Atmos. Chem. Phys. Discuss., 11, 8747–8776, 2011 www.atmos-chem-phys-discuss.net/11/8747/2011/ doi:10.5194/acpd-11-8747-2011 © Author(s) 2011. CC Attribution 3.0 License.



This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

# Spatial features of rain frequency change and pollution and associated aerosols

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Received: 1 February 2011 – Accepted: 9 February 2011 – Published: 14 March 2011

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Published by Copernicus Publications on behalf of the European Geosciences Union.





# Abstract

A spatial-temporal analysis has been conducted using satellite observed distributions of rain frequency, NO<sub>2</sub> concentration and aerosol, with focus on the spring season in East Asia. As NO<sub>2</sub> is a key precursor of secondary aerosols, especially in urban <sup>5</sup> areas, an increase of NO<sub>2</sub> emission is generally accompanied by an increase of fine aerosol particles. Comparison between trends in rain frequency and in precipitation amount shows that the changes in precipitation are more due to changes in precipitation occurrence than in precipitation amount. The overall feature emerged from the

region-by-region analyses is that there is an inverse relationship between the rain frequency and the pollution and associated aerosols at continental scale in spring. The change in rain frequency is associated with changes in pollution-produced aerosols and long-range transport mineral dust. The inverse relationship at large temporal and spatial scales illustrates potential climatological consequence of changed pollution and aerosols on precipitation.

# 15 **1** Introduction

Human-induced climate change has caused a redistribution of precipitation (Alpert et al., 2008; Changnon et al., 1981; Halfon et al., 2009; Lowry, 1998; van den Heever and Cotton, 2007; Zhang et al., 2007). Besides the greenhouse gases-induced global warming that intensifies the hydrological cycle (Allen and Ingram, 2002; Dell et al., 2008; Fowler and Hennessy, 1995), regional and local changes associated with urban heat island, land use change, the injection of moisture from industrial sources, and pollutant aerosols also have significant impacts on precipitation (Cotton and Pielke, 1995; Landsberg, 1981; Bornstein and Lin, 2000; Shepherd and Burian, 2003; Shepherd et al., 2002). While the former three factors tend to promote precipitation by enhancing
the atmospheric updrafts and evaporation or increasing the roughness of urban surface (Chow and Chang, 1984; Hand and Shepherd, 2009; Lowry, 1998; Shepherd,





2006), the possible impacts of pollutant aerosols on precipitation remain the greatest controversial (Stevens and Feingold, 2009). Therefore, it is crucial to indentify and explain precipitation changes associated with regional and local influences, particularly pollution and associated aerosols.

- Anthropogenic aerosols that increase concentrations of cloud condensation nuclei (CCN) and ice-forming nuclei (IN) alter the main path of precipitation-forming microphysical processes and the precipitation amount (Cotton and Pielke, 1995; Ramanathan et al., 2005; Rosenfeld et al., 2008). Albrecht (1989) found that the increasing of aerosol concentration may reduce the drizzle over ocean and increase the
- amount of low-level cloudiness. There were less drizzle in polluted regions than in pristine regions (Heymsfield and McFarquhar, 2001; Yum and Hudson, 2002). Over urban area, the precipitation may shut off or delayed due to the urban and industrial air pollution (Rosenfeld, 2000). Also carbonaceous aerosols absorb radiation and enhance atmospheric heating, thereby reducing the strength of updraft and associated precipi-
- tation (Ramanathan et al., 2005; Zhao et al., 2006). Small et al. (2009) suggested an evaporation-entrainment feedback of and polluted aerosol, which may shift marginallyprecipitating clouds to the non-precipitating regime. Besides most of studies that show aerosol will inhibit precipitation, there are some other studies report that aerosol may promote precipitation through the convection invigorating and the accelerating of cloud
- <sup>20</sup> water convert to precipitation (Koren et al., 2005; Rosenfeld et al., 2008; Stevens and Seifert, 2008; Williams et al., 2002). Furthermore, the influences of anthropogenic pollutants on precipitation are confounded by dynamic processes in various temporal and spatial scales, which heighten the need for accurate information about temporal and spatial variations in precipitation and aerosols (IPCC, 2007a; New et al., 2001; Qian
- et al., 2009; Yang and Lau, 2004). Few, if any, studies have reported directly observational linkage between the rain frequency and the pollution and associated aerosols at continental scale.

Heterogeneous spatial distribution of anthropogenic aerosols, which results from their short lifetime, may provide spatial signatures of anthropogenic pollution on





precipitation. East Asia is characterized by a rapid increase of energy consumption accompanied by a rapid growth of population and economic activities, resulting in significant enhancement in the concentration of aerosols and pollutants (Luo et al., 2001; van der A et al., 2006). East Asia also acts as the receptor of dust from arid and semiarid regions (Sun et al., 2005; Wang et al., 2006). The incoming mineral aerosol 5 particles mixing with local emission may accelerate the gas-particle interaction as well as serve as giant CCN. Since East Asia is the most populous region and one of the largest grain producing regions in the world, climate change, especially precipitation change, may have great consequences for the ecosystem and residents. The severe anthropogenic pollution over Asia provides the possibility and urgency to study the 10 anthropogenic forcing on precipitation at a large scale.

Due to the large spatial and temporal variability of aerosols and precipitation, remote sensing from satellites delivers the most reliable information about their regional and global distribution. This study investigates the linkage between rain and air pollutants

- and associated anthropogenic aerosols from the spatial-temporal perspective by uti-15 lizing multi-satellite observations over East Asia. It is believed that precipitation in the spring is less influenced by the monsoon dynamics of atmospheric general circulation (Gong and Wang, 1999). Also any precipitation change in spring will significantly impact stable crop production in the region (Tirado et al., 2010). Therefore, we will focus our study on the spring season.
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#### Measurements 2

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The Tropical Rainfall Measuring Mission (TRMM) satellite provides the first detailed and comprehensive dataset on the four-dimensional distribution of rainfall within about 36° latitude. To highlight spatial-temporal characteristics of rainfall distribution, monthly rain rate dataset from TRMM Precipitation Radar (PR) at 0.5° × 0.5° spatial grid (TS-DIS, 2007; version; 3A25; source: http://daac.gsfc.nasa.gov/data/) from 1998 to 2009 are used in this study. In addition to the TRRM PR estimated precipitation, the





surface rain gauge precipitation data (the Chinese National Meteorological Center: http://cdc.cma.gov.cn) are used to verify the satellite measurements and investigate the relationship between precipitation and air pollutants. As shown in Fig. 1, the seasonal precipitation amount estimated from TRMM PR over a  $1^{\circ} \times 1^{\circ}$  grid is consistent with

- <sup>5</sup> precipitation measurements from a surface rain gauge (31°10′ N, 121°26′ E) in Shanghai, with correlation coefficient of 0.81 at a 95% confidence level. The comparisons at some other typical sites during 1998–2009 are list in Table 1. Good correlations are observed between the two datasets, although the precipitation amount derived from TRMM PR are more or less lower than the surface rain gauge measurement, which
- <sup>10</sup> could be probably due to the sensitivity of PR that limits its detection of precipitation over 0.4 mm h<sup>-1</sup>. Those comparisons illustrate that precipitation estimated from PR is representative at seasonal scale or longer time scales. Given the same instrument and retrieval algorithm for TRMM PR measurements, we expect that the trend of precipitation estimated from TRMM PR is reliable.
- As a marker of air pollution, tropospheric nitrogen dioxide (NO<sub>2</sub>) has been monitored by both the Global Ozone Monitoring Experiment (GOME) and SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) satellites. Hence, monthly NO<sub>2</sub> vertical column concentration from combined GOME (1998–2002) and SCIAMACHY (2003–2009) measurements (Richter et al., 2002; source: http://www.iup.uni-bremen.de/doas/data\_products.htm) are used to quantify air pollution changes over the same period of PR dataset. NO<sub>2</sub> vertical column concentration
- from GOME and SCIAMACHY have been validated by ground-based and airborne measurements, indicating that satellite observed  $NO_2$  vertical column concentration is a good marker of local air pollution (Heue et al., 2005; lonov et al., 2006).
- Precipitation can be influenced by anthropogenic aerosols associated with pollution through their roles in cloud condensation nuclei and ice nuclei, and through the direct effect of pollution on the stability of the atmosphere. To assess the changes of aerosol loading in the atmosphere directly, aerosol optical depths from MODerate resolution Imaging Spectroradiometer (MODIS) on board the Terra Satellite (King et al., 2003)





are also used. The extensive validation of MODIS aerosol products confirmed that the uncertainty of MODIS optical retrievals within  $\Delta \tau = \pm 0.03$  or  $\pm 0.05 \tau$  over ocean and  $\Delta \tau = \pm 0.05$  or  $\pm 0.15 \tau$  over land, which suggests that MODIS aerosol products can be used in the analysis of aerosol distribution (Chu et al., 2002; Remer et al., 2005). Cloud fraction from MODIS/Terra is also used to identify the changes of cloud amount

# 5 Cloud fraction from MODIS/Terra is also used to identify the changes of cloud amount.

# 3 Results

Due to the relatively short lifetime of NO<sub>2</sub> and the vertical distribution of NO<sub>x</sub> sources, NO<sub>2</sub> columns observed from space are dominated by the NO<sub>2</sub> concentration in the boundary layer and at the source (Richter et al., 2005). As shown in Fig. 2a, satellite retrieved NO<sub>2</sub> column concentration in spring in Shanghai increased substantially from 1998 to 2009. The linear trend in NO<sub>2</sub> column concentration is  $1.6 \times 10^{15}$  molec cm<sup>-2</sup> per year. With respect to the reference value of  $6.8 \times 10^{15}$  molec cm<sup>-2</sup> in spring 1998, air pollution in Shanghai was almost tripled from 1998 to 2009. Nitrogen dioxide is an effective absorber of visible and near-ultraviolet solar radiation. At wavelengths below ~400 nm, photodissociation of NO<sub>2</sub> generates NO and O atoms that quickly attach to molecular oxygen to form ozone. Back-reactions of NO with ozone and/or other radicals establish a steady state between NO and NO<sub>2</sub> in the troposphere. The photodissociation of NO<sub>2</sub> is of major importance to atmospheric chemistry in addition to that of ozone, as this process is also involved in the production of many oxidants, such as radicals OH HO<sub>2</sub> BO<sub>2</sub>, which could oxidize SO<sub>2</sub> in addition to NO<sub>2</sub> and leads to

- as radicals OH, HO<sub>2</sub>, RO<sub>2</sub>, which could oxidize SO<sub>2</sub>, in addition to NO<sub>2</sub>, and leads to the formation of nitric acid and sulfuric acid and, in turn, the subsequent neutralization conversions to nitrate and sulfate, the major parts of the secondary aerosols. Therefore, NO<sub>2</sub> is a key precursor of secondary aerosols, especially in urban areas. Thus, the dramatic increase in NO<sub>2</sub> concentration implies a substantial enhancement of at-
- <sup>25</sup> mospheric aerosol loading. Satellite retrieved AOD includes locally generated aerosols that are associated with pollution and small in size, and transported aerosols, such as dust with large size. As shown in Fig. 2a, the retrieved AOD from MODIS showed an





increase in recent years, although there was large interannual variability which was mostly associated with the spring dust events and local construction activities (Wang et al., 2006). The trend of fraction ratio of fine mode aerosols increased consistently with the increasing trend in NO<sub>2</sub> concentration.

Most particles over urban areas are composed of hygroscopic salts, i.e., sulfates and nitrates (Givati and Rosenfeld, 2004), which can rapidly reach their critical size under relatively low supersaturations and act as effective CCN (Levin et al., 1996). The huge local anthropogenic emission resulting from rapid economic growth and urban development mixing with long range transported dust, therefore, lead to a high concentration of cloud condensation nuclei (CCN). It is plausible that the observed trend of NO<sub>2</sub> in Shanghai implies an increasing trend in CCN concentration from 1998 to 2009.

TRMM PR estimated precipitation in spring at  $1^{\circ} \times 1^{\circ}$  spatial domain centered in Shanghai correlated well with the measured precipitation from a single surface rain gauge (Figs. 1 and 2b). Although there is a spatial-temporal mismatch between the

- two, the consistency of decreasing trends of precipitation in spring is evident. A similar conclusion can be drawn from comparisons at other surface sites, illustrating precipitation estimates from PR are representative at seasonal or longer time scales. Both PR and rain gauge measurements in spring show that precipitation amount was reduced from 1998 to 2009. Reduction in precipitation could be either a decrease in rain
- frequency or in rain rate within the 1° × 1° grid. Small footprint and high sensitivity of TRMM PR allows us to evaluate the seasonal rain frequency at 1° × 1° grids, defined as the ratio of raining pixels to total sampling pixels. Using such a relative parameter also minimizes the systemic bias and retrieval uncertainties of PR rain rate retrievals. Clearly, the decreasing trend of 4.04% per year in rain frequency (0.21% per year in
- <sup>25</sup> absolute rain frequency) is slightly greater that the decrease trend of 2.49% per year in rain amount (5.75 mm per year in absolute rain amount). It suggests that reduction in precipitation is mainly due to the suppression of rain occurrence with a slight enhancement of rain intensity.





Cloud formation is strongly controlled by meteorological conditions, such as temperature and atmospheric convection. The increased NO<sub>2</sub> and aerosols (soot particles in particular) affect the radiative processes in the atmosphere through enhancing absorption of solar radiation and heat the atmosphere, which lead to changes in the air temperature and atmospheric stability (Ramanathan et al., 2005). If the atmosphere 5 becomes more stable, the upward motions are depressed, and cloud formation is reduced, resulting in reduction of precipitation (Zhao et al., 2006). Furthermore, if the moisture in the atmosphere is not altered by the increase in pollution particle number concentration, the cloud droplet radius will decrease, resulting in a decrease in the precipitation efficiency (IPCC, 2007b; Ramanathan et al., 2001). The opposite trends of 10 precipitation and air pollutants imply the possibility that the increased particles over urban areas suppress the local precipitation, particularly the rain frequency. However, the inverse relations of rain frequency and precipitation to the concentrations of NO<sub>2</sub> and aerosols at a single site for past decades can be casual, as precipitation changes are

- <sup>15</sup> strongly influenced by changes of large scale dynamics and by other human-induced local effects. In addition to air pollutant emissions, other human-induced local changes, such as land use-change, may independently or synergistically, altering the local and regional atmosphere dynamic, modifying the precipitation clouds (Hand and Shepherd, 2009; Junkermann et al., 2009; Lin et al., 2006; Zhang et al., 2009). To exclude the pos-
- sible influence of meteorological factor changes on specific sites, the spatial-temporal distribution of rain frequency, NO<sub>2</sub> concentration, and aerosol loading are investigated. Many studies suggested that there were strong increasing trends of NO<sub>2</sub> in some

regions of China and India for the past decade (Richter et al., 2005; van der A et al., 2006). As illustrated in Fig. 3a–c, those regions include the North Chinese Plain, Yangtze River Delta, Pearl River Delta, Sichuan Basin and India Ganges region where

Yangtze River Delta, Pearl River Delta, Sichuan Basin and India Ganges region where economy has been developed substantially in recent years. Since Asia monsoon is in a transition phase in spring, the stable atmospheric structure prevents dispersion of air pollutants. As expected, most of those regions have a high mean AOD with a positive trend in fine mode AOD (Fig. 3d–f). One exceptional region is around the Nepal-India





border, where agriculture is the dominant economy. The NO<sub>2</sub> concentration is very low with no significant changes and fine mode AOD showing slightly increasing trend. However, the coarse mode AOD, which is the major contribution of aerosol loading during MAM (pre-monsoon) (Gautam et al., 2009b), showing a slightly decreasing trend <sup>5</sup> due to reduction of the long-range transport of aerosols ranging from India and the

Middle East (Carrico et al., 2003; Prasad et al., 2007; Singh et al., 2006).

The spatial distribution of mean rain frequency in spring of 1998–2009 (Fig. 3g) in China was consistent with the precipitation distribution measured by the surface rain gauge network (Liu et al., 2005; Yang and Lau, 2004; Zhai et al., 2005). Precipitation occurred more frequently south of Yangtze River and along the India-Myanmar bor-

- occurred more frequently south of Yangtze River and along the India-Myanmar border. The spatial distributions of rain frequency trends were different from the mean rain frequency distribution (Fig. 3h and i). It suggests that changes in rain frequency are not mainly caused by possible rain band shifts associated with large scale dynamical changes. The most significant reductions in rain frequency were observed over Eastern
- <sup>15</sup> China, while no significant trends were detected over western China and even increasing trends were detected over some regions around the Nepal-India border. Based on the threshold of statistical significant level of 95%, three regions show a distinguished trend in rain frequency: Eastern China, India-Myanmar region, and Nepal-India region (Fig. 3c). The first two regions showed a significant decreasing trend and the last region showed an increasing trend.

In Eastern China, the significantly decreasing trends in precipitation frequency were detected at the industrial areas with rapid economic growth, rather than the areas with high mean rain frequency. Each region exhibited its own local characteristics of geography, pollution, aerosols, and precipitation frequency. For Eastern China, there were two rain frequency reduction bands: one in the Yellow River region and the other along the Yangtze River region. In the Yellow River rain frequency reduction band, where the largest coal-producing and consumption areas are located, the NO<sub>2</sub> concentrations increased substantially over the past decade, accompanied by an increase of the fine mode AOD. In the Yangtze River rain frequency reduction band, there are many





mega-cities, such as Shanghai, Nanjing, Wuhan, and Changsha. For the past decade the economic development resulted in severe pollution as indicated by the increasing trends in  $NO_2$  concentration and in the fine mode AOD. Also, the warm cloud fraction increased with fine aerosol loading is found in both bands, shown in Fig. 4. In contrast,

the two variables showing no correlation in background area with pristine atmosphere. The results corroborates that aerosol loading increases the amount of low-level cloudiness and inhibits the precipitation (Nakajima et al., 2001; Ramanathan et al., 2001). The spatial correlation between the increasing trend of NO<sub>2</sub> concentrations (and the positive fine mode AOD trend) and the decreasing trend of rain frequency suggests
 that the two have some fundamental linkage.

The India-Myanmar Region is located to the south of Hengduan Mountain. Moisture air mass from the Indian Ocean will form orographic precipitation, which contributes to the high rain frequency in the region. Although the trend in  $NO_2$  concentration was not significant in the region, the enhancement of the coarse mode AOD (and  $NO_2$ 

- <sup>15</sup> concentration) in the upwind region was clearly evident. The observed decreasing trend in rain frequency along Hengduan Mountain reflects the impacts of enhanced aerosols on the orographic precipitation (Givati and Rosenfeld, 2004; Rosenfeld et al., 2007). In the upwind region, the decreasing trend of rain frequency coincided well with the increasing trend in coarse mode AOD (Alpert et al., 2008; Halfon et al., 2009;
- van den Heever and Cotton, 2007). Also as shown in Fig. 5, there was an increasing trend of ice cloud fraction. As some of coarse aerosols may be insoluble particles, increased coarse mode aerosols may enhance heterogeneous nucleation processes and ice clouds, suppressing precipitation (DeMott et al., 2003; Rosenfeld et al., 2001; Min et al., 2009).
- The only region with an increased precipitation frequency is located at the Nepal-India region, bordered by the Himalaya mountain range to the north. As discussed previously, since the high values of total mean AOD in this region during MAM are mostly contributed by the dust transport driven by pre-monsoon westerlies (Gautam et al., 2009b), it can be inferred that aerosol loading diminished during the study period





when coarse mode AOD showed a negative trend. Thus there is an inverse relationship between the rain frequency increase and the aerosol reduction, which is consistent with the relationship of aerosol loading and rain frequency that revealed by the former two regions. However, the changes of both fine mode AOD and coarse mode AOD is much

- <sup>5</sup> smaller than the trend of rain frequency, suggesting there may some other mechanism response for the rain frequency increment. Gautam et al. (2009a) suggested that the high value of aerosol loading during the pre-monsoon period (MAM) may induce a wetter season in early summer, since the land-sea temperature gradient was strengthened through the aerosol solar absorption. So the potential role of aerosol loading in rain frequency changes may also support in this region with rain frequency increasing despite
- a more rigorous analysis of the reciprocal relation between aerosol and precipitation is required by model studies and specific case investigation.

To further illustrate the relationship of pollution and associate aerosols with rain frequency in the entire domain, we selected pixels with statistically significant trends in

- fine mode AOD, NO<sub>2</sub>, and rain frequency. As shown in Figure 6a, there is a positive correlation coefficient of 0.46 at a 95% confidence level between the fine mode AOD trend and the NO<sub>2</sub> trend. As discussed above, NO<sub>2</sub> is a key precursor of secondary aerosols, especially in urban areas. Thus, an increase of NO<sub>2</sub> emission is generally accompanied by an increase of fine aerosol particles. Precipitation may directly re-
- <sup>20</sup> duce the aerosol concentration in the atmosphere through the wet scavenging process at short time scales. However, the wet scavenging has little impact on NO<sub>2</sub> column concentration. The positive correlation between the fine mode AOD trend and the NO<sub>2</sub> trend also suggests that the wet scavenging process associated with precipitation has no dormant effect in the observed trend of AOD at long-term scales.

As shown in Fig. 6b, the grids with an increase of aerosol loading have a significant rain frequency reduction, with a correlation coefficient of 0.44 at 95% confidence level. Also, a strong positive correlation (0.71 at 95% confidence level) between the aerosol loading and the rain frequency trend was found in the grids with a significant increase of rain frequency. Most of these grids are located in the Nepal-India region.





To further illustrate the potential linkage of aerosols and rain frequency, the distribution of correlation coefficient between the two is presented in Fig. 7. Due to the limited time span of Terra MODIS measurements, a low confidence level of 75% is set to filter out insignificant grids. As shown in Fig. 7, many negatively correlated grids between total AOD and rain frequency appeared in Factors. China, India Myanmar region, and

- total AOD and rain frequency appeared in Eastern China, India-Myanmar region, and Nepal-India region, which is consistent with our trend analysis. In detail, stronger negative correlations of rain frequency with fine mode AOD are detected over the Sichuan basin, the Pearl River delta, and the areas approximately overlapped with the two rain frequency reduction bands in Eastern China, where pollution emission substantially in-
- <sup>10</sup> creases over years. However, in India-Myanmar region, the rain frequency is negatively correlated with the coarse mode AOD, and to some extent positively correlated with the fine mode AOD. The negative correlation with the coarse mode AOD is similar to that with the total AOD, as the coarse mode AOD dominates the component of aerosols in the region.
- However, changes in large-scale atmospheric circulation could result in observed changes in precipitation. The large-scale factors that correlate well with precipitation are the column precipitable water (PW) and divergence of water vapor transport (DWVT) in the atmosphere (Park et al., 2007; Qian et al., 2009). We used NCEP reanalysis data to investigate trends of the two factors in the selected regions. Al-
- though the resolution of NCEP reanalysis data is coarse at 2.5° × 2.5°, the regional features are evident. As shown in Figure 8, the spatial distribution of the PW in spring shows statistically insignificant trends in all selected regions. Similarly, most regions have statistically insignificant trends in DWVT integrated from 1000 mb to 500 mb in spring, except for a few grid-points near north and south boundaries. It illustrates that
- the observed changes in precipitation were not related to the dynamical changes in the atmosphere.

Similar spatial-temporal analysis of precipitation amount from TRMM PR illustrates much weaker regional features than those in rain frequency. It corroborates our finding in Shanghai that the changes in precipitation are more due to changes in precipitation





occurrence than in precipitation amount (not shown here). Further, extensive studies on other seasons have been conducted. The spatial-temporal features of rain frequency in both summer and winter seasons showed a major cluster of decreasing trend pixels, associated with the mean rain frequency. It suggests those changes is rain frequency may be deminated by showned in measure dynamics.

in rain frequency may be dominated by changes in monsoon dynamics. In fall, the spatial-temporal features of rain frequency had some but weaker coherence to the regional features of NO<sub>2</sub> and aerosol trends than in spring. It may be partially due to some influences of monsoon dynamics, as the monsoon transit in fall is relatively short.

# 4 Conclusion and discussion

- A spatial-temporal analysis has been conducted using satellite observed distributions of rain frequency, NO<sub>2</sub> concentration, and aerosols in spring over East Asia. The growing anthropogenic emissions have led to increased air pollution, i.e., anthropogenic aerosol and its precursor gases which have reported in many previous studies (Lu et al., 2010; Zhang et al., 2007) and also present in this study. An increase of NO<sub>2</sub> emission is generally accompanied by an increase of fine aerosol particles. The wet scavenging process associated with precipitation may have no dormant effect in the observed trend of AOD at long-term scales. More importantly, the overall feature emerged from the region-by-region analyses, including Eastern China, India-Myanmar border, and Nepal-India region, is that there is an inverse relationship between the rain
- frequency and the pollution and associated aerosols in spring. The climatically significant reduction of rain frequency is observed in the first two regions, where pollution and aerosols exhibit an increase trend. Also an increase trend of rain frequency is detected in the last region where aerosols decrease. Comparison between trends in rain frequency and in precipitation amount shows that the changes in precipita-
- tion are more due to changes in precipitation occurrence than in precipitation amount. The spatial-temporal inverse relationship of pollution and rain frequency at continental





scale suggests possible indirect climate effects of anthropogenic pollution on precipitation. Two possible pathways are speculated: (1) the increased  $NO_2$  and aerosols (soot particles in particular) enhance the absorption of solar radiation and stabilize the atmosphere, resulting in reduction of cloud formation and rain frequency; and/or

- (2) the enhancement of pollution-produced CCN in addition to mineral dust from longrange transport further suppresses the rain frequency, as favored by topography, wind, and other meteorological conditions. However, aerosol effects on precipitation follow a chain of microphysical and thermodynamical processes that occur at shorter time scales. Also other effects, such as urban effects, may contribute to the reduction of rain frequency at ameli local cools.
- rain frequency at small local scale. Hence, more robust statistical study at various temporal and spatial scales and detailed modeling investigation are warranted to further understand the observed relationship between the rain frequency and the pollution and associated aerosols.
- As the large-scale precipitation is controlled by evaporation, aerosols might influence it by surface cooling. In particular, aerosol microphysical effects can actually affect precipitation characteristics. Recent studies in North America also showed that the rain frequency was increased (Karl and Knight, 1998) while the tropospheric NO<sub>2</sub> column was decreased (Richter et al., 2005). It further corroborates our finding that changes in rain occurrence may be associated with changes in pollution and asso-
- ciated aerosols. Furthermore, the suppression of precipitation leads to an increase in moisture and hygroscopic particles in the atmosphere. The increased amount of moisture and hygroscopic particles enhances regional haze if the moisture is relatively limited, or results in intense precipitation if water vapor in the atmosphere exceeds a threshold. This hypothesis is supported by the surface observations in China, i.e., in-
- <sup>25</sup> creasing haze days (Chan and Yao, 2008; Ma et al., 2009); and an increasing trend of intensive precipitation frequency over the Yangtze River Basin (Zhai et al., 2005; Su et al., 2007).

These findings highlight the threat to vital water resources in polluted regions of the world, as in some industrialized areas of China and India, not only locally but also in





the downwind regions. The importance of that is underlined by the realization that it is not high temperatures due to global warming but rather the lack of water that makes a region into an unlivable land. Particularly, any precipitation change in spring will significantly impact the stable crop production in the regions.

Acknowledgements. Many thanks to the TRMM, MODIS data processing team members and IUP-Bremen DOAS group for GOME and SCIAMACHY data. This research was supported by the Office of Science (BER), US Department of Energy, Grant DE-FG02-03ER63531, and by the NOAA Educational Partnership Program with Minority Serving Institutions (EPP/MSI) under cooperative agreements NA17AE1625 and NA17AE1623. This work was also supported by the oversea scholar foundation of the China Scholarship Council and partially by the National Key Project of Basic Research of China (Grant Nos. 2006CB403704).

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CC II

**Table 1.** The location and correlation of rain gauge and TRMM PR measured precipitation amount at various sites over China.

	Location	Altitude	Function	R
Shanghai	31°10′ N, 121°26′ E	2.8 m	PR = 0.78 × Gauge -5.60	0.81
Hangzhou	30°14′ N, 120°10′ E	41.7 m	PR = 0.44 × Gauge +65.6	0.68
Changsha	28°12′ N, 113°05′ E	44.9 m	PR = 0.66 × Gauge +41.8	0.69
Wuhan	30°37′ N, 114°08′ E	23.3 m	PR = 0.76 × Gauge +122.1	0.74
Zhengzhou	34°43′ N, 113°39′ E	110.4 m	PR = 0.57 × Gauge +17.6	0.84
Nanjing	32°00′ N, 118°48′ E	8.9 m	PR = 0.94 × Gauge -41.5	0.88
Xi'an	34°18′ N, 108°56′ E	396.9 m	PR = 0.57 × Gauge +16.2	0.79
Lasa	29°40′ N, 91°08′ E	3648.7 m	PR = 0.22 × Gauge +6.20	0.77



Fig. 1. Seasonal precipitation amount estimated from TRMM PR and measured from surface rain gauge from 1998 to 2009 in Shanghai.



















**Fig. 3.** Spatial distributions in spring during 1998–2009: (a) mean tropospheric  $NO_2$  column density, (b) tropospheric  $NO_2$  column density annual trend, (c) tropospheric  $NO_2$  column density annual trend with significant level above 95%, (d) MODIS mean total Aerosol Optical Depth (AOD), (e) MODIS fine mode AOD annual trend, (f) MODIS coarse mode AOD annual trend, (g) mean TRMM PR rain frequency, (h) TRMM PR rain frequency annual trend, and (i) TRMM PR rain frequency annual trend with significant level above 95%.



**Fig. 4.** The warm cloud fraction and fine AOD over Yangtze River Region  $(30-34^{\circ} N, 118-122^{\circ} E, grid over ocean is excluded)$ , Yellow River Region  $(32-36^{\circ} N, 111-115^{\circ} E)$ , and background region  $(32-36^{\circ} N, 100-104^{\circ} E)$  with the pristine atmosphere.







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**Fig. 6. (a)** Scatterplot of the  $NO_2$  column concentration trends and the fine AOD trend; **(b)** scatterplot of the rain frequency trend and the fine AOD.



Fig. 7. The spatial distribution of correlation coefficient between rain frequency with AOD (a, b with only significant correlation coefficients shown, a 75% confidence level was taken); fine mode AOD (c, d same with b); coarse mode AOD (e, f same with b).



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**Fig. 8.** Spatial distributions of column precipitable water (PW) and divergence of water vapor transport (DWVT), and their corresponding trends in spring during 1998–2009. The shaded grids are the trend with significant level above 95%.



