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LETTER

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Minghui Tao^{1,2}, Liangfu Chen^{1,2}, Zifeng Wang^{1,2}, Jun Wang³, Jinhua Tao^{1,2} and Xinhui Wang¹¹ State Key Laboratory of Remote Sensing Science, Institute of Remote Sensing and Digital Earth, Chinese Academy of Sciences, Beijing 100101, People's Republic of China² Joint Center for Global Change Studies, Beijing 100875, People's Republic of China³ Department of Earth & Atmospheric Sciences, University of Nebraska—Lincoln, NE 68588, USAE-mail: chenlf@radi.ac.cn and wangzf@radi.ac.cn**Keywords:** haze, aerosol, MODIS, ChinaSupplementary material for this article is available [online](#)**Abstract**

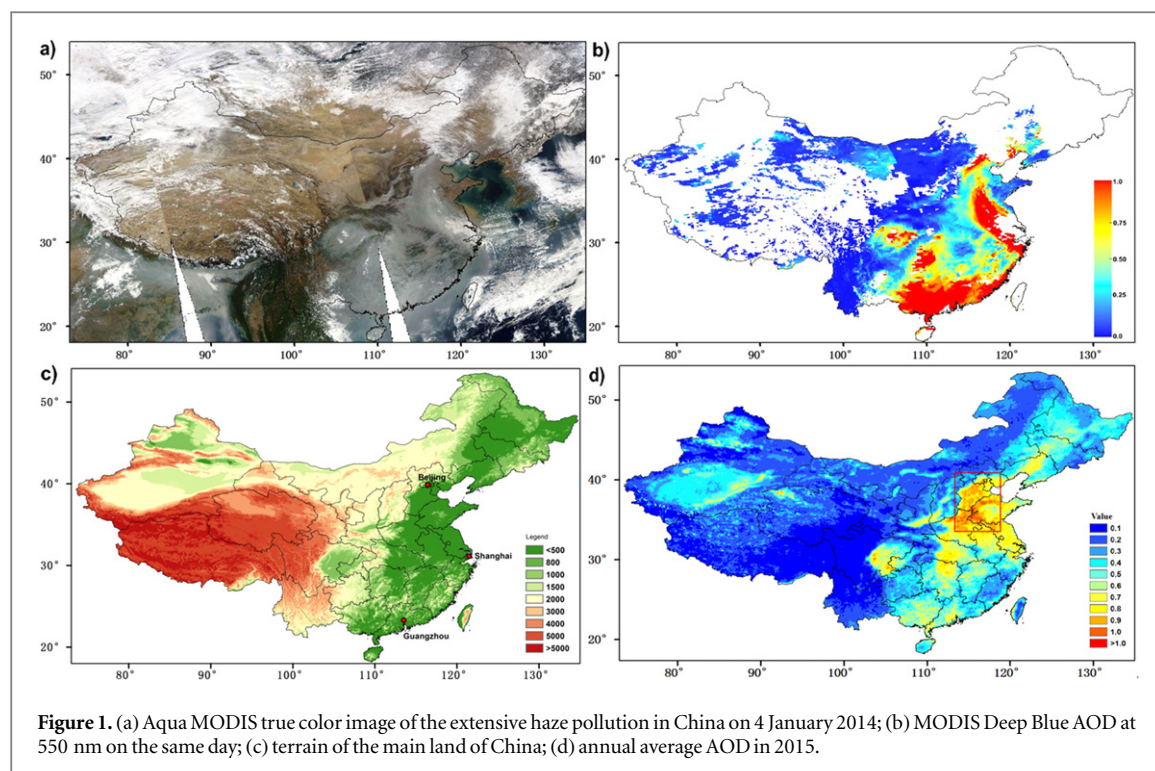
Widespread haze layers usually cover China like low clouds, exerting marked influence on air quality and regional climate. With recent Collection 6 MODIS Deep Blue aerosol data in 2000–2015, we analyzed the trends of regional haze pollution and the corresponding influence of atmospheric circulation in China. Satellite observations show that regional haze pollution is mainly concentrated in northern and central China. The annual frequency of regional haze in northern China nearly doubles between 2000 and 2006, increasing from 30–50 to 80–90 days. Though there is a marked decrease in annual frequency during 2007–2009 due to both reduction of anthropogenic emissions and changes of meteorological conditions, regional pollution increases slowly but steadily after 2009, and maintains at a high level of 70–90 days except for the sudden decrease in 2015. Generally, there is a large increase in the number of regional-scale haze events during the last decade. Seasonal frequency of regional haze exhibits distinct spatial and temporal variations. The increasing winter haze events reach a peak in 2014, but decrease strongly in 2015 due partly to synoptic conditions that are favorable for dispersion. Trends of summer regional haze pollution are more sensitive to changes of atmospheric circulation. Our results indicate that the frequency of regional haze events is associated not only with the strength of atmospheric circulation, but also with its direction and position, as well as variations in anthropogenic emissions.

1. Introduction

Under the background of large increase in anthropogenic emissions during the last decades, haze pollution has been a common problem over China with the degradation of visibility and air quality (Zhang *et al* 2012, Han *et al* 2014). Satellite observations show that haze pollution not only exists in the urban regions, but also usually appears in the extensive rural areas, which form widespread haze layers like low clouds over China (Tao *et al* 2012). Such regional heavy aerosol loading not only threatens public health near the surface, but also exerts marked influences on radiation budget and cloud properties (Fan *et al* 2008, Li *et al* 2011). In addition, thick haze layers can further

aggravate air pollution near surface by radiative feedback (Gao *et al* 2015).

Recently, several extreme haze events over eastern China have led to wide concern due to their extensive spatial coverage and record-breaking particle pollution (Tao *et al* 2013, Yang *et al* 2013, Wang *et al* 2014). Observations at meteorological stations shows a large increasing trend in haze days in China since 2000 (Qu *et al* 2015). Moreover, interannual trends of the haze days have a close relationship with regional circulation (Cao *et al* 2015). However, visibility mainly reflects information of horizontal extinction near surface, which can be impacted by local emissions and meteorology, especially in urban regions. By contrast, columnar extinction of aerosols exhibited different spatial



characteristics (Tao *et al* 2015, Qu *et al* 2016). Formation and variation of the thick haze layers is mostly connected with regional transport and moist airflows (Tao *et al* 2014), which can be more sensitive to variations of atmosphere circulation. So far, it is still not clear how these common haze plumes change over space and time as well as their connection with regional circulation.

The recent Collection 6 (C6) Moderate Resolution Imaging Spectroradiometer (MODIS) Deep Blue (DB) aerosol retrieval has expanded to entire land areas (Hsu *et al* 2013), and performs well in the heavy aerosol loading conditions of China (Tao *et al* 2015). Based on the daily DB aerosol optical depth (AOD) in 2000–2015 and meteorological data, we investigated interannual variations of the haze clouds over China, and analyzed the potential influence of regional atmospheric circulation.

2. Data and methods

By measuring reflected and emitted radiance of the Earth-atmospheric system in a broad spectral range with 36 bands between 0.4–14.4 μm , the MODIS sensor can provide near global detection of diverse atmospheric parameters with a wide swath ~ 2330 km and relatively fine resolution at 250–1000 m. MODIS has been flying on Terra satellite since 2000, and on Aqua satellite since 2002, with equatorial overpassing times around 10:30 am and 13:30 pm, respectively.

The MODIS aerosol retrieval is first realized over dense vegetation regions utilizing the linear

relationship between visible and shortwave bands (Levy *et al* 2007). The MODIS DB algorithm is designed and implemented to obtain aerosol information over bright-reflecting regions by assuming that surface reflectance in near-UV blue channel (0.41 μm) is much lower than those in longer visible bands (Hsu *et al* 2006). By considering scattering angles and vegetation density in the estimate of surface reflectance, the C6 DB retrieval has extended to all cloud-free and ice/snow-free land areas (Hsu *et al* 2013).

MODIS aerosol data is provided at a normal spatial resolution of $10 \times 10 \text{ km}^2$ in Level 2 (L2) data (MOD04 for Terra and MYD04 for Aqua), and with a coarser resolution of $1^\circ \times 1^\circ$ in aggregated L3 products (<https://ladsweb.nascom.nasa.gov/data/search.html>). In addition to this, associated parameters such as cloud mask and quality assurance (QA) are also included in L2 aerosol data for examining origins of retrieval uncertainties. Ground-based evaluation shows that the expected error envelope of C6 DB AOD is generally within $\pm (0.03 + 0.2\text{AOD}_{\text{MODIS}})$ (Sayer *et al* 2013). Since DB aerosol retrieval is available in both bright and dark surfaces, DB AOD can well reveal spatial coverage of the regional haze pollution in China (Tao *et al* 2015). To further interpret spatial and temporal variation of the haze pollution, wind fields from the National Centers for Environmental Prediction (NCEP) Reanalysis were used to analyze variations of meteorological conditions (<http://www.esrl.noaa.gov/psd/data/reanalysis/reanalysis.shtml>).

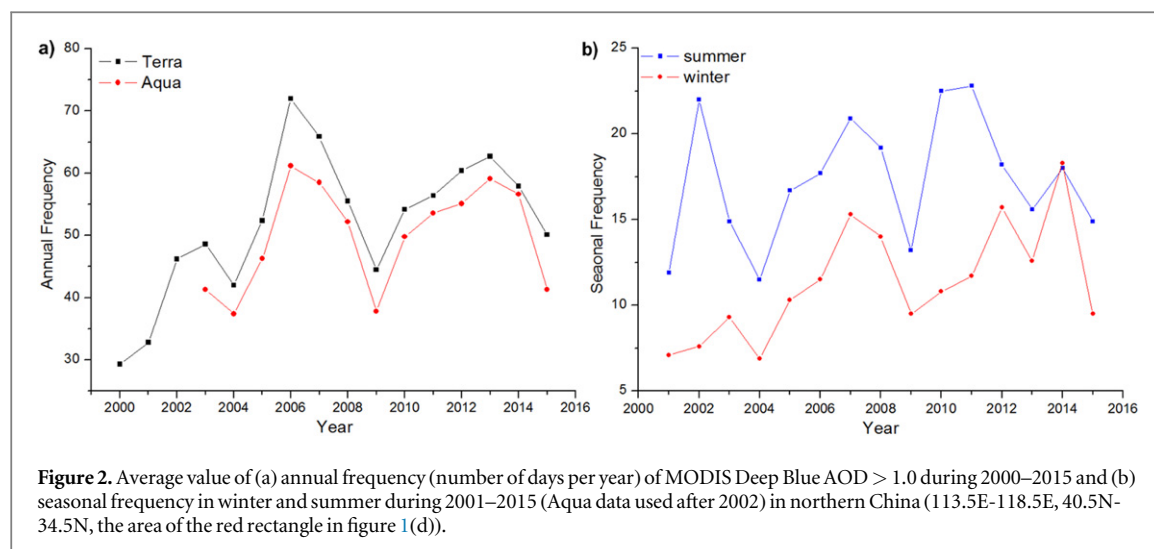


Figure 2. Average value of (a) annual frequency (number of days per year) of MODIS Deep Blue AOD > 1.0 during 2000–2015 and (b) seasonal frequency in winter and summer during 2001–2015 (Aqua data used after 2002) in northern China (113.5E–118.5E, 40.5N–34.5N, the area of the red rectangle in figure 1(d)).

3. Results and discussion

3.1. Satellite view of the widespread haze pollution over China

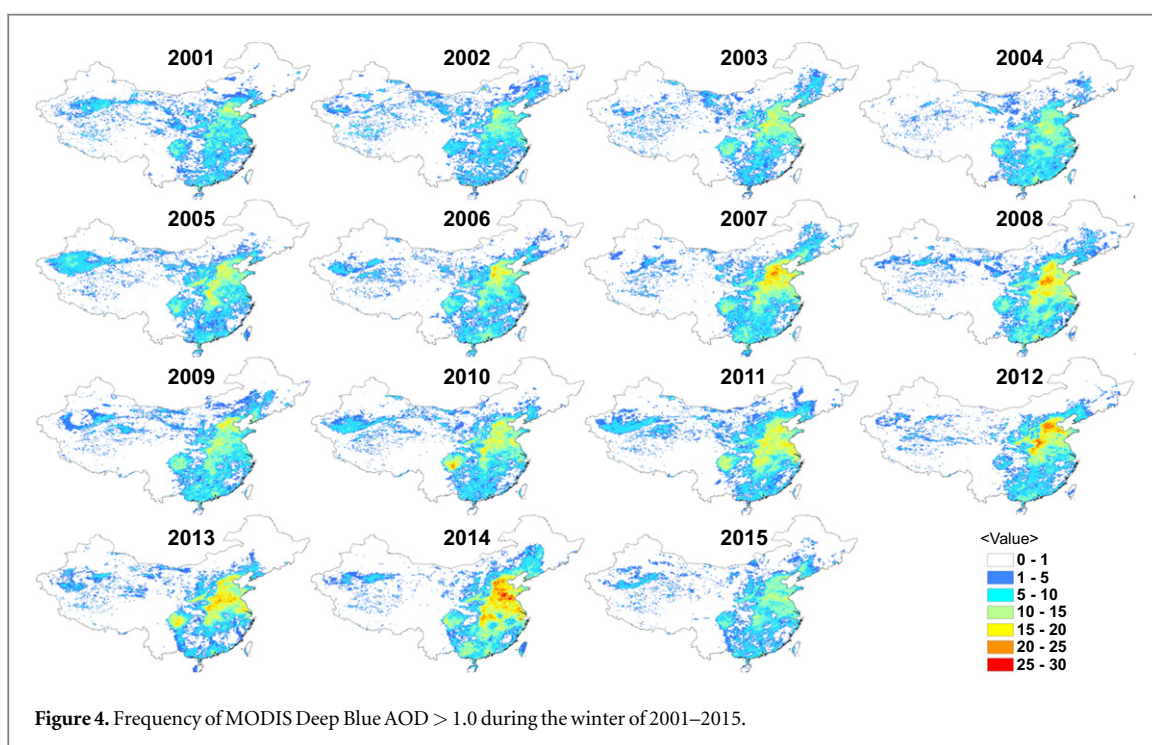
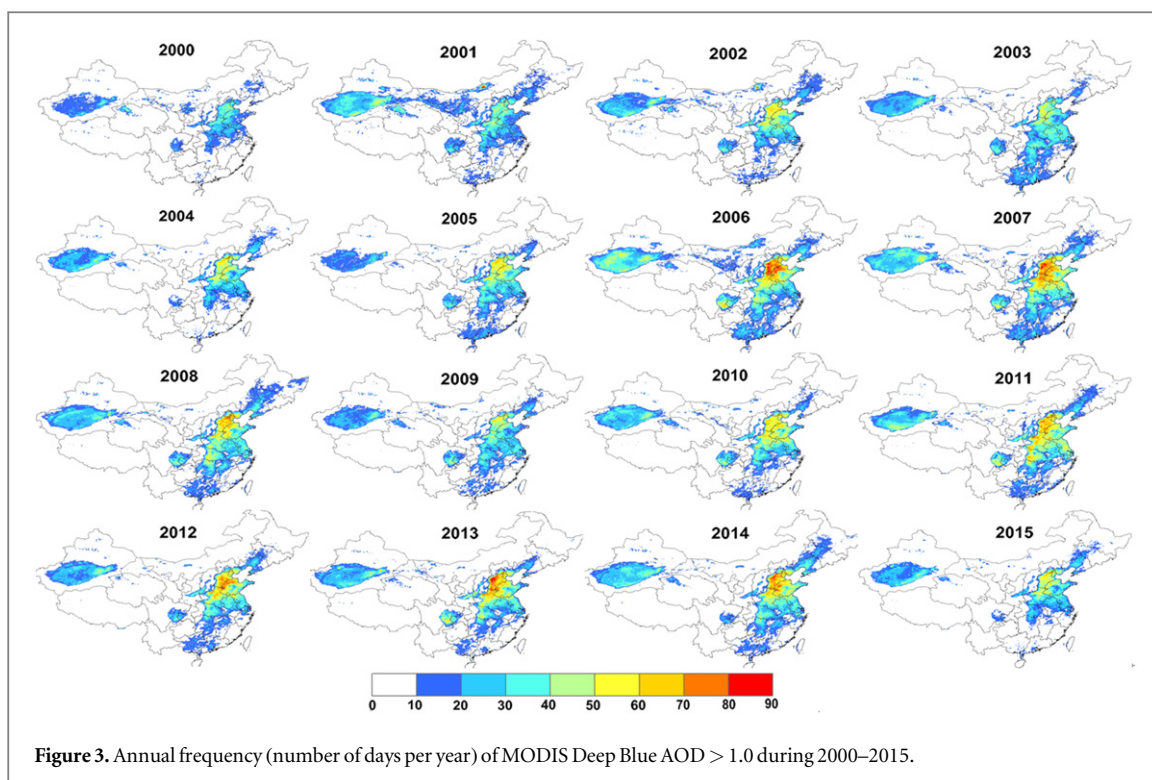
Large-scale haze pollution, as shown in figure 1(a), often appears over China. Such dense haze layers not only exist in urban regions, but also cover extensive rural areas. Satellite observations show that AOD at $0.55 \mu\text{m}$ of the haze layers is far above 1.0 in most areas (figure 1(b)). A haze day is usually defined as a day with low visibility <10 km and relative humidity <80% near surface. Here we mainly focus on regional variations of heavy aerosol loading rather than the usual haze with, low visibility observed from ground sites. Though there is no fixed connection between AOD and visibility, high regional AOD is a good indicator of serious air pollution (tao *et al* (2013), Jia *et al* 2015) and various past studies have shown that low visibility is commonly associated with high AOD except in cases of high altitude aerosol layer (Kessner *et al* 2013). Considering that AOD of the large-scale haze plumes usually far exceed 1.0, we select AOD > 1.0 as threshold of regional haze pollution in this study.

The annual average value of AOD is >0.6 in most areas of eastern China, and exceeds 0.8 in the main polluted region of northern China (figure 1(d)). To get a direct view of temporal variations of the haze pollution, we count linear variations of spatial mean values of the frequency of AOD > 1.0 in the main polluted region of northern China (figure 2). Frequency from Terra AOD is slightly higher than from Aqua due to diurnal changes of aerosol loading and cloud amount. Ground observations show that aerosol loading in the morning is usually higher than in the afternoon in northern China (Kuang *et al* 2015). Meanwhile, there tends to be somewhat more clouds over land in the afternoon (King *et al* 2013). Considering that Aqua MODIS has a higher accuracy in calibration than Terra, Aqua data is used when it is available after 2002.

Frequency of high AOD in summer is consistently larger than in winter, which can be mostly due to intense hygroscopic growth of aerosol particles under common moist weather conditions in summer (Qu *et al* 2016).

Figure 3 shows the annual frequency of regional haze pollution in China during 2000–2015. Occurrence of large-scale haze events is mainly concentrated in northern and central China, where most anthropogenic emissions are located (Lu *et al* 2011). High aerosol loading also exists in the cloudy Sichuan Basin, which can be related to the moist and foggy environment. There are generally 30–40 days with AOD > 1.0 in the Taklimakan Desert every year. Despite the same trends in figures 2 and 3, spatial average obviously decreases the peak values of frequency due to the inhomogeneous distribution of haze pollution. Occurrence of large-scale haze events in the main polluted regions of northern China nearly doubled during 2000–2006, increasing from 30–50 days to 80–90 days. Although there is a marked decrease during 2007–2009, regional pollution increases slowly but steadily after 2009, and maintains at a high level of 70–90 days except sudden decrease in 2015. The annual available days of having Aqua MODIS DB aerosol retrieval in northern China ranges around 180 days (Tao *et al* 2015), indicating that the regional heavy haze pollution nearly accounts for half of the cloud-free days.

Despite the increase of anthropogenic emissions such as carbonaceous aerosol, decline in sulfur dioxide (SO_2) emissions in China appears from 2006 (Lu *et al* 2011), which may partly explain the decrease of haze pollution. The temporary decline in nitrogen oxides (NO_x) emissions in China during 2009 due to economic downturn can further promote the reduction of regional haze (Lin and McElroy (2011)). When the reduction of SO_2 emissions are offset by the increase of other pollutants such as NO_x and carbonaceous aerosol, frequency of the haze pollution can rise again. Compared with the decrease of annual frequency in



2007, increase in haze pollution during winter and summer indicates that meteorological conditions can have also an important contribution.

3.2. Interannual variation of regional haze in winter and summer

To further examine annual variations of the regional haze, we analyzed the seasonal frequency of high AOD in winter and summer. Figure 4 shows changes in trends of AOD > 1.0 in winter during 2001–2015. The

winter of one year here denotes months of January, February, and December of last year. Despite of slight differences, as shown in figure 2, annual trends of the winter haze are generally consistent with that of the yearly frequency. By comparison, regional characteristics of seasonal heavy pollution are clearer, with notable spatial changes in major haze areas. The high frequency of haze pollution during the year tends to be concentrated in northwestern part of the northern China, where the industry belt is located (Lu

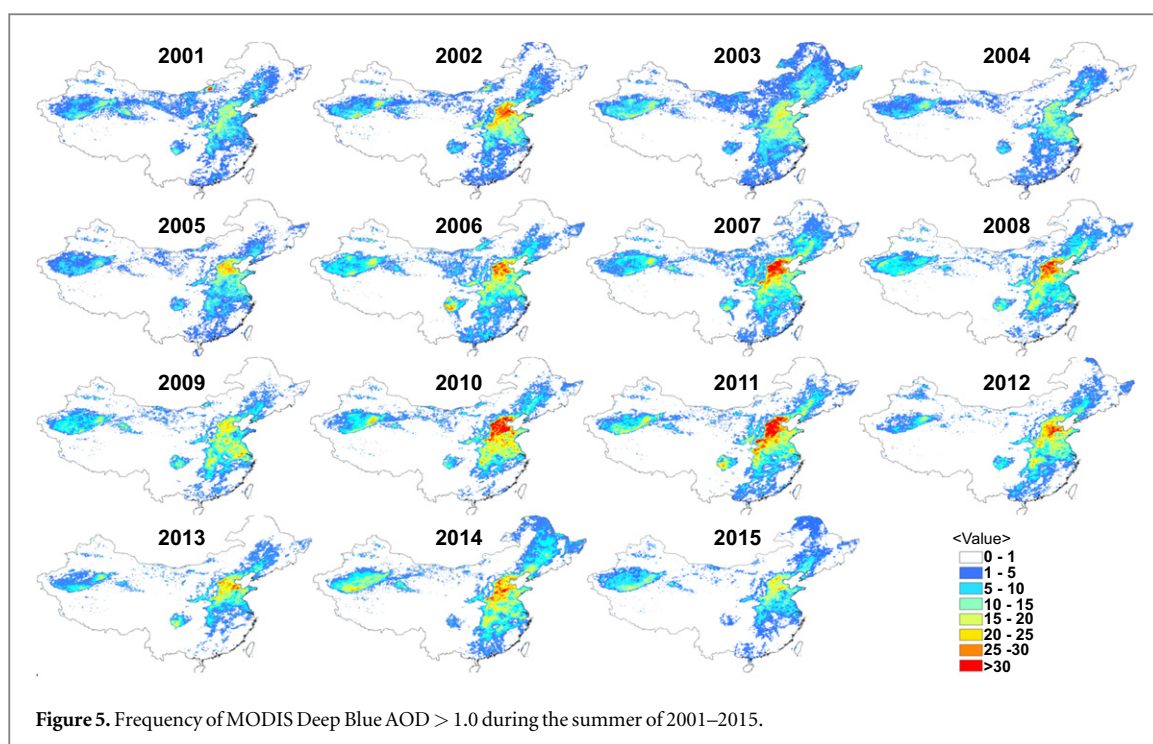


Figure 5. Frequency of MODIS Deep Blue AOD > 1.0 during the summer of 2001–2015.

et al 2011). During the winter, haze areas are more extensive, and major regions move southward obviously in 2013 and 2014, which can be caused by southward transport. The most polluted period occurs in the winter of 2014, with heavily hazy days in northern China reaching one month. However, regional winter haze pollution declines substantially in 2015, due largely to changes in atmospheric circulation (seen in section 3.3).

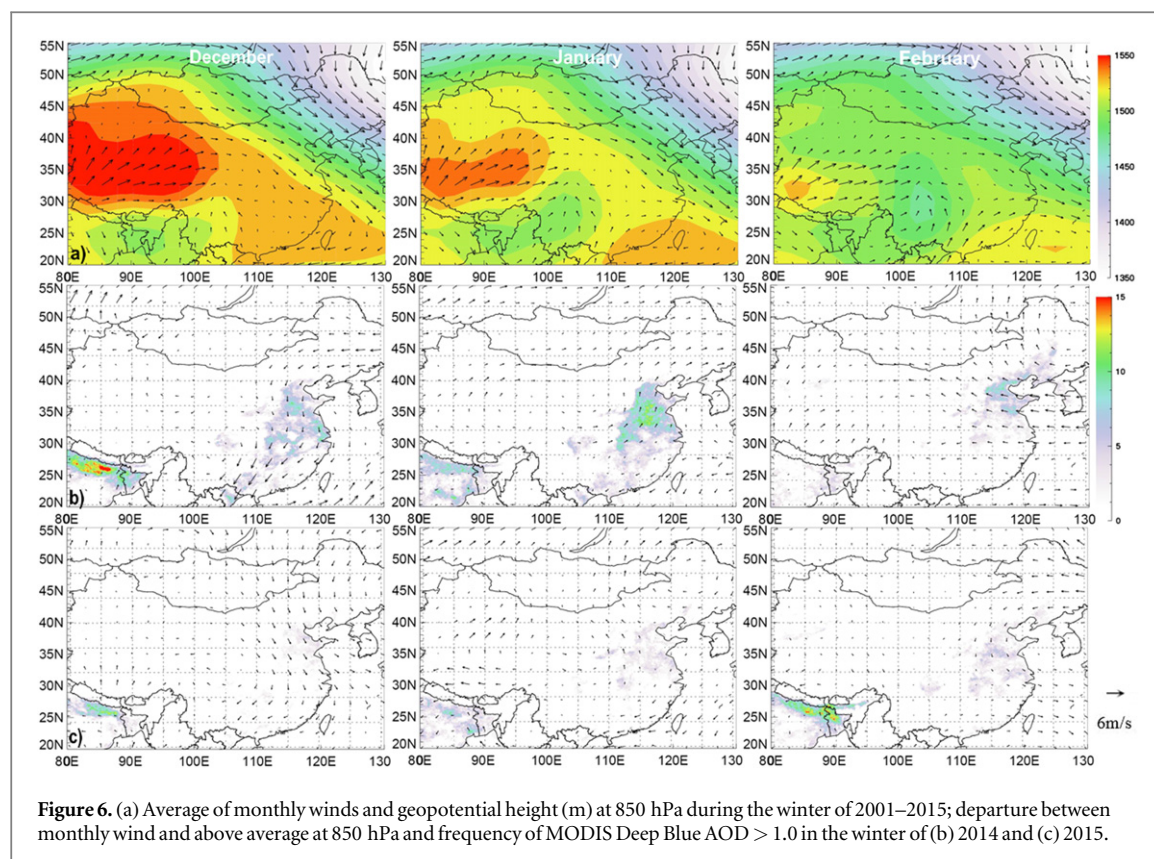
Figure 5 shows the trends of regional haze events in summer during 2001–2015. Similar to the annual frequency, summer haze is mainly concentrated in the northern part of northern China. The amount of regional haze in summer generally increases, but its trend is different as compared to the annual frequency (figure 2). Compared with the low frequency in winter, there is a sudden peak of summer haze events during 2002 due mostly to the variations of the Asian monsoon. Then summer haze increases from 2004, and reached high values during 2007 and 2008. Despite of an obvious decrease in 2009, the most summer haze events occurred in 2010 and 2011, with the number of heavy haze pollution exceeding one month. Though major haze areas in northern China get smaller in 2012–2014, the frequency of summer haze is still at high levels.

The frequency of high AOD in summer is much larger than in winter. Severe particle pollution in northern China usually appears in winter associated with low boundary layer and more coal burning for heating. Although strong convection in summer can reduce particle concentration near surface, agricultural biomass burning and intense moist airflows greatly enhance the columnar extinction of the

elevated aerosol layers. The number of low visibility events in meteorological sites exhibit drastic annual changes in winter (Cao *et al* 2015), which is consistent with the large yearly spatial variations of major haze areas. Different from the slow growth in low visibility events near the surface, the annual frequency of regional dense haze layers varies largely in northern China in summer, indicating that elevated aerosols are more sensitive to changes of the atmospheric circulation. Despite of the abrupt large decrease in 2015, variations of regional haze in winter are generally consistent with that of anthropogenic emissions (Lu *et al* 2011). Cloud amount can impact comparability of the haze frequency, but its annual variation is slight (Ma *et al* 2014).

3.3. Potential influence of atmospheric circulation on regional haze in China

Besides changes of anthropogenic emissions, the marked interannual variations of regional haze pollution in northern China can be largely associated with changes in atmospheric circulation. Several studies have shown that variations of the visibility are closely related to atmospheric circulation such as the El Niño Southern Oscillation (ENSO) or the Asian monsoon (Cao *et al* 2015, Li *et al* 2016). However, the strength of such circulation in certain years is uncorrelated with the frequency of hazy days. Different from previous studies that focus on the general relationship between visibility and atmosphere circulation in seasonal scale, here we examine the role of atmospheric circulation by comparing and analyzing meteorological conditions in specific years with large difference in haze frequency.

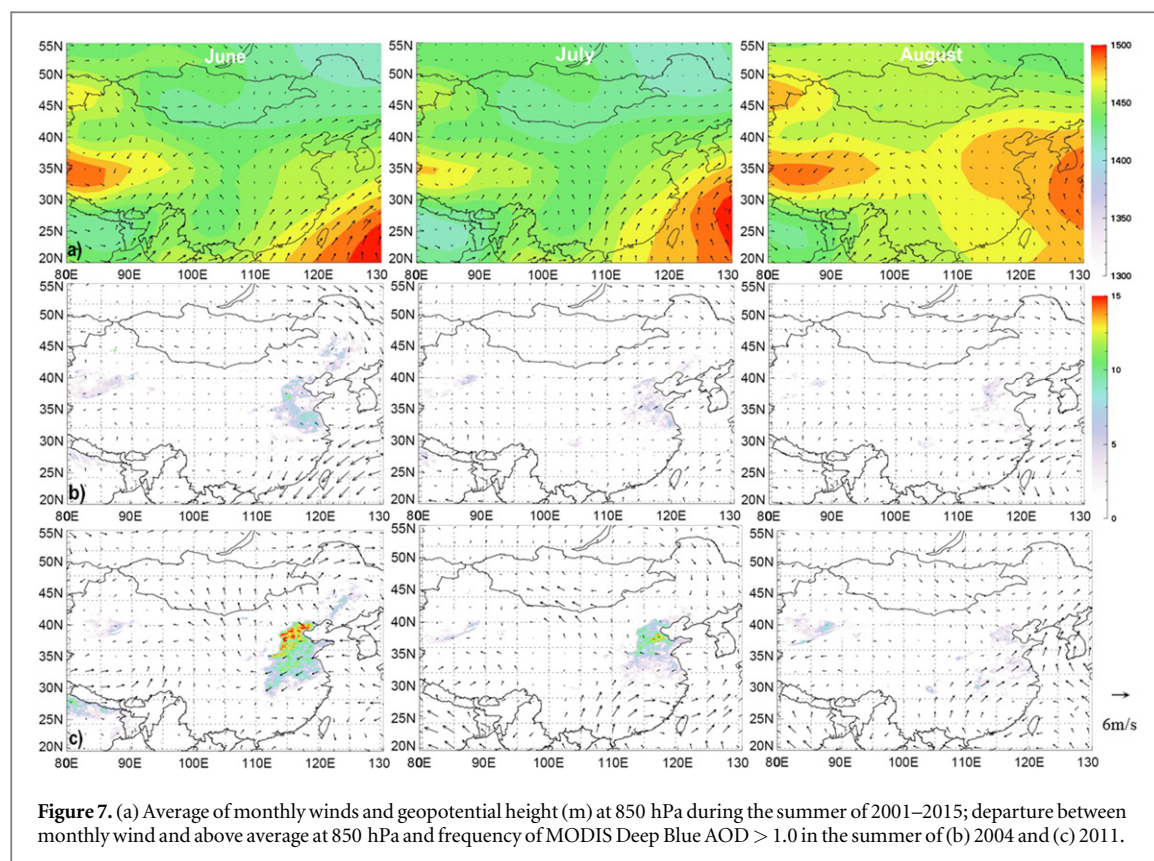


As shown in figure 6(a), dry and cold northwesterly winds prevail in northern China during winter, which can blow the anthropogenic pollutants away. Recent studies emphasize that the weakening of the east Asian winter monsoon favors increase of fog and haze (Qu *et al* 2013, Li *et al* 2016). Regional haze pollution in winter reaches its peak of the last decade in 2014, with haze pollution exceeding 25 days in most areas of northern China (figure 5). By contrast, there is a large decrease in the amount of haze events in 2015. In general, regional circulation gets slower in the winter of 2014 with notable strengthening of southerly airflows in January and February (figure 6(b)). During the same period in 2015, northern winds weaken slightly in northern China in January and become stronger in February. Despite the similar monthly wind fields, there are large differences in the frequency of regional haze. Enhancement of the southwesterly winds in January 2014 is accompanied with more haze events than that of the southeastern direction in February. Though northern winds strengthen in January of both 2014 and 2015, much more haze events occur in northern China when the winds originate from northeastern China rather than northwestern China. Therefore, the influence of atmospheric circulation on regional air pollution is associated with several complex factors besides its strength.

Southerly moist and warm air masses are prevalent in northern China during summer (figure 7(a)). Model simulations suggest that the weakening of the eastern Asian summer monsoon can increase the

aerosol concentration in eastern China (Zhu *et al* 2012). However, satellite observations show that regional aerosol loading increases largely in northern China when southeasterly winds strengthen (figure 7(c)). By comparison, when strong southwestern airflows appear in August 2011, there are no obvious changes in the amount of haze events. Several factors such as changes in direction and position of the monsoon belt can influence the spatial distribution and transport of anthropogenic pollutants. When southeasterly humid airflows from the ocean prevail in northern China, dispersion of regional aerosols and gaseous precursors can be obstructed by the mountains in the north and west of the North China Plain (figure 1(c)). Similar results also exist in 2008 and 2009 (seen in figures 1S and 2S in the supplementary material), indicating that changes in atmospheric circulation have an important contribution in the decrease of haze pollution besides reduction of anthropogenic emissions.

Extensive thick haze layers can linger over northern China for nearly one week before northern or southern winds become strong enough to blow them away (Tao *et al* 2014). Satellite observations show that prevalent dust plumes from deserts in northwestern China can mix with anthropogenic pollutants and cause regional pollution (Tao *et al* 2012). Formation of regional heavy haze can be driven by several factors such as southerly moist airflows and northwesterly dust transport, which varies with seasons and locations. Connection between air pollution and



atmospheric circulation is usually analyzed in yearly or seasonal scales. The factual circulation for one season is composed of numerous processes, which can be very different in conditions such as origin, wind direction, locations of the air masses. When the circulation mainly includes air masses from opposite directions, seasonal or monthly mean value of meteorological data may consider it as stagnant weather even common regional transport exist. To further understand the role of atmospheric circulation in regulating variations of regional haze pollution, more detailed studies are needed.

4. Conclusion

The extensive haze pollution in China has received wide attention due to its marked influence on air quality and public health. By analyzing MODIS Deep Blue AOD in 2000–2015 and associated meteorological data, we provide the first large-scale insight into the trends of the common regional haze pollution in China and their connection with atmospheric circulation. Frequent regional haze pollution is found in northern and central China. During the last decade, regional haze pollution in northern China generally increases from 30–50 to 80–90 days, which almost accounts for half of the cloud-free days. However, there is a decrease in the amount of haze pollution from 2007 to 2009 due to reductions in anthropogenic emissions and changes of meteorological conditions.

In addition, a sudden large decrease appears in 2015, which can be mostly attributed to variations of atmospheric circulation.

Compared with the annual frequency, regional haze pollution in winter and summer exhibits much larger temporal and spatial variations. The frequency of regional winter haze pollution reaches peak in 2014 with a sudden decline in 2015 due partly to the strengthening of cold and clean northern winds. Despite similar trends with the annual amount, major haze areas of the winter haze pollution has notable spatial changes caused by regional transport. By contrast, frequency of the summer haze pollution has notable temporal variations. Seasonal variations of the amount of regional haze events exhibit sensitive response to changes in atmosphere circulation, especially in summer. Our results demonstrate that direction and position of the atmospheric circulation can also play an important role in its influence on regional air quality besides its strengths.

Acknowledgments

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