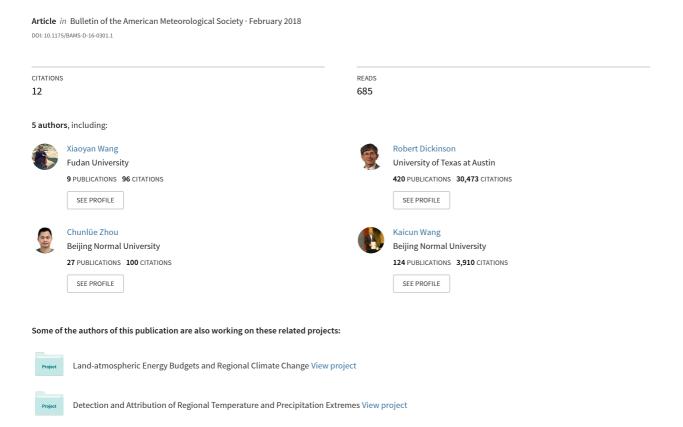
# PM 2.5 Pollution in China and How It Has Been Exacerbated by Terrain and Meteorological Conditions



# PM<sub>2.5</sub> POLLUTION IN CHINA AND HOW IT HAS BEEN **EXACERBATED BY TERRAIN** AND METEOROLOGICAL **CONDITIONS**

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Frequent air stagnation events due to the interaction of meteorological conditions and terrain in China exacerbate its severe air pollution.

ecause of its dense population and rapid industrial development, China has become extremely polluted, a focus of public concern (Ding and Liu 2014; Qiu 2014; Xin et al. 2016b; Zhang et al. 2012). A good metric for this pollution is  $PM_{2.5}$  (i.e., the mass concentration of atmospheric particles with

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diameters less than 2.5  $\mu$ m). These particles have received considerable attention owing to their effects on human health and public welfare (Jerrett 2015; Lelieveld et al. 2015; Tai et al. 2010). In addition, they are efficient in scattering and absorbing solar radiation, resulting in a reduction of atmospheric visibility (Li et al. 2016; Wang et al. 2009; Wang et al. 2015) and significant climatic impacts (Che et al. 2015; Li et al. 2016; Liao et al. 2015; Xia 2015).

Extensive studies have been carried out to establish the severity and frequency of recent air pollution in China. The increase of emissions with rapid urbanization and economic development are generally considered as the primary reason for the increase of polluted days in China (Wang and Chen 2016). However, the observed air pollution has significant interannual and decadal variability, although the increase of the local total energy consumption has been persistent and rapid (Li et al. 2016; Wang et al. 2016).

Evidently, these variabilities must depend on the effects of meteorological conditions (Mao et al. 2016; Xu et al. 2015). For example, Zhang et al. (2016) show that the reduction of horizontal advection and boundary layer height due to the shallow East Asia trough and weak Siberian high have increased the frequency of air pollution over the Beijing–Tianjin–Hebei region in recent years. Wang and Chen (2016) show that the decline of Arctic sea ice extent intensified the haze pollution of central north China after 2000. Other studies have demonstrated that the recent weakening of the East Asia winter monsoon, reduction of wind shear of horizontal zonal winds, increase of humidity, decrease of precipitation, anomalous stabilization in the lower troposphere, increase of calmer winds, and the decrease of cold-air outbreaks in wintertime have also contributed to the increase in episodes of pollution over eastern China (Chen and Wang 2015; Ding and Liu 2014; Li et al. 2016; Niu et al. 2010; Zhang et al. 2015; Zhang et al. 2016).

The formation mechanisms of specific cases of heavy air pollution in recent years over China have been thoroughly studied (Mu and Zhang 2014; Wang et al. 2014; Wang et al. 2014; Zhang et al. 2014), these studies indicating that the formation of severe air pollution episodes in northern China is closely linked with the high emissions, large secondary particulate matter (PM) formation, effective transport of air masses from highly polluted areas, and unfavorable and stagnant meteorological conditions (Bi et al. 2014; Guo et al. 2014; He et al. 2014; Zhang et al. 2016). It has been estimated that secondary aerosol formation contributed 30%-77% of PM<sub>2.5</sub> in episodes of severe haze pollution in northern China (Huang et al. 2014). Sun et al. (2016a) emphasized the roles of regional transport and downward mixing during the formation and evolution of a haze episode in winter 2015. Zhang et al. (2014) found that atmospheric dynamic and thermodynamic factors could explain two-thirds of the evolution of daily haze over eastern China in 2013. Stable anticyclonic synoptic conditions at the surface, leading to low boundary layer height, dominated the formation and evolution of the haze episode that occurred in Beijing in 2011 (Liu et al. 2013). Moreover, some feedbacks between atmospheric boundary layer processes and air pollutants have also been evaluated to estimate their contribution to the haze evolution (Gao et al. 2015; Leng et al. 2016; Ye et al. 2016).

In sum, air pollutant emission is the initial cause of air pollution episodes, but their daily fluctuations and evolution are exacerbated by unfavorable meteorological conditions. The recent severe air pollution in China has generally been attributed to its higher emissions compared to those of developed countries, but a quantitative evaluation of the differences of the capability of the atmospheric dispersion to remove pollutants between China and other countries has been lacking. Such effects of meteorological factors

on the variation of air quality should be considered in evaluation of the effect of emission reduction and development of further mitigation policies. The quantitative assessment of the effects of atmospheric conditions on air quality over the whole country has been far from sufficient.

The Chinese government has made tremendous efforts to control air pollution and improve air quality but mostly by emission reduction at its source (State Council 2013). In January 2013, China began to deploy instruments to measure  $PM_{2.5}$  nationally and released hourly observational data to the public, giving us an opportunity to investigate the effect of meteorological conditions on air quality on a countrywide basis. Using this unique dataset, we have addressed the following questions:

- 1) What are the PM<sub>2.5</sub> concentrations in major cities of China, and how do these compare to those in Europe and the United States?
- 2) How can the atmospheric dispersion capability for the air pollutants be evaluated? And what is the difference of this capability between China and developed countries?
- 3) How does the terrain interact with meteorological conditions to affect the air quality in China?

**DATASET.** The hourly observational PM<sub>2.5</sub> concentration data released since 2013 was obtained from the website of the Ministry of Environmental Protection (http://106.37.208.233:20035) (He et al. 2016; Zhang and Cao 2015). Because the observational system is under development and improving gradually, the duration of available datasets is different for each station. This study uses only the 512 stations that have had more than two years of valid data.

Hourly mass concentrations of PM<sub>2.5</sub> data in the United States and Europe were also collected for the comparisons of this study. The European PM<sub>2.5</sub> database was downloaded from AirBase, an air quality database maintained by the European Environment Agency (EEA) through its European topic center on air pollution and climate change mitigation. It contains metainformation on those monitoring networks involved, their stations, and their measurements. This database covers geographically all European Union (EU) member states, the EEA member countries, and some EEA collaborating countries.

The AirData website gives access to hourly PM<sub>2.5</sub> data collected at outdoor monitors across the United States and contains ambient air quality data collected by the Environmental Protection Agency and state and local air pollution control agencies from thousands

of monitoring stations. Its data come primarily from the air quality system (AQS) database and contain information about each monitoring station and data quality assurance/quality control information.

All the hourly PM<sub>2.5</sub> concentration data were averaged for longer periods (i.e., daily, monthly, and annual) with missing data no more than 40% in each step. Only stations with effective PM<sub>2.5</sub> data for at least two years and at least three months for the warm or cold seasons, respectively, were included (i.e., 271 stations in the United States and 549 in Europe were used); their spatial distribution and data duration are given in Fig. ES1 (see https://doi.org/10.1175/BAMS -D-16-0301.2).

European Centre for Medium-Range Weather Forecasts (ECMWF) interim reanalysis (ERA-Interim) four-times-daily wind speed and boundary layer height (gridded at  $0.25^{\circ} \times 0.25^{\circ}$ ) were bilinearly interpolated to the PM<sub>2.5</sub> stations. Station-based daily observed synoptic phenomena from the U.S. National Climatic Data Center [NCDC, now known as National Centers for Environmental Information (NCEI)] Global Surface Summary of the Day (GSOD) database were used to define the occurrence of precipitation (i.e., the occurrence of either rain, hail, or snow), rather than the daily precipitation amount because of the large fraction of missing data for the latter in China. Each PM<sub>2.5</sub> station was paired to the nearest available weather station. Daily records of the Global Precipitation Climatology Project (GPCP;  $1^{\circ} \times 1^{\circ}$ ) were used in all grid-based figures.

**RESULTS.** Comparison of the annual PM<sub>2.5</sub> concentrations in China with those in the United States and Europe. The annual mean average of PM<sub>2.5</sub> concentrations in China for the period examined is 61  $\mu$ g m<sup>-3</sup>, compared to 16 µg m<sup>-3</sup> for Europe and 10  $\mu$ g m<sup>-3</sup> for the United States. The annual lowest level at which total cardiopulmonary and lung cancer mortality have been shown to increase with more than 95% confidence in response to long-term exposure to PM<sub>2.5</sub> is  $10 \,\mu g \, m^{-3}$  according to the air quality guideline (AQG) of the World Health Organization (World

Health Organization 2014). As shown in Fig. 1, the annual PM<sub>2.5</sub> level at 116 of the 271 U.S. stations (43%) and 96 of the 549 Europe stations (12%) are less than this level, but none of the 512 Chinese stations meet this criterion, and only 9% of them satisfy even the weaker Interim target-1 level (i.e., an annual PM<sub>2.5</sub> of 35  $\mu$ g m<sup>-3</sup>).

The spatial and seasonal differences of PM<sub>2.5</sub> in the United States are small, with relatively high concentrations over the Southeast in summer (as shown in Fig. 2). Most of the stations in Europe had higher PM<sub>2.5</sub> concentrations in winter than in other seasons, but their averages for all seasons were less than  $25 \,\mu \mathrm{g m^{-3}}$ with the exception of those of the Czech Republic, Slovakia, and Poland. Significant seasonal variation of PM<sub>2.5</sub> concentration occurred over China, with its highest concentration of  $86 \pm 28 \,\mu\mathrm{g}$  m<sup>-3</sup> in winter and lowest of  $43 \pm 16 \,\mu \text{g m}^{-3}$  in summer. There is a distinct contrast in concentrations between those of China's coastal area and of its central-northern inland region, in particular those of Beijing, Tianjin, and Hebei Province [i.e., the Jing-Jin-Ji area (J3)] and the Sichuan basin (marked in Fig. 2 and Fig. ES1). The annualmean PM<sub>2.5</sub> concentration in J3 is 87 μg m<sup>-3</sup> (i.e., approximately 1.5 times that of the national mean).

Relationship of annual PM<sub>2.5</sub> concentrations to annual polluted days. The U.S. Environmental Protection Agency has proposed an air quality index (AQI) to report daily air quality in terms of its effects on human health; that is, air quality is classified into

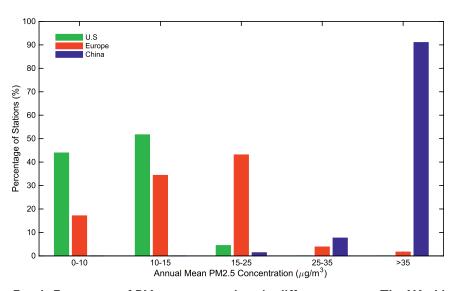


Fig. 1. Frequency of PM<sub>2.5</sub> concentrations in different ranges. The World Health Organization AQG for annual PM<sub>2.5</sub> is 10  $\mu$ g m<sup>-3</sup> and interim target I (IT-1) is 35  $\mu$ g m<sup>-3</sup>, adding an about 15% higher long-term mortality risk relative to the AQG level, IT-2 is 25  $\mu$ g m<sup>-3</sup>, and IT-3 is 15  $\mu$ g m<sup>-3</sup>, adding a 9% and 3% risk level comparing to the AQG level, respectively.

six categories, from good to hazardous (cf. Table ES1 for more detailed information). We further summarize these categories as two classes: unpolluted (good or moderate quality) and polluted (unhealthy to sensitive groups, unhealthy, very unhealthy, and hazardous) conditions; Fig. 3 shows that 97% of the stations in Europe and 98% in the United States have more than 200 days that are unpolluted during a year, while 78% of the stations in China have more than 200 days that are polluted.

We calculate the sensitivity of the total number of annual polluted days to the annual-mean  $PM_{2.5}$  concentrations. Figure 3 shows the number of annual unpolluted (good, moderate, and their sum) and polluted days (i.e.,  $PM_{2.5}$  concentration > 35.5  $\mu$ g m<sup>-3</sup>) versus annual-mean  $PM_{2.5}$  concentrations. The latter (Fig. 3d) is a "sigmoid" curve. When its annual  $PM_{2.5}$ 

concentrations exceed about  $80 \,\mu g \, m^{-3}$  level, a station is polluted nearly the entire year, and so there can be little further increase of its annual number of polluted days. In other words, a slight decrease of the annual-mean PM<sub>2.5</sub> concentrations does little to decrease the annual number of polluted days for the currently highly polluted cities. This concept is quantified by the derivative of the sigmoid curve (shown in Fig. 3d) that indicates the sensitivity of the number of annual polluted days to the annual-mean pollution.

China's State Council released its Air Pollution

China's State Council released its Air Pollution Prevention and Control Action Plan on 12 September 2013, which set the road map for air pollution and control for the next five years in China with a focus on three key regions: the J3 area, the Yangtze River delta and the Pearl River delta, requiring these regions to reduce their atmospheric levels of PM<sub>2.5</sub> by 25%, 20%,

and 15%, respectively, by the year 2017 (State Council 2013). However, since most stations in China are located in the insensitive range, especially those in J3, with annual PM<sub>2.5</sub> concentration exceeding 80  $\mu$ g m<sup>-3</sup>, such air quality control as planned cannot significantly reduce the number of polluted days during a year. The "sigmoid" curve in Fig. 3 shows that a reduction of 25% of the current PM<sub>2.5</sub> annual-mean concentration would decrease the current annual polluted days by 42%, 21%, 6%, and 1% if the current annual PM<sub>2.5</sub> concentrations were 40, 60, 80, and 100  $\mu$ g m<sup>-3</sup>, respectively (based on the fitting equation used for Fig. 3d; cf. its legend).

Figure ES2 shows the number of annual polluted days at China's stations that would occur if their current annual-mean PM<sub>2.5</sub> concentrations were reduced by 15%, 20%, and 25%, respectively. It shows that this number of days for most of inland highly polluted stations is not

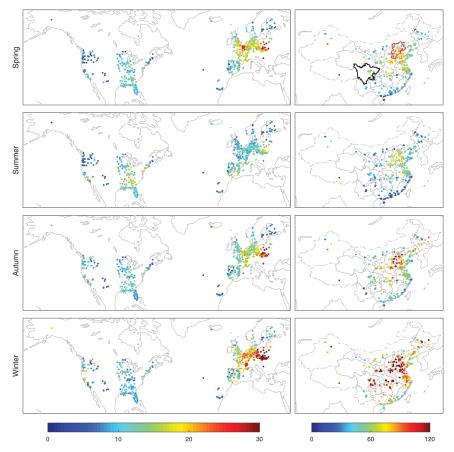


Fig. 2. Seasonal-mean PM<sub>2.5</sub> mass concentration (left) in Europe and the United States (20°-70°N, 170°W-40°E) and (right) in China (18°-55°N, 72°-136°E) ( $\mu$ g m<sup>-3</sup>). Different color bar scales were used for the United States/Europe and China to show the regional distribution of PM<sub>2.5</sub> concentrations more clearly. The data duration of each station is given in Fig. ESI. The months Mar-May were defined as spring in this study. Regional averages of seasonal-mean PM<sub>2.5</sub> concentrations are shown in Table 1. The red and black lines in China indicate the location of Jing-Jin-Ji region and Sichuan basin, respectively.

sensitive to such reduction of annual-mean PM<sub>2.5</sub> concentrations. The annual polluted days over the J3 region would decrease by less than 10% if its current annual PM<sub>2.5</sub> concentrations were reduced by 25%.

Evaluation of the atmospheric dispersion capability. PM<sub>2.5</sub> variations with METEOROLOGICAL CONDITIONS DURING THE 2014 APEC. Evidently, a significant improvement of air quality in China would require more extreme controls than have been planned. For example, in preparation for the Asia-Pacific Economic Cooperation (APEC) summit in Beijing during 10-12 November 2014, the Chinese government applied special controls to improve air quality for the period 1-12 November. The results of this emis-

sions reduction achieved the desired reduction of pollution, with the shade of the clear skies being dubbed "APEC blue." The APEC blue period was ideal for assessing the effect of emission reduction. The air pollutant concentration in Beijing decreased by 60% compared to its monthly mean value before and after the emission reduction period (Tang et al. 2015). The decreases of local emission and regional transport suppressed the formation of secondary aerosols (Chen et al. 2015; Sun et al. 2016b). However, weak pollution episodes occurred on 4-5 and 7-11 November in Beijing during the emission reduction period (Huang et al. 2015; Wang and Dai 2016). Figure ES3 shows the variation of 10-m wind speed (Wsp) and boundary layer height (BLH) during the APEC emission reduction period. Both the Wsp and BLH were lower than normal when the periods of weak pollution occurred. The end of the last one came with a rapid decrease of PM<sub>2.5</sub> concentrations at midnight of 10-11 November over Beijing, Tianjin, and Shijiazhuang accompanied by a sudden increase of wind speed and well-developed boundary layer and a change of wind direction (Sun et al. 2016b). The APEC blue experience shows that such a large

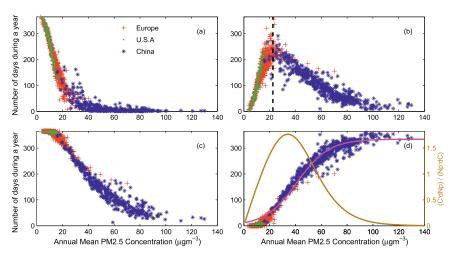


Fig. 3. Scatterplots of station climatological PM<sub>2.5</sub> concentrations vs their annual total number of days in each AQI category: (a) good, (b) moderate, (c) the sum of good and moderate levels (considered as unpolluted air quality), and (d) the sum of levels of unhealthy to sensitive groups, unhealthy, very unhealthy, and hazardous (i.e., polluted air quality). The dashed line in (b) indicates the inflection point at which moderate days stop increasing and begin to decrease with further increase of annual-mean PM<sub>2.5</sub> concentrations. The magenta solid line in (d) is the sigmoid fitting of station-mean PM<sub>2.5</sub> concentrations vs annual polluted days [ $N_p = 334.3/(1 + 28.08e^{-0.081C})$ , where  $N_p$  is the number of polluted days in a year and C is the annual-mean PM<sub>2.5</sub> concentrations ( $\mu$ g m<sup>-3</sup>) of each station]. The brown curve, corresponding to the right coordinate, indicates the sensitivity of annual polluted days with the variation of station annual-mean PM<sub>2.5</sub> concentration [(dN/N)/(dC/C) = CdN/NdC; dN is the variation of annual polluted days for a given variation of annual-mean PM<sub>2.5</sub> concentration dC].

emission reduction is highly effective. However, such emission mitigation comes at a cost to the economy so large that it is not sustainable.

THE CONCEPT OF AIR STAGNATION. Emissions from a particular area do not normally change much over a short period, but local meteorological patterns can strongly affect the accumulation, removal, and transport of air pollutants (Tai et al. 2011; Zhang 2017) and thus the day-to-day variation of air pollutants. Evidently, we need to exclude the effect of such meteorological factors on the variation of air quality when evaluating the observed effect of emission reduction for making further mitigation policies. Indeed, major pollution episodes are usually related to the presence of air stagnation; that is, the near-surface circulation is insufficient to disperse accumulated pollutants in the horizontal and vertical directions. The United States (i.e., NOAA/NCEI; www.ncdc.noaa.gov/societal -impacts/air-stagnation/overview) has defined an air stagnation day as occurring when the daily 10-m wind speed is less than 3.2 m s<sup>-1</sup> (so that the nearsurface circulation is insufficient to disperse accumulated air pollutants), the 500-hPa midtropospheric

wind is less than 13 m s<sup>-1</sup> (i.e., a high pressure ridge at 500 hPa, implying weak vertical mixing), and moreover, no precipitation occurred during the day (i.e., excluding the effect of wet deposition) (Horton et al. 2014; Wang and Angell 1999). An air stagnation event indicates that the air stagnation days last for at least three more days. Using this definition, authors have reported higher air pollutant concentrations in the United States on air stagnation days than on days without stagnation (Dawson et al. 2014; Hou and Wu 2016), but we are not aware of any such studies that have used this definition of air stagnation for China.

Figure 4 shows the frequency of weak 10-m and 500-hPa winds (less than the wind speed thresholds) and the frequency of air stagnation events using the definition of NOAA. Air stagnation so defined is nearly absent over the east-central United States and east China during the wintertime, a result of the strong midtropospheric winds over those areas. However, in reality much pollution is observed to occur in China during this time. Why do the NOAA criteria not predict it? If the midtroposphere weak wind threshold was dropped but the other criteria retained, China would have frequent air stagnation conditions at this time.

To interpret this unusual pattern of weak 10-m wind but relatively strong 500-hPa midtropospheric wind over east China in wintertime, the seasonal patterns of the vertical shear of horizontal wind and vertical distribution of wind fields from the surface to midtroposphere are shown in Fig. 5 and Fig. ES4, respectively. Although strong wind shear exists in the upper to midtroposphere (i.e., 700-500 hPa) over east China in winter, which is favorable for the downward transport of momentum, weak wind shear occurs in the lower troposphere (i.e., 925-850 hPa) because of the blocking by large mountains (i.e., the Tibetan Plateau in the west, Yen Mountains, and Taihang Mountains) suppressing the farther downward transport of momentum (Figs. 5 and ES4) and so resulting in both the low 10-m winds and weak vertical mixing. These weak near-surface winds reduce the dispersion of air pollutants in the horizontal direction. In conclusion, because of the poor agreement during winter in China between the occurrence of pollution episodes and NOAA's air stagnation threshold, the NOAA air stagnation definition appears not to be applicable for China.

A NEW APPROACH TO DEFINING AIR STAGNATION CONDITIONS. We propose a new method, seemingly more broadly

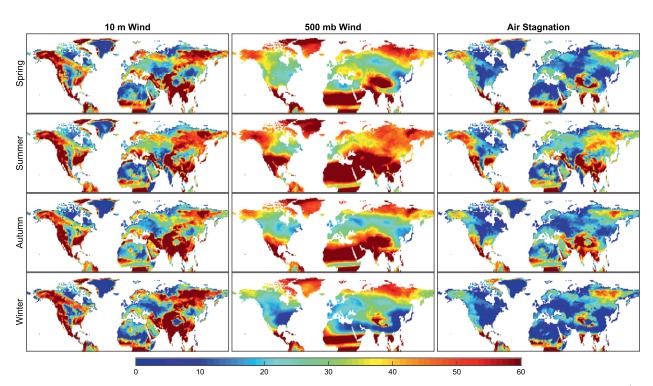


Fig. 4. Climatology of the frequency of (left) no near-surface 10-m wind (10-m wind speed less than 3.2 m s<sup>-1</sup>), (middle) no midtropospheric wind (500-hPa wind speed less than 13 m s<sup>-1</sup>), and air stagnation frequency based on the NOAA air stagnation definition from 2000 to 2014 (%). The presence of no near-surface wind and no midtropospheric wind along with the absence of precipitation was defined as the occurrence of air stagnation. GPCP daily precipitation amounts larger than 1 mm day<sup>-1</sup> were defined as the occurrence of precipitation.

applicable, differing from that of NOAA in its index of downward mixing, to quantify the presence of air stagnation conditions. It retains the 10-m wind speed and the occurrence of precipitation as indices of atmospheric horizontal dispersion capability and wet deposition, respectively. However, it uses the atmospheric BLH rather than midtropospheric winds to indicate the strength of atmospheric vertical mixing. The BLH determines the volume in which the emitted pollutant is dispersed (Wang and Wang 2014, 2016). Extensive studies have been conducted to investigate the effect of BLH on air quality (Tang et al. 2016). To eliminate the effect of seasonal, spatial, and long-term variation of PM<sub>2.5</sub> concentration, monthly means of days without precipitation are determined. Daily PM<sub>2.5</sub> concentrations of those rain-free days are normalized by their current so-defined monthly mean value. For example, for a specific station with the available dataset from 2000 to 2010, the daily PM<sub>2.5</sub> concentrations of days without precipitation during January 2000 were normalized by the mean PM<sub>2.5</sub> concentration of rain-free days in January of 2000 rather than by a monthly mean for January during the whole study period. Such normalized daily PM<sub>2.5</sub> concentrations of all the available stations are divided into different bins based on their contemporaneous daily 10-m wind speed and BLH. Figure 6 shows the average value of the normalized PM<sub>2.5</sub> concentrations for each wind-BLH bin. Only the wind-BLH bands with sample sizes larger than 100 are shown here.

Generally, weak 10-m winds along with shallow boundary layer heights tend to restrict the diffusion of PM<sub>2.5</sub>. The wind and BLH locations, in which the normalized PM<sub>2.5</sub> is closest to 100% in each row, are used to fit an equation of the quantitatively related wind and BLH (i.e., the dashed line in Fig. 6). To the lower-left side of the fitting line occur the wind-BLH conditions with daily PM<sub>2.5</sub> concentrations higher than normal (i.e., conditions of air stagnation as defined). A specific day is defined as an air stagnation day provided it has no precipitation and the wind-BLH are below the fitting line. An air stagnation event is defined as the occurrence of at least three consecutive air stagnation days in this study as in the NOAA definition. Details about the duration of air stagnation events are shown in the online supplement (Fig. ES5).

EVALUATION OF THE NEWLY DEFINED AIR STAGNATION CONDITIONS. This new criteria for an air stagnation event depends on the strength of atmospheric horizontal and vertical dispersion and on the absence of wet deposition. Figure ES6 shows a case of the occurrence of air stagnation events and the variation of PM<sub>2.5</sub> concentrations, indicating that air stagnation events, as just defined, track the daily variation of air pollutants. Figure 7 evaluates the performance of our definition of air stagnation events and measures its influence on the ambient air quality by comparing the relative difference of PM<sub>2.5</sub> concentrations between

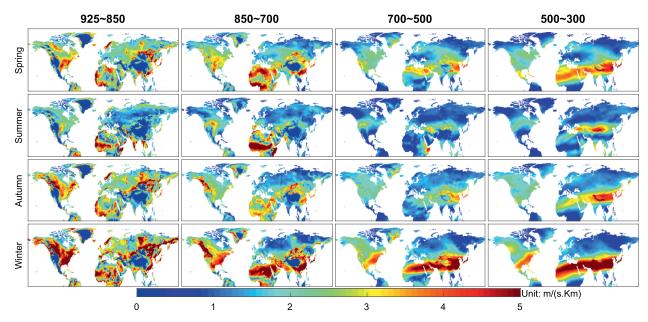


Fig. 5. Seasonal mean of vertical shear of horizontal wind between 925 and 850, 850 and 700, 700 and 500, and 500 and 300 hPa (m s<sup>-1</sup> km<sup>-1</sup>). The 0.5° × 0.5° ERA-Interim monthly wind field and geopotential from 2000 to 2014 were used here. The wind shear was calculated as  $[(u_{z_1} - u_{z_2})^2 + (v_{z_1} - v_{z_2})^2]^{(1/2)}/(z_1 - z_2) \times 1,000$ , where  $u_{z_1}$  indicates the zonal wind at the height of  $z_1$ .

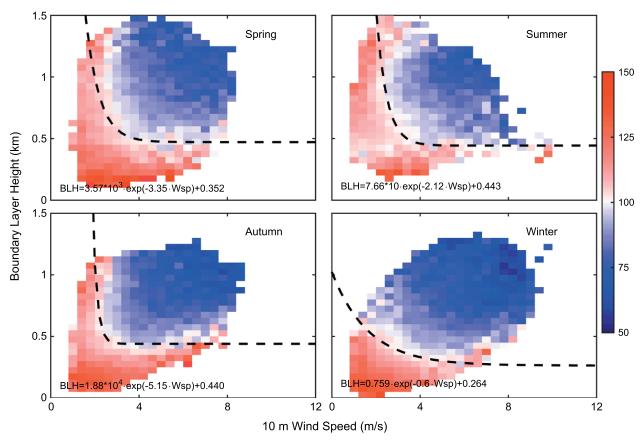


Fig. 6. Seasonal dependence of normalized daily PM<sub>2.5</sub> concentrations on the 10-m wind speed and boundary layer height (unit of normalized PM<sub>2.5</sub> is %). For a specific station, daily PM<sub>2.5</sub> mass concentrations were normalized by its monthly mean to avoid the effect of seasonal, spatial, and long-term variation of PM<sub>2.5</sub>. The dashed lines fitting the wind and BLH at 100% normalized PM<sub>2.5</sub> concentrations were used as the thresholds of air stagnation (unit of BLH is km, and unit of wind is m s<sup>-1</sup>). All the available data in Europe, the United States, and China were used in this figure. Rainy days were excluded in these statistics; that is, the occurrence of precipitation was considered as a no-stagnation condition by default. Grids with sample sizes larger than 100 are shown here.

air stagnation and no-stagnation events (i.e., relative to the latter). It shows that most stations have higher PM<sub>2.5</sub> concentrations during air stagnation events than during no-stagnation events. Air stagnation events have the largest impact on PM<sub>2.5</sub> dispersion in winter (i.e., with PM<sub>2.5</sub> concentrations higher by 46%, 68%, and 60% under air stagnation events for the United States, Europe, and China, respectively), followed by autumn. European PM<sub>2.5</sub> concentrations are most sensitive to air stagnation in winter with a relatively weak effect in summer, possibly due to an increase of sea salt aerosol carried by summer sea breezes or the transport of dust aerosols from the North Africa desert during heavy wind (Megaritis et al. 2014; Moulin et al. 1997; Pey et al. 2013). Air stagnation effects are significant over central and north China during autumn and winter. However, some negative effects appear over northwest China and the central United States in springtime; that is,

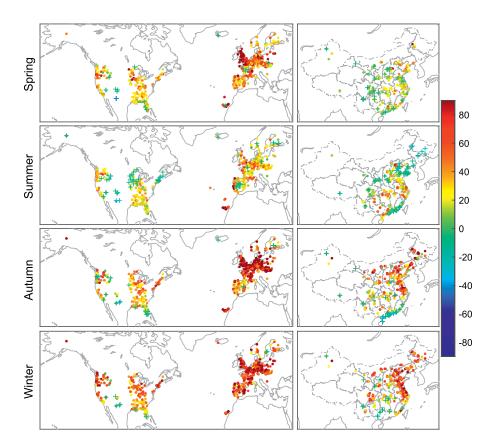
even higher PM<sub>2.5</sub> concentrations occur in the absence of stagnation conditions, possibly from the occurrence of dust storms with strong winds. In the summer, air stagnation effects are small or even reversed, especially over the United States and China (cf. Figs. ES7 and ES8 for details as to reasons).

FREQUENCY OF THE AIR STAGNATION EVENTS. Figure 8 demonstrates the seasonal occurrence of air stagnation events. Europe has the strongest atmospheric dispersion conditions over the study area, with the lowest annual air stagnation frequency of 20%, compared to 24% and 29% in the United States and China, respectively. In the United States, air stagnation events are more frequent in summer and autumn, with a higher air stagnation frequency over the West Coast and Southeast. Europe has more frequent air stagnation events in autumn and winter, 24% and 20%, respectively. Figure 8 shows that a significant northwest–southeast increase

Fig. 7. Seasonal effects of air stagnation events on PM<sub>2.5</sub> mass concentration (%). A day can be defined as an air stagnation day or no-stagnation day based on the wind-BLH thresholds of Fig. 6. Three continuous stagnation days are defined as an air stagnation event. The relative difference of PM<sub>2.5</sub> concentrations between the air stagnation events and no-stagnation events (relative to the latter) was considered as the effect of air stagnation on PM<sub>2.5</sub> dispersion. Regional air stagnation effects are summarized in Table 1. The reason for some negative air stagnation effects occurring in summer is discussed in Figs. ES7 and ES8.

of air stagnation occurs over Europe, with the most frequent stagnation over the Mediterranean basin. The air stagnation frequency shows obvious seasonal and spatial differences over China. The average stagnation frequency is 33% in winter over China, approximately 10% more than in springtime. Strong atmospheric dispersion occurs in the eastern coastal region. Central and north China have strong air stagnation conditions from autumn to winter, especially in the J3 region, with 35% and 42% air stagnation condition in these respective seasons. The Sichuan basin (marked in Fig. ES1) experiences quite frequent air stagnation conditions (more than 40%) over the whole year.

Terrain and meteorological conditions exacerbate  $PM_{2.5}$  pollution in China. As shown in Fig.



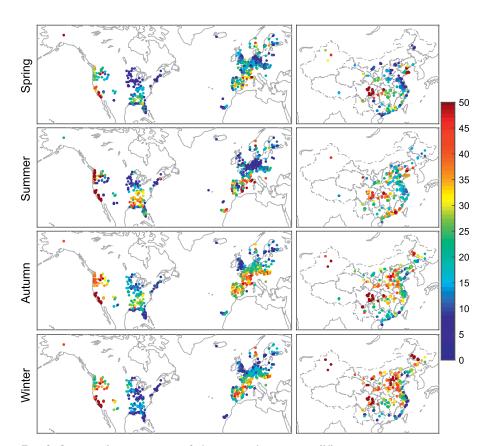


Fig. 8. Seasonal occurrence of air stagnation events (%).

9, the magnitude of 10-m winds is clearly tied to local terrain; for example, weak or calm winds occur over the low elevations of eastern China, the Mediterranean basin, and U.S. West Coast all year-round, while relatively strong 10-m winds occur over the central Great Plains of the United States. High altitudes block surface wind, triggering more frequent air stagnation over basin regions than over the surrounding regions (e.g., the Sichuan basin and Mediterranean basin). Weak winds accompanied by infrequent precipitation lead to frequent stagnation events over the U.S. West Coast, especially in summer. The frequency of precipitation over eastern China has an obvious seasonal difference due to the effect of the Asian monsoon. Perennial lack of near-surface wind in conjunction with the seasonal variation of precipitation and BLH determines the persistent stagnation over central and north China during autumn and winter.

The frequent air stagnation events that occur in China in autumn and winter exacerbate the air pollution that occurs with its high emissions background. If China had the same meteorological dispersion conditions as those of Europe or the United States, what would happen to its air quality? We replaced the air stagnation frequencies of China during autumn and winter by the average of Europe with the following method [the average air stagnation frequency of the United States is almost the same as that of Europe (cf. Table 1); therefore, just the European case is shown here].

The average PM<sub>2.5</sub> concentrations of a specific station are designated as  $C_1$  and  $C_0$  during air stagnation events and their absence, respectively. We applied the average seasonal air stagnation frequency of Europe  $F_{\rm eur}$  (i.e., 24.12% and 19.66% in autumn and winter as shown in Table 1) to each station in China using the following equation:  $C' = C_1 \times F_{\rm eur} + C_0 \times (1 - F_{\rm eur})$ , where C' is what the climatological PM<sub>2.5</sub> concentration of the station would be if its meteorological condition (i.e., air stagnation frequency) was converted into that of Europe.

The relative differences between C' and the original observed PM<sub>2.5</sub> concentrations are shown in

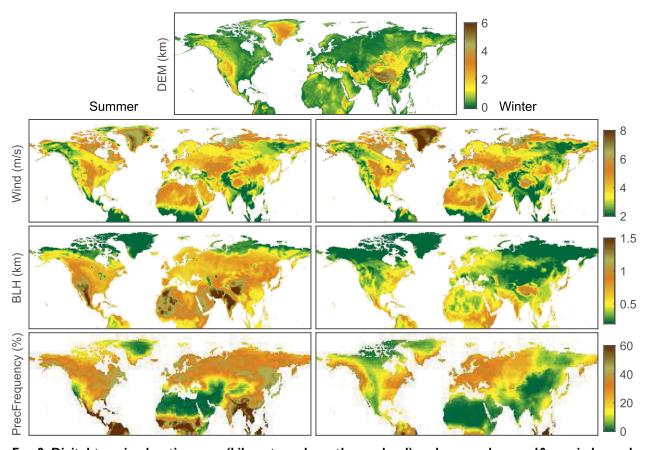


Fig. 9. Digital terrain elevation map (kilometers above the sea level) and seasonal-mean 10-m wind speed, boundary layer height, and the occurrence of precipitation. The terrain data used were from the Global 30 arc s elevation dataset (GTOPO30). The 0.25° × 0.25° daily ERA-Interim 10-m wind and boundary layer height and 1° × 1° GPCP precipitation frequency from 2000 to 2014 were used to illustrate the seasonal frequency of precipitation. A daily total precipitation amount exceeding 1 mm was defined as the occurrence of precipitation.

Fig. 10. They indicate that the local PM<sub>2.5</sub> concentrations would substantially decrease if China had the same air stagnation frequency as Europe, with 67% and 82% of stations having lower autumn and winter PM<sub>2.5</sub> concentrations, respectively. The average PM<sub>2.5</sub> concentrations over the J3 area would decrease by 5.2% and 11.6% during autumn and winter (5.8% and 12.3% for the Sichuan basin).

**DISCUSSION.** Atmospheric conditions on days without precipitation with daily PM<sub>2.5</sub> concentrations higher than their monthly average are defined as the occurrence of air stagnation (i.e., the 100% normalized daily PM<sub>2.5</sub> concentration was taken as the threshold of the presence of air stagnation). But a daily PM<sub>2.5</sub> concentration higher than its monthly average (i.e., air stagnation) does not necessarily mean a polluted day or vice versa. It is quite possible to have an unpolluted day in the United States and Europe with air stagnation; that is, although a daily PM2.5 concentration exceeds its monthly mean, it can still have a low daily PM2.5 concentration because of its

relatively low monthly average, and in China, absence of stagnation, as defined, can occur during a pollution episode. In other words, different thresholds for air stagnation will determine the occurrence of pollution in different locations, depending on the local monthly average of the  $\rm PM_{2.5}$  concentration.

To address this issue, sensitivity tests of the thresholds of air stagnation conditions (i.e., different relative values of  $PM_{2.5}$ ) were carried out as summarized in Fig. ES9 and Table ES2. Figures ES10 and ES11

Table I. Regional average of the seasonal  $PM_{2.5}$  concentrations, the effect of air stagnation on  $PM_{2.5}$ , and the occurrence frequency of air stagnation events.

	Spring	Summer	Autumn	Winter
$PM_{2.5}$ concentration ( $\mu g m^{-3}$ )				
United States	9.67	11.25	10.34	10.65
Europe	16.17	11.65	15.09	19.31
China	54.15	43.39	61.20	83.88
J3	77.77	65.60	92.13	111.60
Sichuan	59.51	40.68	49.63	94.13
Effect of air stagnation events (%)				
United States	31.48	20.04	34.97	46.47
Europe	58.47	37.77	75.08	75.39
China	17.68	17.23	44.33	49.00
Frequency of air stagnation events (%)				
United States	18.88	29.77	26.30	20.63
Europe	17.00	17.61	24.12	19.66
China	23.53	27.04	29.87	33.39
J3	9.67	21.77	35.48	42.16
Sichuan	56.17	39.18	58.72	47.23

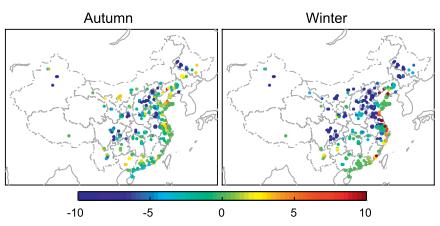


Fig. 10. Relative differences between  $PM_{2.5}$  concentrations that would be expected if China had the same meteorological diffusion conditions as Europe and the actually observed concentrations (percent, relative to the actual observation).

show the occurrence of air stagnation events taking 80% and 120% normalized PM<sub>2.5</sub> as dividing lines to determine occurrence of stagnation, respectively. The 120% threshold might be most appropriate to define pollution for the United States and Europe and 80% for China. The most frequent air stagnation occurs over the western United States, the west coast of Europe, and central China during autumn to winter, consistent with the location of mountainous terrain and consequent high air pollutant

concentrations. Application in autumn and winter of the 80% threshold to China would greatly increase its occurrence of stagnation, and application of the 120% threshold to the United States and Europe correspondingly reduce theirs (cf. Figs. ES10 and ES11). Consequently, the estimated decrease of the autumn and winter  $\rm PM_{2.5}$  concentrations over China shown in Fig. 10 if it had conditions of Europe or the United States would be much larger.

The PM<sub>2.5</sub> producing pollution would be expected to introduce absorptive aerosols to the middle and low troposphere, which would provide a positive feedback on PM<sub>2.5</sub> pollution; that is, they would absorb solar radiation to heat the atmosphere and would reduce the amount of solar radiation reaching the Earth's surface, and so cool the surface (Gong et al. 2014; Petäjä et al. 2016; Xin et al. 2016a). The consequent increase of atmospheric stability would suppress the development of atmospheric boundary layer, and this more shallow boundary layer height would increase the surface PM<sub>2.5</sub> concentrations by compressing their dispersion volume (Wang et al. 2016; Zhu et al. 2016). In addition, enhanced atmospheric stability would impair the downward transport of momentum and lead to weaker dispersion conditions with lowvelocity near-surface winds (Bell et al. 2008; Jacobson and Kaufman 2006; Lin et al. 2015).

This study reports the effect of atmospheric dispersion on the dispersion of air pollutants and estimates how much  $PM_{2.5}$  concentrations are elevated with air stagnation conditions in Europe or the United States. Although fit to mean conditions, its threshold estimates will be somewhat inaccurate on a day-to-day basis since the air pollutant concentrations are significantly different on different air stagnation days (or days without stagnation), and each air stagnation period may differ in air temperature, humidity, and wind direction, which will influence the emission, formation, and transport of air pollutants and so increase the uncertainty of our estimation of what  $PM_{2.5}$  concentrations China would have if it had the same air stagnation as Europe.

**CONCLUSIONS.** China has severe pollution, with its annual-mean PM<sub>2.5</sub> concentrations for the year analyzed to be 61  $\mu$ g m<sup>-3</sup> compared to 16 and 10  $\mu$ g m<sup>-3</sup> for Europe and the United States. None of the 512 Chinese observation stations met the PM<sub>2.5</sub> annual quality guideline of the World Health Organization of 10  $\mu$ g m<sup>-3</sup> during the study period. The quantitative threshold of air stagnation conditions proposed by NOAA does not apply to China because it does not consider the effect of terrain on meteorological

conditions. Thus, a new quantitative threshold of air stagnation events is proposed in this study based on the 10-m wind speed, boundary layer height, and the occurrence of precipitation. China has more frequent air stagnation events than the United States and Europe, especially during winter and autumn, during which time the local emission is usually higher than normal.

The Sichuan basin is exposed to the air stagnation conditions for approximately half of the year, compared to 20% in Europe and 24% in the United States. Over the J3 region, 42% of the winter atmospheric conditions are unfavorable for air pollutant dispersion (35% for autumn). If China had the same atmospheric dispersion conditions as those of the United States and Europe, 67% and 82% of stations in China would improve their current air quality to a certain extent during autumn and winter (e.g., by a 12% decrease of PM<sub>2.5</sub> concentrations for J3 and Sichuan basin in wintertime). Though many emission reduction measures have been taken in China, its severe pollution and frequent unfavorable meteorological conditions make this air quality control less effective, so that further controls would be necessary to reach unpolluted conditions. The presence of unfavorable atmospheric diffusion conditions in China should not be neglected when evaluating the effects of emission reduction and developing further mitigation policies.

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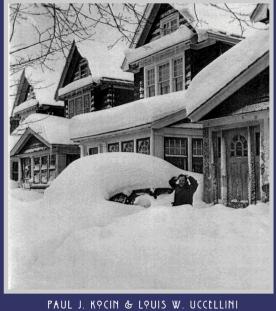






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