

**Spatial features of  
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change**

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

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## 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. As revealed by measurements from 1998–2009 over Shanghai, China, the suppression of rain is mainly attributed to the reduction of rain occurrence rather than changes in rain intensity. The overall features emerge from the region-by-region analyses that there is an inverse relationship between the rain frequency and the pollution and associated aerosols at continental scale in spring. The enhancement of pollution-produced CCN in addition to mineral dust from long-term transport suppresses the rain frequency, as favored by topography, wind, and other meteorological conditions.

## 1 Introduction

Human-induced climate change has caused a redistribution of precipitation (Zhang et al., 2007). Besides the greenhouse gases-induced global warming, anthropogenic aerosols increase concentrations of cloud condensation nuclei (CCN) and ice-forming nuclei (IN), which alter the main path of precipitation-forming microphysical processes and the precipitation amount (e.g., Cotton and Pielke, 1995; Lohmann et al., 2005; Rosenfeld et al., 2008). The response of the hydrological cycle to the aerosol indirect effect is different to the greenhouse effect, and the hydrological cycle is expected to be weakened due to aerosol effects (Ramanathan et al., 2001; IPCC, 2007). 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, 2007; New et al., 2001; Yang et al., 2004; Qian et al., 2009). 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

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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 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 variations of 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 impacts of air pollutants and associated anthropogenic aerosols on precipitation from the spatial-temporal perspective by utilizing 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 et al., 1999). Also any precipitation change in spring will significantly impact stable crop production in the region. Therefore, we will focus our study on the spring season.

## 2 Measurements

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 precipitation distribution, monthly rain rate dataset from TRMM Precipitation Radar (PR) at 0.5° × 0.5° spatial grid (TSDIS, 2007; version; 3A25; source: <http://daac.gsfc.nasa.gov/data/>) from 1998 to

2009 are used in this study. As a marker of air pollution, tropospheric nitrogen dioxide ( $\text{NO}_2$ ) has been monitored by both the **Global Ozone Monitoring Experiment (GOME)** and **SCanning Imaging Absorption SpectroMeter for Atmospheric CHartography (SCIAMACHY)** satellites. Hence, monthly  $\text{NO}_2$  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](http://www.iup.uni-bremen.de/doas/data_products.htm)) are used to quantify air pollution changes over the same period of PR dataset. Precipitation can be influenced by anthropogenic aerosols associated with pollution through their roles in cloud condensation nuclei and ice nuclei. 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. In addition to the above three combined satellite datasets that provide the spatial-temporal variation of pollution, aerosols, and precipitation, the surface rain gauge precipitation data are used to verify the satellite measurements and investigate the relationship between precipitation and air pollutants.

### 3 Results

Due to the relatively short lifetime of  $\text{NO}_2$  and the vertical distribution of  $\text{NO}_x$  sources,  $\text{NO}_2$  columns observed from space are dominated by the  $\text{NO}_2$  concentration in the boundary layer and at the location (Richter et al., 2005). As shown in Fig. 1, satellite retrieved  $\text{NO}_2$  column concentration in spring at Shanghai increased substantially from 1998 to 2009. The linear trend in  $\text{NO}_2$  column concentration is  $1.9 \times 10^{15}$  molec/cm<sup>2</sup> per year. With respect to the reference value of  $6.0 \times 10^{15}$  molec/cm<sup>2</sup> in spring 1998, air pollution in Shanghai was tripled from 1998 to 2009. Nitrogen dioxide is an effective absorber of visible and near-ultraviolet solar radiation. At wavelengths below  $\sim 400$  nm, photodissociation of  $\text{NO}_2$  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  $\text{NO}_2$  in the troposphere. The photodissociation of  $\text{NO}_2$

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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>, 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 atmospheric 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. 1a, the retrieved AOD from MODIS showed an increase in recent years, although there was large interannual variability which was mostly associated with the sprint dust events. The trend of fraction ratio of fine mode aerosols increased consistently with the increase trend in NO<sub>2</sub> concentration.

Most particles over urban areas are composed of hygroscopic salts, i.e., sulfates and nitrates (Givati et al., 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°×1° spatial domain centered at Shanghai correlated well with the measured precipitation from a single surface rain gauge (Fig. 1b). Although there is a spatial-temporal mismatch between the two, the consistency of decreasing trends of precipitation 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 trend 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  $1^\circ \times 1^\circ$  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 decrease trend of 4.04% per year in rain frequency (0.21% per year in absolute rain frequency) is slightly greater than 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  $\text{NO}_2$  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 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, 2007; Ramanathan et al., 2001). The opposite trends of precipitation and air pollutants imply the possibility that the increased particles over urban areas suppress the local precipitation, particularly the rain frequency.

The inverse relations of rain frequency and precipitation to the concentrations of  $\text{NO}_2$  and aerosols at a single site for past decades can be casual, as precipitation changes are strongly influenced by changes of large scale dynamics. To exclude the possible influence of meteorological factor changes on specific sites, the spatial-temporal distribution of rain frequency,  $\text{NO}_2$  concentration, and aerosol loading are investigated.

Many studies suggested that there were strong increase trends of  $\text{NO}_2$  in some regions of China and India for the past decade (Richter et al., 2005; and van der A et al., 2006). As illustrated in Fig. 2a–c, those regions include the North Chinese Plain, 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

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5 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. 2d–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 and has no significant changes. However, both fine and coarse AODs show a negative trend in the region, which could be in part due to reduction of the long-range transport of aerosols ranging from India and the Middle East (Carrico et al., 2003; Singh et al., 2006; Prasad et al., 2007).

10 The spatial distribution of mean rain frequency in spring of 1998–2009 (Fig. 2g) in China was consistent with the precipitation distribution measured by the surface rain gauge network (Yang et al., 2004; Liu et al., 2005; and Zhai et al., 2005). Precipitation 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. 2h and i). It suggests that changes in rain frequency are not  
15 caused by possible rain band shifts associated with large scale dynamical changes. The most significant reductions in rain frequency were observed over Eastern 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  
20 trend in rain frequency: Eastern China, India-Myanmar region, and Nepal-India region (Fig. 2c). The first two regions showed a significant decreasing trend and the last region showed an increasing trend.

25 In general, the significant decrease 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

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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 megacities, such as Shanghai, Nanjing, Wuhan, and Changsha. For the past decade the economic development resulted in severe pollution as indicated by the increase trends in  $\text{NO}_2$  concentration and in the fine mode AOD. The spatial correlation between the increase trend of  $\text{NO}_2$  concentrations (and the positive fine mode AOD trend) and the decrease 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  $\text{NO}_2$  concentration was not significant in the region, the enhancement of the coarse mode AOD (and  $\text{NO}_2$  concentration) in the upwind region was clearly evident. The observed decrease trend in rain frequency along Hengduan Mountain reflects the impacts of enhanced aerosols on the orographic precipitation (Givity and Rosenfeld, 2004; Rosenfeld et al., 2007). In the upwind region, the decrease trend of rain frequency coincided well with the increase trend in coarse mode AOD.

The only region with an increase precipitation frequency is located at the Nepal-India region, bordered by the Himalaya mountain range to the north. As discussed previously, both fine and coarse mode AODs showed a negative trend in the region. Thus there is an inverse relationship between the rain frequency increase and the aerosol reduction, which is consistent with our hypothesis that aerosols play a key role of modulating rain frequency.

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. Although the resolution of NCEP reanalysis data is coarse at  $2.5^\circ \times 2.5^\circ$ , the regional features are evident. As shown in Fig. 3, 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 air pollution and associated aerosols suppress precipitation occurrence rather than 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 decrease trend pixels, associated with the mean rain frequency. It suggests those changes 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  $\text{NO}_2$  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,  $\text{NO}_2$  concentration, and aerosols over East Asia. The overall feature emerging from the region-by-region analyses was that there is an inverse relationship between the rain frequency and the pollution and associated aerosols in spring. The spatial-temporal consistency of pollution and rain frequency at continental scale provides further evidences that precipitation could be changed possibly due to the pollution effects. Comparison between trends in rain frequency and in precipitation amount shows that air pollution tends to suppress precipitation occurrence more than precipitation amount. The growing anthropogenic activities have led to increased air pollution, i.e., anthropogenic emission of aerosol and its precursor gases, in springtime due to

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the rapid urbanization and motorization. We speculate that 1) the increased NO<sub>2</sub> 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 2) the enhancement of pollution-produced CCN in addition to mineral dust from long-term transport further suppresses the rain frequency, as favored by topography, wind, and other meteorological conditions. Certainly, more robust statistical study 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 et al., 1998) while the tropospheric NO<sub>2</sub> column was decreased (Richter et al., 2005). It further corroborates the hypothesis that the high concentration of anthropogenic emission of aerosol and its precursor gases suppresses rain occurrence. 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., increasing haze days (Zhuang et al., 2007); and an increasing trend of intensive precipitation frequency over the Yangtze River Basin (Jiang et al., 2007; 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.

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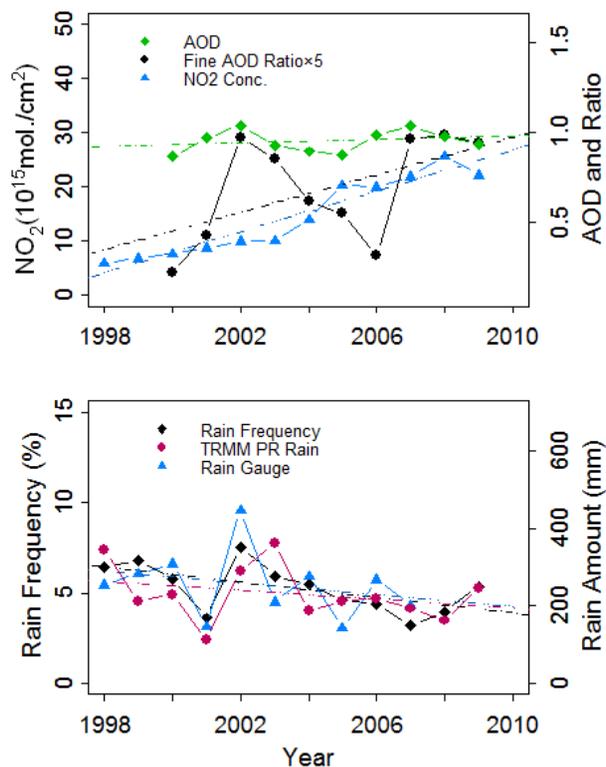
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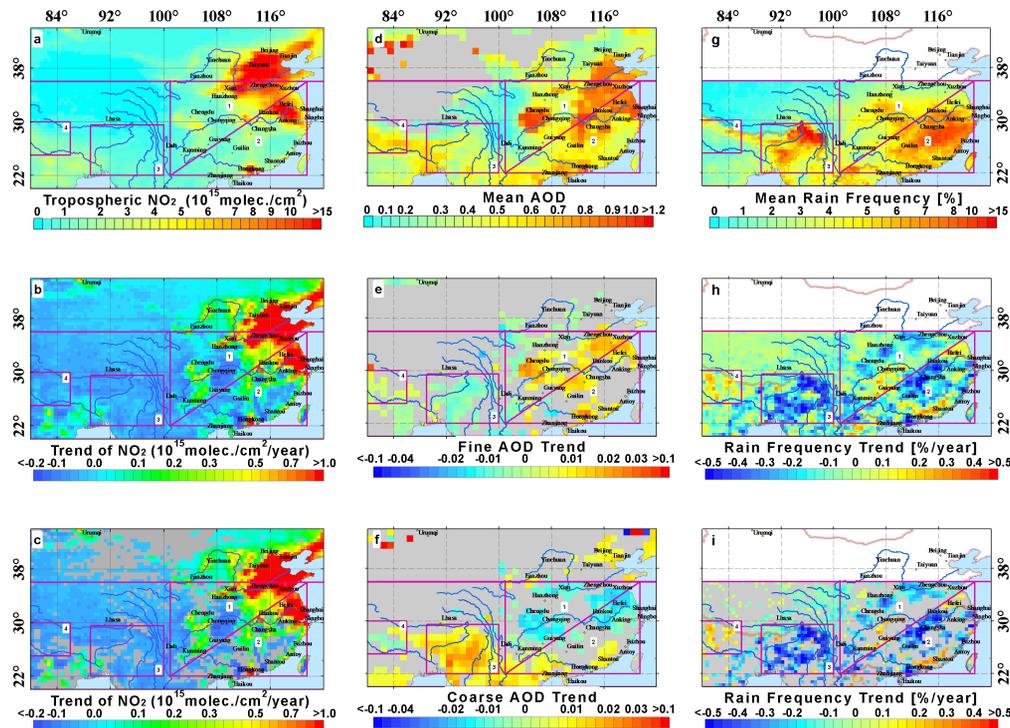


**Fig. 1.** (a) Time series of MODIS aerosol optical depth and satellite measured tropospheric NO<sub>2</sub>; (b) time series of TRMM PR rain frequency and rain amount, and surface rain gauge measured precipitation from 1998 to 2009 for the 1° × 1° grid near Shanghai.

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**Fig. 2.** Spatial distributions in spring during 1998–2009: **(a)** mean tropospheric  $\text{NO}_2$  column density, **(b)** tropospheric  $\text{NO}_2$  column density annual trend, **(c)** tropospheric  $\text{NO}_2$  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%.

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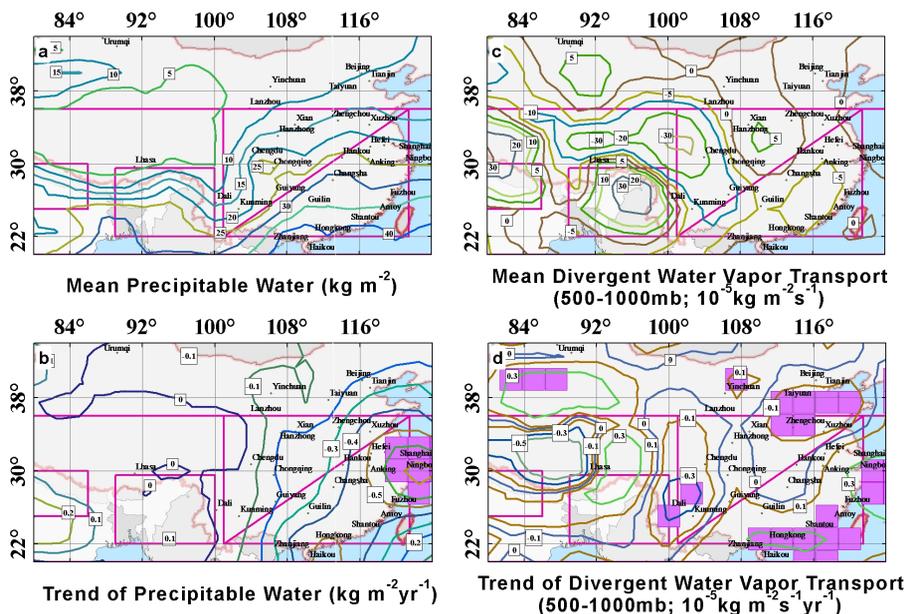
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**Fig. 3.** Spatial distributions of column precipitable water (PW) and divergence of water vapor transport (DWVT), and their corresponding trends in spring during 1998–2009. The shaded grids are the trend with significant level above 95%.

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