Inter-Annual Variations of Cloud and Precipitation and Their Possible Relationships with Surface Aerosols in Shanghai

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ABSTRACT

Aerosol-cloud-precipitation interactions have attracted much more attention for decades, but there still remain many uncertainties in assessing global climate. Long-term ground-based measurements of aerosol, cloud and precipitation in Shanghai were used to examine their inter-annual variations and possible relationships. During 1990–2010, the yearly-averaged total cloud cover (TCC) and low cloud cover (LCC) decrease on average by 0.58% and 2.49% per year. LCC correlates to surface aerosols (e.g., PM₁₀), with a correlation coefficient (R) of 0.67. Aerosol optical depth (AOD), as an indicator of columnar aerosol loading, shows a non-significant correlation with cloud cover. The yearly-aggregated heavy and extreme rain days and their rainfall amount increase gradually. The moderate rain day enhances but its annual rainfall amount declines year by year, while the light rain exhibits an opposite pattern to the moderate rain. These results imply that local aerosols maybe exert somewhat enforcing on low cloud and light rain through possible entrainment or updraft that can bring up surface particles into free troposphere, whereas its influence to total cloud and precipitation is negligible at a small scale. Future studies are needed to ensure whether local aerosols to directly affect low cloud, and to explore how surface aerosols to enter into higher atmospheric layers and impact cloud and precipitation at larger scales.

Keywords: Rainfall; Aerosol; Cloud cover; Climate change.

INTRODUCTION

The changes of global climate system such as temperature, precipitation, water vapor, cloud and winds have significant impacts on the earth’s environment such as drought and wetness (Karl et al., 1995), as well as aerosol spatio-temporal distribution. Owing to climate changes, cloud properties such as cloud thickness, height and geographical distribution etc., which further affect precipitation, have changed and this situation will continue (Eastman and Warren, 2013). The climate change can cause the variations of atmosphere aerosols either from anthropogenic or natural sources (Levin and Cotton, 2008). Ren et al. (2005) reported that the aggravating sulfate and weakening wind can lead to the decrease of evaporation and sunshine duration, which probably further influence cloud cover. Inversely, aerosols also play an important role in global and regional climate changes. Increased absorptive aerosols (e.g., black carbon) not only result in larger diabatic heating and columnar atmosphere warming, but also contribute to lower relative humidity (Perlwitz and Miller, 2010). More studies are needed as the complicated implication of pollution to global climate change.

To date, the effect of aerosols on cloud remains highly uncertain. Perlwitz and Miller (2010) proposed that the relationship between aerosols and clouds varies with aerosol absorptivity, atmospheric circulation, and specific humidity caused by aerosols. The cloud-aerosol interaction differs from region to region, partly due to the spatial variations of pollutants (Wild, 2009). Based on satellite retrievals during Asian dust storm, East Asia has experienced a reduction of cloud cover (Huang et al., 2006). Dark haze can reduce cloud
cover significantly, and thus neutralize aerosol-induced radiative cooling at the top of atmosphere (Ackerman et al., 2000). Zhang et al. (2007) found an increase of deep convective clouds in Pacific storm relevant to Asian pollution. In the west coast of North Africa where the soil dust is abundant, the amount of thin low cloud and mineral dust show a positive correlation (Mahowald and Kiehl, 2003).

Precipitation can be affected by aerosols through multiple processes. The aerosol direct radiative forcing tends to suppress precipitation as they decrease the amount of solar radiation downward on surface, which can reduce heat available for emerging water vapor and energizing convective rain clouds. Absorptive aerosols can heat air masses above the surface by absorbing solar radiation to stabilize the low atmosphere and to inhibit the generation of convective clouds (Rosenfeld et al., 2008). Furthermore, the microphysical effects of aerosols tend to change cloud cover, prolong cloud lifetime, and consequently influence precipitation (Devara et al., 2013). Aerosols tend to reduce rainfall for warm or shallow clouds (Ackerman et al., 2003; Rosenfeld et al., 2008), however, aerosols can increase precipitation by enhancing convection and accelerating the conversion of clouds to rainfall (Koren et al., 2005; Lin et al., 2006). Many researchers have studied the impact of aerosols on rainfall using long-term data, but the conclusions is not consistent to date. Because of the indirect effect, aerosols can suppress the precipitation of shallow cloud and extend cloud lifetime (e.g., the shallow cloud development over the Atlantic Ocean) (Kaufman et al., 2005). A possible positive feedback between reduced precipitation and accumulated aerosols over eastern central China has been reported by Zhao et al. (2006). Rosenfeld et al. (2007b) proposed that aerosol and orographic precipitation have an inverse relationship over central China. In contrast, the observational and modeling studies indicate when deep convective clouds are prevailing, the delay of precipitation can invigorate the storm dynamics by freezing super-cooled water transported aloft (Levin and Cotton, 2008). More efforts are still needed to clarify the response of rainfall to accumulated aerosols.

Since 1970, the enhanced human activities, such as fossil fuel combustion and industrial emissions, have led to rapid and continuous increase in aerosols and their precursors over the Yangtze River Delta region (YRD) (Tie and Cao, 2009). In the YRD region, there is a rising trend in hazy days since 1990 (Che et al., 2007; Chang et al., 2009). Annual mean aerosol optical depth (AOD) has increased from 2000 to 2007 (He et al., 2012). Higher AODs are attributed to the existence of persistent haze layers. Meanwhile, a sharp decrease of visual range from 13.2 to 10.5 km has been observed in the YRD region during 1980–2000, followed by a fluctuating variation around low values during 2001–2011 (Cheng et al., 2013).

In this study, the long-term variations of aerosol, cloud and precipitation over Shanghai are investigated through analyzing ground-based measurements. The aim is to elucidate the possible relationships between surface aerosols, cloud and precipitation, and to understand whether local particulate pollutants is potential to influence cloud and precipitation at a small scale of urban environment.

PARAMETERS

**DATA**

Long-term meteorological data of precipitation, cloud cover and atmospheric sounding were collected from ground-based in situ measurements at a basic meteorological station (Baoshan, Lat: 121.48°E, Long: 31.41°N) in Shanghai. The daily precipitation data spanned from 1981 to 2013, and the daily cloud cover data was from 1990 to 2010. Atmospheric sounding data was used to calculate atmospheric stability for 1992–2013. Additionally, available pressure, atmospheric visibility, temperature and wind speed were used to analyze climate changes for 1981–2010. AODs at 550 nm for 2000-2013 were derived from the retrieval dataset of MODIS (Level 2-Collection 5) on board the Terra satellite, which overpasses Shanghai at approximately 10:30 a.m. local time (Chu et al., 2002). In addition, we also derived cloud parameters (e.g., cloud fraction, liquid water path, cloud optical depth and water particle radius) from CERES SYN1deg. The hourly PM10 concentrations during 2000–2012 were obtained through averaging the measurements at 9 air quality observation stations of the Shanghai Environmental Monitoring Center (http://www.semc.com.cn).

The tendency of stable-day occurrence frequency can reveal the variation of atmospheric stability. This frequency is calculated as the ratio of days when the difference of convective available potential energy (CAPE) and convective inhibition (CIN) is negative to the available sounding days over a year. Among them, CAPE refers to maximum energy available to an anabatic parcel, while the energy consumed in the process of air parcel pseudo-adiabatically lifting from its original level to free convection level is defined as CIN. Both CAPE and CIN are available from the atmospheric sounding.

**RESULTS AND DISCUSSION**

The Trend of Climate Changes

According to IPCC reports, the climate change is defined as any change in climate system over time generally for 10 years or longer, either due to natural variability or human activity (Ding et al., 2001). Fig. 1 shows the temporal variations of annual mean temperature, vapor pressure, wind speed and sunshine duration over 1981–2010. Overall, the temperature grows significantly by 2.4°C with a mean rate of 0.079 °C/yr. It increases slightly by 0.5°C during the first decade. However, a more rapid increase by about 2°C appears in the following period. Both extreme low and high temperatures also experience an obvious ascending. In fact, Shanghai area undergoes a great extension of urban heat island during 1981–2010 (Li et al. 2013), which can be significantly enhanced by the increased pollutants (Czarnecka and Nidzgorska, 2014). Vice versa, the urban heat island can change turbulence, which further impacts on the primary and secondary regional pollutants, promoting transport of aerosols, water and gaseous pollutants to the middle and upper troposphere (Crutzen, 2004; Sarrat et al., 2006).

The annual mean wind speed displays a declining tendency (~0.02 m/s per year) from 1981 to 2010. This variation can be divided into three stages: 1981–1995, 1998–2002 and
Fig. 1. Long-term variations of yearly mean wind speed, vapor pressure, temperature and sunshine duration over 1981-2010, and the dash lines in the figure indicate the linear trend of wind speed, vapor pressure, temperature and sunshine duration during 1981–2010. The thin solid and thick solid lines indicate the linear trend of vapor pressure for the period of 1981–2000 and 2000–2010, respectively.

The wind speed gradually drops from 4 to 2.9 m/s during 1981–1995, while a sudden increase occurs in 1996 and 2002, which is similar with the result of Zhou and Liang (2013). Following these relatively short periods of increasing winds, the wind speed decreases by 0.08–0.09 m/s per year. Overall, the wind speed declines simultaneously with the temperature rise, consistent with the report of Chen (1998). Global warming and winter/summer monsoon weakening in East Asian are probably responsible for wind slowing in Shanghai (Jiang et al., 2010). The vapor pressure presents a gradual increase trend before 1998 and a steady decline trend afterwards. The sunshine duration shows a decreasing trend during 1980–2010, which agrees with the previous finding in China (Qian et al., 2007). The changes of cloud fraction is reported as a crucial factor to impact sunshine duration in many studies (Cutforth and Judiesch, 2007; Li et al., 2011). In this study, the decline of sunshine duration can partly explained by the change of total cloud cover (TCC), a correlation coefficient of −0.41 between them. However, there is no correlation existing between sunshine duration and low cloud cover (LCC). Xia (2010) proposed that the reduction of sunshine duration in southern China is strongly associated with increasing LCC, while TCC shows non-correlation with sunshine duration. You et al. (2010) reported that sunshine duration is negative correlated with changes of both LCC and TCC. The indirect effect of aerosols can change surface solar radiation and sunshine duration by acting as cloud condensation nuclei (CCN) to influence cloud and precipitation processes. The increasing aerosol loading is likely major contributor to impair sunshine duration not only under cloud-free conditions but also under cloudy conditions (Xia, 2010). In addition, different research times and areas are likely another factors for these discrepancies.

Relation between Cloud and Aerosol

Fig. 2 shows the inter-annual variations of PM$_{10}$ and AOD during 2000–2012, and the number of haze event (i.e. atmospheric visibility < 10 km and relative humidity < 90%) during 1981–2012. A gradual decrease of annual mean visibility in Shanghai can be ascertained by Chang et al. (2009). The number of haze days keeps a fluctuant rising trend during 1981–2010, especially during 2000–2010. AOD increases gradually since 2000 and is generally over 0.6, except in 2010 when the multiple measures of air pollution control were taken in Shanghai during the world Expo 2010. PM$_{10}$ shows a decreasing trend during 2000–2012. These changes in recent years may result from the large quantities of fine particles or even ultrafine particles, which are the most important contributor to low visibility weather such as haze (Fu et al., 2008; Pan et al., 2010; Huang et al., 2012). Shanghai is intensively influenced by human activity to emit a large amount of carbonaceous and secondary species (sulfate, nitrate, and ammonium) that can contribute to the formation of fine particles (< 2.5 µm) (Ye et al., 2003). Generally, the fine mode particles in Shanghai keep an increasing tendency in recent years (Chen et al., 2003; Ye et al., 2003; Wang et al., 2006; Kan et al., 2007; Senlin et al., 2008; Feng et al., 2009; Kang et al., 2014). The aerosol mass extinction efficiency (Chin et al., 2002), relative contributions of different aerosol types to columnar AOD, and the fractional AOD of surface layer
Fig. 2. Series of annual mean PM$_{10}$ (from 2000 to 2012), aerosol optical depth (AOD) (from 2000 to 2012) and haze days (from 1981 to 2010), and the straight line in the figure indicates the linear trend of haze days from 1981 to 2010.

(Van Donkelaar et al., 2006) are the major factors that influence the relationship of AOD and surface PM$_{10}$ concentration. PM$_{10}$ is in low mass extinction efficiency, resulting in a low correlation between PM$_{10}$ and AOD. The AOD value is more sensitive to the mass concentration of PM$_{2.5}$ rather than PM$_{10}$ (Song et al., 2009).

To exclude the impact of aerosol obscuring on the ground observations of cloud cover, we compare TCC and LCC measured by CERES satellite and ground from 2000 to 2010. The cloud cover measured from space and surface shows a similar variation. Correlation analyses show that annual mean LCC measured by satellite is significantly related to the surface measured LCC (R = 0.64), while TCC shows a weak correlation (R = 0.47). Meanwhile, ΔTCC (defined as TCC measured by satellite minus TCC observed in ground) is negative correlated with AOD (R = -0.44), while ΔLCC (defined as LCC measured by satellite minus LCC observed in ground) is positive correlated with AOD (R = 0.6). These evidences indicate that aerosols impact little on the ground observation of LCC, but they can affect the ground observation of TCC (Sun et al., 2014). Combining cloud covers measured by ground (1990–1999) and derived from satellite (2000–2010) are used to discuss the variation of cloud cover in Shanghai. Decadal analysis shows a slight increase of TCC and a rapid increase of LCC from 1990 to 2000, but both of them decrease from 2001 to 2010 (Fig. 3). According to previous studies, enhanced pollution might be the contributor to the variation of cloud cover (Rosenfeld, 2000; Jacobson, 2001; Lal et al., 2013). Black carbon concentrations increase significantly due to industrialization and urbanization in recent years in China, especially in Shanghai (Qian et al., 2009; Huang et al., 2012). Black carbon can facilitate low atmosphere heating to dispel low cloud and to impair warm-rain efficiency by semi-direct effect of aerosols (Menon et al., 2002), i.e. the phenomenon that aerosols can inhibit cloud formation through absorbing solar radiation (Jacobson, 2001; Perlwitz and Miller, 2010), heating atmosphere, and causing cloud droplet evaporation (Kaufman and Koren, 2006; Zhao et al., 2006). The frequency of low TCC (< 2.0), high TCC (> 8.0) and high LCC (> 8.0) exhibit a downward trend during 1990–2010, except a slight increase in low LCC (< 2.0). In general, high TCC and LCC enhance before 2000 and decline in the late decade, while low TCC and LCC experience an opposite pattern. There are more significant changes in LCC than TCC.

In polluted environments, owing to aerosol indirect effect, cloud tends to have more abundant but smaller droplets, which can reflect more sunlight to space and cool the Earth (Twomey, 1977). The smaller cloud droplets tend to reduce droplet growth by collision coalescence, and therefore extend cloud lifetime and decrease precipitation (Warner, 1968; Rosenfeld, 2000). Correspondingly, the increasing cloud optical depth is observed during 2003–2012 (Fig. 4), which is similar with the variation of AOD. However, cloud cover follows an inverse trend. The increased fine particles are more likely to produce a large number of CCN, which then effectively reduce cloud droplet radius (as shown in Fig. 5). According to Kelvin effect, small cloud droplet radius favors water evaporation which in further suppresses cloud formation and eventually leads to less cloud cover, thus cloud droplets have enough time to reach higher altitudes (Rosenfeld, 2007a). Correspondingly, cloud top pressure shows a decreasing trend in 2003–2010 (Fig. 5). This enlarges cloud liquid water path and ascends non-precipitating water to high levels, and then results in reducing LCC formation (Lal et al., 2013). Meanwhile, cloud is also driven by atmospheric humidity and stability.
The ratio between condensation and evaporation of water vapor determines precipitation (Lal et al., 2013). Before 1998, vapor pressure keeps rising in Shanghai. Therefore, condensation is dominant and low clouds experiences rapid formation. This process results in the development of abundant low clouds before 2000. After that, vapor pressure decreases and evaporation increases rapidly. Precipitation is restrained and more non-precipitation water is transported upward. As a result, the attenuation of LCC is observed since 2000.

In addition, Zhao et al. (2006) argued when the atmosphere becomes more stable, the upward motion is depressed and cloud formation rate is inhibited, resulting in reduction in cloud cover. Eastman and Warren (2013) proposed that variations of tropospheric stability can impact cloud amount and type. According to their study, heavy pollution...

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**Fig. 3.** Series of (a) annual mean total cloud cover (TCC) and low cloud cover (LCC), and the dash and solid lines in the figure indicates the linear trend of TCC/LCC during the period of 1990–2000 and 2000–2010. (b) frequency of daily TCC lower than 2.0 and TCC larger than 8.0, and the dash line in the figure indicate the linear trend of TCC lower than 2.0 or TCC larger than 8.0 in the period of 1990–2000, while the solid line in the figure indicate the linear trend of TCC lower than 2.0 or TCC larger than 8.0 in the period of 2000–2010, (c) frequency of daily LCC lower than 2.0 and LCC larger than 8.0 from 1990 to 2010, and the dash line in the figure indicate the linear trend of LCC lower than 2.0 and LCC larger than 8.0 during the period of 1990–2000, while the solid line in the figure indicate the linear trend of LCC lower than 2.0 and LCC larger than 8.0 during the period of 2000–2010.
can stabilize atmosphere and then depress cloud formation. Here we find the frequency of stable days keeps intensifying since 2000 in Shanghai, indicating a reduction of LCC since 2000. It should be noted that the interactions between aerosol and cloud cover is complicated.

Linear regression analyses are conducted to investigate the relationship of TCC (LCC) and PM$_{10}$ (AOD) (Table 1) in Shanghai. We find AOD and TCC (LCC) is weakly correlated ($R = -0.24$ and $-0.25$). The wet scavenging of aerosols by precipitation in clouds, namely the predominant sink of CCN, could presumably explain the negative relationship between AOD and cloud cover. Meanwhile, positive correlation between TCC and AOD derived from satellite data has been found in the previous studies (Loeb et al., 2005). Quaas et al. (2010) proposed six main hypotheses to interpret the relationship between AOD and TCC. Among these wet scavenging is the only one showing negative correlation. However, the correlations between PM$_{10}$ and TCC (LCC) are fairly different ($-0.26$ and $0.67$), indicating that PM$_{10}$ exerts somewhat contribution to low cloud formation and lifetime through possible entrainment or updraft that can bring up surface particles into free troposphere. Choi et al. (2008a) also reported a possible link between cloud formation and PM$_{10}$. Different from AOD,
PM$_{10}$ has a strong positive correlation (0.71) with high LCC (> 8.0), but a negative relation (−0.43) with low LCC (< 2.0). According to the urban turbidity island theory, with high PM$_{10}$ loadings, more hygroscopic nuclei and strong thermal mechanical turbulence in urban atmosphere can lead to the increase low cloud cover, along with reduced number of clear days (Shuzhen et al., 1991). As fine particles increase in recent years in Shanghai, they accounted for a larger fraction in decreasing PM$_{10}$. The aggravated urban heat may lead to stronger entrainment or updraft, and then to change turbulence and cause more primary and secondary regional pollutants into high tropospheric layers (Crutzen, 2004; Sarrat et al., 2006).

**Relation between Precipitation and Aerosol**

Fig. 6(a) shows the variations of annual rainfall amount and rainy days during 1981–2013, and Fig. 6(b) presents the trends of precipitation intensity for 1981–2013 and precipitable water amount for 1992–2013. The annual rainfall amount and rainy days increase by 14 mm/yr and 0.12 days/yr for 1981–2013, respectively. In general, the variations of rainfall amount and rainy days can be divided into two time periods: 1981–2003 and 2003–2013. The annual rainfall amount increases by 0.58 mm per year, and the annual number of rainy days decreases by 0.27 days per year during 1981-2003. The sharp increase appears after 2003 for both rainfall amount and rainy days, which is probably impacted by increased pollution (haze days and AOD) since 2003. More intensified convection and efficient mixed phase processes in the polluted aerosol condition can influence precipitation (Wang et al., 2011). Zhou et al. (2013) contributed the decadal change of precipitation over eastern China during the summer monsoon season to heavy pollution. The increase of rainy days during 2003-2013 can be explained by aerosol self-cleansing effect, that is, the accumulation of aerosol loading (AOD and haze days) tends to increase rainy days, which consequently eliminates aerosols from the atmosphere. Then aerosol loadings are quickly accumulated again within a few days to duplicate the cycle (Choi et al., 2008a). We find the precipitation intensity increases by 0.09 per year, with a sharp increase occurs after 2003. Previous studies have demonstrated that the restrain of early rainfall caused by aerosols can lead to greater precipitation intensity and cloud water amount at later stage (Tao et al., 2007; Philips et al., 2007). With high aerosol concentration, the non-precipitating water is more readily to reach high altitudes and converts into ice which releases the latent heat when freezing aloft (Rosenfeld et al., 2008). This can increase the cloud buoyancy and result in the formation of deeper clouds (Rosenfeld, 2000; Rosenfeld et al., 2008). Stevens (2007) pointed out that the deeper cloud with higher liquid water content may enhance precipitation. Furthermore, the clouds with different dimensions generate various precipitation intensities, i.e., deeper cloud may lead to more intense rainfall (Nuijens et al., 2009). As the key factor of precipitation, precipitable water does not appear an elevating trend during 1992–2013 as excepted. A rising tendency during 2003–2013 is consistent with total rainfall and pollution. This result indicates that precipitable water may only play a minor role in altering precipitation characteristics.

According to the China Meteorological Administration (CMA), the daily rainfall intensity is classified into four
Fig. 6. Series of (a) annual rainy days and rain amount, and the dash, thin solid and thick solid lines in the figure indicate the linear trend of rainy days (rain amount) during the period of 1981–2012, 1981–2003 and 2003–2012, respectively. (b) precipitation intensity and precipitable water from 1981 to 2013, and the dash, thin solid and thick solid lines in the upper figure indicate the linear trend of precipitation intensity during the period of 1981–2012, 1981–2003 and 2003–2012, respectively. The thin dash, thick dash, thin solid and thick solid lines in the under figure indicate the linear trend of precipitable water for 1992–2012, 1992–2000, 2000–2004 and 2004–2012, respectively.

grades, i.e., small rain (0–9.9 mm/d), moderate rain (10–24.9 mm/d), heavy rain (25–49.9 mm/d) and extreme rain (> 50 mm/d). Fig. 7 displays the inter-annual variations of light rain and moderate rains. Although the light rainy days decreases by 0.09 days per year, the light rainfall amount increases by 0.22 mm per year in Shanghai, which is different from the declined frequency and amount of light rain in eastern China for 1956–2005 (Qian et al., 2009). Notably, the decadal analysis presents a slight decrease of light rainy days in 1981–2003, and a rapid increase in the following years. The correlation coefficient (~0.54) indicates that somewhat relationship is exist between PM<sub>10</sub> and light rainy days (Table 2). PM<sub>10</sub> is easier to act as CCN and can impact low warm cloud. The enhancement of low-level warm cloud caused by lower PM<sub>10</sub> enriches warm-rain efficiency (Kaufman et al., 2005; Choi et al., 2008b). No statistically
Fig. 7. Series of annual days and amount of (a) light rain and (b) moderate rain from 1981 to 2012. The dash, thin solid and thick solid lines in the figure indicate the linear trend of light rain (events and amount) or moderate rain (events and amount) during the period of 1981–2012, 1981–2003 and 2003–2012, respectively.

Table 2. Correlation coefficients (R) between rainy days and aerosol optical depth (AOD) and PM$_{10}$.

|          | Light rain | Moderate rain | Heavy rain | Extreme rain | Total rain |
|----------|------------|---------------|------------|--------------|------------|
| AOD      | –0.09      | –0.42         | –0.13      | –0.22        | –0.36      |
| PM$_{10}$| –0.54      | 0.06          | –0.05      | –0.34        | –0.43      |

meaningful relationship exists in light rain days and AOD. The light rainfall amount increases at the rate of 1.2 mm/yr for 1981–2003, while decreases by 3.55 mm/yr for 2003–2013. The change of moderate rain is insignificant, with the rainfall amount falling by 0.016 mm/yr and the rainy days increasing by 0.008 days/yr. A rapid increase of moderate rain occurs during 2003–2013, consistent with AOD and reverse to PM$_{10}$. The relationship between PM$_{10}$
and moderate rain is negligible, while the weak correlation coefficient shows that AOD and moderate rain have some relations. These results show very weak correlations between surface aerosols (e.g., PM$_{10}$) and moderate rain. However, higher aerosol loadings could allow more moisture to reach the freezing level (Khain et al., 2005; Lau et al., 2005). Ice nucleation in the mid-troposphere occurs immediately above the freezing level and/or the influence on initial updrafts ruins the low-level warm clouds by aerosol indirect or semidirect effects (Menon et al., 2002; Kaufman et al., 2005; Zhao et al., 2006). This strengthens the midlevel ice clouds and leads to more moderate events.

Overall, during 1981–2013, heavy rainy days and rainfall amount increase by about 0.11 days/yr and 5.4 mm/yr, respectively (Fig. 8(a)). The change of heavy rainy days and rainfall amount is insignificant during 1981–2003, but has a noticeable increase in 2003–2013. A significant increasing trend of extreme rain is observed over the whole period, especially during 2003–2013, with 0.09 days/yr for rainy days and 8.4 mm/yr for rainfall amount (Fig. 8(b)).

Fig. 8. Series of annual days and amount of (a) heavy rain and (b) extreme rain from 1981 to 2012. The dash, thin solid and thick solid lines in the figure indicate the linear trend of heavy rain (events and amount) or extreme rain (events and amount) during the period of 1981–2012, 1981–2003 and 2003–2012, respectively.
Together with the shrinkage of light rain, these evidences suggest a shift of precipitation from light to heavy and extreme rain. The increase of extreme rainy days has been attributed to increasing aerosols loadings (Li et al., 2008; Fan et al., 2012). It was observed that the number concentrations of rain droplets are slightly higher in clean environment. In contrast, pollution cases tend to result in much larger size of rain droplets (Wang et al., 2011). This conduces to the development of heavy even extreme rainfall. In addition, high aerosol concentrations tend to promote the formation of deeper cloud, which increases the possibility of more intense rainfall (Nuijens et al., 2009). Aerosol particles can transport to upper levels due to the impact of suppression of warm-cloud, resulting in intense thunderstorm and more transport to upper levels due to the impact of suppression of nocturnal drizzle in stratocumulus polluted by haze. Geophys. Res. Lett. 30: 1381, doi: 10.1029/2002GL016634. Andreea, M.O., Rosenfeld, D., Artaxo, P., Costa, A.A., Frank, G.P., Longo, K.M. and Silva-Dias, M.A.F. (2004). Smoking Rain Clouds over the Amazon. Science 303: 1337–1342. Chang, D., Song, Y. and Liu, B. (2009). Visibility Trends in Six Megacities in China 1973-2007. Atmos. Res. 94: 161–167. Che, H., Zhang, X., Li, Y., Zhou, Z. and Qu, J. (2007). Horizontal Visibility Trends in China 1981-2005. Geophys. Res. Lett. 34: L24706, doi: 10.1029/2007GL031450. Chen, L., Zhu, W., Wang, W., Zhou, X. and Li, W. (1998). Study on Climate Change in China in Recent 45 Years. Acta Meteorol. Sin. 12: 1–17. Chen, M., Li, D. and Chen, C. (2003). Survey and Analysis on the Status Quo of Fine Particulates Pollution in Shanghai. Shanghai Environ. Sci. 22: 1038–1055 (in Chinese with English Abstract). Cheng, Z., Wang, S., Jiang, J., Fu, Q., Chen, C., Xu, B., Yu, J., Fu, X. and Hao, J. (2013). Long-term Trend of Haze Pollution and Impact of Particulate Matter in the Yangtze River Delta, China. Environ. Pollut. 182: 101–110. Chin, M., Ginoux, P., Kinne, S., Torres, O., Holben, B.N., Duncan, B.N., Martin, R.V., Logan, J.A., Higurashi, A. and Nakajima T. (2002). Tropospheric Aerosol Optical Thickness from the GOCART Model and Comparisons with Satellite and Sun Photometer Measurements. J. Atmos. Sci. 59: 461–483. Choi, Y.S., Ho, C.H., Chen, D., Noh, Y.H. and Song, C.K. (2008a). Spectral Analysis of Weekly Variation in PM\text{\textsubscript{10}} Mass Concentration and Meteorological Conditions over China. Atmos. Environ. 42: 655–666. Choi, Y.S., Ho, C.H., Kim, J., Gong, D.Y. and Park, R.J. (2008b). The Impact of Aerosols on the Summer Rainfall Frequency in China. J. Appl. Meteorol. Climatol. 47: 1802–1813. Chu, D., Kaufman, Y.J., Ichoku, C., Remer, L.A., Tanré, D. and Holben, B.N. (2002). Validation of MODIS Aerosol Optical Depth Retrieval over Land. Geophys. Res. Lett. 29, doi: 10.1029/2001GL013205. Crutzen, P.J. (2004). New Directions: The Growing Urban Heat and Pollution “Island” Effect—Impact on Chemistry and Climate. Atmos. Environ. 38: 3539–3540. Cuthbert, H.W. and Judiesch, D. (2007). Long-term Changes to Incoming Solar Energy on the Canadian Prairie. Agric. For. Meteorol. 145: 167–175. Czarnecka, M. and Nidzgorska-Lencwicz, J. (2014). Intensity of Urban Heat Island and Air Quality in Gdańsk during 2010 Heat Wave. Pol. J. Environ. Stud. 23: 329–340.

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