The Impact of Aerosols on the Summer Rainfall Frequency in China

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ABSTRACT

The authors investigate the short-term relationship between aerosol concentrations and summer rainfall frequency in China using the daily surface observations of particulate matters with a diameter of less than 10 μm (PM10) mass concentration, rainfall, and satellite-observed cloud properties. Results in this study reveal that on the time scale of a few days aerosol concentration is positively correlated with the frequency of moderate-rainfall (10–20 mm day^{-1}) days but is negatively correlated with the frequency of light-rainfall ($<$5 mm day^{-1}) days. Satellite observations of cloud properties show that higher aerosol concentrations are positively correlated with the increase in mixed cloud amount, cloud effective radius, cloud optical depth, and cloud-top heights; this corresponds to the decrease in low-level liquid clouds and the increase in midlevel ice–mixed clouds. Based on this analysis, the authors hypothesize that the increase in aerosol concentration results in the increase in summer rainfall frequency in China via enhanced ice nucleation in the midtroposphere. However, over the past few decades, observations show an increasing long-term trend in aerosol concentration but decreasing trends in summer rainfall frequency and relative humidity (RH) in China. Despite the short-term positive relationship between summer rainfall frequency and aerosol concentration found in this study, the long-term variations in summer rainfall frequency in China are mainly determined by other factors including RH variation possibly caused by global and regional climate changes. A continuous decrease in RH resulting in less summer rainfall frequency may further enhance aerosol concentrations in the future in conjunction with the increase in the anthropogenic emissions.

1. Introduction

Aerosols of both natural and anthropogenic origins play a crucial role in cloud formation and the atmospheric hydrologic cycle. During the past few decades, anthropogenic emissions of aerosol precursors have increased considerably in China because of the increase in industrial activities (Cheng et al. 2005; Richter et al. 2005; Zhao et al. 2006; Gong et al. 2007). The corresponding increase in anthropogenic aerosols would act as a superfluous forcing to alter atmospheric physical processes such as radiation, the size spectrum of cloud particles, cloud cover, atmospheric stability, and precipitation (Ramanathan et al. 2001; Kaufman et al. 2005; Gong et al. 2007, and references therein).

The effect of aerosols on precipitation may operate via manifold processes. Previous studies showed that aerosols from biomass burning and industrial emissions observed in Indonesia, the Amazon, and elsewhere reduce the warm-rain efficiency in tropical clouds (Rosenfeld 1999, 2000; Ramanathan et al. 2001) by in-

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creasing cloud condensation nuclei (CCN) number concentration and reducing the size and coalescence of cloud droplets (aerosol indirect effect; Twomey 1977). The aerosol semidirect effect (Hansen et al. 1997), that is, absorbing aerosols such as black carbon heat the atmosphere, can hinder cloud formation and stabilize the low atmosphere (Koren et al. 2004). Andreae et al. (2004) also found that the reduction in cloud droplet size and the delay in precipitation onset within smoky clouds in the Amazon allow more aerosols and moisture to reach higher altitudes by updrafts. This effect tends to enhance precipitation from intense convective storms, opposite to the impact of indirect aerosol effects on precipitation. The net effect of aerosols on precipitation remains a subject of research.

Recent studies suggested that the increase in aerosol concentrations, via aerosol indirect and semidirect effects, may affect the long-term variations in precipitation in China, in conjunction with other climatic forcing such as global warming. Menon et al. (2002) noted in a global modeling study that increased semidirect effect of black carbon aerosols may have caused the precipitation changes in the past decade, that are characterized by increased (decreased) rainfall in southeast (northeast) China. Cheng et al. (2005) suggested on the basis of 30-yr surface-observed aerosol optical depth data that aerosol indirect effects contribute to the long-term trend of decreasing precipitation in southern China, but not severely in eastern China. Zhao et al. (2006) analyzed the long-term decrease in precipitation in eastern-central China to suggest a positive feedback between aerosol concentration and precipitation; higher aerosol concentrations reduce precipitation, thereby further enhancing aerosol concentrations. These studies concerned only rainfall amount, but rainfall frequency has not received any attention.

The relationship between aerosol concentrations and the total precipitation involving both warm- and cold-rain processes (Lin et al. 2006) on short time scales of less than two weeks is directly affected by aerosol–precipitation feedback; however, details of such a feedback are not well established. Quantitative examinations of the changes in rainfall characteristics via the aerosol concentration–rainfall relationship are complex because they are highly interactive. Also, precipitation is closely linked to atmospheric circulations on various spatiotemporal scales. Hence, long-term regional climate variations can also alter the characteristics of rainfall in China over decadal or longer time scale (e.g., Gong and Ho 2002; Ho et al. 2005; Zhai et al. 2005).

This study investigates the short-term relationship between aerosol concentration and summer rainfall characteristics in China using daily surface observations

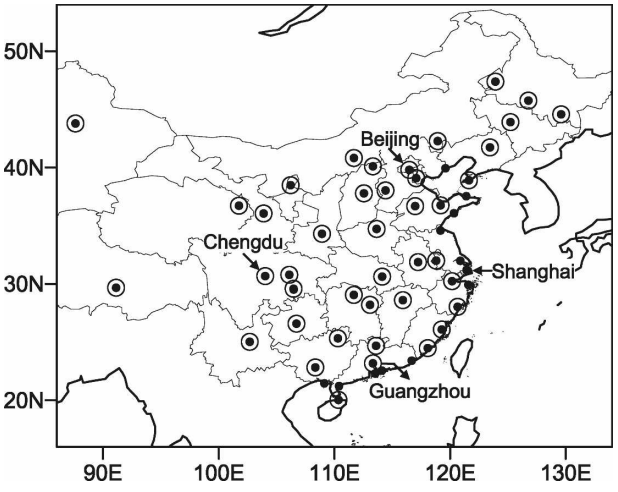

FIG. 1. The locations of the 55 pollution measurement stations over China (dots). The 42 stations with the records of both pollution and precipitation within the period of 2000–06 are selected for this study (distinguished by bigger open circles).

of particulate matters with a diameter of less than 10 μm (PM10) mass concentration from newly compiled air pollution index (API) records and rainfall in conjunction with satellite-observed cloud properties. It is the first attempt to apply the API data to investigate such a relationship. This paper is organized as follows. Section 2 describes the details of the surface data and analysis methods used to examine the aerosol concentration–rainfall frequency relationship in China. In the section, we also present the method to decouple wet scavenging effects from aerosol concentrations employed in the analysis. Section 3 presents the results showing the positive relationship between aerosol concentrations and summer rainfall frequency in China. Section 4 suggests the possible role of enhanced ice-nucleating activity in midlevel clouds and other aerosol effects in the positive aerosol–rainfall frequency relationship by using satellite-based observations of the midtropospheric cloud properties and the corresponding rainfall intensity. In section 5, we discuss the aerosol–precipitation interaction on weekly and decadal time scales. Conclusions are given in section 6.

2. Surface observations and analysis method

a. Air pollution index and precipitation observations

China has been measuring mass concentrations of the three leading air pollutants, PM10, SO_2, and NO_2. The measured highest pollutant concentrations have been archived in terms of daily-mean API in 55 major cities in China since 2000 (Fig. 1). The API is defined as

TABLE 1. National standard of the ambient air quality in China.

Index	Concentration (μg m^{-3})		
	SO_2 (daily mean)	NO_2 (daily mean)	PM10 (daily mean)
50	50	80	50
100	150	120	150
200	800	280	350
300	1600	565	420
400	2100	750	500
500	2620	940	600

API = max(I_{PM10}, I_{SO2}, I_{NO2}) (Gong et al. 2007). The subindices, I_{PM10}, I_{SO2}, and I_{NO2}, are converted from observed mass concentrations (M) of the three pollutants as shown in Eq. (1):

$$I = (I_U - I_L)(M - M_L)/(M_U - M_L) + I_L, \quad (1)$$

where I_U and I_L (M_U and M_L) are the upper and lower standard indices (mass concentrations), respectively (Table 1). Provided that the practical monitoring concentration of PM10 is 215 μg m^{-3}, the value is between 350 μg m^{-3} and 150 μg m^{-3} (i.e., M_U and M_L, respectively) and the corresponding subindex values I_U and I_L are 200 and 100, respectively; then I_{PM10} = (200 − 100)(215 − 150)/(350 − 150) + 100 = 132. Note that if the API is below 50 (PM10 < 50 μg m^{-3}, SO_2 < 50 μg m^{-3}, and NO_2 < 80 μg m^{-3}), the day is defined as a clean day and no pollutant type is recorded.

The API dataset used in this study consists of two parameters: the final API values and the corresponding pollutant types. Thus the daily API on a given day originates from a single pollutant type. Among the three types of air pollutants, PM10 is most dominant in China; the API of PM10 including clean days accounts for over 90% of the total API days. The majority of PM10 consists of fine particulates (diameter ≤2.5 μm; Shi et al. 2003; Cao et al. 2004; Wang et al. 2006), whose number and size distributions play a more important role in the nucleation of cloud particles and precipitation formation than their compositions (Dusek et al. 2006). Excluding the cases where API is caused by SO_2 and NO_2, we use the daily PM10 mass concentrations measured in terms of API for the 7-yr period from 5 June 2000 to 31 July 2006 (available online at http://www.sepa.gov.cn/quality/air.php3). We focus on summers because of sufficient moisture supplies by large-scale circulation and less frequent dust storms. Since our analysis is based on mass concentrations between 0 and 200 μg m^{-3}, the aerosol mass concentrations are calculated using the inverse formula of Eq. (1):

$$M = M_L + (I - I_L)(M_U - M_L)/(I_U - I_L). \quad (2)$$

Daily precipitation observations archived at the Chinese Meteorological Administration are used to examine the relationship between aerosol concentrations and rainfall frequency for the same 7-yr period. We selected 42 surface stations (large open circles in Fig. 1) at which aerosol concentrations and precipitation are observed simultaneously. Since daily precipitation data are available from 1955 to the present at over 193 stations within China, the long-term trend of rainfall frequency is calculated from this dataset as well. The observable range of precipitation is over 0.1 mm day^{-1}, but the occurrence of trace precipitation (less than 0.1 mm day^{-1}) is also indicated in the dataset. In this study, the trace precipitation is counted toward rainy days, but major results in this study are insensitive to the inclusion of trace precipitation events.

b. Rain-day frequency calculation

We first denote the observations of aerosol concentration and precipitation on the ith day as $M(i)$ and $P(i)$, respectively. Then 5-day segments of the precipitation time series are expressed as $P(i + j)$ where the day-lag j is an integer from −3 to +1 [i.e., $P(i - 3)$ to $P(i + 1)$]. We consider precipitation on the next day of ith aerosol-measured day to examine the cause [i.e., $M(i)$] and effect [i.e., $P(i + 1)$] relationship between aerosol concentration and rainfall. Since we are interested only in the occurrence of rainfall events, values of $P(i + j)$ represent either a rainy day ($P = 1$) or a rainless day ($P = 0$). To examine the effect of $M(i)$ on $P(i + 1)$ on the next day, we have implemented the following two conditions: 1) in order to exclude the removal of aerosols by precipitation on the same day, aerosol concentrations only on rainless day are used; 2) as aerosols continue to accumulate over consecutive rainless days, the number of successive rainless days are also considered and are denoted as *RC*R*, *RCC*R*, and *RCCC*R* for consecutive rainless days of one, two, and three, where *R*, *C*, and *C** are rainy, rainless, and aerosol-measured rainless days, respectively.

The concentration $M(i)$ is binned at 20 μg m^{-3} intervals between 0 and 200 μg m^{-3} [i.e., $0 \le M_1(i) < 20$ μg m^{-3}, . . . , $180 \le M_{10}(i) < 200$ μg m^{-3}]. The total number of days corresponding to the kth category of aerosol concentration M_k (simply, n_k) is 300−500 days for both very small aerosol concentrations (<20 μg m^{-3}; n_1) and very large aerosol concentrations (≥160 μg m^{-3}; n_9 and n_{10}) and over 1000 days for the aerosol concentrations of the remaining categories (n_2–n_8). Finally, we calculated the rain-day frequency (RDF), defined as the ratio in percent of the number of days for *RCCC*R* (or *RCC*R*, or *RC*R*) in kth category ($n_{k,RCCC^*R}$) to n_k:

$$\mathrm{RDF}_{k,RCCC^*R} = \frac{n_{k,RCCC^*R}}{n_k}. \quad (3)$$

In the same manner, RDF_{k,RCC^*R} and RDF_{k,RC^*R} can be obtained.

Both the removal and the accumulation of aerosols rely on a number of factors such as wet/dry scavenging, atmospheric transport and mixing, chemical transformations, and emissions (Garrett et al. 2006). Presumably, stronger atmospheric stability and drier boundary layer may induce effects on clouds similar to those of the more polluted area (Andreae et al. 2004). The *RC*R, RCC*R,* and *RCCC*R* cases, in fact, have individual meteorological settings, as originating from different regions and dates. However, a composite of sufficiently large cases in terms of M_k, as introduced in Eq. (3), can offset complex meteorological settings and noises, which is thereby commonly applied in climate studies. An analysis of the National Centers for Environmental Prediction–National Center for Atmospheric Research (NCEP–NCAR) reanalysis data (Kanamitsu et al. 2002) shows that the averaged equivalent potential temperature profiles from 850 to 1000 hPa for M_k have almost the same values within ±0.2 K deviation (not shown). This indicates that the 10 RDFs (i.e., M_1–M_{10}) are equally treated with the same thermal and moisture structures in the low troposphere. Consequently, the use of daily aerosol concentration and rainfall observations at coincident stations is expected to give a great advantage to analytic studies.

3. Relationship between surface PM10 concentration and rainfall frequency

Most aerosols emitted into and produced within the atmospheric boundary layer are transported into the free atmosphere via boundary layer turbulence and convective updrafts (Andreae et al. 2004), and affect the physical processes involved in the formation of clouds and precipitation. Precipitation is the leading mechanism for removing aerosols from the atmosphere. This suggests that aerosol concentrations and precipitation are closely related with each other (Jacobson 2002; Zhao et al. 2006).

Figure 2 shows the RDF calculated following Eq. (3) for the cases of one, two, and three consecutive rainless days (i.e., *RC*R, RCC*R*, and *RCCC*R*, respectively). The total number of cases for *RC*R, RCC*R*, and *RCCC*R* is about 3500, 2600, and 2000, respectively. The figure shows that higher aerosol concentrations are closely correlated with higher rainfall frequencies, especially for *RCC*R* and *RCCC*R*. Spearman's rank correlation coefficients (the two-sided significance of its deviation from zero) between aerosol concentrations and the RDF are 0.62, 0.95, and 0.87, significant at the 90%, 99.9%, and 99.9% confidence levels, for *RC*R, RCC*R*, and *RCCC*R*, respectively. In the case of *RC*R*, aerosol concentrations can fluctuate because of rainfall on the preceding day resulting in relatively lower correlations than *RCC*R* and *RCCC*R*.

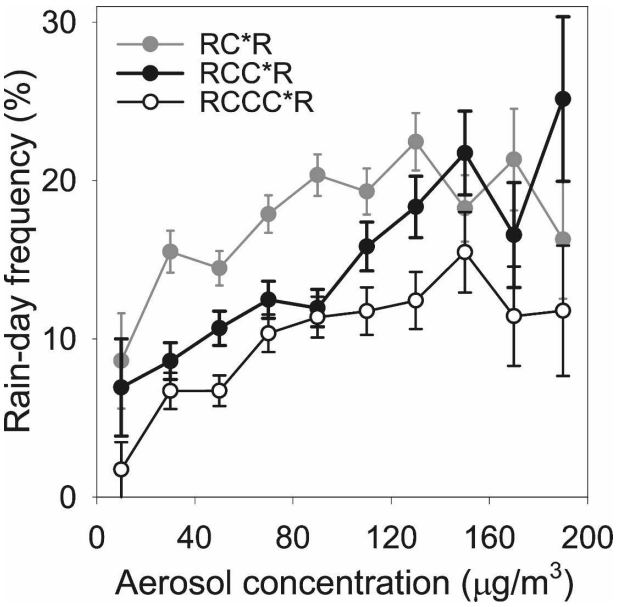

FIG. 2. Rain-day frequencies as a function of aerosol concentrations. The samples are categorized by successive rainless and rainy days as well as aerosol measurements over China during summer. Here *R, C*, and *C** denote rainy, rainless, and aerosol-measured rainless days, respectively; *RC*R, RCC*R*, and *RCCC*R* indicate the cases of one, two, and three consecutive rainless days between rain days, respectively. The error bar corresponds to ±1 std error.

Standard errors ($\sigma_k/\sqrt{n_k}$) for RDF_k varies from 1 to 5.2, with values for M_1, M_9, and M_{10} due to small n_1, n_9, and n_{10}; note error bars in Fig. 2. Because the standard errors are smaller than the differences between RDF_k, the increase in RDF_k with increasing k is statistically significant for all cases of *RC*R, RCC*R*, and *RCCC*R*. The correlation coefficients for four or more consecutive rainless days are small (≤0.42) and the corresponding statistical significance is low (≤77%) because of small sample sizes (<150 days for seven categories; not shown). Further investigation shows that rainfall events satisfying the *RCC*R* condition are almost evenly distributed in this study, with up to 25 days at each measurement site, despite higher total rainfall frequencies in southern China. The time-lag correlation coefficients between the two time series $M(i)$ and $P(i)$ which are computed without considering $M(i)$ measured for *C** of such *RC*R, RCC*R*, and *RCCC*R* do

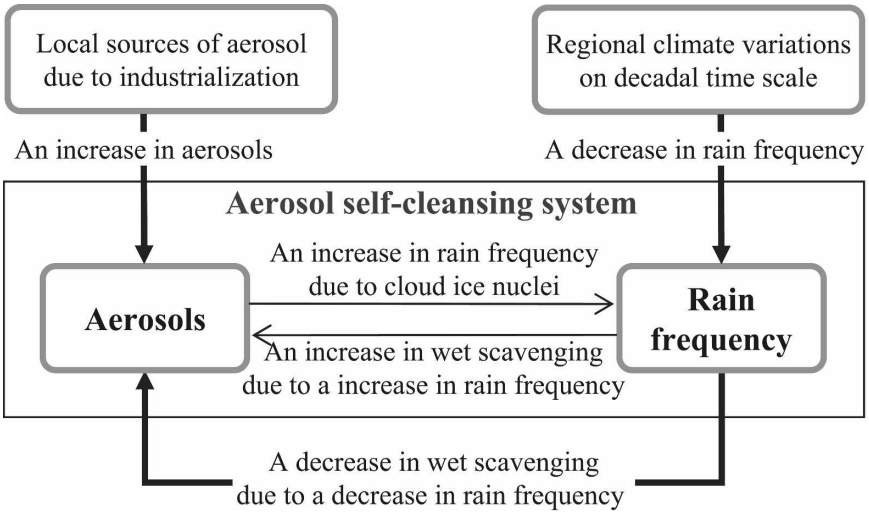

FIG. 3. Possible long-term influence of anthropogenic effects such as the oversupply of aerosol particles via industrialization and regional climate variations on the short-term aerosol self-cleansing system for summer of China. The thin (thick) arrow indicates the short-term (long term) effect on aerosols and rain frequency.

not show significant positive correlations for five or longer time-lagged comparison [$M(i)$ versus $P(i - 5$ or less)] either. This indicates that aerosols are rapidly restored within a few days after a rainfall event.

In summary, the significant correlation between the observed aerosol concentration and rainfall frequencies suggests that the increase in aerosol concentration increases rainfall frequency, which in turn removes aerosols from the atmosphere. Following the removal by rainfall, aerosol concentrations are rapidly restored within a few days to repeat the above cycle. This process is referred to "aerosol self-cleansing effect" hereafter. A schematic illustration of the aerosol self-cleansing effect and the influence of climate variations on this mechanism are presented in Fig. 3; the detailed discussion on the influence of long-term variations will be provided in section 5. This impact of aerosols may be particularly effective under atmospheric conditions of high RH, which is typical during summers in China (Ackerman et al. 2004). Under low- and moderate-RH conditions, increased aerosol concentrations may reduce rainfall via the first aerosol indirect effect; that is, a limited amount of cloud water is allotted to a larger number of smaller cloud droplets, thus reduced precipitation efficiency, for a larger number of cloud condensation nuclei (Twomey 1977; Rosenfeld 1999; Ramanathan et al. 2001).

4. The possible role of aerosols in rainfall frequency

Aerosols also act as ice nuclei (IN) to trigger the formation of ice crystals (DeMott et al. 2003; Sassen et al. 2003). The ice nucleation due to the presence of IN can play an important role in generating clouds and precipitation in atmospheric conditions that may be too warm or too dry, or both, for liquid condensation to occur, especially in the region immediately above the top of midlevel liquid clouds where temperature is moderately below freezing and humidity is between water- and ice-saturated values. The generation of ice clouds immediately above midlevel warm clouds can foster rain drop creation via the seeder–feeder mechanism (e.g., Mitchell et al. 1990) and accelerated precipitation formation due to the rapid growth of ice crystals at the expense of liquid droplets. In addition, the formation of midlevel ice clouds can enhance precipitation via cooling of lower-level stratiform clouds or the lower part of convective clouds, or both, as shown in previous studies (Toon 2003; Khain et al. 2005; Lau et al. 2005).

We examine the occurrence of additional ice nucleation under high aerosol concentration episodes using satellite measurements of cloud properties including the thermodynamic phase of cloud particles, cloud-top pressure, cloud optical depth, and effective particle radius. The satellite data are the $1° \times 1°$ gridded Moderate Resolution Imaging Spectroradiometer (MODIS) data averaged using quality assessment weights in version 4 of the MODIS/*Terra* level-3 daily atmosphere product (Platnick et al. 2003). The mean and the frequency distribution of MODIS cloud properties for a given M_k are computed using the data nearest to the surface sites. One concern in this approach is the distance between the locations of the gridded satellite data and the surface sites. Fortunately, this turned out to be

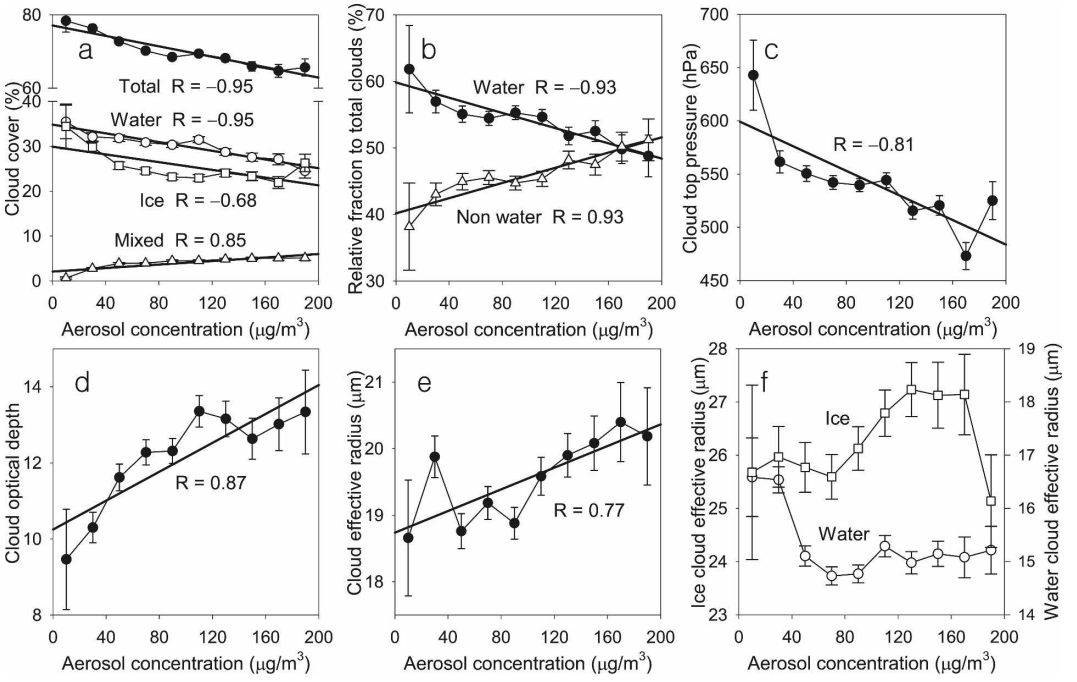

FIG. 4. Satellite-retrieved cloud properties as a function of aerosol concentrations. (a) Cloud cover, (b) relative contributions of water vs nonwater phase clouds to total clouds computed by the ratio of the number of cloudy pixels with different phases to that of the total cloudy pixels in a given grid, (c) cloud-top pressure, (d) cloud optical depth, (e) cloud effective radius, and (f) effective radii of water vs ice phase clouds. The last four are the averages of mean values in individual aerosol concentration bins. The error bar plotted in (a)–(f) represents ±1 std error. (Note that in this figure R denotes correlation coefficient *r* rather than rainy day.)

a minor problem because of relatively homogeneous spatial distribution of aerosol concentrations within a spatial scale of 1° (~90 km day^{-1} for the case of 5 m s^{-1} wind speed). Another concern is that the surface aerosol concentration is not always proportional to the column-total aerosol loading. However, the surface-observed aerosol data facilitate the comparison with nearly coincidental satellite-observed cloud properties on daily basis. This is not possible with spaceborne aerosol optical depth data because they are obtained only for the clear-sky field of view (Remer et al. 2005). Since the MODIS/*Terra* data over China are snapshots between 1030 and 1130 local time, the MODIS cloud properties observed in the next morning of the aerosol-measured day are selected in the analysis. The relationship between cloud properties and aerosol concentrations gradually decreases as the time lag between the MODIS cloud property measurements and surface aerosol measurements increases. All of the data used in this analysis were sampled during two or more consecutive rainless days in order to eliminate the influence of rainfall on aerosol concentration and cloud properties (Ackerman et al. 2004).

The relationships between the observed aerosol concentrations and the MODIS measurements (Fig. 4) provide a number of implications about the impact of aerosol concentrations on midlevel clouds. The cloudiness associated with both liquid- and ice-phase clouds is reduced by similar amounts (~−0.5%/10-μg m^{-3}) with increasing aerosol concentrations (Fig. 4a). The cloudiness of both liquid and ice clouds is closely correlated with the aerosol concentration at correlation coefficient (r) value of −0.95 and −0.68, respectively. The reduction in liquid cloud cover is consistent with the previous studies over the Mediterranean and Amazon regions by Lelieveld et al. (2002) and Koren et al. (2004). However, the mechanism for the reduction of ice cloud cover is unclear. The cloudiness associated with mixed clouds increases with increasing aerosol concentration, but the magnitude is small (~0.2% 10-μg m^{-3}).

Aerosol concentrations may also alter cloud properties such as the phase of cloud particles, the vertical extent of clouds, and cloud optical properties. To examine the variations in cloud properties according to aerosol concentration, the relative fractions in different phases are computed for liquid and nonliquid (i.e., ice and mixed) clouds by dividing the number of liquid and nonliquid cloudy pixels by the number of total cloudy pixels. Figure 4b shows the resulting fractions as a function of aerosol concentrations. Nonliquid cloud fraction

increases with increasing aerosol concentration, whereas that of liquid clouds decreases. The correlation coefficients are exceptionally high ($r = 0.93$ for nonliquid clouds and -0.93 for liquid clouds). Higher aerosol concentrations are also strongly correlated with higher mean cloud-top heights (or lower mean cloud-top temperatures) and larger cloud optical depths on the following day, with $r = 0.81$ and 0.87, respectively (Figs. 4c,d). These results imply that the reduction in low-level liquid clouds and the increase in midlevel ice/mixed clouds occur in response to increased aerosol concentrations.

The results in Figs. 4b–d are consistent with the satellite-based studies over Amazonia during the dry season by Lin et al. (2006). One notable difference between the results of Lin et al. (2006) and this study is that the total cloudiness decreases with increasing aerosol concentration in China (Fig. 4a). This discrepancy between these two studies may originate from the differences in RH because summertime RH over China is much higher than RH over the Amazonia during the dry season. Ackerman et al. (2004) pointed out that the response of cloud water (or cloud cover) to aerosol loading can be determined by the competition between surface precipitation and the RH of the overlying air. Our results also suggest that the semidirect effect may play an important role in aerosol–cloud interaction during summer in China; black carbon aerosols, which are abundant in this region, can induce significant heating of the low atmosphere to dissipate low-level clouds and to lower warm-rain efficiency (Menon et al. 2002). We do not have data for the analysis of the underlying detailed physical processes, which should be further investigated in the future.

The satellite-observed mean cloud-top pressure varies between the 400- and 600-hPa levels (Fig. 4c). NCEP–NCAR reanalysis data show that high RH ($\geq$80%) with respect to liquid water frequently occurs in this elevation range on the days following high aerosol concentrations (not shown). During summer, the freezing level over China occurs around the 600-hPa level. This implies that additional ice cloud particles can be formed at an altitude as low as the 600-hPa level due to higher aerosol concentrations (Andreae et al. 2004; Khain et al. 2005). Recent analyses of Saharan dust storms (DeMott et al. 2003; Sassen et al. 2003) also showed that liquid cloud particles can freeze at temperatures between -5°C and -8°C because of enhanced IN concentration during high aerosol concentration events. The MODIS data show that the size of ice crystals increases substantially with higher aerosol loading except M_{10}, but the size of water droplets shows the opposite behavior (Fig. 4f). Thus, the increase in the cloud-top heights and the relative cloudiness of nonliquid clouds with increasing aerosol concentration found in this study can originate from the formation of midlevel ice or mixed clouds, or both, via the enhanced supply of IN into the midtroposphere during high aerosol concentration episodes. The increase in the mean cloud effective radius in response to higher aerosol concentrations also supports the formation of ice clouds and the subsequent growth of ice crystals during high aerosol concentration episodes (Rosenfeld et al. 2001) because ice crystals are larger than liquid droplets of the same mass by an order of magnitude (Fig. 4e). This relationship between the observed cloud properties and aerosol concentrations strongly suggest that additional ice clouds can form at an altitude as low as the 600-hPa level during high aerosol concentration episodes. Rainfall originating from ice or mixed clouds, or both, in this level may be of moderate-to-heavy intensity (e.g., $\geq$10 mm day^{-1}) because the presence of ice crystals can enhance precipitation efficiency significantly.

The impact of the additional formation of the midlevel ice clouds during high aerosol concentration episodes may appear in rainfall intensity. We examine this on the basis of the differences in RDF of daily rainfall intensity between the large (100–190 μg m^{-3}) and the small ($<$90 μg m^{-3}) aerosol concentration episodes (Fig. 5). The sample size corresponding to these cases is approximately 50% of the total. A significant positive anomaly of moderate rainfall (10–20 mm day^{-1}) occurs for two and three consecutive rainless days (*RCC*R* and *RCCC*R* in Fig. 5) when aerosol concentration is high. The increase in moderate rainfall events during the high aerosol concentration periods is accompanied by a significant decrease in light rainfall ($<$5 mm day^{-1}) frequency, especially in the case of *RCC*R* when the aerosol concentration is highest. The statistical significance of the anomaly of the heavy rainfall ($\geq$20 mm day^{-1}) episodes is low because of a small sample size. The rainfall intensity anomalies for *RC*R* are also small because of significant wet scavenging (i.e., aerosols have not accumulated significantly immediately after a rainfall event).

The invigoration of midlevel clouds via the initial delay in warm-rain formation (Rudich et al. 2003; Andreae et al. 2004; Khain et al. 2005; Lau et al. 2005) may also contribute to the observed increase in moderate rainfall events accompanied by the decrease in light rainfall events during the high-pollution episodes. Khain et al. (2005) showed in a modeling study that initial weakening of warm-rain processes due to elevated aerosol loading decreases the drag on updrafts and allows additional moisture to reach the freezing level. This can result in the enhancement of updrafts

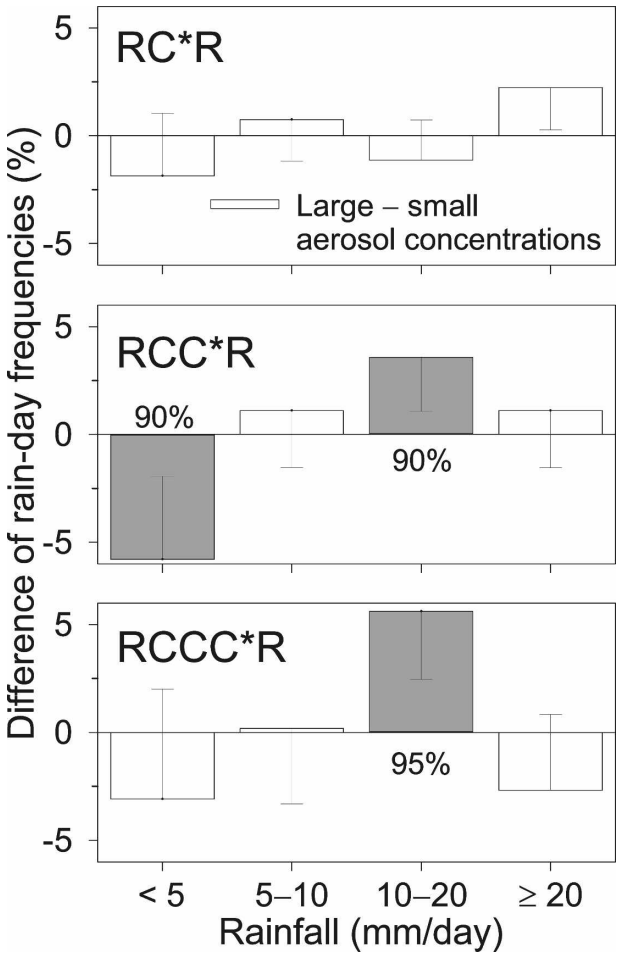

FIG. 5. The difference of the rain-day frequencies in terms of rainfall intensity with respect to aerosol concentrations: the cases of high aerosol concentrations (100–190 $\mu g\ m^{-3}$) minus the cases of low aerosol concentrations (0–90 $\mu g\ m^{-3}$). The error bar corresponds to the std error of the difference. Bars significant at the 90% and 95% levels are shaded.

and ice nucleation in the midtroposphere. The initial delay of warm-rain process can also be partially due to the semidirect effects by black carbon aerosols in the region. This process will also result in reduced light rainfall events due to the weakening of the low-level warm clouds and enhanced moderate rainfall events due to the development of midlevel ice clouds.

In summary, the relationship between the observed aerosol concentration, the generation or enhancement, or both, of midlevel ice clouds, and rainfall intensity found in this study supports the premise that higher aerosol concentrations enhance moderate rainfall events, possibly via additional ice nucleation in the midtroposphere immediately above the freezing level and/or the influence on initial updrafts that weakens the low-level warm clouds and strengthens the midlevel ice clouds by aerosol indirect or semidirect effects (Jacobson 2002; Menon et al. 2002; Kaufman et al. 2005; Zhao et al. 2006).

5. Discussion on long-term trends in aerosol concentration and rainfall frequency

Reliable precipitation records exist from 1955 to the present in China, although the period of observation varies across its 193 stations. To investigate long-term rainfall trend in China, the trends in the number of summer rainy days are calculated at the stations with more than 10-yr precipitation records (Fig. 6). The statistical significance of the trend is estimated using a two-tailed t test under the null hypothesis of no trend.

On the basis of the aerosol self-cleansing effect found in this study, it is expected that the significant increase in aerosol concentration during the past two decades would result in increased rainfall frequency. The results, however, show that the rainfall frequency in China has notably decreased since the beginning of its industrialization in the 1980s; the rates of change are -1.3 and -2.8 days $(10\ yr)^{-1}$ for 1955–79 and 1980–2005, respectively. Our results also attribute the decreasing trend mainly to the reduction in light rain days, which constitute over 80% of the total rainy days during summer (Zhai et al. 2005). The observed decrease in light rainfall frequency may result from indirect and semidirect aerosol effects that reduce warm-rain efficiency (Qian et al. 2006; Zhao et al. 2006) or the ongoing long-term climate variations, or both (Gong and Ho 2002; Ho et al. 2005; Zhai et al. 2005).

To answer this question, the trend in 850-hPa RH is calculated using the NCEP–NCAR reanalysis data for the same period of the station precipitation data. The resulting analysis (Fig. 7) clearly shows that the summertime RH in China has decreased over this period. This suggests that the decreasing rainfall trend in China may be primarily caused by the decrease in RH, instead of aerosol indirect and/or semidirect effects. The decreasing rainfall trend could also have resulted in further enhancement of aerosol concentrations via reduced wet scavenging, in addition to increased emissions (see Fig. 3).

Because the aerosol self-cleansing effect operates on short time scales of no longer than two weeks, this effect is imbedded within the long-term RH trend. Previous studies inferred the impact of aerosols on clouds and precipitation on the basis of their long-term variability (Menon et al. 2002; Cheng et al. 2005; Zhao et al. 2006), but they did not notice the intricate processes working on a variety of time scales. The impact of aero-

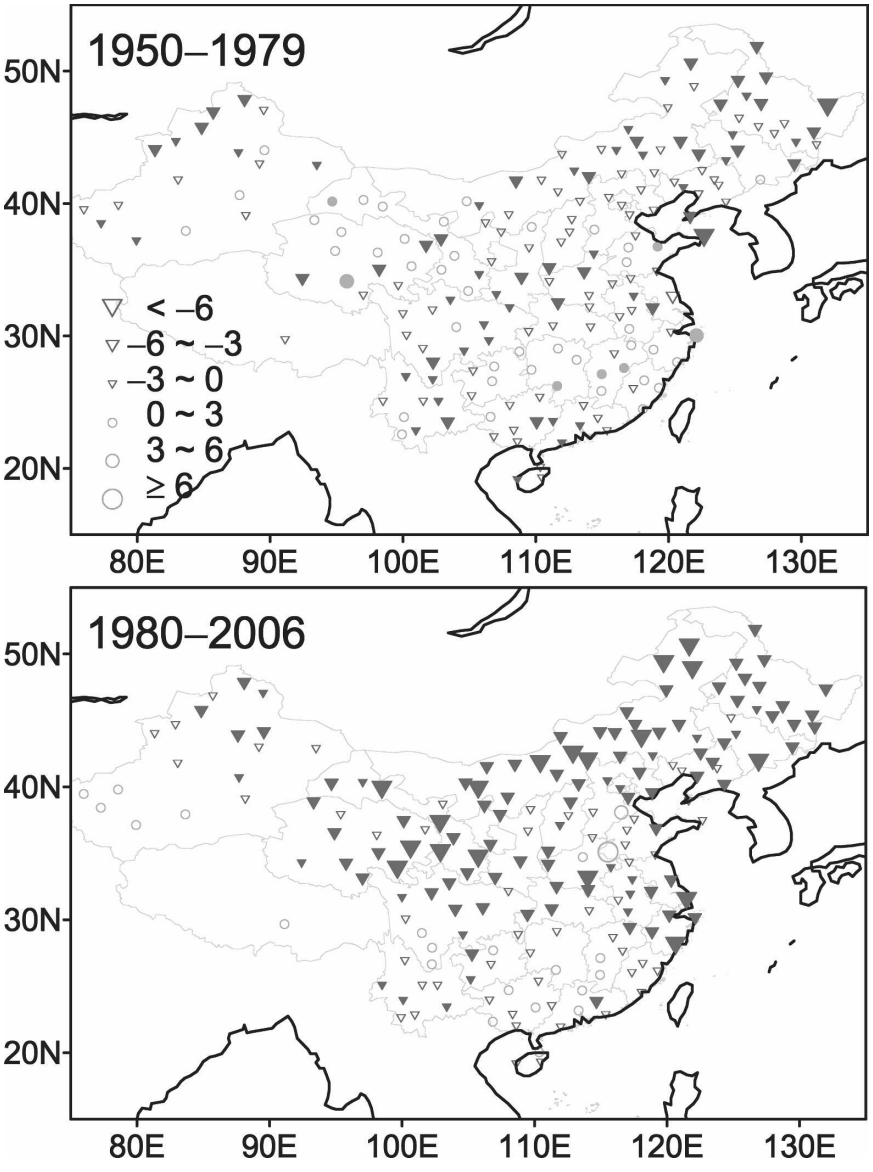

Fig. 6. The trend of the rain frequency [days (10 yr)$^{-1}$] in summer for (top) 1955–79 and (bottom) 1980–2005. Stations significant at the 90% level are indicated by filled symbols. In contrast to the situation before 1979, the rain frequency has rapidly decreased since 1980.

sols on the regional hydroclimate involves a number of processes on a wide range of time scales including aerosol–cloud interaction (hourly or shorter time scales), aerosol accumulation–rainfall and wet scavenging (a few days), and the long-term variations in the atmospheric RH that modulate the former two effects (decadal or longer). This study presents an important perspective on the separation of the aerosol self-cleansing effect, working on approximately weekly time scales, from the long-term trends in rainfall and aerosol concentrations associated with regional climate variations and increased emissions (Fig. 3).

6. Conclusions

It has been commonly believed that the increase in aerosol concentrations is likely to inhibit rainfall via aerosol indirect effect, which tends to reduce rainfall efficiency of the low-level warm clouds (Rosenfeld 1999, 2000; Ramanathan et al. 2001; Ackerman et al. 2004; Kaufman et al. 2005). Results in this study also suggest that aerosol indirect effect is responsible for the reduction in the light rainfall events during high aerosol concentration episodes; however, the aerosol self-cleansing effect is another important mechanism in-

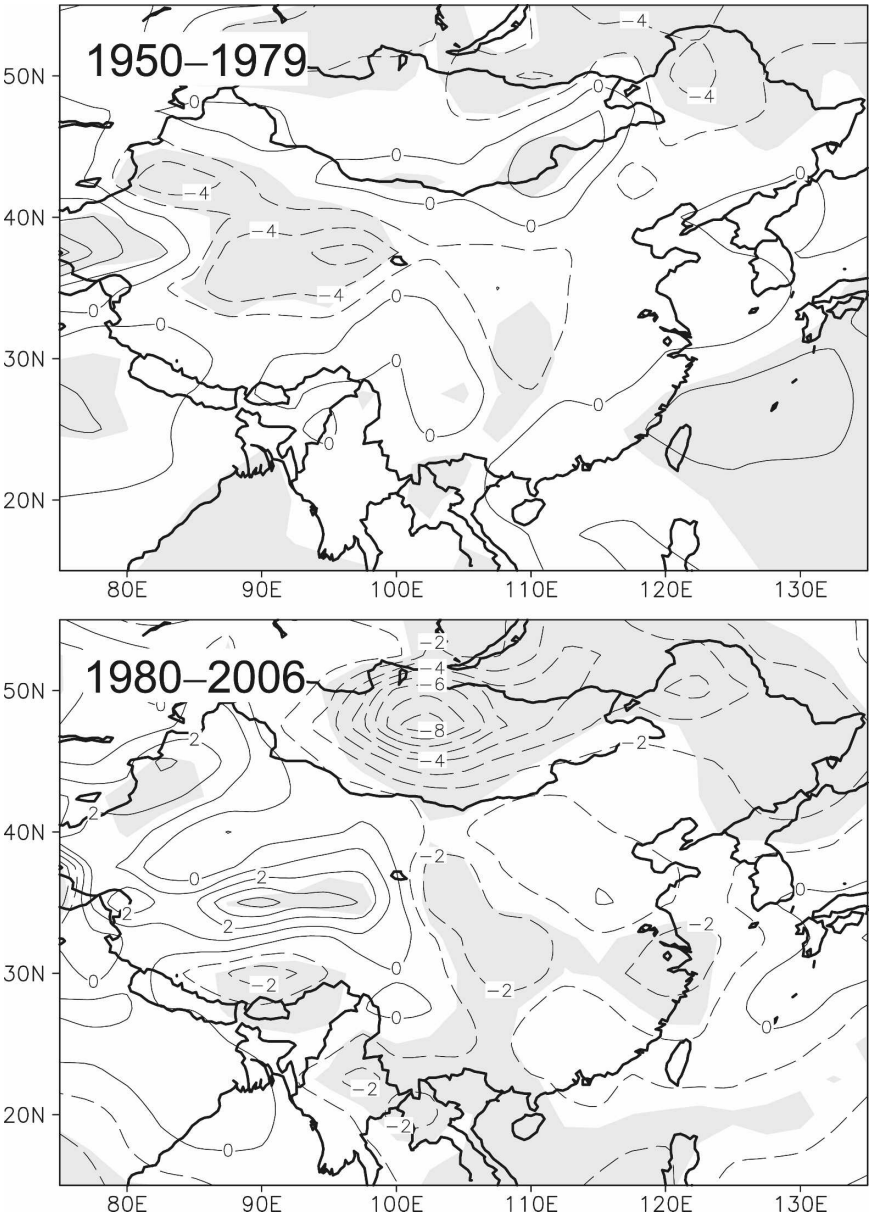

FIG. 7. As in Fig. 6, but for the 850-hPa RH [% (10 yr)$^{-1}$] obtained by NCEP–NCAR reanalysis data. Values significant at the 95% level are shaded.

volved in aerosol–cloud–rainfall interaction in the climate conditions corresponding the summertime in China. It is found that higher aerosol concentrations are significantly correlated with the increase in moderate rainfall events in China. The significant correlation between the aerosol concentration and ice clouds is consistent with the increase in moderate rainfall events since the formation of cloud ice is an important mechanism for enhancing precipitation (Mitchell et al. 1990; Pruppacher and Klett 1997). Satellite measurements of cloud properties and their correlations with the observed aerosol concentration and precipitation strongly support the aerosol self-cleansing effect.

Investigating the detailed microphysical process of faster glaciations and its impact on rainfall due to added ice requires elaborate work with high quality and high-resolution data, which are not available for China. Passive radiometer measurements such as MODIS cannot provide the details necessary for analyzing microphysical processes (e.g., those involved in the effects of glaciogenic cloud seeding on rain formation). The measurements using a spaceborne depolarization lidar on

board the *Calipso* satellite may be useful for analyzing more detailed physical processes in the future.

Despite the presence of aerosol self-cleansing effect, aerosol concentration has been increasing in China, accompanied by a decrease in rainfall events, over the last few decades. This shows that there must be two mechanisms of significantly different time scales that control summer rainfall frequency in China: the self-cleansing mechanism working on a time scale of a few days tends to increase rainfall events from midlevel clouds in response to the increases in aerosol concentration, and the long-term decrease in RH over China tends to reduce rainfall events. Results in this study show that the impact of reduced RH dominates the impact of the aerosol self-cleansing mechanism in determining the long-term trend in rainfall frequency. The effects of the decreasing trend in RH may also contribute to the increase in aerosol concentrations in conjunction with increased emissions of anthropogenic aerosol precursors since the beginning of the industrialization of China in 1980s.

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