Title:

Long-term Changes in Extreme Air Pollution Meteorology and the Implications for Air Quality

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Abstract:

Extreme air pollution meteorological events, such as heat wave, temperature inversion and atmospheric stagnation, can significantly affect air quality. Based on observational data, we have analyzed the long-term evolution of extreme air pollution meteorology on the global scale and their potential impacts on air quality, especially the high pollution episodes. We have identified significant increasing trends for the occurrences of extreme air pollution meteorological events in the past six decades, especially over the continental regions. Statistical analysis combining air quality data and meteorological data further indicates strong sensitivities of air quality (including both average air pollutant concentrations and high pollution episodes) to extreme meteorological events. For example, we find that in the United States, the probability of severe ozone pollution would be enhanced by up to six times when there are heat waves occurring in summer. We have also identified significant seasonal and spatial variations in the in the sensitivity of air quality to extreme air pollution meteorology.

Main Text:

Besides affecting the mean values of various meteorological variables, a critical implication of climate change is to alter the frequency and intensity of a suite of extreme meteorological events [1-7]. Some of these extreme events such as heat wave, temperature inversion and atmospheric stagnation have important implications for atmospheric chemistry and air quality [8-10]. There have been many studies on the potential impacts of climate change on air quality [11-20], but they have generally focused on the impacts associated with the changes in the mean values of air pollution meteorological variables such as temperature, humidity, precipitation, etc. The long-term evolution of extreme air pollution meteorology and the potential impacts on air quality have not been investigated.

We first examine the evolution of extreme air pollution meteorology in the past six decades. We follow the World Meteorological Organization method [21] on the definition of heat waves with some modification - A heat wave is defined when the daily maximum temperature at a given location exceeds the “climatological" daily maximum temperature (averaged over the reference period of 1961-1990) by at least 5 K for more than two consecutive days. Fig. 1a shows the average annual occurrences of heat wave in the first 30-year (1951-1980) period as well as the changes when compared with the more recent 30-year (1981-2010) period. Significant increases in heat waves in the more recent decades are observed over most continental regions, especially the high latitude regions. The annual average frequency of heat wave for the global non-polar continental regions is found to increase by 25.8±3.3% (Table 1). The largest increases (around 40%) are found during Northern Hemisphere spring (March-May) and summer (June-August) seasons.

For temperature inversion, we examine the atmospheric temperature profile below 800 hPa which is most relevant for air quality. A temperature inversion event is defined when the temperature at a higher level is at least 0.1 K higher than the temperature below. On the global scale, a general increase in the occurrences of temperature inversion is found, except over the high latitudes (Fig. 1b). A warmer climate is expected to increase the evapotranspiration, releasing more latent heat in the upper troposphere and hence reducing the temperature lapse rate in the troposphere. As a consequence, the stability of the atmosphere is expected to increase with climate change leading to more temperature inversions. The decreases in temperature inversion over polar regions reflect the strong surface warming there in the past decades, partly driven by the positive feedback associated with snow/ice albedo [22]. For non-polar continental regions in the Northern Hemisphere, the trends in temperature inversion events show clear seasonal variations: the strongest increases are observed in summer (by 17.4±7.0%) while little changes are found in winter.

The definition of atmospheric stagnation used in this study follows the National Climatic Data Center (NCDC) methodology [23] with a relative threshold to focus on the local changes: A stagnation event is defined when the 10 m wind speed, 500 hPa wind speed, and precipitation at a given location are all less than their climatological values for the reference period (1961-1990) by at least 20%. Fig. 1c shows that the occurrences of atmospheric stagnation have increased over most continental areas. For non-polar continental regions, the annual average atmospheric stagnation events have increased by 4.5±0.8%. This increase is partly due to the weakening of surface winds driven by climate change [24]. In addition, the more intense but less frequent precipitation in a warmer climate could also contribute to the increased frequency of atmospheric stagnation events [25].

To examine the impacts on air quality from each specific extreme meteorological event (heat wave, temperature inversion or atmospheric stagnation), we first compare the average air quality on “event days” with those on “non-event days”. The statistical significance of the differences between these two groups are evaluated with t-tests with a 95% confidence interval. Fig. 2a shows the percentage change of seasonal average afternoon ozone concentrations on days with heat waves compared to those on days without heat waves for each season. The highest sensitivity of surface ozone to heat waves is found during summer and fall. The low sensitivity in winter and spring reflects the weaker photochemical ozone production in those seasons [26,27]. From Fig. 2a we can also see large spatial variations in the sensitivity of ozone to heat waves: The strongest sensitivities are found in the eastern United States and the west coast, where the mixing ratios of afternoon ozone are enhanced by more than 40% on days with heat waves, reflecting the strong emissions of ozone precursors [28] and hence high ozone production there.

The impacts of temperature inversion on seasonal average concentrations of $PM_{2.5}$ are shown in Fig. 2b. The strongest impacts from temperature inversion are observed in winter time with daily average $PM_{2.5}$ concentrations enhanced by 40% or more for large areas in the United States. The impacts are much weaker in summer and fall, mainly limited to the northeast and northwest states. In contrast, significant impacts on $PM_{2.5}$ concentrations associated with atmospheric stagnation are found for all seasons throughout the United States (Fig. 2c), with the largest increases in $PM_{2.5}$

concentrations exceeding 40% over large areas.

We further examine the impacts of extreme air pollution meteorology on the cumulative probability distributions of ozone and $PM_{2.5}$ concentrations (Fig. 3). For each season, the cumulative probability distributions of ozone mixing ratios for days with heat waves were compared with those without heat waves. We can see that heat waves have the greatest impacts on the high end of the distributions, which represent the high ozone pollution episodes. For example, during summer time, the 95th percentile ozone is increased by about 25% while the 50th percentile ozone is only increased by about 19% due to heat waves (Fig. 3a). Similar feature is found for the impacts on $PM_{2.5}$ from temperature inversion and atmospheric stagnation. For winter time, the 95th percentile $PM_{2.5}$ concentration is increased by 65% while the 50th percentile $PM_{2.5}$ concentration only increases by 28% in response to temperature inversion (Fig. 3b). Similarly, atmospheric stagnation is found to have little effects on the low end of $PM_{2.5}$ distributions (which represent the clean conditions) but significant impacts on the high pollution episodes for each season (Fig. 3c).

For a specific air pollutant (i.e. ozone or $PM_{2.5}$), we define the high pollution days as the top 10% most polluted days for each season and examine their sensitivities to various extreme air pollution meteorological events. To better quantify the impacts from extreme events on high pollution episodes and their relative importance, we define an impact factor as the enhancement in the probability of high pollution episodes due to extreme meteorological events (see Materials and Methods section for details). The impact factors for high ozone pollution days in summer associated with the three types of extreme events for each of the 48 contiguous states in the United States are shown in Fig. 4a. We find that heat wave is the most important meteorological event in leading to high ozone pollution days in summer for most areas in the United States. The impact factors for ozone pollution associated with heat waves are particularly high in the eastern United States (such as Louisiana, Alabama and Georgia), with values up to 6, which indicates the probability of severe ozone pollution would be enhanced by up to 6 times when there are heat waves over those areas. The highest impact factors for temperature inversion are found over the eastern United States and the Northwest region, while the highest impact factors for atmospheric stagnation are found over the Midwest.

Fig. 4b shows the impact factors for $PM_{2.5}$ in winter associated with the three types of extreme events. The highest impact factors (up to 1.6) are found for temperature inversion over the western region. The impact factors for atmospheric stagnation are generally higher in the eastern United States, and consistently positive (indicating positive correlation between stagnation and high $PM_{2.5}$ pollution episodes) throughout the United States. In contrast, some negative impact factors are found for heat waves. One likely reason is the decrease of ammonium nitrate (a major component of $PM_{2.5}$ in winter time) at higher temperatures. In addition, during warmer days in winter, there would be less residential biomass burning, which is a major source for aerosols in the Western United States [29]. This could also contribute to the negative correlation between heat waves and $PM_{2.5}$ in winter.

To account for the interactions between different types of extreme meteorological events and their synthetic effects on air quality, we also calculate the impact factors for high pollution days associated with multiple events occurring simultaneously. The impact factors for U.S. high ozone and $PM_{2.5}$ days in different seasons are summarized in Table 2. With the increase in the number of simultaneously occurring extreme events (from 0-3), the probability of high pollution episodes almost always increase (with the notable exception of the winter season). The highest impact factor (3.3) is found for summer ozone associated with the combination of three extreme events. This implies that, on average over the whole United States, the probability of high ozone pollution would be enhanced by more than 3 times when the three extreme events happen at the same time in summer.

Methods

We first examine the evolution of extreme air pollution meteorology in the past six decades based on the National Centers for Environmental Prediction (NCEP) reanalysis dataset [30]. The dataset covers the 1951-2010 period with a horizontal resolution of 2.5° latitude by 2.5° longitude and temporal resolution of 6 hours (http://www.esrl.noaa.gov/psd/). To identify the long-term changes in extreme air pollution meteorology (heat waves, temperature inversion and atmospheric stagnation), we compare the climatological data for extreme events for two 30-yr periods: 1951-1980 vs. 1981-2010.

To quantify the impacts of extreme air pollution meteorology on air quality, we analyze air quality data (focusing on ozone and $PM_{2.5}$) from the U.S. Environmental Protection Agency (EPA) AQS (http://www.epa.gov/airdata/) database for 2001-2010 together with the meteorology data for the same period. The air quality data are processed into the same spatial resolution as the meteorology data (2.5°x2.5°). Daily average concentrations for $PM_{2.5}$ and afternoon (1-4pm local time) concentrations for ozone (derived from hourly ozone data) are used in the analysis.

To compare the relative importance of various extreme air pollution meteorological events in leading to high pollution episodes for various regions, we carry out further analysis focusing on the high pollution days, which are defined as the top 10% most polluted days for that season during the 2001-2010 period at that location. For days with a specific meteorological event (heat wave, temperature inversion, or stagnation) occurring, we calculate the probability of those days falling in the top 10% high pollution days (i.e. having the top 10% highest concentrations for a given pollutant – ozone or $PM_{2.5}$ in this case). This probability (P_{event}) is then compared with the average probability ($\overline{P}$) of a given day during the same season (whether or not it has any extreme meteorological event) falling into the top 10% high pollution days ($\overline{P}$ should be equal to 10% following the definition). We also define an impact factor (I) for a specific meteorological event as

$$I = \frac{P_{event} - \overline{P}}{\overline{P}} \tag{1}$$

where

$$\overline{P} = \frac{\#\ of\ days\ with\ high\ pollution}{\#\ of\ total\ days\ in\ a\ given\ period} = 10\% \tag{2}$$

$$P_{event} = \frac{\#\ of\ days\ with\ both\ high\ pollutoin\ and\ extreme\ meteorological\ event}{\#\ of\ days\ with\ extreme\ meteorological\ event} \tag{3}$$

We use the impact factor to quantify the impacts of extreme air pollution meteorology on high pollution episodes. It clearly shows the changes in the probability of severe air pollution associated with certain extreme air pollution meteorological events. .

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Author Contributions:

S. W. and P. H. designed the study and wrote the manuscript. P. H. performed analysis of the meteorological data and air quality data.

Figure and table:

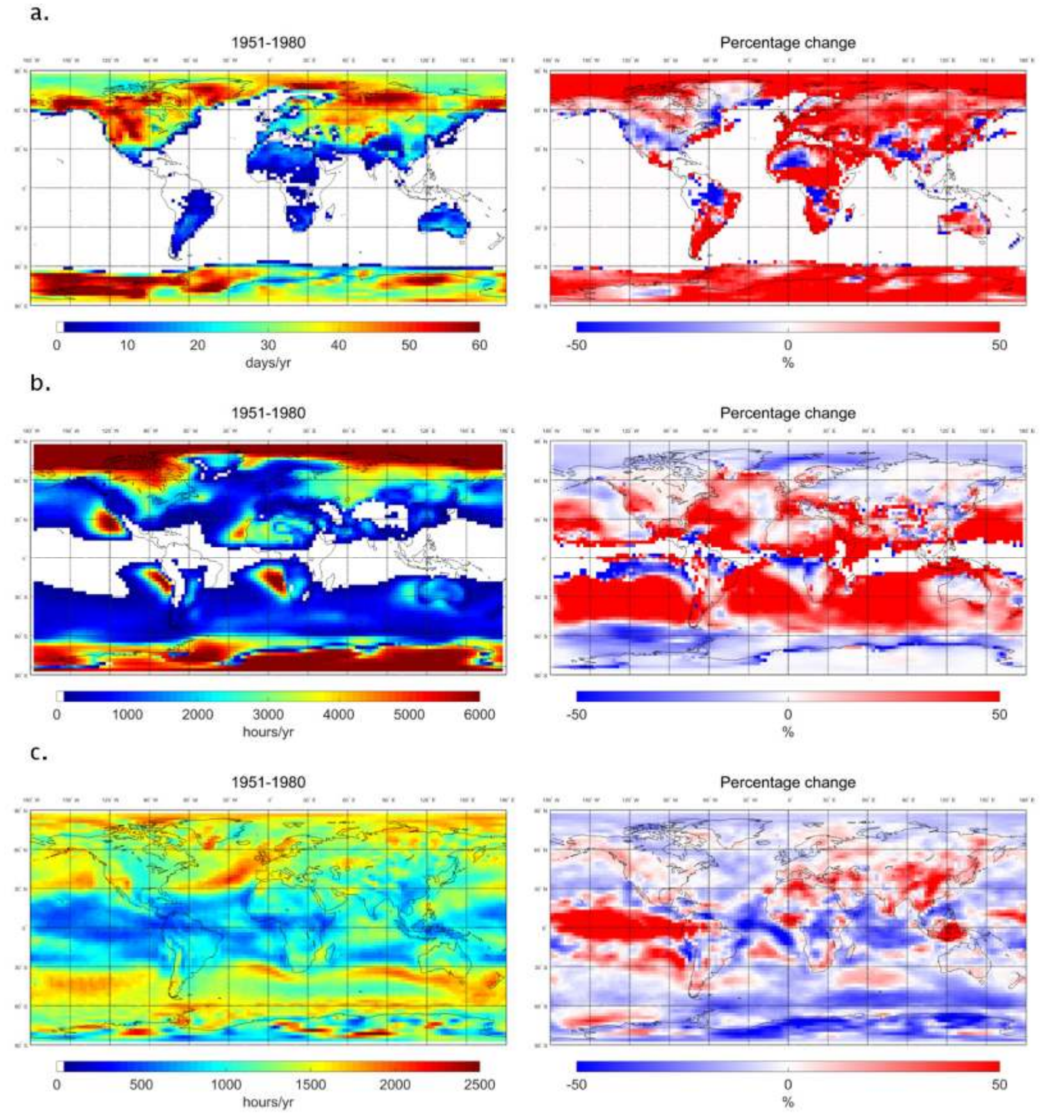

Fig. 1. Changes in the frequency of extreme air pollution meteorological events in the past six decades (based on the NCEP reanalysis data): A. heat wave (days/yr); B. temperature inversion (hrs/yr); C. atmospheric stagnation (hrs/yr). Left: 1951-1980 average; right: percentage change (%) between 1951-1980 and 1981-2010.

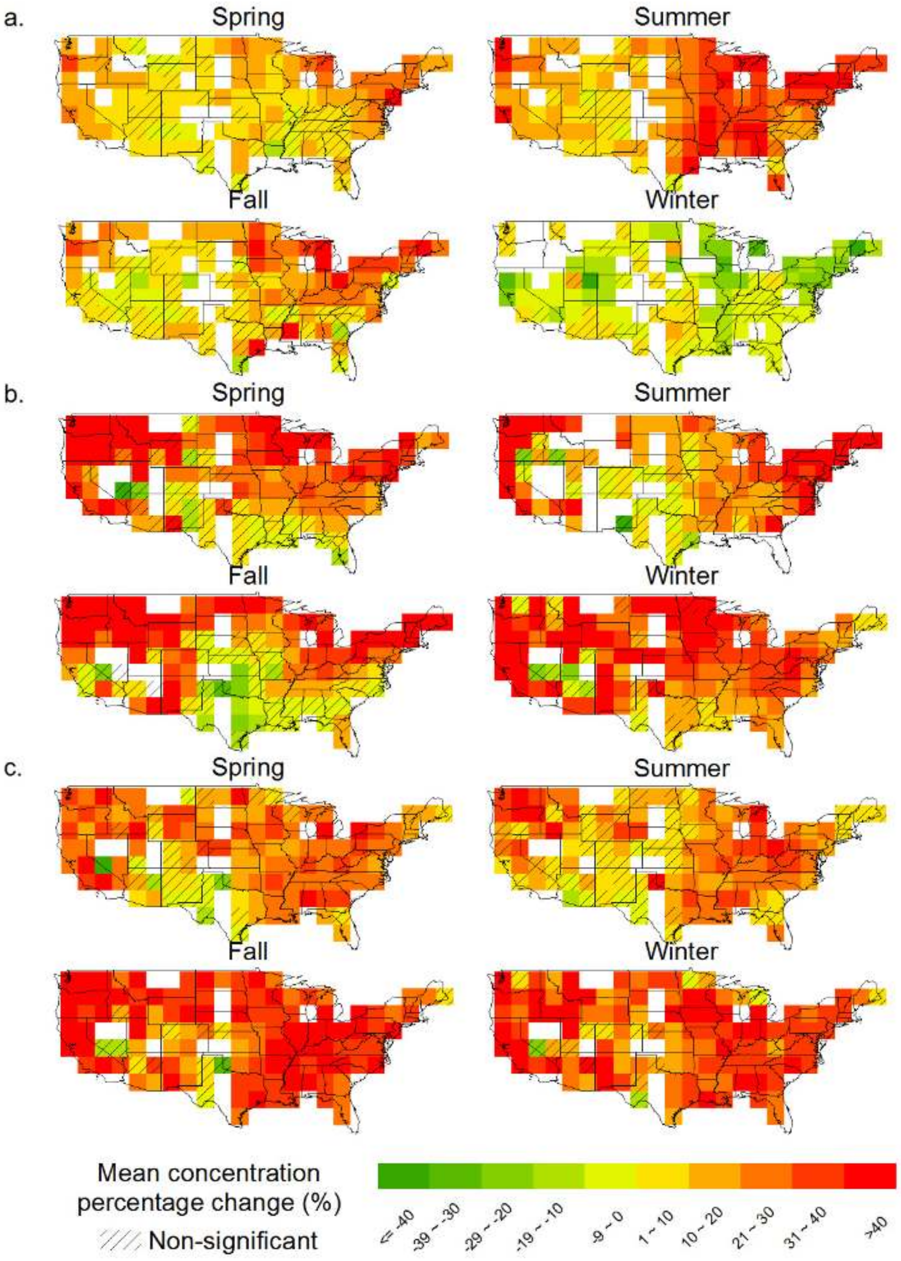

Fig. 2. Enhancement in the mean air pollutant concentrations by extreme meteorological events. Shown as the percentage change (%) of mean concentrations (for either ozone or $PM_{2.5}$) on days with a specific meteorological event (event groups) compared to those on days without event occurrence (non-event groups): A. ozone vs. heat wave; B. $PM_{2.5}$ vs. temperature inversion; C. $PM_{2.5}$ vs. atmospheric stagnation. Shadowed regions indicate that the differences between the two groups are statistically non-significant at the 95% confidence interval. Blank regions indicate those with less than 3 data points for either group.

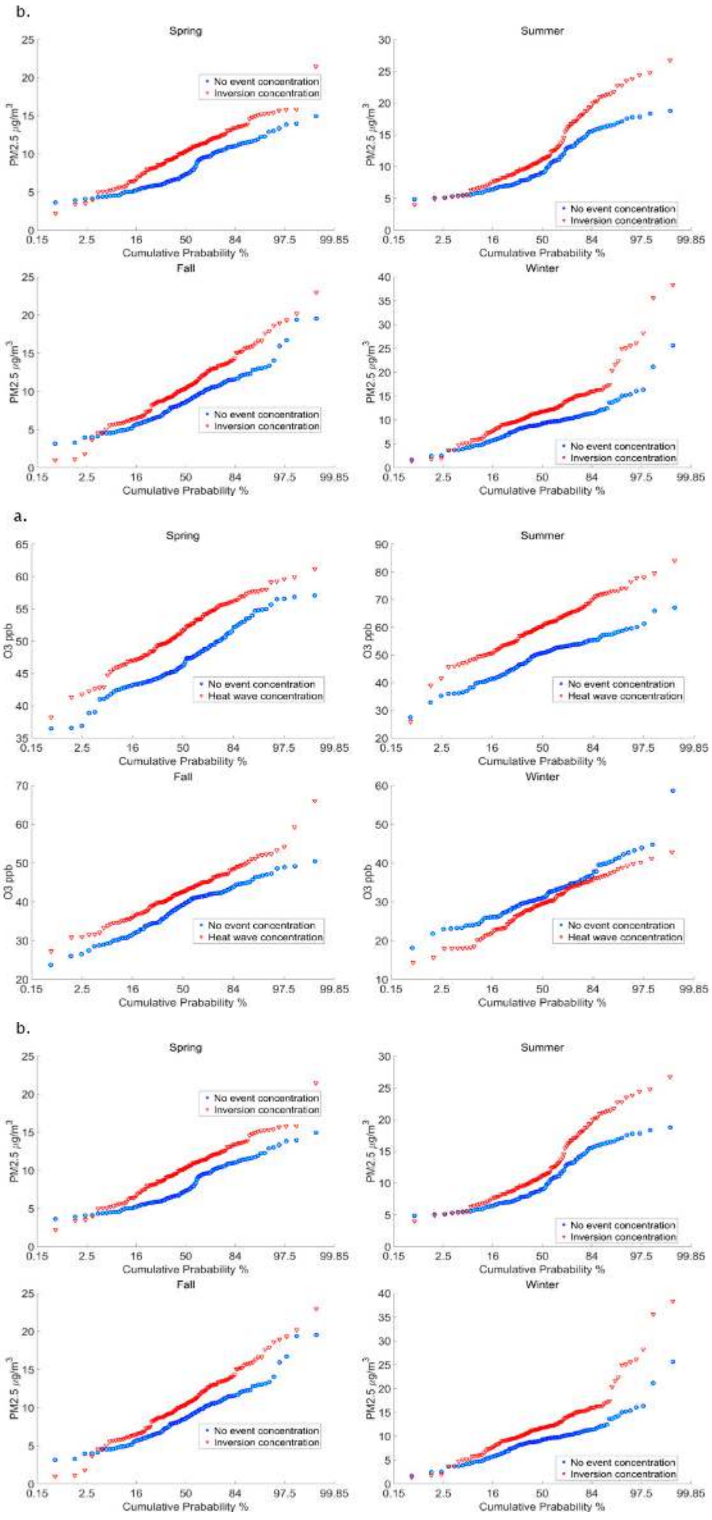

Fig. 3. Cumulative probability plot for concentration of air pollutants. Red triangle: event group; blue circle: no event group. A. ozone mean concentration of heat wave group and no heat wave group; B. $PM_{2.5}$ mean concentration of temperature inversion group and no temperature inversion group; C. $PM_{2.5}$ mean concentration of atmospheric stagnation group and no atmospheric stagnation group.

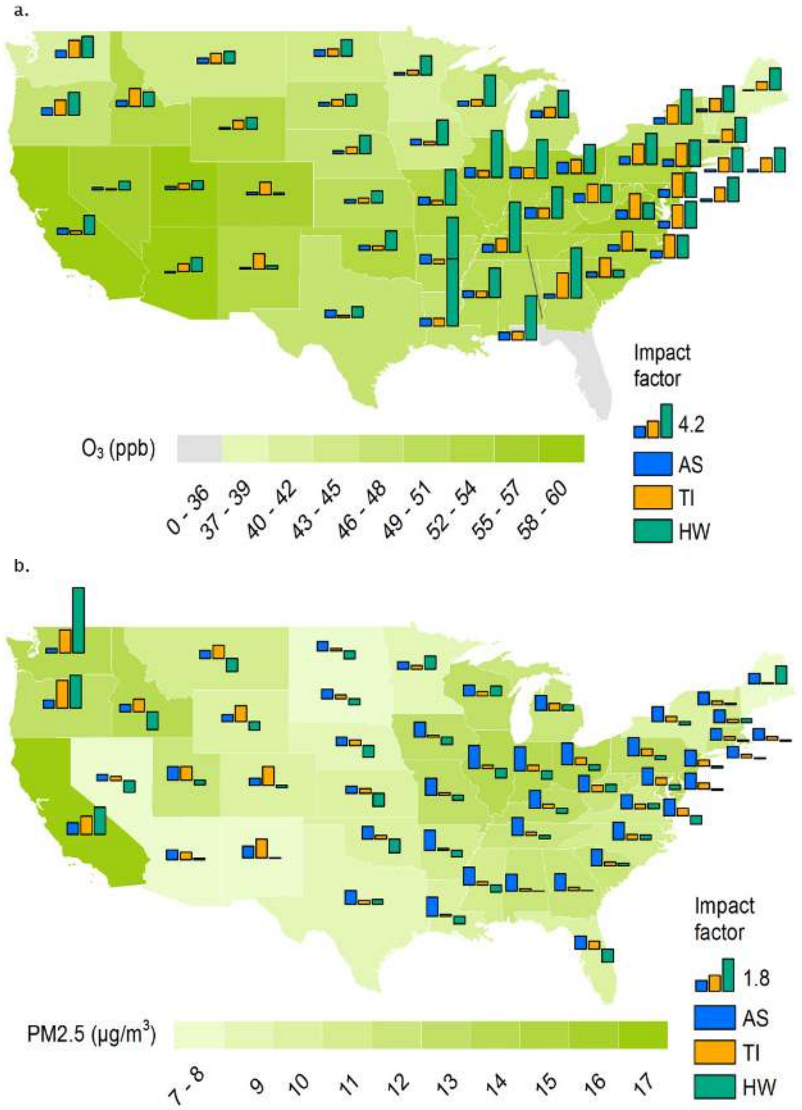

Fig. 4. Enhancement in the probability of high pollution episodes by extreme air pollution meteorological events. Shown as the impact factor for (A) summer ozone and (B) winter $PM_{2.5}$ associated with various meteorological events (heat wave, temperature inversion and atmospheric stagnation; indicated by the green, yellow and blue bars respectively). The impact factor is defined as the enhancement in the probability of high pollution episodes due to extreme meteorological events. Background color indicates the mean concentration for that pollutant.

Table 1. The percentage change ± s.e.m. (%) in the average frequencies of extreme events (HW: heat wave; TI: temperature inversion; AS: atmospheric stagnation) for the global non-polar continental regions between the two 30-yr periods: 1981-2010 vs. 1951-1980. '*' indicates statistically non-significant results at the 95% confidence interval.

Event	Season	Global	Northern Hemisphere
HW	Annual	25.8±3.3	24.5±3.1
	March-May	45.4±4.3	44.9±4.1
	June-August	40.3±5.0	40.9±5.3
	September-November	9.9±3.6	6.5±3.7*
	December-January	17.9±3.5	16.2±3.3
TI	Annual	6.2±3.2	6.7±3.4*
	March-May	9.1±3.9	9.0±4.3
	June-August	10.3±5.3*	17.4±7.0
	September-November	8.8±3.6	10.6±3.9
	December-January	1.8±3.4*	1.6±3.0*
AS	Annual	4.5±0.8	6.8±0.9
	March-May	7.2±0.9	9.8±1.1
	June-August	6.7±1.1	11.8±1.3
	September-November	3.6±0.9	5.5±1.0
	December-January	0.5±0.9*	1.0±1.1*

Table 2. The impact factor for high pollution days (ozone and PM2.5) over the United States associated with various extreme meteorological events (None: no event; AS: only atmospheric stagnation; TI: only temperature inversion; HW: only heat wave; All: three kinds of events happened at the same time). High pollution days are defined as the top 10% most polluted days for each season during 2001-2010. The impact factor is defined as the enhancement in the probability of high pollution episodes due to extreme meteorological events.

Species	Season	None	HW	TI	AS	HW&TI	HW&AS	TI&AS	All
O3	Spring	-0.5	0.1	0.0	0.0	1.2	1.0	1.1	3.0
	Summer	-0.5	1.2	0.4	0.2	3.0	2.1	1.3	3.3
	Fall	-0.5	-0.1	-0.2	0.4	0.8	0.8	0.6	2.1
	Winter	0.1	-0.4	-0.1	0.2	-0.1	0.1	0.1	0.0
$PM_{2.5}$	Spring	-0.4	0.0	0.1	0.2	0.6	0.7	0.8	1.9
	Summer	-0.3	0.8	0.2	0.2	1.9	1.2	0.7	2.5
	Fall	-0.4	0.2	-0.3	0.4	1.0	1.0	0.5	2.0
	Winter	-0.5	-0.7	-0.1	0.2	-0.2	0.3	1.2	0.8