Are precipitation anomalies associated with aerosol variations over Eastern China?

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Abstracts. In Eastern China (EC), the strong anthropogenic emissions deteriorate atmospheric environment, building a south-north zonal distribution of high aerosols harbored by the upstream Tibetan and Loess Plateaus in China. This study climatologically analyzed the interannual variability of precipitations with different intensities in association with aerosol variations over the EC region from 1961 to 2010 by using precipitation and visibility data in of more than 50 years and aircraft and surface aerosol data in recent years in China, and the impacts of aerosol variations on interannual variability of various precipitation intensities of precipitation events and their physical causes are investigated. We found that the frequency of light rain significantly decreased and the occurrence of rainstorm, especially the extraordinary rainstorm significantly increased over recent decades. The extreme precipitation events presented the similar interannual variability pattern with the frequent haze events over the EC. Accompanied with the frequent haze events in EC, the light rain frequency significantly decreased and the extremely heavy precipitation event have occurred more frequently. During the 1980s, the regional precipitation trends in EC showed an obvious "transform" from more light rain to more extreme rainstorms. The running correlation analysis of interdecadal variation further verified that the correlation between the increasing aerosols and frequencies of abnormal precipitation events tended to be more significant in the EC. The correlation between atmospheric visibility and low cloud amounts, which are both closely related with aerosol concentrations, had a spatial distribution of "northern positive and southern negative" pattern, and the spatial distribution of the variability of regional rainstorm frequency was "southern positive and northern negative". After the 1990s, the visibility in summer season deteriorated more remarkably, and light rain frequency decreased obviously while the rainstorm and extraordinary heavy rainfall occurred more frequently. There were significant differences in the interdecadal variation trends in light rain

and rainstorm events between the high aerosol polluted area in the EC and the relatively “clean area” in the western plateaus of China. The aircraft measurements over the EC confirmed that the diameters of cloud droplets decreased under high aerosol concentration condition, thereby inhibiting weak precipitation process.

1. Introduction

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). Precipitation is not only influenced by atmospheric circulation related with land-sea discrepancy and land-sea water vapor exchange, but also by local cloud microphysical processes. Atmospheric aerosols might add cloud droplets number concentrations, and change cloud lifetime, and modify 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 an important issue with large uncertainties for climate change (IPCC, 2013).

Since the middle 1980s, China has been experiencing a rapid development in industry and agriculture. As a result, a huge amount of anthropogenic emissions and biomass burning increasingly released particulate matters into the atmosphere. There was no obvious change in annual precipitation in China, but the extremely heavy rainfall area, mainly in the EC, had expanded (Zhai et al., 1999). However, the regional annual precipitation, summer precipitation, and extreme precipitation events had obvious rising tendencies in middle and lower Yangtze River Basin of EC (Wang and Zhou, 2005).

The numerical simulations also presented that the increase of aerosols could decrease the summer convective precipitation in the intensity under 30 mm h^{-1}, and increase summer strong convective precipitation in the rates above 30 mm h^{-1} in China (Guo et al., 2014). With a rapid increase of aerosols, not only light rain over wide areas could decrease, but also local extremely heavy rain could be triggered, inducing frequent flood (Guo et al., 2014; Fan et al., 2015). Light rain tended to decrease and at the same time the extremely heavy precipitation had increasing tendency in the EC (Choi et al., 2008; Qian et al., 2007; Qian et al., 2009). This phenomenon might be the strong signal of climate variability connecting to global warming together with the increased emissions of anthropogenic aerosols.

The previous investigations of this issue primarily focused on limited cases with large discrepancies (Rosenfeld et al., 2007; Rosenfeld and Coauthors, 2008; Stevens and Feingold, 2009; Li et al., 2011; Fan et al., 2013). The climatic forcing of aerosols on precipitation in a large-scale region and its physical causes has been poorly understood. The long-term visibility data can be used to climatologically assess the air quality change (Wang et al. 2009; Che et al. 2009; Xia et al. 2006), as the atmospheric visibility is a good indicator of air pollutant levels in the environmental atmosphere (Zhao et al., 2016). By using precipitation and visibility data in a 50-year period and aircraft and surface aerosol observational data in recent years in China, the climatic impacts of aerosols on interannual variability of various precipitation intensities and their physical links were investigated in this study. In addition, the high aerosol concentrations are accumulated in the north-south direction over EC in connection with the terrain effect of the Tibetan plateau and the Loess plateau in China (Xu, et al, 2016).The polluted EC region and the clean plateaus in China may be the ideal places to identify the climate forcing of aerosols with comparing the interannual variation trends in various precipitation intensities for exploring .the relation of precipitation anomalies and aerosol variations.

2. Data

In this study, we classified extraordinary storm, large rainstorm, rainstorm, large rain, moderate rain and light rain respectively with daily precipitation >200 and ranging between 100-200mm, 50-100mm, 25-50 mm, 10-25mm and 0.1-10mm in this study. The monthly data of precipitation events of extraordinary storm large rainstorm, rainstorm, large rain, moderate rain, light rain from 601 stations in China over 1961-2010 were adopted from the National Meteorological Information Center of China Meteorological Administration. In addition, the meteorological and environmental data including haze days daily visibility and low cloud cover in 1961-2010 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 EC, 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 in 1961-2010 were used in this study. The equivalent visibility was corrected VIS (dry) based on the following formula (1) under the relative humidity from 40% to 90%.

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

The vertical changes of aerosol and cloud droplets size were comprehensively analyzed based on the aerosol-cloud data observed from aircraft flights over Beijing and its surrounding regions during 2008-2010. The flight observed clouds were mainly stratus cloud, stratocumulus and cumulus clouds, and the maximum detection altitude was 7000 m. There were 40 flights carried out in 2-6 h before precipitation. The flight area and tracks were shown in Fig. 1. The Passive Cavity Aerosol Spectrometer Probe (PCASP-200，DMT Co.) was used for observing aerosol particle size in 0.1-0.3μm. The probe of Cloud, Aerosol and Precipitation Spectrometer (CAPS，DMT Co.) was used for observing cloud droplets in 0.6-50μm. The probes were returned to the DMT for standard calibration before starting measurements in each year. In addition, the probes were calibrated using the spheres of polystyrene latex (PSL) of Duke Scientific Corporation for each month. Considering the influence of cloud droplets on aerosol probing, the averaged aerosol concentration below 300m of cloud base was calculated to represent aerosol concentration in clouds. The cloud droplet measurements were made within clouds at 100m height intervals. The data were processed into two or more samples when the clouds were multiply layered.

3 Haze distributions in Eastern China

Due to the influence of the terrain on the typical westerly, the air flowing from the windward plateaus descended between about 110^{o}E and 125^{o}E (upper panel of Fig. 2). Accompanying this strong downward current were weak winds in the near-surface layers. The wind condition leads to accumulating air pollutants in EC. The weak wind and downward current areas coincide well with the centers of frequent haze events in EC (lower panel of Fig. 2). The frequent haze pollution over EC is associated with the "harbor" effect of the topography under specific meteorological conditions that trap air pollutants (Xu et al., 2016). The EC is climatologically a region with frequent haze events over recent decades, where high aerosol pollution could exert an impact on the regional variation in precipitation.

4. Change trends in various precipitation intensities

The trends interannual variation of precipitation events with various intensities of light rain, moderate rain, heavy rain, rainstorm, the large rainstorm, extraordinary rainstorm over EC were comparatively analyzed in Figure 3. Regionally averaged over EC, the trends in light rain frequency had significantly decreased, while the events of rainstorm including large and extraordinary storm had increased significantly (Fig. 3a), although the moderate rain frequency trend slightly declined, and the interannual change trend of large rain frequency was not significant (Fig. 3a). Especially since 1980s, the extremely heavy precipitation events have become more frequent, showing frequent occurrences of disastrous rainstorm along with the frequent haze in EC. Overall rainstorm extreme events were on the rise trend, but light rain tended to decline significantly. In contrast, stations in the Tibetan Plateau (TP) with altitude >4000m, a relative clean area in China, were selected for a statistical analysis of interannual variation trend of light rain frequency. The characteristic of the decreased trend of light rain frequency was not significant in the TP over recent decades (Fig. 3b), which could imply aerosols anomalies restraining light rain frequency over EC...

5. Regional changes of precipitation events, haze and visibility

We calculated the trends in interannual variations of precipitation and visibility at all the site in China (Fig. 4). The area with the negative trends in light rain frequency quite well matched with the areas of negative trends in and positive trends in haze frequency in EC (Figs. 4a, b and c). 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 covered the most sites in EC (Fig. 4c). This might be also closely related with temporal and spatial variations of East Asian summer monsoon which offered a suitable dynamic background for the effect of aerosols on clouds and precipitation. Figure 5a shows that a spatial distribution of the trends in rainstorm frequency 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.

There were obvious differences in the precipitation change rate of various precipitation intensities in the EC region (Fig.6a), where the negative variability stations of light rain made up the majority (about 87.6%), the positive variability stations of moderate rain were approximately equal to the negative ones (about 51%), the positive variability stations of large rain (about 71.3%) were much more than the negative ones indicating the reverse trend. The positive variability stations of rainstorm with daily precipitation >50mm, including catastrophic rainstorm over 100mm occupied obvious majority (about 78.9%). The increase of the anthropogenic aerosol particles in the atmosphere may suppress light rain (Qian et al., 2009), and also enhance the rainstorm precipitation with more frequent events in EC. The anomalous changes of regional precipitation from less light rain changed to more heavy rain and even the catastrophic rainstorm along with the frequent haze pollution in EC.

Although precipitation events depended on dynamical and thermodynamic processes and water vapor source in the atmosphere, aerosol's "Albrecht effect" with increasing cloud droplet concentrations and decreasing cloud droplet size i could suppress cloud precipitation process and extend cloud life time. The extension of the cloud life time might save the potential that triggering the abnormal severe precipitation extreme events. This mechanism could partly explain the precipitation degrading from light rain to severe extreme events (Fig. 6a) in the polluted EC region. Furthermore, the region (west of 110°E, south of 40°N) of TP, a relative clean area in western China was selected as the reference area to comparatively analyze the effects of aerosol pollution in hazy EC on regional precipitation change. we calculated percentages of sites with negative frequency trends of light rain and positive trends of rainstorm events in total sites with the positive and negative trends in haze over the EC and TP regions during the three interdecadal periods (1961-1980, 1971-2000, 1981-2010) (Fig. 6b). During past more than 5 decades, the light rain and rainstorm were receptively steady declined and augmented in the polluted EC, while there were no obvious positive and negative variability trend of light rain and rainstorm in the clean TP region (right panel of Fig. 6b).

6. interannual anomalies between visibility and precipitation

Figure 7 further verified the relation of interannual variability of regional visibility and precipitation in EC over recent years. Regionally averaged, less light rain events and more rainstorms varied significantly from year to year in association with enhanced aerosol levels with declining visibility over EC (Figs. 7a, 7b, 7c and 7d). , Taking summer months (June, July and August) as examples, the 20-year running correlation coefficients of visibility and precipitation were presented in Figure 7e. It is very interesting that the interannual variations of visibility and precipitation over EC were evolved from positive correlations in the early 1960s to negative correlations in the 1970s and 1980s (Fig. 7e), reflecting the interaannul variation of the aerosol and precipitation interaction in changing climate.

In order to investigate the interannual variations of monthly correlation pattern between regional visibility with light rain and extremely precipitation events in EC, we illustrated the cross-section of monthly anomalies of visibility and number of days with light rain and rainstorm, in Figure 8.in. Through a comprehensive comparison of Figs. 8 a, b, c and d, we could find significant positive correlation between visibility and light rain, indicating that the poor visibility suppressed light rain frequency. Moreover, there was a significant difference between the changing extremely rainstorm and light precipitation occurrences. The changes of large and extraordinary rainstorm frequency from 1960s to 1980s were not as prominent as those at the latter period of 1990s, during which time visibility deteriorated remarkably, heavy and extremely heavy rainstorm occurred frequently. Compared to other seasons, the influence effect of poor summer visibility was more significant in EC, showing summertime disastrous rainfalls happened more often with less light rain over recent years. .

The increased atmospheric aerosol concentration may reduce the solar radiation to surface and decrease surface temperature. forming a temperature inversion structure (Bollasina et al, 2011; Zhang et al., 2009; Bond et al., 2013; Bond et al., 2011; Grant et al., 2014; Seinfeld et al., 2008). This temperature inversion structure with the stability of atmospheric boundary layer provides an important condition for the frequent occurrence of haze events. The stable low-level structure also inhibits the weak convection development of atmospheric boundary layer, reducing the formation of low-level clouds and weak precipitation process. However, the strong dynamic convergence disturbance could destroy the stability of atmospheric boundary layer and cause the formation and development of severe rainstorms.

To further clarify the relation between aerosols and light rain frequency, the light rain frequency distribution from 601 stations in July, 2013 is displayed (Fig.9). The light rain events have significantly declined in EC with high aerosol concentrations have and but enhanced in the relative clean TP region (Fig. 9).

To reveal the relationship between aerosols and atmospheric vertical thermal structure, the correlation between surface $PM_{2.5}$ concentrations 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 respectively were selected to represent the less light rain region while those of Linzhi and Dingri of TP were 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 L-band sounding were calculated. The correlations at Changsha and Hongjia stations (Figs.10a-b) show that the correlation between $PM_{2.5}$ and temperature profiles presented an "inverse phase" pattern, reflecting the high aerosol concentrations in a thermal stable structure similar to temperature inversion layers with "cold at low-layer and warm at upper-layer" in the EC. On the contrary, the correlations in Linzhi and Dingri stations in the TP (Fig. 10c-d) indicate that an unstable atmospheric structure with "warm at low-layer and cold at upper-layer" with a favorable condition for the occurrence and development of convection and light rain events in the TP.

7. Physical connection between aerosols and precipitation

According to the results of observation and modeling studies, the increased aerosol concentrations could reduce effective particle radius and increased number concentration of cloud droplets (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) for the negative correlation between aerosols and light precipitation in China (Choi et al., 2008).

In order to further confirm the relationship between aerosols and cloud droplets, the cloud droplet data observed by aircraft in north China during 2008-2010 were used. The vertical profiles of cloud droplets under different aerosol state obtained by 40 aircraft are shown in Figure 11. Aerosol Albrecht "cloud lifetime effect" was significant in the northern EC (Figs. 11a and 11b). Under the background of high aerosol concentrations, the cloud droplets sizes were smaller and increased slowly with the increasing altitude (red profile of Fig. 11b). In addition, from the cloud base to 2000m, the cloud droplets size remained less than 20 microns (Fig.11b); resulting in precipitation delay, in favor of cloud system development to form heavy rain easily. Cloud droplets diameter enlarged quickly with the increase of height, and reached 30 microns easily to forming light rain at 1000m altitude under low aerosol concentrations (green profile in Fig.11b). The aircraft observation analysis showed that high aerosol concentrations could reduce cloud droplets size, increase cloud droplets concentrations, extend cloud lifetime, which could restrict the light rain process.

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 small spatial and temporal scales. The climate forcing of aerosols on precipitation in large-scale region and physical causes remain uncertain. By using precipitation and visibility data of more than 50 years, 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. In the 1960s, the monthly visibility and light rain presented a significantly positive correlation, while the visibility was in good condition. In recent 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.

The investigation of relation between aerosol concentrations and light rain frequency distributions in July, 2013 in China shows that that the light rain appeared significantly low frequency in the EC region with high aerosol concentrations , and but high-frequency in the relative clean region of Tibetan plateau presented significantly. High aerosol concentrations was strongly correlated to warming low-level atmospheric to forming a stable structure suppressing the occurrence and development of light rain events in EC. The aircraft measurements over the EC confirmed that the diameters of cloud droplets decreased under high aerosol concentration condition, thereby inhibiting weak precipitation process.

The findings from this study have important implications for aerosol and precipitation interaction. The frequent haze events in EC not only cause regional environment deterioration, but also induce the long-term change of regional water cycle with the effect on regional climate change.

Acknowledgements

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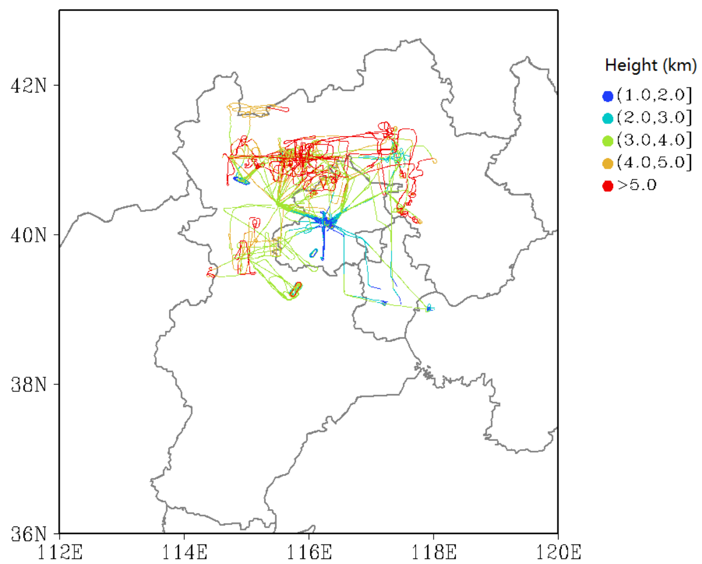
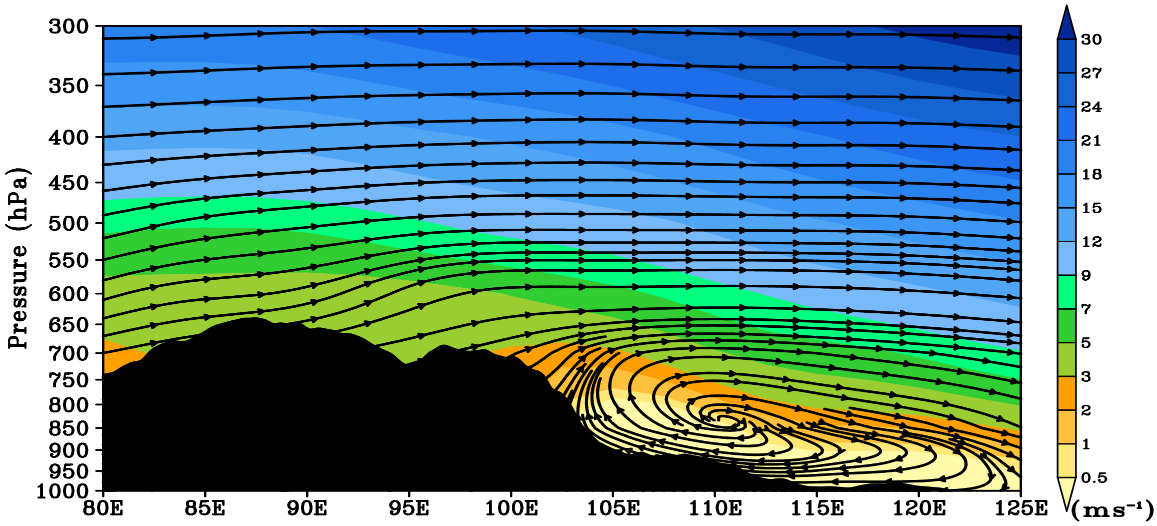


Figure 1 Area and tracks of 40 aircraft flights carried out in Beijing and its surrounding regions during aerosol-cloud experiment from 2008 to 2010 by the Beijing Weather Modification Office. China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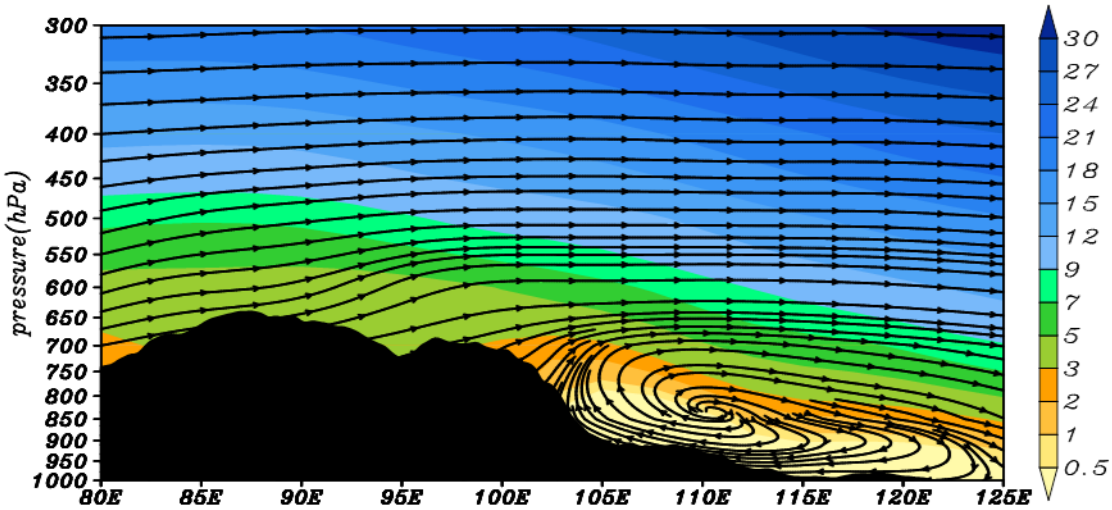
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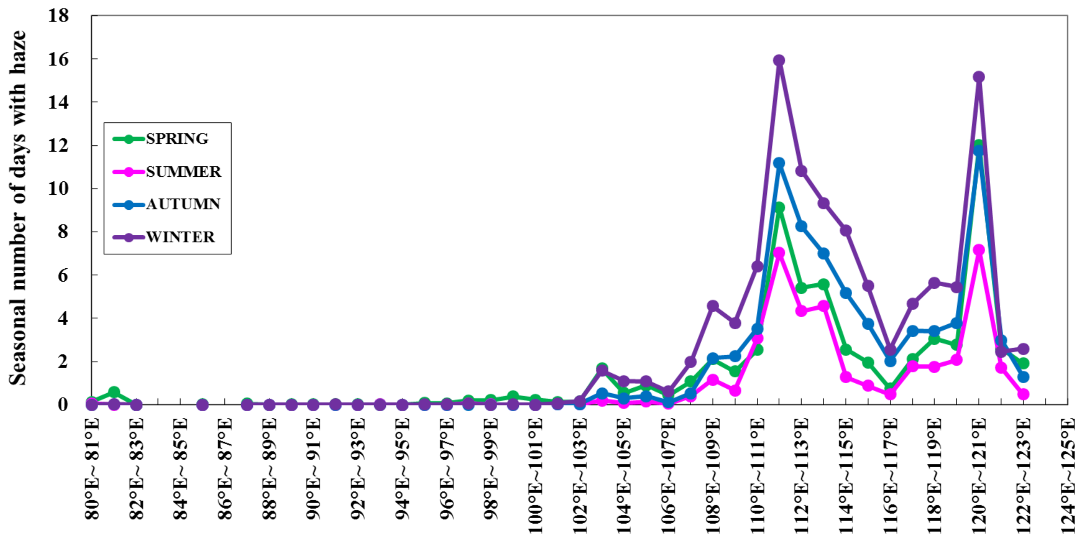
Seasonal number of days with haze
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88°E~89°E
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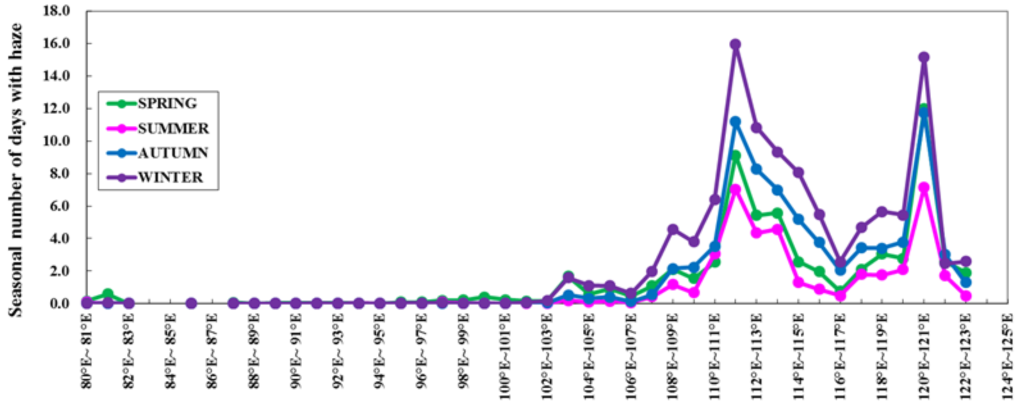

Figure 2. Cross sections of vertical circulations illustrated by stream lines (upper panel) with the horizontal wind speed (m s^{-1}; color contours) and zonal variations of annual haze event frequency (lower panel) at 27 °N-41 °N averaged in spring, summer, autumn and winter over 1961-2012. Note that 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.

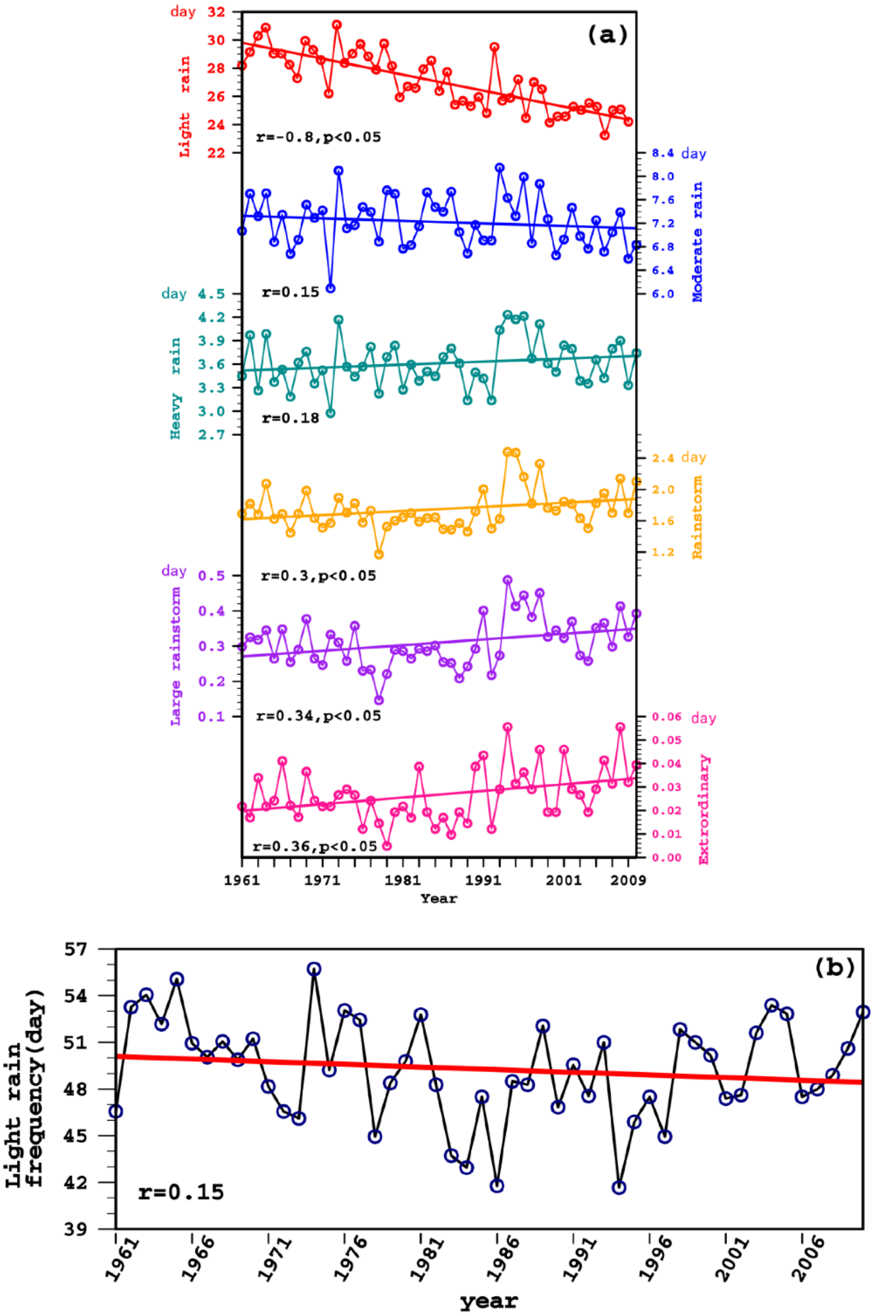

Figure 3 Interannual variations with their anomalies (broken lines) and trends (straight lines) in (a) various precipitation intensities in the high aerosol concentration area in the EC region and (b) light rain in the relative clean area of Tibetan Plateau.

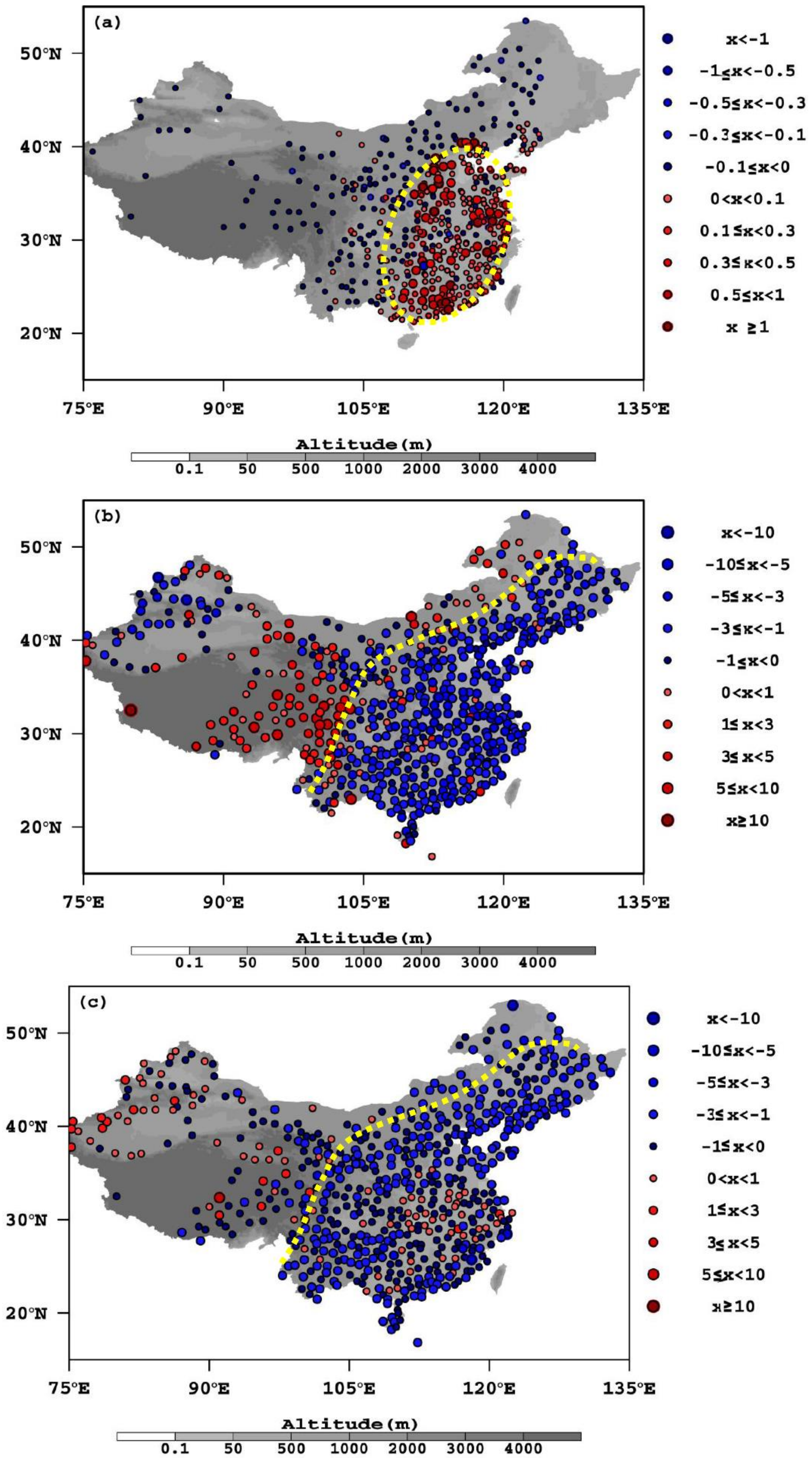

Figure 4 Distribution of interannual change trends (day per 10 years) in (a) haze frequency, (b) visibility and (c) light rain frequency in summer in mainland China in 1961-2010. The yellow dash lines mark the borders of frequent haze area or the

eastern borders of plateaus in China.

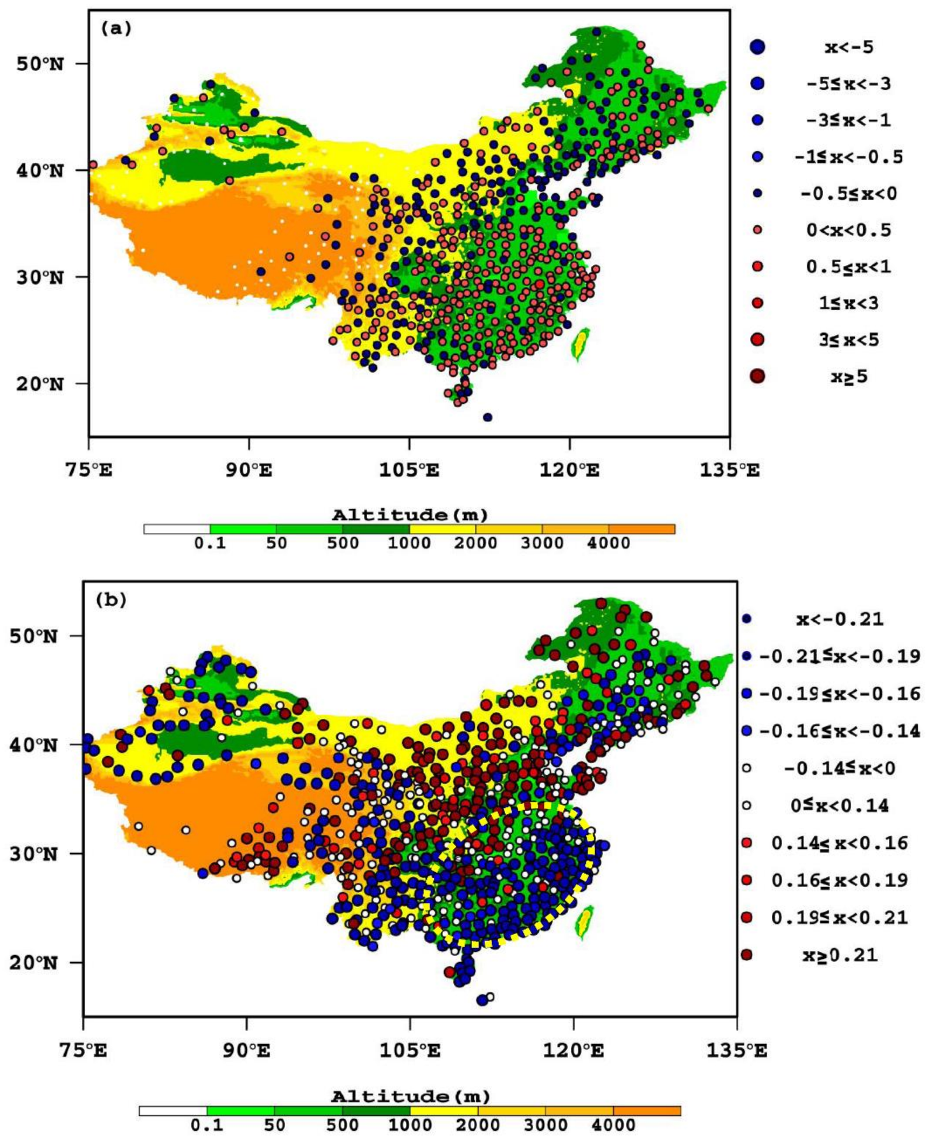

Figure 5 The spatial distributions of (a) trends (day per 10 years) in summertime rainstorm frequency over 1961-2010 in China and (b) correlation coefficients between visibility and low cloud amount in summer of 1961-2010 With the yellow dash line marking the border of negative correlation area.

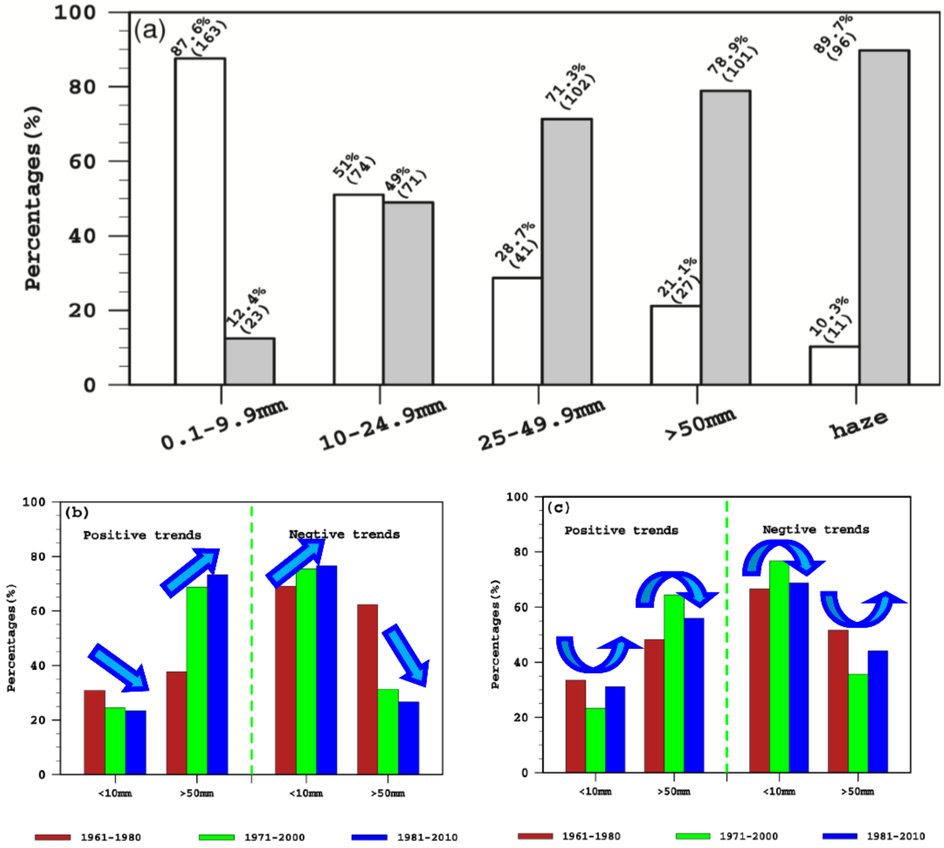

Figure 6 (a)percentages of stations with by positive and negative trends in number of day in various precipitation grades from 1961 to 2010 over the EC stations ,(.The percentages of sites with negative frequency trends of light rain and positive trends of rainstorm events in total sites with the and 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). The arrows indicate the interdecadal change patterns.

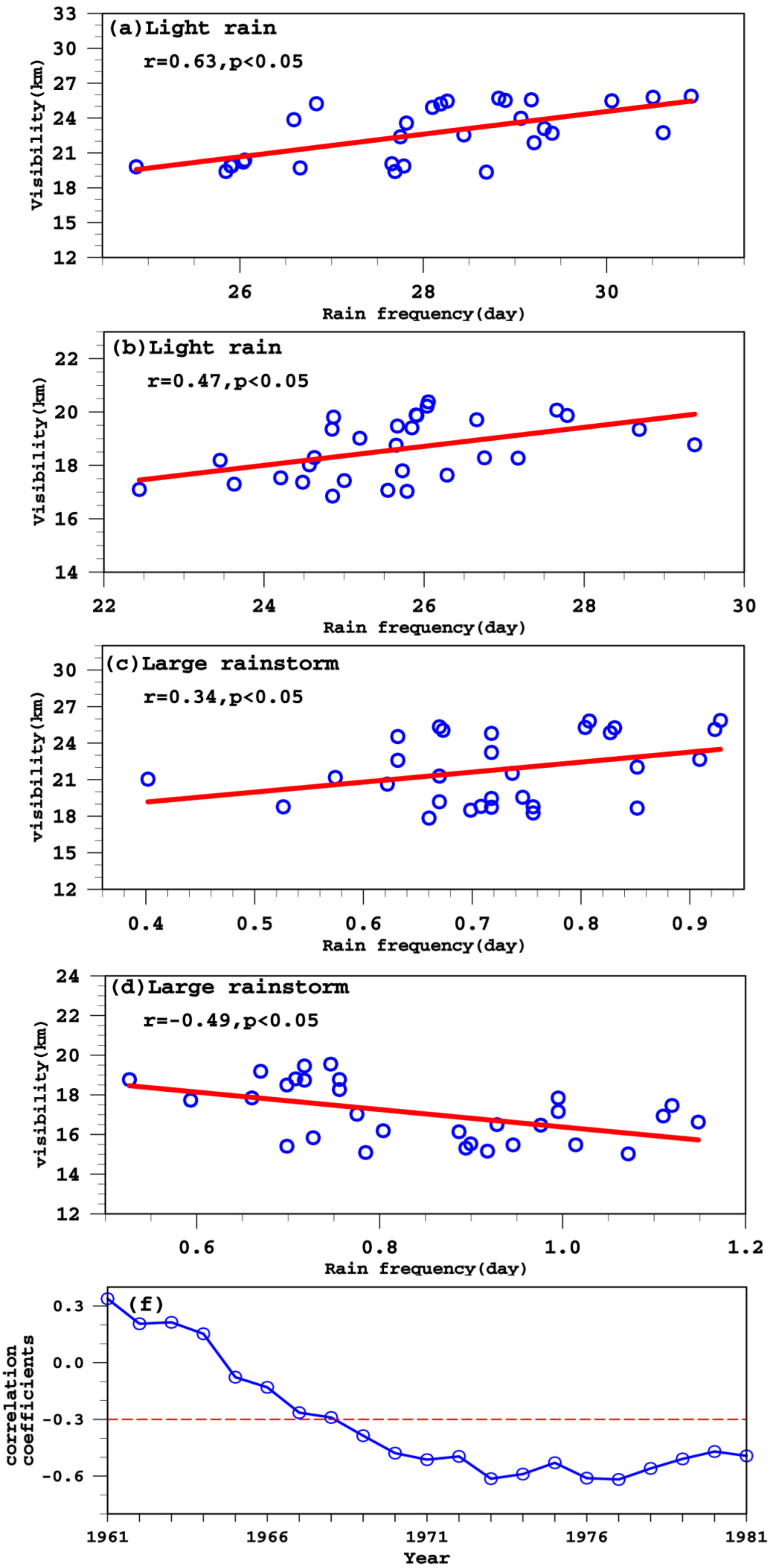

Figure 7 Correlation between summer average visibility (June, July and August) with light rain frequency (a) in 1961-1990, (b) light rain frequency in 1981-2010, (c) extremely heavy rain event frequency in 1961-1990, and (d) extreme heavy rain event

frequency in 1981-2010; (e) The 20-year running correlation coefficients of visibility and precipitation over EC with dash line standing for the correlation passing the confidence level of 90%.

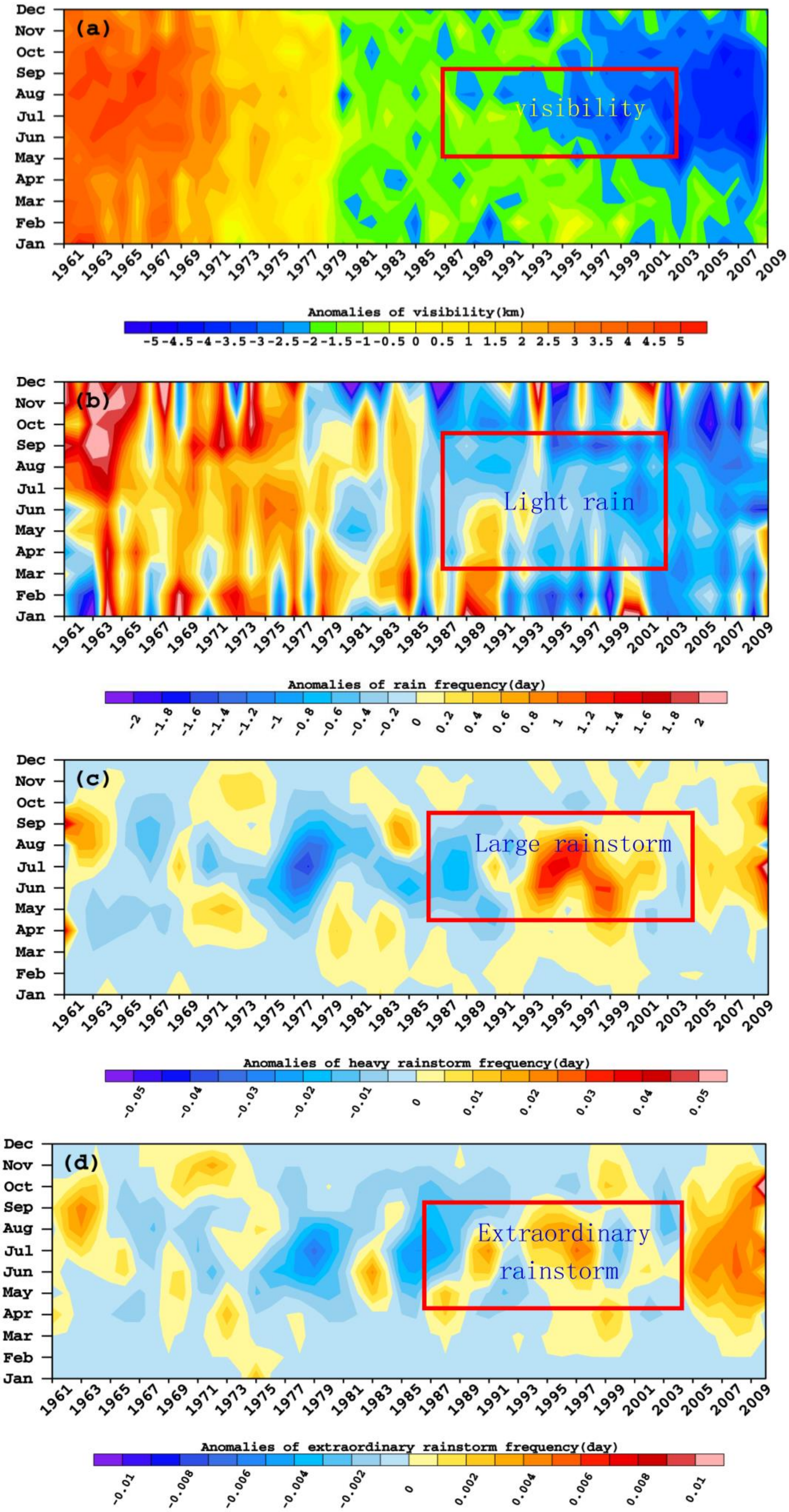

Figure 8 Monthly anomalies of number of days with (a) visibility, (b) light rain, (c) heavy rain, and (d) extremely heavy rain

from 1961-2010 averaged over EC. The red rectangles mark the areas with significant changes.

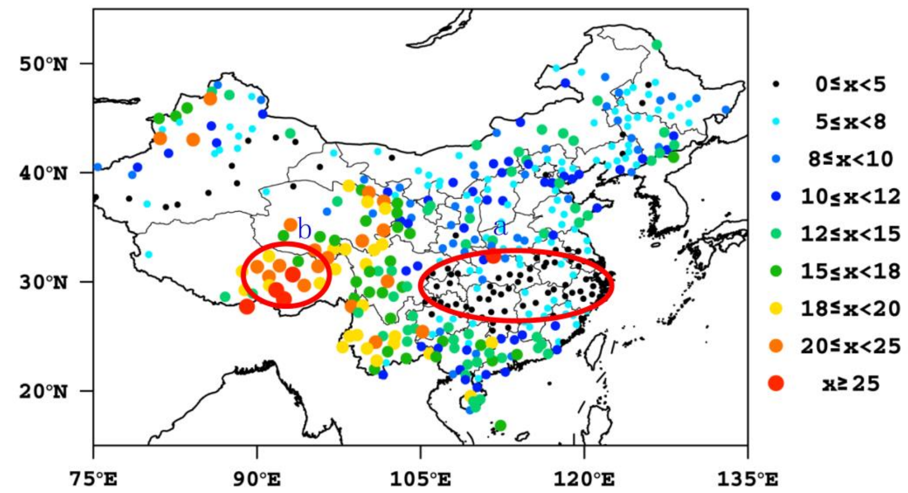

Figure 9 Light rain frequency distribution of 601 stations in China in July of 2013. The circled region on right is the low-frequency light rain region in middle and downstream region in EC, and that on left is the high-frequency light rain region in relative clean region over the TP.

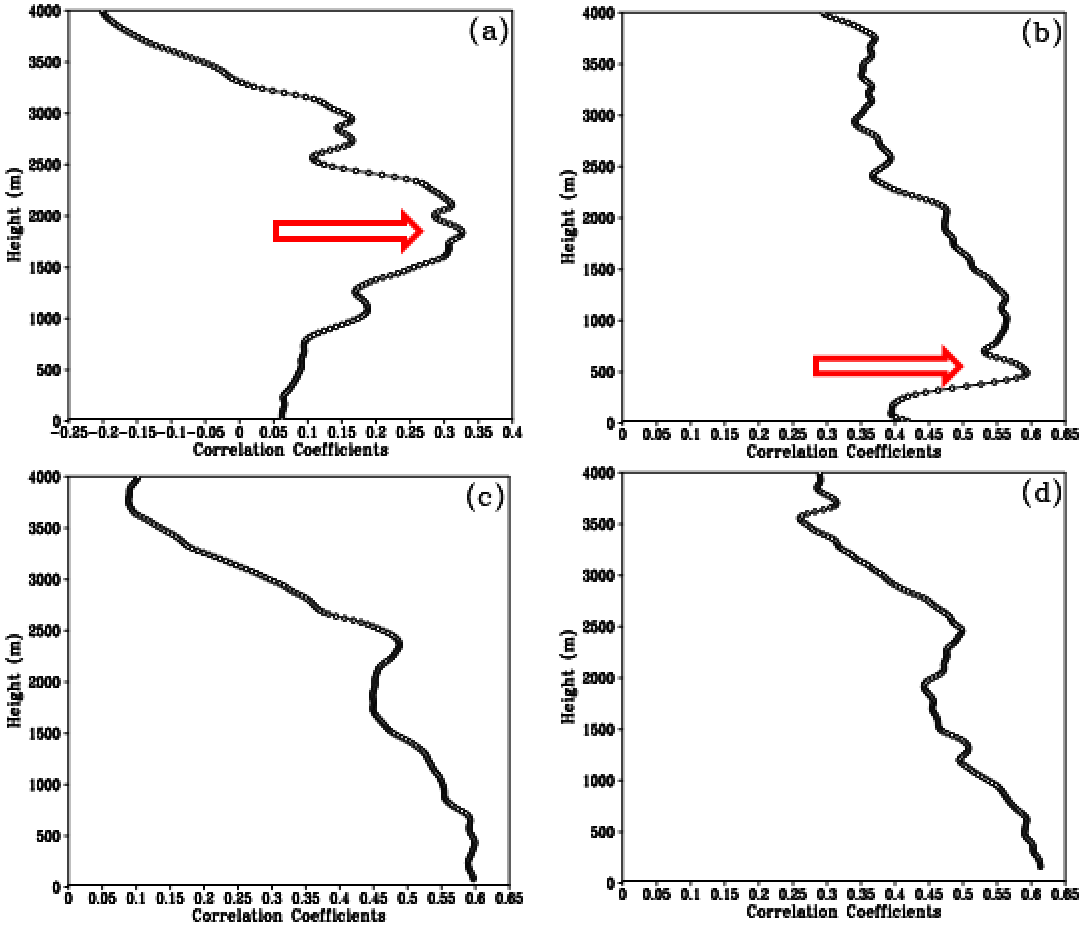

Figure 10 Correlation coefficient profiles between the surface $PM_{2.5}$ concentration (12 hour intervals) and atmospheric temperature from L-band sounding for representing low-frequency light rain regions at stations of (a) Changsha and (b) Hongjia in EC, and for representing high-frequency light rain regions at relative "clean area" at stations of (c) Linzhi and (d) Dingri over the TP with correlation coefficients of 0.21, 0.25 and 0.32 passing the confidence level of 90%, 95% and 99%.

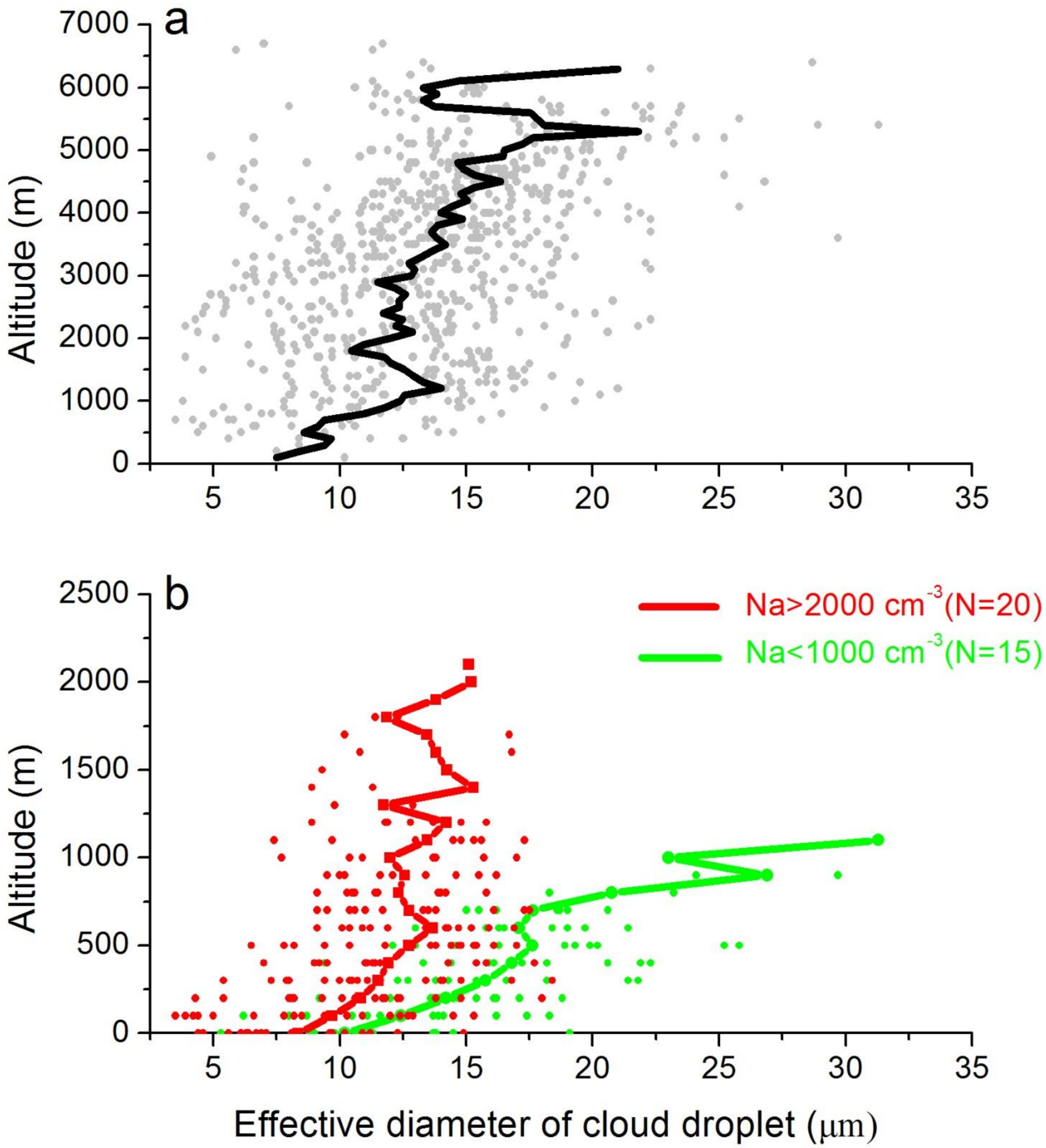

Figure 11 the vertical profiles of (a) sampling cloud droplet size detected by 40 aircraft, and (b) changes and number and diameters of cloud droplets under low and high aerosol number concentrations.