1), and (c) hourly and monthly averaged δ values for fixed-size particles: Dp = 1 μm and 5 μm from 29 November 2015 to 29 July 2016. Error bars for monthly averaged δ in Figure 2c depict the monthly averaged standard deviation of value.

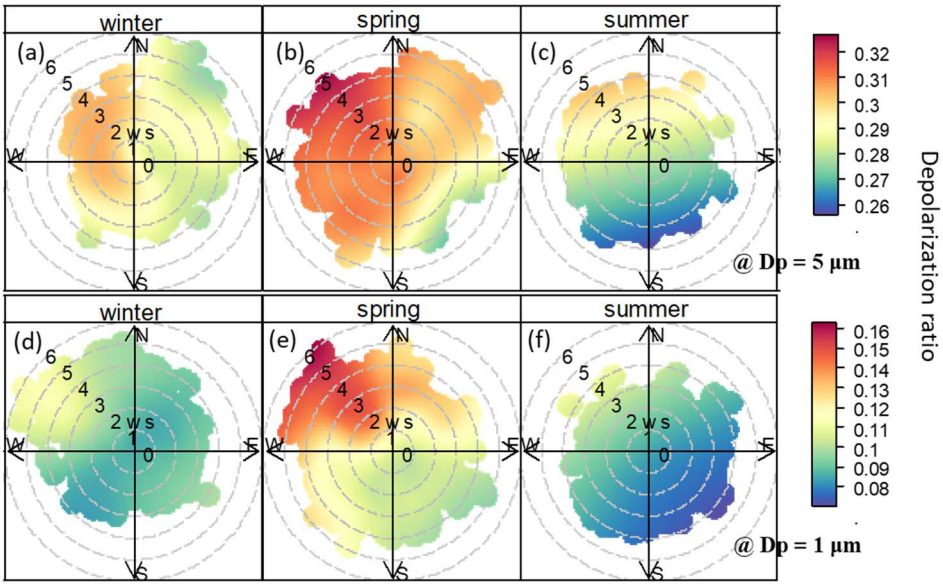

Figure 3. Dependence of the hourly-averaged δ value of particles at Dp = 5 µm and Dp = 1 µm on the wind speed and direction in winter (December, January and February: DJF) 2015, spring (March, April and May: MAM) and summer (June and July: JJ) 2016.

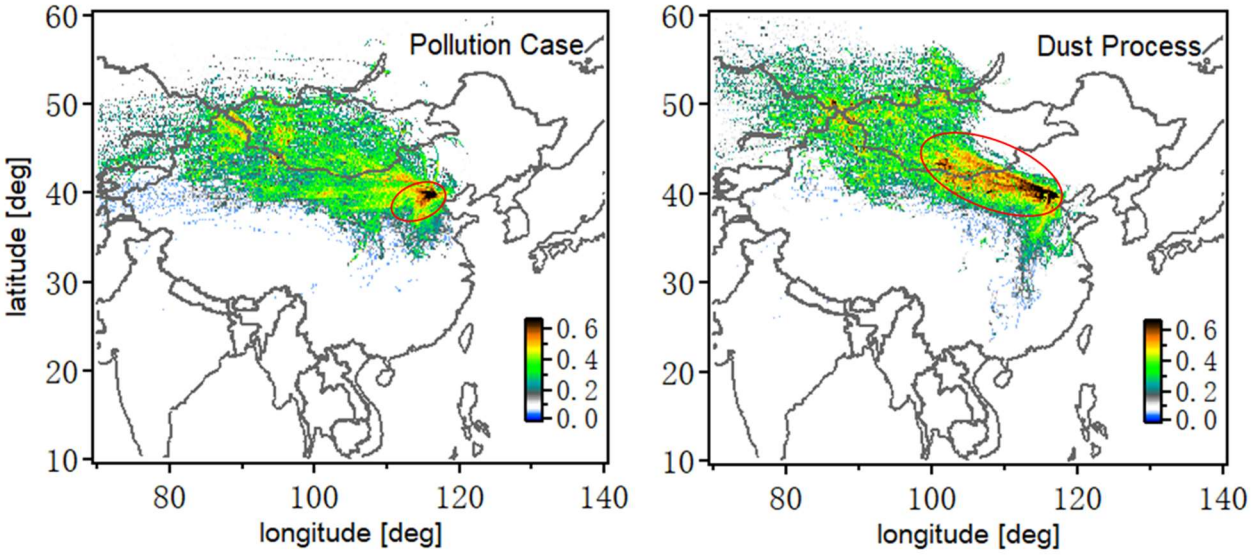

Figure 4: Proportion of different directions from which the air mass over Beijing originated in varying pollution types: severe anthropogenic pollution-dominant case (left), and dust-dominant pollution case(right). The red ellipse represents the major source region of air mass arriving at the site.

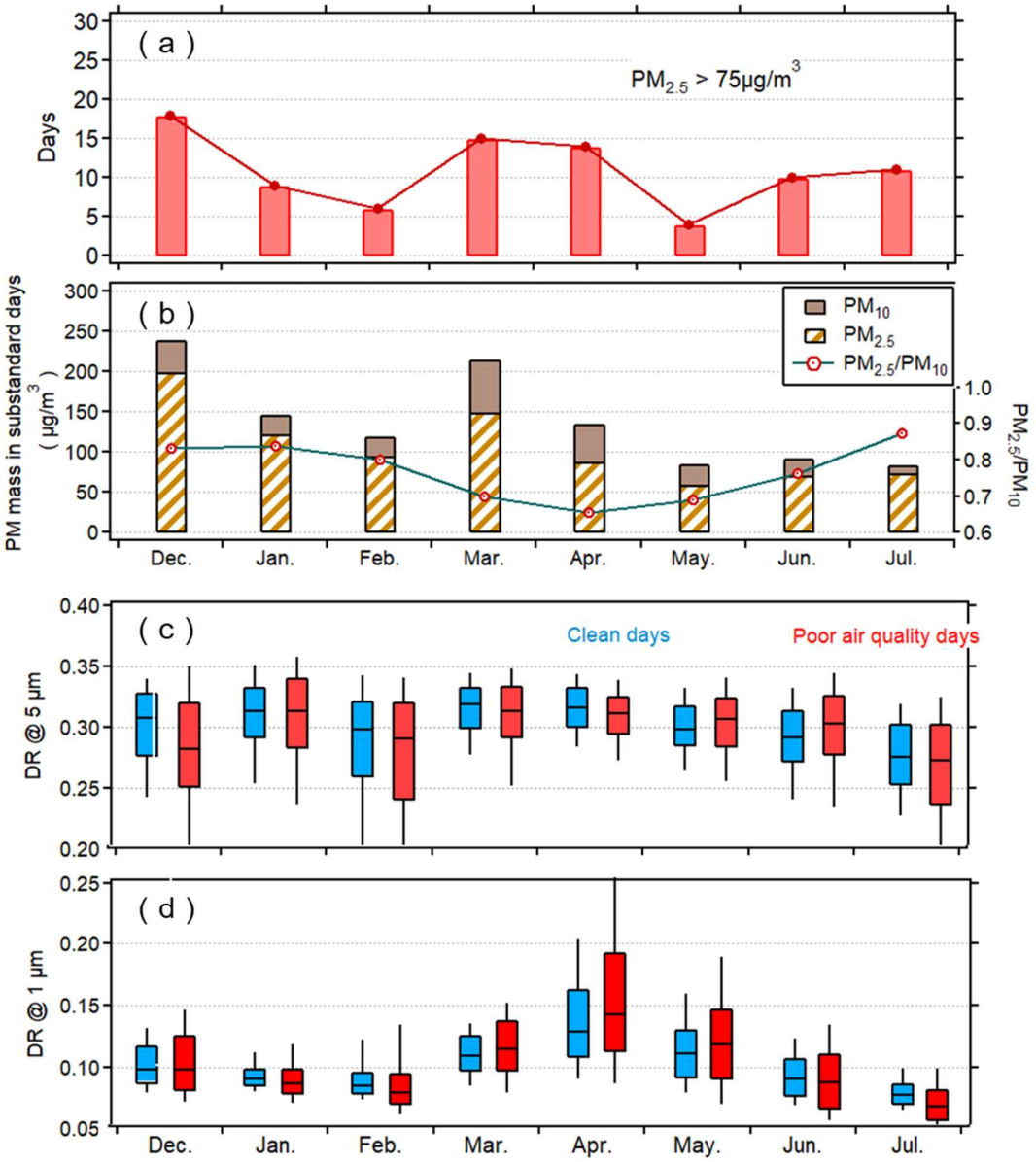

Figure 5. (a) The number of poor air quality days (daily-averaged $PM_{2.5}$ exceeding 75 μg/m^3, the secondary standard of Chinese Ambient Air Quality Standard of $PM_{2.5}$, (b) daily averaged mass concentration of $PM_{2.5}$ and PM_{10} in the poor air quality days, and ratio of $PM_{2.5}/PM_{10}$, box plot of particles δ value at (c) Dp = 5 μm and (d) Dp = 1 μm in clean days ($PM_{2.5}$ < 35 μg/m^3) and poor air quality days.

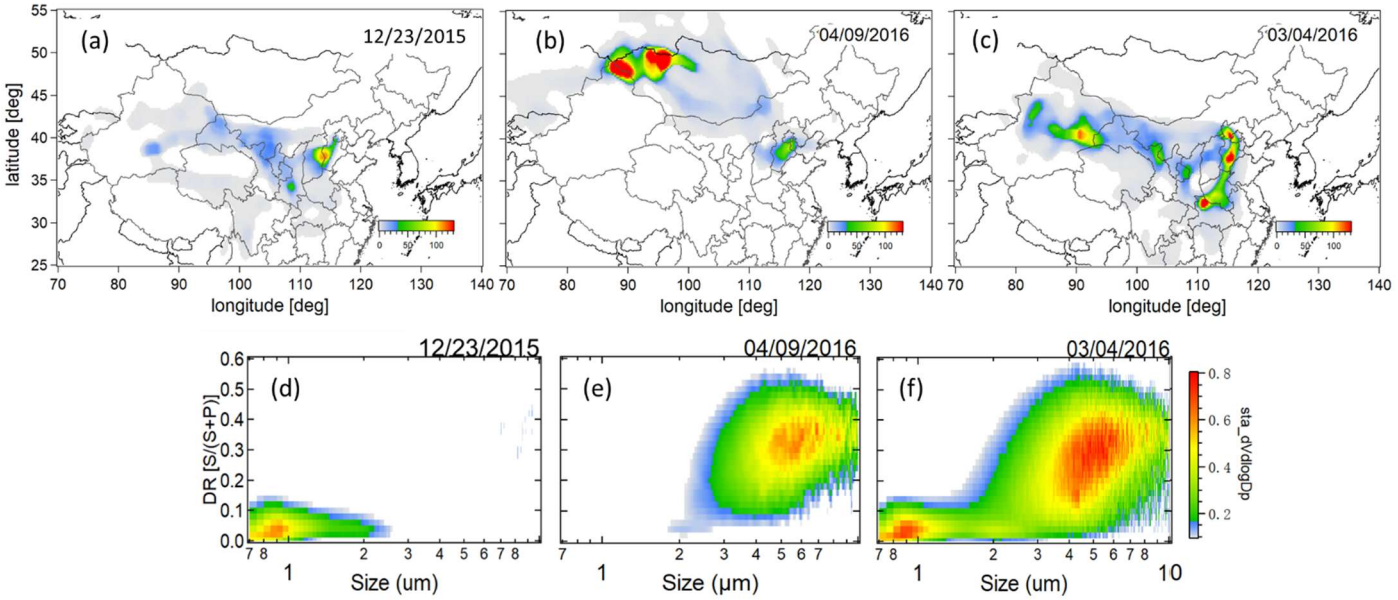

Figure 6. Backward trajectories from Beijing calculated by the FLEXPART dispersion model for (a) the anthropogenic pollution case, (b) the dust-dominant case, and (c) the mixed pollution period. Variation in the standard δ value as a function of particle size for corresponding episodes: (d), (e), and (f).

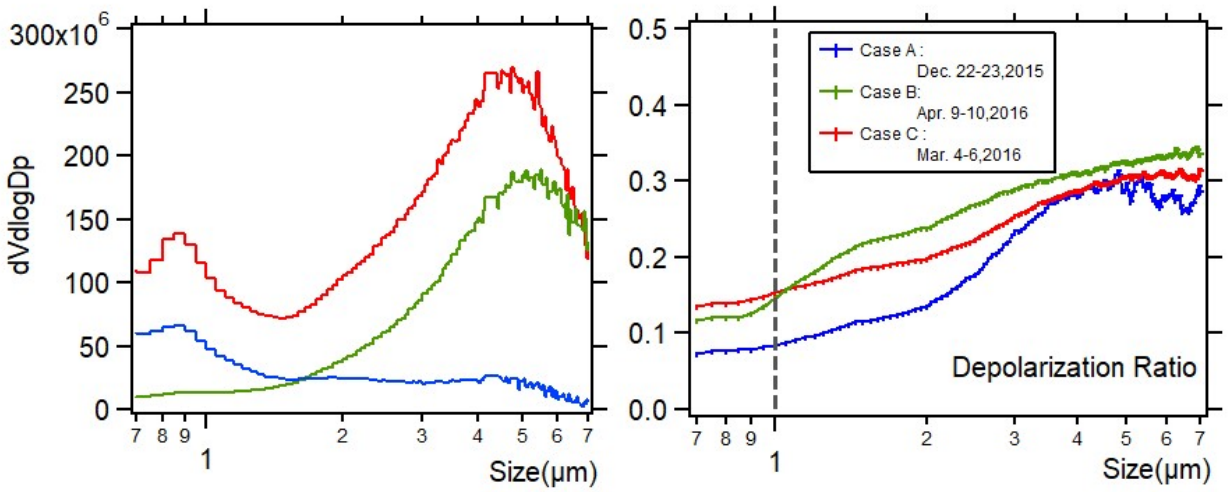

Figure 7. Volume (left) and δ value (right) size distribution of aerosols observed in the study cases.

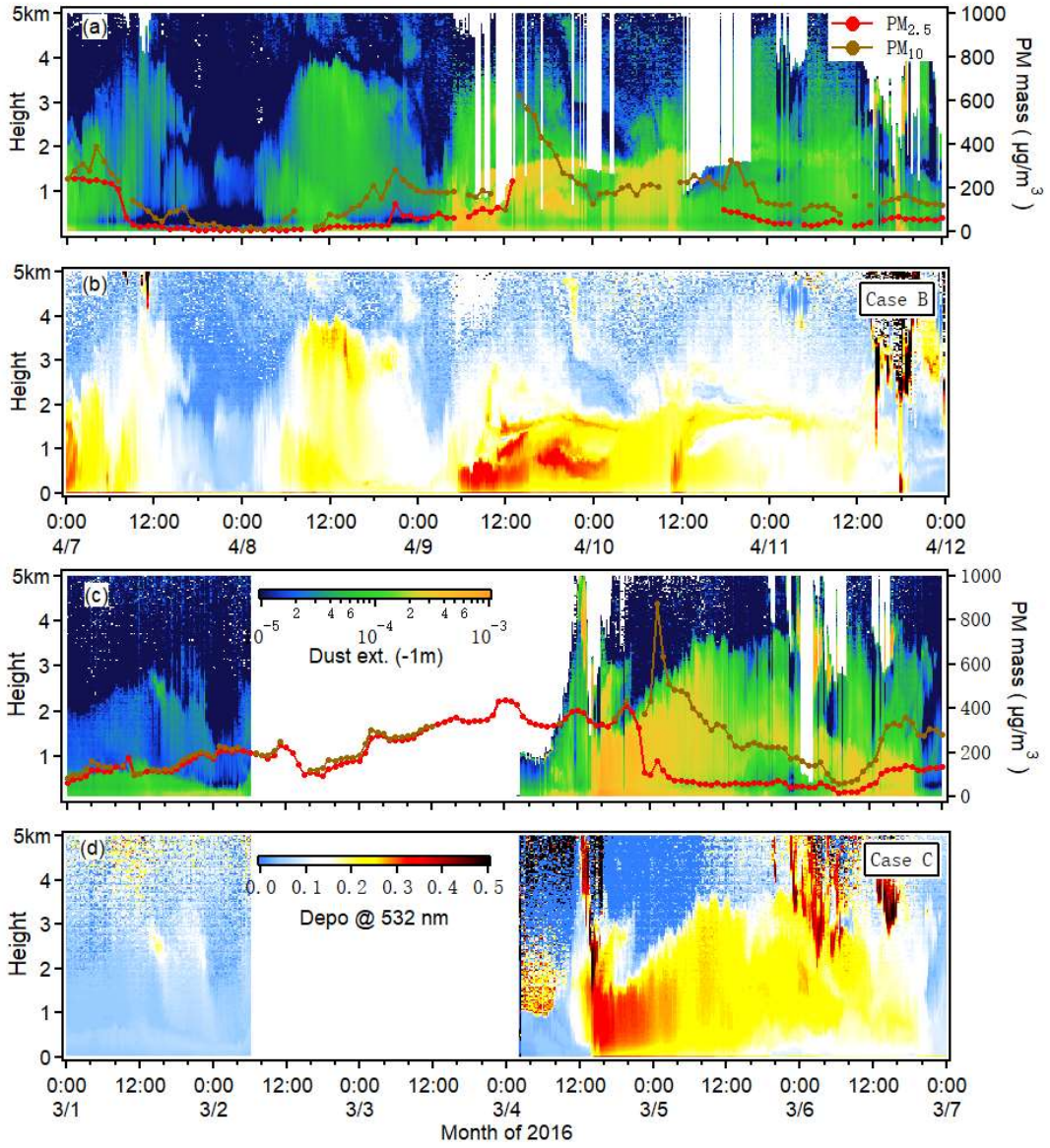

Figure 8. Time–height indications of the dust extinction coefficient and the δ value at 532 nm derived from polarization-sensitive Lidar measurement in Beijing in case B and case C

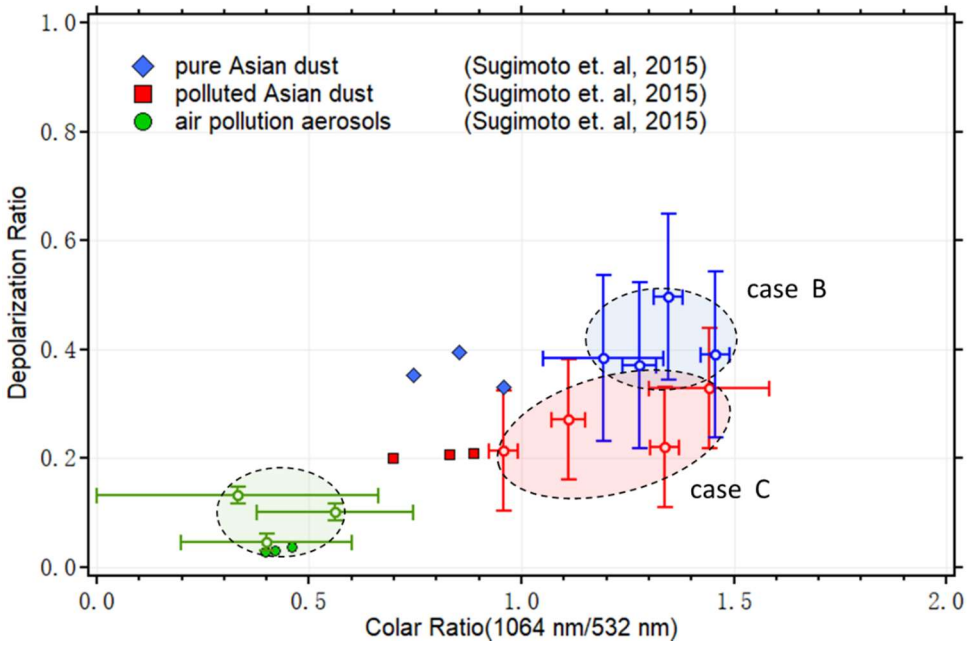

Figure 9. Scatter diagram between the backscattering color ratio (1064 nm/ 532 nm) and the particle depolarization ratio at 532 nm for case B (dust-dominant case) and C (polluted dust case) and an anthropogenic-dominant case. The error bars indicate estimates of statistical error. The observations results in Seoul in previous study are displayed in the plot.

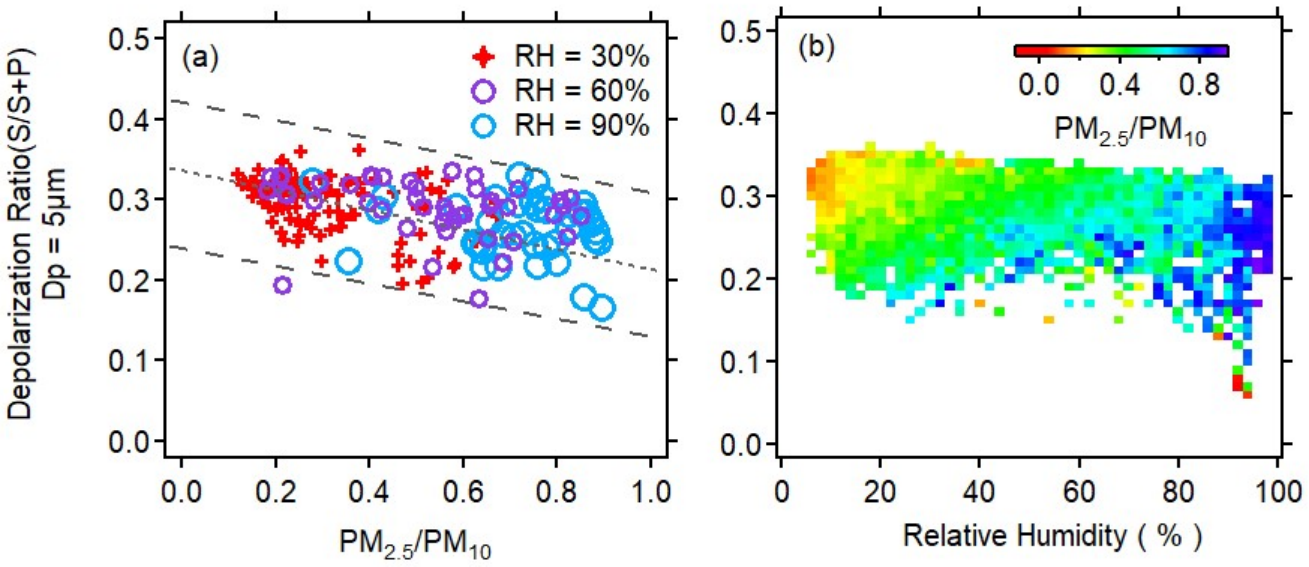

Figure 10. Scatter diagramof the relationship between the δvalue of dust particles (at DP = 5 μm), vaper content (RH) and $PM_{2.5}/PM_{10}$ in the air.