1 Reply to Referee 1

2 We are grateful to the referee for the encouraging comments and careful revisions, which helped to 3 improve the quality of our paper. In the following we quoted each review question in the square brackets 4 and added our response after each paragraph.

5

6 [The paper can be improved with more in-depth discussion. In the introduction, the authors can write a more 7 comprehensive literature review on the precipitation-aerosol relationship, such as, what are controversy issues, 8 what are the research gaps and what are the possible underlying mechanisms for various processes. Then the 9 review can lead to what the authors will address in this paper. The conclusions and abstract seem to have 10 different emphasis. What do the authors really like to emphasize? Should the authors also discuss, in conclusion, 11 the spatial correlation pattern between visibility and rainstorms and other issues emphasized in the abstract?]

Reply 1: Many thanks to the reviewer for the great suggestions. Following the suggestions, we have substantially improved the manuscript with more in-depth discussion with modifying the introduction about a more comprehensive literature review on the precipitation-aerosol relationship and highlighting our study results in both conclusions and abstract.

16

17 Please find the detailed revisions in the uploaded marked-up manuscript version with track changes.

18

19 [The presentation can be greatly improved. Please pay close attention on the presentation because poor 20 presentation can hamper the readers from understanding the contents in the paper. There are numerous places 21 requiring polishing on presentation and corrections for grammatical errors. Some examples are provided in 22 Specific. The authors can take advantage of the editing service provided by the journal.]

Reply 2: We are grateful to the referee for encouraging comments. We have greatly improved the presentation
with rewriting the sentences and correcting the grammatical errors in the revised manuscript. Please find the
detailed revisions in the uploaded marked-up manuscript version with track changes.

26 [Figure 3 shows trends for different rain intensity. Have the authors looked into the total precipitation? What is
27 the trend? What that trend tells us?]

Reply 3: Thanks for the suggestions. We have looked into the total precipitation averaged over Eastern China.
The trend in the interannual variations of the total precipitation from 1961 to 2010 is insignificant, which
indicates that the impact of aerosols on precipitation could be complicated by different rain intensity.

32 [Figure 10 can be improved in the presentation and discussion. How significant is the correlation at each level?
33 The statement in line 204 "indicating they were negatively correlated at low boundary layer" is not supported by
34 Figure 10a and 10b.]

35

Reply 4: Following the referee's suggestion, we have modify Fig. 10 and the corresponding text in the revised
manuscript as follows:

38

39 To reveal the relationship between aerosols and atmospheric vertical thermal structure, the correlation between 40 surface PM_{2.5} concentration and atmospheric thermal structure in both polluted and clean areas in July, 2013 was investigated (Fig. 10). The stations of Changsha and Hongjia located in Hunan and Zhejiang provinces in EC 41 respectively were selected to represent the less light rain region while those of Linzhi and Dingriin of Tibet were 42 43 selected to represent the high-frequency light rain region. The correlation coefficient profiles between the observed surface daily PM_{2.5} concentration and atmospheric temperature profiles derived from high-resolution 44 45 L-band sounding were calculated. The correlations at Changsha and Hongjia stations (Figs.10a-b) show that the correlation between PM2.5 and temperature profiles presented an "inverse phase" pattern, reflecting the high 46 aerosol concentrations in a thermal stable structure similar to temperature inversion layers with "cold at low-layer 47 48 and warm at upper-layer" in the eastern China. On the contrary, the correlations in Linzhi and Dingri stations in 49 the Tibetan Plateau (Fig. 10c-d) indicate that an unstable atmospheric structure with "warm at low-layer and cold at upper-layer" for a favorable condition for the occurrence and development of convection and light rain events 50 in the Tibetan region. 51 52

53

54 [Line 33, delete "It is widely acknowledged that".]

- 55 **Reply 5:** it has been deleted in the revised manuscript.
- 56
- 57 [Line 46, give the full expression of CCN IN.]

Reply 6: The full expressions of CCN and IN have been given with "cloud condensation nuclei" and "icenuclei" in the revised muanscript.

- 60
- 61 [Line 51, use "An earlier study showed" to replace "The study shows".]
- 62 **Reply 7:** It has been changed. .

64 [Line 75-82 Data, some descriptions on quality control would be helpful.]

65 **Reply 8:** The precipitation data are archived at the China Meteorological Administration (CMA) with the 66 conventional quality control of global climate data.

67

[Line 76-77, some description on MODIS data would be helpful, for example, what is the resolution of the
MODIS data? How are the data used in this study?]

Reply 9: because the MODIS aerosol products are not used in the result analysis of revised manuscript, we have
deleted the sentence "annual average AOD data in 2001-2010 from Moderate Resolution Imaging
Spectroradiometer (MODIS)" at the beginning of Section 2. Data. Therefore, we have not given any information
on MODIS data in the revised manuscript..

74

[leave space between 200 and mm. Correct the same problem in the rest of the paper. For example, in Lines 78,
79, 94 and 95.]

77 **Reply 10**: Thank the referee for careful review. It has been corrected.

78

79 [Line 108, Xu et al. (2016) is missing in Reference]

80 **Reply 11:** we have added the following ACP-paper into References:

81 Xu, X., Zhao, T., Liu, F., Gong, S. L., Kristovich, D., Lu, C., Guo, Y., Cheng, X., Wang, Y., and Ding, G.: Climate

82 modulation of the Tibetan Plateau on haze in China, Atmos. Chem. Phys., 16, 1365-1375,
83 doi:10.5194/acp-16-1365-2016, 2016.

84

85 [Line 130, delete "trends" and "extreme".

86 Line 112, use "the differences in the trends between" to replace "the interannual variation trend differences for".

87 Line 117, use "rainstorm, especially large rainstorms, have presented a significant increase trend" to replace

88 "rainstorm and especially large rainstorm extreme events presented significantly an increased trend".

89 Line 118, delete an extra comma.]

90 Reply 12: The careful reviews are greatly appreciated. All the errors have been corrected in the revised91 manuscript.

- 92
- 93 [Line 130, delete "trends" and "extreme".]

94 **Reply 13:** The have been deleted.

95

96 [Line 112, use "the differences in the trends between" to replace "the interannual variation trend differences
97 for".]

98 **Reply 14:** It has been done in the revised manuscript.

99

100 [Line 117, use "rainstorm, especially large rainstorms, have presented a significant increase trend" to replace

101 "rainstorm and especially large rainstorm extreme events presented significantly an increased trend".]

102 **Reply 15:** Following the referee's comments, it has been revised.

103

104 [Line 118, delete an extra comma.]

105 **Reply 16:** It has been deleted.

106

107 [Line 123-126, the sentence can be rephrased as "The areas with negative trends in light rain frequency almost 108 matched with areas with positive trends in visibility and haze frequency in EC (Fig. 4a,b and c), which are well 109 consistent with the area of high aerosol concentrations and frequent haze events (Fig.2a,b). The light rain 110 frequency reduction in China was closely associated with the enhancement of aerosol levels in the atmosphere 111 (Qian et al., 2009)."]

112

Reply 17 : Following the suggestion, the lines 123-126 have be rephrased as "The areas with negative trends in light rain frequency almost matched with areas with positive trends in visibility and haze frequency in EC (Fig. 4a,b and c), which are well consistent with the area of high aerosol concentrations and frequent haze events (Fig.2a,b). The light rain frequency reduction in China was closely associated with the enhancement of aerosol levels in the atmosphere (Qian et al., 2009)." in the revised manuscript.

118

[Line 123-126, what is light rain frequency? Is it the number of days with light rain in a year? What are
visibility and haze frequencies? Please define them clearly in the paper.]

Reply 18: It has been clarified with "The light rain frequency is the number of days with light rain in a year; andthe visibility is in unit of km, haze frequency is the number of days with haze" in the revised manuscript.

123

124 [Line 127, The sentence can be rephrased as "The areas with negative trends in light rain almost covered eastern

125	China and a large part of China".]
126	Reply 19: The sentence has been rephrased as "The areas with negative trends in light rain almost covered
127	eastern China and a large part of China" following the referee's suggestion
128	
129	[Line 142-143, change the phrase as "make the number of cloud droplets increase but the size of cloud droplets
130	decrease".]
131	Reply 20: Thanks for the suggestion. Following the referee's suggestion, it has been changed.
132	
133	[Line 159-160, change the phrase as "As shown in Fig. 6b (left), in the three periods".]
134	Reply 21: It has been changed.
135	
136	[Line 171, use "significant increasing trends" instead.]
137	Reply 22: It has been done.
138	
139	[Line 176, delete "could".]
140	Reply 23: Thank the referee for the kind suggestions. It has been done.
141	
142	[Line 197, use "PM2.5" instead.]
143	Reply 24: It has been done.
144	
145	[Lines 198, 219, 225, 226, add "s" after "concentration".]
146 147	Reply 25: It has been done.
147	[Lines 220, 225, 226, delete "s" after "droplet".]
149	Reply 26: It has been done.
150	
151	[Line 224, use "These aircraft observations showed" instead.]
152	Reply 27: It has been changed in the revised manuscript.
153	
154	[Line 229, use "and the effects depend" to replace "depending".]
155	Reply 28: It has been done in the revised manuscript.
156	

- 157 [Line 235, delete "trend".] Reply 29: The "trend " has been deleted in the revised manuscript. 158 159 160 [Line 236, delete extra space between occurrence and more. Add a space between "with" and "an"] Reply 30: It has been done in the revised manuscript. 161 162 [Line 237, add "the" before "1960"] 163 **Reply 31:** It has been changed in the revised manuscript. 164 165 [Line 234, delete "of precipitation events" before "and".] 166 Reply 32: It has been deleted in the revised manuscript. 167 168 [There are various problems in the figures, their captions and annotations. The following are some examples for 169 170 the authors to take into consideration. 171 1. Be consistent with the figure format; 172 2. Use the consistent fonts and font size; 173 3. Use correct term to label x-axis and y-axis. 174 4. Use capitalized words to label x-axis and y-axis; 5. Label sub-plots using letters (usually at the top, top-left, or top-right of a sub-plot); 175 176 6. Use superscripts and subscripts when necessary; 7. Provide the unit for the variable displayed if no unit, indicate with dimensionless or "(-)"; 177 178 8. Indicate the unit for the color bar. 179 9. Remove zeros for the most insignificant digit after a decimal. 10. Add significant level (p value) on trends. 180 181 11. It is better to indicate latitude/longitude in the China maps in Figures 4 and 5.] Reply 33: We are very grateful to the referee for the encouraging comments and careful revisions. All the 182 183 mentioned problems in the figures, their captions and annotations have been corrected in the revised manuscript. 184 All the figures have been redrawn following the referee's suggetsions.. 185 [Figure 2. Labelling sub-plots (a) and (b). Capitalize "pressure" for the label for the y-axis in Figure 2a. It 186
 - 187 should be "Pressure (hPa)" so to leave a space between "pressure" and its unit. In the caption, wind speed

188 should have a unit of m s-1. Please use correct superscripts.]

189 **Reply 34:** Following the referee's suggestion, Figure 2 has been modified as follows:



190

191

192 [Figure 3. Use the same font and font size to label sub-plots. No need for zeros after a decimal point in y-axis.
193 Label "Precipitation" or "Rain" for the y-axis in Figure 3a. Use "Year" to label x-axis (not "date"). Add
194 significant level (p-value).]

Reply 35: Following the suggestion, has Fig. 3 suggested been modified in the revised manuscript.

196

197 [Figure 4. Label (a), (b), (c) for the subplots. Provide the unit for haze frequency, visibility, and light rain
198 frequency. Indicate what the dots and the background stand for. Indicate the unit for the color bar.]

Reply 36: We have changed Figure 4. Label (a), (b), (c) for the subplots. In the revised caption of Fig. 4, we have added "haze frequency (day), visibility (km), and light rain frequency (day). The dots stand for observation sites with shading by the variation trends, the background presents the terrain height in mainland China".

202

[Figure 5. Label (a) and (b) for the subplots. Provide the unit for the trend of the rainstorm frequency. Indicate
what the dots and the background stand for. Indicate the unit for the color bar.]

205 Reply 37: We have redrawn Fig. 4 as follows and revised the caption as "the trend (day per year) of the rainstorm



206 frequency and the background (shaded colors) stands for the terrain height (m) in mainland China

[Figure 6. Label (a), (b) and (c) for the subplots. In Figure 6a, no color is needed as this will cause confusion
with Figure 6b and 6c. Good titles for each figure will help readers to understand the differences between Figure
6a and Figure 6b and 6c. Otherwise, the figure can be quite confusion. In the caption, it is better to use "the
positive (negative) trend" than "the positive (negative) variability".]

213 **Reply 38:** We have redrawn Fig. 5 as follows, and following the suggestiuon, the caption has been revised.



216 [Figure 7. Label sub-plots at the top, top-left, or top-right. Remove zeros for the most insignificant digit after a

217 decimal. Keep sub-plots (a) -(d) the same size.]

218 Reply 39: Following the referee's suggestion, in the revised manuscript, Figs. 7a -7f have been redrawn as219 follows:







226	[Figure 9. Provide the unit for the dots.]
227	Reply 40: The revised caption of Figure 9 has provided the unit of day for the dots.
228	
229	[Figure 10. For the label for the c-axis, remove "index".]
230	Reply 41: It has been removed in the revised manuscript.
231	
232	[Figure 11. What are the different marks in Figure 11a?]
233	Reply 42: In the revised caption of Figure 11a, we have added "the different marks represent the different
234	flights".
235	
236	Reply to Referee 2
237	
238	We are grateful to the referee for the encouraging comments and careful revisions which helped to
239	improve the quality of our paper. In the following we quoted each review question in the square brackets
240	and added our response after each paragraph.
241	
242	[The manuscript is thorough, clear, compelling, very well written, and presents the results
243	with good figures and tables. I recommend publication after attending to the following detailed comments.]
244	
245	Reply 1: the referee's encouraging comments are great appreciated. We have revised our manuscript following
246	the detailed \ comments of referee.
247	
248	[Line 71: Please give references for the previous investigations of this issue primarily
249	focused on limited cases (references).]
250	
251	Reply 2: Thanks for the suggestion. Accordingly, we have given the following references in the revised
252	manuscript:
253	
254	Li, Z., Niu, F., Fan, J., Liu, Y., Rosenfeld, D. and Ding, Y.: Long-term impacts of aerosols on the vertical
255	development of clouds and precipitation, Nat. Geosci. 4, 888-894, 2011.
256	

257	Rosenfeld, D., Dai, J., Yu, X., Yao, Z., Xu, X., Yang, X. and Du, C.: Inverse relations between amounts of air
258	pollution and orographic precipitation, Science 315,1396-1398, 2007.
259	
260	Zhao, T. Liu, D., Zheng, X., Yang, L., Gu, X., Hu, J., Shu, Z., Chang, J., Wu, X.: Revealed variations of air
261	quality in industrial development over a remote plateau of Southwest China: an application of atmospheric
262	visibility data, Meteorol Atmos Phys, doi:10.1007/s00703-016-0492-7, 2016.
263	
264	[Please give a website or reference for MODIS data.]
265	
266	Reply 3: because the MODIS aerosol products are not used in the result analysis of revised manuscript, we have
267	deleted the sentence "annual average AOD data in 2001-2010 from Moderate Resolution Imaging
268	Spectroradiometer (MODIS)" at the beginning of Section 2. Data. Therefore, we have not given any information
269	on MODIS data in the revised manuscript
270	
271	[For all of Chinese map, if the author can use LambertEqualArea projection
272	(http://ncl.ucar.edu/Applications/maponly.shtml), that would be nice.]
273	
274	Reply 4: Thanks for the suggestion, In order to more clearly present the regional distributions of our results over
275	mainland of China, we have used cylindrical map projection for all of Chinese map
276	
277	[Fig. 10: Please restrict the four panels in the same size.]
278	
279	Reply 5: Following the referee's suggestion, we have restricted the four panels of Fig. 10 in the same size. Please
280	see the modified Fig. 10 in the revised manuscript.
281	
282	
283	
284	
285	
286	
20/	

Are precipitation anomalies associated with aerosol variations over Eastern China?

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298 Abstracts. In Eastern China (EC), the strong anthropogenic emissions deteriorate the atmospheric environment harbored by 299 the upstream Tibetan and Loess Plateaus, building a south-north zonal distribution of high anthropogenic aerosols harbored 300 by the upstream Tibetan and Loess Plateaus in China. This research study climatologically analyzed the interannual 301 variability of precipitations with different intensities in association with aerosol variations in over the EC region from 1961 302 to 2010.,- by using precipitation and visibility data in of more than 50 years and satellite, aircraft and surface aerosol data in 303 recent years in China, the impacts of aerosol variations on interannual variability of various precipitation intensities of 304 precipitation events and their physical causes are investigated. We found that the frequency of light rain significantly 305 decreased and the occurrence of rainstorm, especially the extraordinary rainstorm significantly increased over the recent 306 decades. The extreme precipitation events presented the same similar interannual variability pattern with the frequent haze 307 events over the EC. Accompanied with the frequent haze events in EC, the light rain frequency trend significantly decreased, 308 Especially, since the 1980s the extremely heavy precipitation event have occurred more frequently. Moreover, the extreme 309 oinfoll. annual variability trend. During the 1980s, the regional 310 precipitation trends in EC showed an obvious "transform" from more light rain to more extreme rainstorms. The running 311 correlation analysis of interdecadal variation further verified that the correlation between the increasing aerosol 312 aerosolsemissions and the frequency of abnormal precipitation events tended to be more significant in the EC. The 313 correlation between atmospheric visibility and low cloud amounts, which are both closely related with aerosol concentrations, 314 had a spatial distribution of "northern positive and southern negative" pattern, and the spatial distribution of the frequency 315 variability of regional rainstorm frequency s was "southern positive and northern negative". After the 1990s, the visibility in summer season deteriorated more remarkably-than other seasons, and the light rain frequency decreased obviously while the 316 rainstorm and extraordinary heavy rainfall occurred more frequently. There were significant differences in the interdecadal 317 318 variation trends in light rain and rainstorm events between the high aerosol concentration-polluted areas in the EC and the 319 relatively "clean area" in western plareau of China. The aircraft measurements over the EC confirmed that the diameters of 320 cloud droplets decreased under high aerosol concentration condition, thereby inhibiting weak precipitation process.

321 1. Introduction

322 It is widely acknowledged that the global mean temperature has been increasing since 1850 and will continue to increase in 323 the following decades (IPCC, 2007). The long term forcing of a warming environment might change precipitation, the 324 regional and global water cycle (Allan and Soden, 2008; Allan and Ingram, 2002). The heavy precipitation events showed a 325 overall increasing trend as the result of global warming (Allan and Ingram, 2002; Trenberth et al., 2003). Since the 1950s, 326 the precipitation has increased remarkably at high latitudes, also increased at tropical marine areas, but it underwent some 327 decrease at tropical mainland (New et al., 2001; Kumar et al., 2006; Bosilvoich et al., 2005).

The variation of aerosols in atmosphere is also an important factor to influence the water cycle in regions where bearing long term high aerosol loading (Ramanathan et al., 2005; Ramanathan et al., 2007; Koren et al., 2008; Levin and Cotton, 2009; Li et al., 2011).-Under the background of global warming, the regional precipitation tends to have more complex temporal and spatial distribution patterns. The variations of precipitation could be reflected by the different-grade precipitation, and even by frequency changes of extreme precipitation events (Lau and Wu, 2007)_, which could threaten the social economy and is seriously concerned by governments, public and scientific community.

Precipitation is not only influenced by atmospheric circulation structure related with land-sea discrepancy and land-sea water vapor exchange, but also by local cloud microphysical processes (e.g., CCN, IN). Studies have shown that atmospheric aerosols might add cloud droplets number concentrations (CDNC), and change cloud lifetime, and restrain or enhancemodify precipitation (Khain et al., 2005; Rosenfeld et al., 2007; Rosenfeld and Coauthors, 2008; Stevens and Feingold, 2009; Fan et al., 2013). Aerosols might also change Asian monsoon system (Bollasina et al., 2011). The interaction of aerosols and cloud-precipitation is still a important issue with large uncertainties for climate change (IPCC, 2013).

340 Since the middle 1980s, China has been experienced experiencing a rapid development in industry and agriculture. As a result, a huge amount of industrial-anthropogenic emissions and biomass burning significantly released particulate matters 341 342 into the atmosphere. The study shows that there was no obvious change in annual precipitation in China, but the extremely 343 heavy rainfall area, mainly in the EC, had expanded (Zhai et al., 1999). However, the regional annual precipitation, summer 344 precipitation, and extreme precipitation events had an obvious rising tendencies in middle and lower Yangtz River Basin of 345 EC (Wang and Zhou, 2005). The numerical simulations also presented that the increase of aerosols could decrease the 346 summer convective precipitation in the intensity under 30 mm h⁻¹, and increase summer strong convective precipitation in 347 the rates above 30 mm h⁻¹ in China (Guo et al., 2014). With a rapid increase of aerosols, not only local-light rain over wide areas could decrease, but also local extremely heavy rain could be triggered, inducing frequent flood (Guo et al., 2014; Fan 348 349 et al., 2015). Light rain tended to decrease and at the same time the extremely heavy precipitation had increasing tendency in

351	variability connecting to global warming together with the increased emissions of anthropogenic aerosols.
352	Aerosols might also change Asian monsoon system (Bollasina et al., 2011). There are great uncertainties in the interactions
353	between the internal influence factors including complex influences of land sea discrepancy, aerosols and cloud and
354	precipitation processes of Asia monsoon system and external forcing factors.
355	The previous investigations of this issue primarily focused on limited cases with large discrpancies (Rosenfeld et al., 2007;
356	Rosenfeld and Coauthors, 2008; Stevens and Feingold, 2009; Li et al., 2011; Zhao et al., 2016 Fan et al., 2013). The climatic
357	forcing of aerosols on precipitation-extremes in a large-scale continent region and its physical causes remain uncertain have
358	been poorly understood. The long-term visibility data can be used to climatologically assess the air quality change (Wang et
359	al. 2009; Che et al. 2009; Xia et al. 2006), as the atmospheric visibility is a good indicator of air pollutant levels in the
360	environmental atmosphere (Zhao et al., 2016). By using precipitation and visibility data in a 50-year period and aircraft and
361	surface aerosol observational data in recent years in China, the climatic impacts of aerosols on interannual variability of
362	various precipitation intensities and their physical links were investigated in this study. The large amounts of anthropogenic
363	aerosols not only deteriorate the environment over large spatial scales, but they might induce the rapid change of regional
364	climate and water cycle. In addition _a , the high aerosol concentrations are accumulated zonein the north-south direction
365	over China is located onover EC in connection with - the eastern side of the Tibetan plateau and the Loess plateau, which
366	might be connected with the leeward slopes of the large-terrain structure effect of the Tibetan plateau and the Loess plateau
367	in China (Xu, et al, 2016), the plateaus. The polluted EC regions from and the clean the plateaus to EC in China may be an the
368	ideal places to identify the climate forcing of aerosols with following questions: with comparing the interannual variation
369	trends in various precipitation intensities between the polluted EC with the clean region over the Tibetan plateau for
370	exploring .the realtion of precipitation anomalies and aerosol variations. The previous investigations of this issue primarily
371	focused on limited cases (Rosenfeld et al., 2007; Li et al., 2011; Zhao et al., 2016). The elimatic forcing of aerosols on
372	precipitation extremes in large scale continent region and its physical causes remain uncertain. By using precipitation and
373	visibility data in a 50 year period and satellite, aircraft and surface aerosol observational data in recent years in China, the
374	climatic impacts of aerosols on interannual variability of various precipitation intensities and their physical links were
375	investigated in this study.

the EC (Choi et al., 2008; Qian et al., 2007; Qian et al., 2009). This phenomena might be the strong signal of climate

376 2. Data

377	In this study, we classifed extraordinary storm, large rainstorm, rainstorm, large rain, moderate rain and light rain
378	respectively with daily precipitation >200 and ranging between 100-200mm, 50-100mm, 25-50 mm, 10-25mm and
379	0.1-10mm in this study. In this work, we adopted annual average AOD data in 2001-2010 from Moderate Resolution

Imaging Spectroradiometer (MODIS);<u>t</u>The monthly frequency_data_of different-gradesintensity of precipitation events including of __extraordinary storm (>200mm, the precipitation intensity classification standard for 24 hours),<u>large</u> rainstorm, rainstorm-(100-200mm), large rainstorm heavy-rain-(25-50 mm), moderate rain-(10-25mm), light rain (0.1-10mm) from 601 stations in China over 1961-2010_were adopted_-(Datasets-from the National Meteorological Information Center of China Meteorological Administration);-_In addition, the meteorological and environmental-data including-monthly haze days of 2513-stations, daily visibility of 598 stations and daily-low cloud cover of 753 stations in 1961-2010 in China as well as the daily PM_{2.5} data of 946 stations in 2013-2014 in China were also used in this study.

In order to analyze the regional variations in aerosols over <u>Eastern ChinaEC</u>, we adopt the equivalent visibility by excluding
the influence of natural factors (Rosenfeld et al., 2007) on the observed visibility based on the meteorological data-observed
from 598 stations in 1961-2010 were used in this study. The equivalent visibility was corrected <u>VIS (dry)</u> based on the the
following_formula (1) under the relative humidity from 40% to <u>9990</u>%.

391

$$\frac{\text{VIS}}{\text{VIS}(\text{dry})} = 0.26 + 0.4285\log(100 - \text{RH})$$
(1)

392 The characteristics of aerosol and cloud droplets size were comprehensively analyzed based on the aerosol-cloud data 393 obtainedobserved from aircraft flights carried out inover Beijing and its surrounding regions during 2008-2010 by the Beijing Weather Modification Office. The scientific detection time was from May to August during 2008 2010. The observed 394 395 clouds were mainly stratus cloud, stratocumulus and cumulus clouds, and the maximum detection altitude was 7000 m. 396 There were 40 flights carried out during the experiment period. Aircraft measurements were usually carried out withinin 2-6 h before the clouds precipitated. The flight area and tracks were shown in Fig. 1. The Passive Cavity Aerosol Spectrometer 397 398 Probe (PCASP-200, DMT Co.) was used for observing aerosol particle size in 0.1-0.3µm. The probe of Cloud, Aerosol and 399 Precipitation Spectrometer (CAPS, DMT Co.) was used for observing cloud droplets in 0.6-50µm. The probes were returned 400 to the DMT for standard calibration before starting measurements in each year. In addition, the probes were calibrated using 401 the spheres of polystyrene latex (PSL) of Duke Scientific Corporation for each month. Considering the influence of cloud 402 droplets on aerosol probing, the averaged aerosol concentration below 300m of cloud base was calculated to represent 403 aerosol concentration in clouds. The cloud droplet -measurements were made within clouds at 100m height intervals. The 404 data were processed into two or more samples when the clouds were multiply layered.

405 **3** Haze distributions in Eastern China harbored by large plteaus

Due to the influence of the terrain on the typical westerly-winds in Eastern China, the air flowing from the windward
plateaus descends sinked in a north south oriented zone between about 110°E and 125°E (upper panel of Fig. 2).
Accompanying this strong downward current are were weak winds in the near-surface layers in the lee side of the plateaus.
These air flow and wind condition lead to development of a "harbor" accumulating air pollutants in EC. The weak wind and

downward current areas coincide well with the centers of frequent haze events in <u>ChinaEC</u> (lower panel of Fig. 2). The
"susceptible region" of <u>frequent</u> haze events <u>pollution</u> over <u>Eastern ChinaEC</u> from the eastern edge of the plateaus to the
lower flatlands is associated with the "harbor" effect of the unique topography under specific meteorological conditions that
trap air pollutants (Xu etl al., 2016). <u>The EC is clomatologically a region with frequent haze events over recent decades</u>,
where high aerosol pollution could excert an impact on the regional variation in precipitation.

415 4. Change trends in various precipitation intensities

416 The interannual variation trends interannual variation of extreme precipitation events with various intensities including light 417 rain, moderate rain, heavy rain, rainstorm, the large rainstorm, extraordinary rainstorm (Fig. 3a) over EC were comparatively analyzed in Figure 3. and it is found that the interannual variation trend differences for the six various precipitation 418 419 significant. Regionally averaged over EC, The the trends in light rain frequency had trend significantly 420 decreased, while the events of rainstorm including large and extraordinary storm had increased significantly (Fig. 3a, A),), 421 althrough the moderate rain frequency trend slightly declined (Fig. 3a, B), and the interannual change trend of heavy large rain frequency was not significant (Fig. 3a, C), the rainstorm and large rainstorm events increased significantly (Fig. 3a, D, E, 422 423 P) in EC., Especially since 1980s, the extremely heavy precipitation events have become more frequent, showing an obvious 424 transforming characteristics of frequent heavy rain and torrential rainfrequent occrences of disatrous. Large rainstorm and 425 especially large rainstorm extreme events presented significantly an increased trend, along with the frequent haze weather-in 426 EC. Overall rainstorm extreme events were on the rise trend, , but light rain tended to decline significantly. In contrast, 427 stations in the Tibetan Plateau (atwith height altitude of >4000m), a relative clean area in China, were selected for a 428 statistical analysis of interannual variation trend of light rain frequency., indicating that tThe characteristic of the decreased 429 trend of light rain frequency was not significant in the Tibetan Plateau over recent decades (Fig. 3b, A), implying an anomaly 430 of aerosols restraining light rain frequency over EC.

431 5. Distribution of frequencyregioanl changes of extreme rainstormprecipitation events, haze and visibility

We calculated the trends in interannual variations of precipitation and visibility at all the site in China (Fig. 4). The negative variability-area with the negative frends in light rain frequency-almostquite well matched with the areas of positive variability negative trends in _-of visibility-and positive trends in haze frequency in EC (Figs. 4a,b and c)₂, which are well consistent with the area of high aerosol concentrations and frequent haze events (Fig. 2a,b). The light rain frequency reduction in China was closely associated with the enhancement --of aerosol levels in the atmosphere (Qian et al., 2009). It is noteworthy that the negative trend areas of light rain almost-covered a large part of areas in China and all the sites in EC eastern China (Fig. 4c). This might be also closely related with temporal and spatial variations of n trends of summerEast Asian summer monsoons-activity which offered a suitable dynamic background for the effect of aerosols on clouds and precipitation._Figure 5a shows that the spatial distribution of the trends in rainstorm frequency variability-was "southern positive and northern negative" in summer during 1961-2010, while the correlations between visibility and low-level cloud amount were distributed with the "northern positive and southern negative" pattern in EC_during 1961-2010 in EC (Fig.5b), indicating that the effect of aerosols on summer convective precipitation was more obvious in southern part than that in northern part of EC.

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446 There were obvious differences in the interdecadal precipitation decreasing change rate of the various precipitation 447 intensities in the EC region (Fig.6a), where the - In this region, the summer interdecadal precipitation variability revealed the 448 on clouds and precipitation process, that negative variability stations of light rain made up the 449 majority (about 87.6%), the positive variability stations of moderate rain were approximately equal to the negative ones 450 (about 51%), the positive variability stations of heavy-large rain (about 71.3%) were much more than the negative ones 451 indicating the reverse trend. The positive variability stations of rainstorm-with daily precipit-ation_->50mm, including 452 catastrophic rainstorm over 100mm occupied obvious majority (about 78.9%). In China, in the recent decades, tThe rapid 453 increase of the anthropogenic aerosol particles in the atmosphere may not only make the cloud droplet number concentration 454 increased but the size of cloud droplet decreased, thus changing the life time of the cloud and suppressing the precipitation, 455 especially for the light rain (Qian et al., 2009), but also enhance, the rainstorm precipitation with more frequent events in EC. mentioned above, the light rain frequency reduced significantly; the moderate rain frequency changed unobvious; heavy 456 rain increased relatively obvious, rainstorm and catastrophic rainstorm increased significantly obvious in eastern China, 457 458 indicating the obvious anomalous change characteristics of regional precipitation from less light rain changed to more 459 heavy rain and even the catastrophic rainstorm along with the frequent haze trend-pollution in EC.

460 Although severe-precipitation events mainly depended on dynamical and thermodynamic processes and water vapor source in the atmospheric circulation and deep convective activitye, aerosol's "Albrecht effect" considered that increased with 461 462 increasing cloud droplet concentrations and decreased decreasing cloud droplet size influenced by aerosol would could 463 suppress cloud precipitation process and extend cloud maintenance-life time. The extension of the cloud life time might save 464 the potential that triggering the abnormal severe precipitation extreme events when the cloud droplets coagulation condition 465 vas mature. This mechanism could partly explain the significant light rain reduction trend (Fig. 6a) and the spatial consistency indicating the precipitation rate transformation characteristics degrading of from light rain to heavy rain or 466 severe extreme precipitation extreme events (blue arrows in Fig. 6a) in the polluted eastern ChinaEC region. Moreover, in 467 research, in order to fEurther-more we comparatively analyzed the effects of aerosol pollution in hazy EC on extreme 468 precipitation events, we selected the <u>region of Tibetan Plateau</u> (west of 110 E, south of 40 N) <u>of Tibetan Plateau (TP)</u>, a 469 470 relative clean area in western China was selected, as the reference area to comparatively analyze the effects of aerosol 471 pollution in hazy EC on regional precipitation change, and, we calculated percentages of sites with negative frequency 472 variability trends of light rain and positive trends of rainstorm events in total sites with the negative and positive and 473 negative trends in visibilityhaze over the EC and TP regions duringin the three different interdecadal periods (1961-1980, 474 1971-2000, 1981-2010) in the (Fig. 6b) east and westEC and TP regions. As-Deuing past more then 5 decades, could be seen 475 from Fig. 6b (left), in the three stages, tThe positive variability of _light rain and reainstorm were _was steady a declining trended and augmented, receptively in the polluted EC, _-while the negative variability was an increasing trend in EC, while 476 477 there were no obvious positive or and negative variability trend of light rain and rainstorm in the clean TP region Tibetan 478 Plateau (double headed arrows inright panel of Fig. 6b-right).

479 6. interannual variability anomalies between atmospheric visibility and precipitation anomalies

480 Figure 7 further verified the relation of interannual variability of regional visibility and precipitation in EC over recent years. 481 Regionally averged, less light rain events and more rainstorm occrences significantly from year to year in association with 482 enhanced aerosol levels with declining vaisibility over EC (Figs. 7a, 7b, 7c and 7d). We calculated the correlation 483 coefficients between regional averages of visibility with the frequency of light rain, heavy rain, extremely torrential rain over 484 region (east to 110 E, south to 40 N), respectively in consideration of correlation pattern between visibility and various 485 recipitation intensities. Therefore, Taking summer months (June, July and August) as examples, the 20-year running 486 correlation coefficients of visibility and precipitation were obtained (presented in Figs. Figure 7e. It is very interesting that 487 the interannual varaitions of visibility and precipitation over EC were evolved from positive correlations in the eraly 1960s 488 to negative correlations in the 1970s and 1980s (Fig. 7e), reflecting the interaannul variation of the aerosol and 489 precipitation interaction in changing climate. - a, b and c). The running correlation coefficients curve of visibility and light confidence level (90%), and the running correlation coefficients curve of visibility and heavy rain and 490 491 xtremely heavy rain reached the confidence level (90%). This statistical analysis results further verified that with the increase of aerosol emissions, visibility and various precipitation exhibited increasing significant correlation trends (Fig.6). 492

In order to investigate the seasonal and interannual variations of monthly correlation pattern between regional visibility and with light rain, as well as regional visibility and extremely precipitation events in EC, we illustrated the annual cross-section of monthly anomaly anomalies of visibility and number of days with light rain, visibility and heavy __rainstrom, as in Figure 8, well as visibility and extremely heavy rainin. Through a comprehensive comparison of Figs. 8 a, b, c and d, we could find significant positive correlation between visibility and light rain, showing indicating that the poor visibility surpressed light rain frequency. Moreover, there was a significant difference between the changing trend of extremely precipitation events frequencyrainstorm and light precipitation frequency occurrences. The changes of heavy precipitation large and extraordinary

rainstorm frequency from 1960s to 1980s was were not as prominent as that thoese at the latter period of 1990s, during
 which time visibility deteriorated remarkably, heavy and extremely heavy rain occurred frequently. Compared to other
 seasons, the influence effect of poor summer visibility was more significant in ChinaEC, showing summertime disastrous
 rainfalls happened more often with less light rain over recent years. - frequency decreasing significantly and sudden heavy
 rain and large heavy rain frequency increasing.

506 The increased atmospheric aerosol concentration may reduce the solar radiation to surface and decrease surface temperature. 507 At the same time, the polluted black carbon aerosols can strongly absorb solar short wave radiation and directly heat 508 el atmosphere, and forming a temperature inversion structure (Bollasina et al, 2011; Zhang et al., 2009; Bond et al., 509 2013; Bond et al., 2011; Grant et al., 2014; Seinfeld et al., 2008). Therefore, the aerosol radiation interaction could change the atmospheric stability and alter local or region atmospheric circulation and precipitation process. This temperature 510 511 inversion structure increases with the stability of atmospheric boundary layer-and provides an important condition for the 512 frequent occurrence of haze and fog events. The stable low-level structure also inhibits the weak convection development of 513 atmospheric boundary layer, so as to reducinge the formation of low-level clouds and weak precipitation process. However, 514 the strong dynamic convergence disturbance could destroy the stability of atmospheric boundary layer and cause the 515 formation and development of severe rainstorms.

To further clarify the relation between aerosols<u>-concentration</u> and light rain frequency, the light rain frequency distribution from 601 stations in July, 2013 is displayed (Fig.9). It shows in Fig. 9 that t<u>T</u>he light rain events have significantly declined in the Yangtze River region of EC with high aerosol concentrations have and but enhaced while t in the relative clean <u>TP</u> region of <u>Tibetan Plateau(Fig. 9)</u>.

520 To reveal the relationship between aerosols and atmospheric vertical thermal structure, the correlation between surface PM2.5 521 concentrations and atmospheric thermal structure in both polluted and clean areas in July, 2013 was investigated (Fig. 10). 522 The stations of Changsha and Hongjia located in Hunan and Zhejiang provinces in EC respectively were selected to 523 represent the less light rain region while those of Linzhi and Dingriin-Dingri of Tibet were selected to represent the 524 high-frequency light rain region. The correlation coefficient profiles between the observed surface daily PM2.5 concentration 525 and atmospheric temperature profiles derived from high-resolution L-band sounding were calculated. The correlations at 526 Changsha and Hongjia stations (Figs. 10a-b) show that the correlation between $PM_{2,5}$ and temperature profiles presented an 527 "inverse phase" characteristic pattern, indicating they were negatively correlated at low boundary layer and positively 528 correlated at upper boundary layer or troposphere, reflecting the high aerosol concentrations in a thermal stable structure 529 similar to temperature inversion layers like-with "cold at low-layer and warm at upper-layer" in the eastern ChinaEC. On the 530 contrary, the correlations in Linzhi and Dingri stations in the Tibetan PlateauTP (Fig. 9e10c, d) indicate that an unstable 531 atmospheric structure with "warm at low-layer and cold at upper-layer" for with a favorable condition for the occurrence and

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533 7. Physical connection between aerosols and precipitation

According to the results of observation and modeling studies, the increased aerosol concentrations could reduced effective particle radius and increased number concentration of cloud droplets, and latent heat release (Khain et al., 2005; Van den Heever et al., 2006; Tao et al., 2007; Altaratz et al., 2014)._The increase of cloud droplets concentrations would delay raindrop formation, thereby lessening light precipitation (Qian et al., 2009) leadingfor_the negative correlation between aerosols and light_precipitation in China (Choi et al., 2008).

539 In order to further investigate confirm the relationship between aerosols and cloud droplets, the cloud dropletobserved data 540 observedcollected by aircraft in north China during 2008-2010 were used. The vertical profiles of cloud droplets data at each 541 level-under different aerosol state obtained by 40 aircraft is shown in Figure - 11.4. From Figures - 11a and Fig. 11b, aerosol 542 Albrecht "cloud lifetime effect" was significant in the Northern northern EC. As shown in red profile of Fig. 11b, uunder the 543 background of high aerosol concentrations, the cloud droplets sizes were smaller and increased slowly with the increasing 544 altitude (red profile of Fig. 11b). In addition, from the cloud base to 2000m, the cloud droplets size remained less than 20 545 microns (Fig.11b), resulting in precipitation delay, in favour of cloud system development to form heavy rain easily. 546 addition, eCloud droplets diameter enlarged quickly with the increase of height, and reached 30 microns easily to forming 547 light rain at 1000m altitude under low aerosol concentrations (green profile in Fig.11b). The above-aircraft observation 548 analysis showed that under the condition of insufficient water vapor in the North China relative to that in the southern China. 549 high aerosol concentrations could reduce cloud droplets size, increase cloud droplets concentrations, extend cloud lifetime, and this which would could restrain the development of low clouds, especially restrict the light rain process. 550

551 8. Discussion and conclusions

Aerosols have complicated effects on clouds and precipitation, depending on many factors such as aerosol properties, topography and meteorological conditions. The most previous investigations of aerosol impacts on clouds and precipitation are primarily based on limited cases in relatively smaller spatial and temporal scales. the climate forcing of aerosols on precipitation in large-scale continent region and physical causes remain uncertain. By using precipitation and visibility data in-of more than_50 years-and-satellite, aircraft and surface aerosol data in recent years in China, the impacts of aerosol variations on interannual variability of various precipitation intensities of precipitation events and their physical causes are investigated.

Accompanied with the frequent haze events in EC, the light rain frequency trend significantly decreased. Especially, since
the 1980s the extremely heavy precipitation event have occurred –more frequently with_an obvious transform from more

light rain to more frequent heavy rain and rainstorm. From 1960s to 1970sIn the 1960s, the monthly visibility and light rain
presented a significantly positive correlation, while the visibility was in good condition, and the light rain frequency was also
in high value. In recent 20-30 years, the dramatically increased aerosols resulted in poor visibility, and the light rain
frequency decreased obviously, and, heavy and extremely heavy rain occurred more frequently.

565 The investigation of relation between aerosol concentrations and light rain frequency distributions from 601 stations in July, 2013 in China shows that the light rain-in the Yangtze River region of EC with high aerosol concentration appeared 566 567 significantly low-frequency in the EC region with high aerosol concentrations, and but while that in the relative clean 568 presented significantly high-frequency in the relative clean region of Tibetan plateau presented significantly. . The physical cause of this relation was investigated, and found that the hHigh aerosol concentrations was 569 570 strongly correlated to the warming low-level atmospheric warming which tended to forming a stable structure that 571 suppressed_suppressing_the occurrence and development of weak convection and light rain events in eastern ChinaEC. 572 aircraft measurements over the EC confirmed that the diameters of cloud droplets decreased under high aerosol 573 concentration condition, thereby inhibiting weak precipitation process. while this was not be found in the relatively clean egion over the Tibetan plateau. 574

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The findings from this study have <u>some</u>-important implications<u>for aerosol and precipitation interaction</u>. - the frequent haze
events in EC not only cause regional environment deterioration, but also threaten the social economy and people life in large
spatial and temporal scales, and possible induce the long-term change of regional water cycle. This may exacerbate with the
effect of on regional climate change.

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581 Acknowledgements

The study was supported by the National Key R & D Program Pilot Projects of China (JFYS2016ZY01002213; 2016YFC0203304), the National Natural ScienceFoundation of China (91544109; 91644223), the project of Environmental Protection (HY14093355; 201509001) in the Public Interest and Chinese Third Tibetan Plateau Atmospheric Experiment (GYHY201406001).

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Figure 2. Cross sections of vertical circulations illustrated by stream lines (upper panel) with the horizontal wind speed (m s_1^{-1} ; color contours) and zonal variations of annual haze event frequency (lower panel) at 27 N-41 N averaged in spring, summer, autumn and winter over 1961-2012. Note that near-surface vertical and horizontal winds are not illustrated well here due to north-south variations in the terrain and approximation of the location of the plateaus (black shaded area) in upper panel. All fields are for the annual-averages.

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clean area of Qinghai-Tibetan Plateau Note: Various precipitation intensities included light rain (A), moderate rain (B), heavy

rain (C), rainstorm (D), large rainstorm (E), and extraordinary storm (F).







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687 Figure 4 Interannual variability dDistribution of interannual change trends (day per 10 years) in (a) haze frequency-(a), (b)

688 visibility (b) and (c) light rain frequency (c) in summer in mainland China in 1961-2010. The yellow dash lines mark the borders of

689 <u>frequent haze area or the eastern borders of plateaus in China.</u>





Figure 5 The spatial distributions of (a) trends (day per 10 years) in summertime rainstorm frequency over 1961-2010 in China
and (b) correlation coefficients between visibility and low cloud amount in summer of 1961-2010. With the vellow dash line
marking the border of negative correlation area.





703 704 705 706 707 708 709 positive (left side) and negative (right side) trends in haze over (b) the EC and (c) TP regions during the three interdecadal periods (1961-1980, 1971-2000, 1981-2010) west of 110 E, south of 40 N (b). 601 precipitation stations and 598 visibility stations in eastern China. The right side is the positive variability and left side is the negative variability). The arrows indicate the interdecadal change patterns.

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Figure 6 The(a) ratiopercentages of summer haze days and the stations eovered with by positive and negative variability trends in number of day inst various precipitation grades from 1961 to 2010 over in the EC stayions east of 110 E, south of 40 N₂ (ab), The percentages of sites with negative frequency trends of light rain and positive trends of rainstorm events in total sites with the and







Figure 7 Correlation between summer average visibility (June, July and August) <u>and-with</u> light rain frequency (<u>a</u>) in 1961-1990, (<u>a</u>), (<u>b</u>) light rain frequency in 1981-2010 (<u>b</u>), (<u>c</u>) extremely heavy rain event frequency in 1961-1990 (<u>c</u>), and (<u>d</u>) extreme heavy



over eastern China (east to 110 E, south to 40 N)EC with dash line standing for the correlation passing the confidence level of





Figure 9 Light rain frequency distribution of 601 stations in China in July of 2013. The circled region on right is the low-frequency
light rain region in middle and downstream region of <u>Vangtze River</u> in <u>eastern ChinaEC</u>, and that on left is the high-frequency
light rain region in relative clean region over <u>Tibet plateauthe TP</u>.



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Figure 11 The vertical profiles of (a) sampling cloud droplets droplet size data at each level under different aerosol state(a) and concentration(b) detected by 40 aircraft, and (b) changes and number and diameters of (Green profile: cloud droplets diameter

757 under low concentration; Red profile: cloud droplets diameter under and high aerosol number concentrations}.