Letter to the referee #1

We thank the Referee #1 for the suggestive comments. We have addressed the comments carefully and revised the manuscript according to these suggestions.

Specific comments:

1. Line 107 - Line 109. The authors claimed that the South Asia Monsoon system is responsible for the transport of pollution from South Asia to HTP. However, many previous researches demonstrated that such long-range transport events actually occurred in winter or pre-monsoon seasons, mainly driven by the westerlies. So is there any more explanation?

   Response: As the referee has mentioned above, the westerlies is dominant for long-range transport of pollutants to western HTP, suggested by atmospheric black carbon measurement such as *Cao et al. (2009)*. Observations of air pollutants including metal elements and POPs also suggested that, their transport pathways to the HTP vary seasonally and regionally *(Liu et al., 2013; Wang et al., 2016)*. Summer monsoon is potentially important in the transport of air pollutants from southern and southeastern Asia, especially for southeastern TP *(Liu et al., 2013; Sheng et al., 2013)*. Additionally, in the tropospheric scale, monsoon system transport of air pollutant to HTP and the surrounding may play a more important role, suggested by satellite observations *(Randel et al., 2010)*. Then, we revised these sentences as bellow:

   "Anthropogenic emissions into this region occur occasionally and are dependent on local/large-scale atmospheric dynamics." (Lines 102, 103). "The Westerlies transport of air pollutant is dominant in the western TP *(Cao et al. 2009)*. Additionally, the South Asian monsoon system is one of the important atmospheric dynamics in the transport of pollutants to the southeastern TP from southern and southeastern Asia *(Liu et al., 2013; Sheng et al., 2013)*." (Lines 107-112).

2. Line 111, actually, for the impact of air pollution on the monsoon system, there are different perspectives by scientists (Ramanathan et al, 2005 and Lau 2006). So here I suggest the authors to rewrite the sentence to reflect this disagreement. Anyway, it highlights the importance to carry out more field observation of aerosols in this region.

   Response: We refreshed the sentences as "Studies have raised the different hypotheses, including

a suppression of the Southern Asian monsoon through a weakening of the meridional surface temperature gradient *(Ramanathan et al., 2005)* and an increase of regional monsoonal rainfall in northern India, the Himalayas, and the southern Tibetan Plateau (TP) through the "elevated-heat-pump" effect *(Lau et al., 2006)*." (Lines 113-117).

3. Line 157 - 159. Why did you think there was an altitude effect? I checked the altitudes of each station, it appears the altitude effect among the four stations is not evident.

   Response: We revised the sentences "The altitude effect may have also influenced the horizontal WS values, which were 2.7±1.1, 4.3±1.6, 3.4±1.4 and 1.1±0.7 m s-1 for the Ngari, QOMS, Nam Co, SET stations, respectively (line 157 – 159)." into the "The average horizontal WS values were 2.7±1.1 s^{-1}, 4.3±1.6 s^{-1}, 3.4±1.4 $s^{-1}$and 1.1±0.7 m s^{-1} for the Ngari, QOMS, Nam Co, SET station, respectively." (Lines 160-161).

4. Line 164 - 165. It is not easy to follow.

   Response: We changed the sentence with "Compared to other stations, the greater seasonal variability in T at the Ngari station, i.e., from the lowest value (-10.6±4.8℃) in December-February to the highest value (14.0±3.1℃) in June-August, can be explained by its far inland position and its attendant climate." (Lines 160-161).

5. Line 226, the authors wrote that ion chromatography (IC) was used to analyze the aerosol composition. However, in the main text, I can not find such description of the major ions data. It seems only the elemental data from ICP-MS was used.

   Response: Yes, only the elemental data measured by ICP-MS was used to calculate the mineral matter fraction. We refreshed the sentence into "We further assessed mineral matter content in fine particles by analyzing elements in $PM_{2.1}$ samples with inductively coupled plasma mass spectroscopy (ICP-MS)." (Lines 229-230). Because aerosol chemical is significant and need to be careful analyzed, detailed discussions are beyond the topic of this manuscript; instead, a following companion paper will present a comprehensive discussion of chemical compositions in size-segregated aerosols at these stations.

6. How did you calculate the crustal materials basing on the elemental data from ICP-MS? More

details should be presented, at least in the supplementary materials. As far as I know, the ICP-MS methods especially the acid digestion in the pretreatment have some difficulty to determine Si contents. If the crustal material account for 26%-29% of the total aerosol mass, so what is the constituent of the remaining fraction? Organic carbon and black carbon?

Response: The details in calculating mineral matter content were implemented as the Table S1 (please also see Table R1). As mentioned above, the acid digestion in the pretreatment has some difficulty in determining Si contents. Then in this manuscript, Si content was calculated according to the ratio of Si to Al in the upper continental crust recommended by *Rudnick and Gao (2014)*. Consequently, dust species account for ~26% and ~29% mass fractions in aerosol matter closure at the Ngari and QOMS stations (other constituents contain organic matters, secondary inorganic aerosol, black carbon and etc.).

Table R1. The method of mineral matter calculation in the fine aerosols at the HTP stations, following *Xin et al. (2015)*.

Source pattern	Abb.	Formula
Mineral matter	MM	$MM=CaO+MgO+Al_2O_3+Fe_2O_3+K_2O+Na_2O+SiO_2$ $=1.89(Al)+1.66(Mg)+1.21(K)+1.40(Ca)+1.43(Fe)+1.35(Na)+2.14(Si)$

Si content was calculated according to the ratio of Si to Al in the upper continental crust recommended by Rudnick and Gao (2014).

7. Line 240-241, what's the possible mechanism or process responsible for this distribution pattern? More explanation is needed.

   Response: We added the explanation "In previous studies, accumulation mode particles were observed at Mt. Waliguan observatory *(Kivekas et al., 2009)* possibly formed by the coagulation or condensation of smaller aerosol particles and the effect of dust emission to the coarser particles have been reported in the TP atmosphere *(Zhang et al., 2001).*" into the text (Lines 244-247).

8. Line258 and Line 339, QOM should be QOMS, and in figure 4, sation should be station.

   Response: We corrected them.

9. Line 296, How did you say the regional dust emissions contributed to the chemical composition of fine aerosols in Ngari station? Currently no data supports this point.

   Response: We changed the "regional" into the "local", according to our observed results (Line 302).

10. Line 315-316, the description of the MODIS aerosol fine-mode fraction data should be presented in the Section 2.3. (i.e. Methods of data analysis). Now only the MODIS land cover product was described in Section 2.3

    Response: We thank the Referee for the suggestion. We moved this sentence to the Methods of data analysis (Line 201-203) and then altered the sequent sentence (Lines 323-325)

11. Line 323, is there any more evidence to support your interpretation about the spread of ABCs in this region. Usually ABCs happened in the winter and pre-monsoon seasons, with much less air pollution in the summer monsoon season.

    Response: Asian tropospheric aerosol layer that covered the HTP was observed at the tropopause level by *Vernier et al. (2011)*, basing on the CALIPSO detection. It was associated with the Asian monsoon season in June, July and August. We refreshed the sentence into "Furthermore, such a spatial pattern was more notable for April-August, coinciding with the appearance of the reported Asian tropospheric aerosol layer during this period *(Vernier et al., 2011)*." (Lines 330-331).

12. AERONET should be acknowledged.

    Response: We thank the Referee for the suggestion and added the acknowledgement.

13. Page 37, the title of Figure 13 should be shortened as brief as possible. Some sentences regarding the satellite data analysis could be moved to the main text.

    Response: We updated the sentences as expanded information about Figure 13 and provide in Supplementary Materials (please see Lines 122-130 in SMs).

References

Cao, J. J., Xu, B. Q., He, J. Q., Liu, X. Q., Han, Y. M., Wang, G. H., and Zhu, C. S.: Concentrations, seasonal variations, and transport of carbonaceous aerosols at a remote Mountainous region in western China, Atmospheric Environment, 43, 4444-4452, 10.1016/j.atmosenv.2009.06.023, 2009.

Kivekas, N., Sun, J., Zhan, M., Kerminen, V. M., Hyvarinen, A., Komppula, M., Viisanen, Y., Hong, N., Zhang, Y., Kulmala, M., Zhang, X. C., Deli, G., and Lihavainen, H.: Long term particle size distribution measurements at Mount Waliguan, a high-altitude site in inland China, Atmos Chem Phys, 9, 5461-5474, 2009.

Lau, K., Kim, M., and Kim, K.: Asian summer monsoon anomalies induced by aerosol direct forcing: the role of the Tibetan Plateau, Clim. Dynam., 26, 855-864, 10.1007/s00382-006-0114-z, 2006.

Liu, B., Kang, S., Sun, J., Zhang, Y., Xu, R., Wang, Y., Liu, Y., and Cong, Z.: Wet precipitation chemistry at a high-altitude

site (3,326 m a.s.l.) in the southeastern Tibetan Plateau, Environ. Sci. Pollut. Res., 20, 5013-5027, 10.1007/s11356-012-1379-x, 2013.

Ramanathan, V., Chung, C., Kim, D., Bettge, T., Buja, L., Kiehl, J. T., Washington, W. M., Fu, Q., Sikka, D. R., and Wild, M.: Atmospheric brown clouds: Impacts on South Asian climate and hydrological cycle, Proc. Natl. Acad. Sci. U.S.A., 102, 5326-5333, 10.1073/pnas.0500656102, 2005.

Randel, W. J., Park, M., Emmons, L., Kinnison, D., Bernath, P., Walker, K. A., Boone, C., and Pumphrey, H.: Asian Monsoon Transport of Pollution to the Stratosphere, Science, 328, 611-613, 10.1126/science.1182274, 2010.

Sheng, J., Wang, X., Gong, P., Joswiak, D. R., Tian, L., Yao, T., and Jones, K. C.: Monsoon-Driven Transport of Organochlorine Pesticides and Polychlorinated Biphenyls to the Tibetan Plateau: Three Year Atmospheric Monitoring Study, Environ. Sci. Technol., 47, 3199-3208, 10.1021/es305201s, 2013.

Vernier, J. P., Thomason, L. W., and Kar, J.: CALIPSO detection of an Asian tropopause aerosol layer, Geophys. Res. Lett., 38, L07804, 10.1029/2010gl046614, 2011.

Wang, X., Ren, J., Gong, P., Wang, C., Xue, Y., Yao, T., and Lohmann, R.: Spatial distribution of the persistent organic pollutants across the Tibetan Plateau and its linkage with the climate systems: a 5-year air monitoring study, Atmos. Chem. Phys., 16, 6901-6911, 10.5194/acp-16-6901-2016, 2016.

Xin, J., Wang, Y., Pan, Y., Ji, D., Liu, Z., Wen, T., Wang, Y., Li, X., Sun, Y., Sun, J., Wang, P., Wang, G., Wang, X., Cong, Z., Song, T., Hu, B., Wang, L., Tang, G., Gao, W., Guo, Y., Miao, H., Tian, S., and Wang, L.: The Campaign on Atmospheric Aerosol Research Network of China: CARE-China, Bull. Amer. Meteor. Soc., 96, 1137-1155, 10.1175/BAMS-D-14-00039.1, 2015.

Zhang, X. Y., Arimoto, R., Cao, J. J., An, Z. S., and Wang, D.: Atmospheric dust aerosol over the Tibetan Plateau, J. Geophys. Res.-Atmos., 106, 18471-18476, 10.1029/2000jd900672, 2001.