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Intraseasonal variation and future projection of atmospheric diffusion conditions conducive to extreme haze formation over eastern China

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\textbf{ABSTRACT}
Future projection of diffusion conditions associated with extreme haze events over eastern China is of great importance to government emission regulations and public health safety. Here, the diffusion conditions and their changes under future warming scenarios are examined. The relative strength of haze events in the Northern China Plain region increase from 150\% during 2006–15 to 190\% during 2090–99 under RCP8.5 scenarios, induced by a stronger and longer-lasting anticyclone anomaly in eastern China. The strengthened anticyclone anomaly is mainly induced by increased northern wave train convergence emanating from the Barents–Kara Sea, and the longer duration of the anticyclone anomaly is mainly induced by stronger local feedback that can extract more energy from the basic state to maintain the anticyclone anomaly in eastern China. Aerosol reduction is found to play a dominant role in strengthening the upstream wave train near the Barents–Kara Sea and the downstream anticyclone in eastern China, while the effects from increased greenhouse gases are small. The results of this study indicate that future aerosol emissions reduction can induce deteriorating diffusion conditions, suggesting more stringent regulations on aerosol emissions in China are needed to meet air quality standards.

1. Introduction
China has experienced deteriorating air quality since the 1990s due to rapid industrial and economic development (An et al. 2019). Although China has implemented stringent emission reduction measures in recent years, which have indeed reduced the average annual level of fine particle pollution (Zheng et al. 2017), extreme and persistent haze events still occur frequently (Pei et al. 2018), especially under stagnant weather conditions.

Stagnant weather conditions can be influenced by natural climate variability and climate change (Sherman et al. 2019). While the effects of climate variability on haze events have been widely investigated (Wang, Chen, and Liu 2015; Yin and Wang 2018), studies about the effects of climate change on haze pollution in China are still limited and results from different studies sometimes do not agree with each other, especially with respect to the mechanisms that cause extreme haze events. Cai et al. (2017) established a haze weather index relating the daily variability of Beijing PM\textsubscript{2.5} concentrations with the daily variation in weather conditions, including the 850-hPa meridional wind (U\textsubscript{850}), the longitudinal gradient of 500-hPa zonal wind (U\textsubscript{500}), and...
the vertical temperature gradient ($\Delta T$). They found the index shifted to larger values under RCP8.5 scenarios, mainly induced by changes in $U_{500}$ and $\Delta T$. Shen et al. (2018) found the combined first principal component of $V_{850}$ and relative humidity can explain 85% of PM$_{2.5}$ monthly change, while its future changes based on the CMIP5 ensemble of climate models are statistically insignificant.

While all the above studies have mentioned the importance of $V_{850}$ and the associated anticyclone anomaly, future projection of the anticyclone anomaly is uncertain and the dynamic processes associated with the anticyclone and its future projection have not been well investigated. An anticyclone anomaly conducive to haze event formation can be induced by an upstream teleconnection wave train (Yin and Wang 2017). The strength and phase structure of the wave train can change with different backgrounds mean flow and surface conditions (e.g. Arctic sea ice and Eurasian snow) under different external forcing (Wang et al. 2019).

For future climate change, warming is not only induced by increases in greenhouse gases (GHGs), but also by aerosol reduction due to emissions control. Previous studies have mentioned the effects of increased aerosol on the northern mode of the East Asian winter monsoon (EAWM) and Hadley circulation in boreal winter (e.g. Jiang et al. 2017; Liu et al. 2019), but the effects of aerosol reduction on the upstream wave train related with the EAWM southern mode have not been investigated in the context of future climate change. In addition, the duration of extreme haze events can be a period of weeks, which means the intraseasonal signal has a greater impact on their formation. The intraseasonal wave train has not yet been investigated with respect to the formation of haze events.

The present study aims to analyze the change in diffusion conditions conducive to haze events and the associated anticyclone anomaly under the RCP8.5 scenario. The effects from two main external forcings—increased GHGs and aerosol reduction—are examined separately. The intraseasonal dynamic processes relating to the upstream teleconnection (Eurasian wave train), the anticyclone, and haze events are analyzed. In the following sections, the data and methods used in this study are described in Section 2, the change in the anticyclone anomaly and its attribution are analyzed in Section 3, and discussion and conclusions are presented in Section 4.

2. Data and methods

2.1. Model experiments

This study uses the daily model data from the Community Earth System Model Large Ensemble (CESM-LENS) project (Kay et al. 2015). All the ensemble members are carried out with the fully coupled CESM1.2 utilizing the Community Atmosphere Model version 5 (CAM5) and the Community Land Surface Model version 4 (CLM4), both of which are run at approximately 1° resolutions with small differences in the initial atmospheric conditions. All the ensemble members share the same emission scenarios (historical before 2005 and emission scenarios from 2006 to 2100). The RCP8.5 warming scenario is one of the worst emission outcomes, under which warming is induced by increased GHGs, decreased aerosol emissions, and other external drivers, with an increased radiative forcing of about 8.5 W m$^{-2}$ in 2100 relative to pre-industrial conditions (Lamarque et al. 2011). Two additional scenarios (RCP8.5_FixAerosol2005 and RCP8.5_FixGHG2005) are used here to compare with RCP8.5. In the RCP8.5_FixAerosol2005 scenario, all emissions of aerosol and atmospheric oxidants are fixed at the present-day level (2005), but GHGs and other factors follow RCP8.5 (Xu, Lamarque, and Sanderson 2018). In the RCP8.5_FixGHG2005 scenario, GHG emissions are fixed at the present-day level (2005), but aerosol and other factors follow RCP8.5. So, the differences between the 2006–15 ensemble mean and the 2090–99 ensemble mean to represent the climate effects from increased GHGs in RCP8.5_FixAerosol2005 and from decreased aerosol in RCP8.5_FixGHG2005. The seven ensemble members from the RCP8.5 experiments and 14 ensemble members each from the other two scenarios with daily aerosol output are used for our analysis.

The CESM-LENS project has been widely used in analyzing future changes of cold-air outbreaks, extreme precipitation, and drought (Herring et al. 2018; Landgren, Seierstad, and Iversen 2019). However, studies about extreme haze events using this dataset are still limited (Chen et al. 2019). Here, we use the daily outputs from the CESM-LENS project to analyze how extreme haze events change under different external drivers. The model output of the project is freely available and easily accessed from the following website: http://www.cesm.ucar.edu/projects/community-projects/LENS/data-sets.html.

2.2. Definition of extreme haze events

We employ a composite analysis to obtain the common features of haze events during the winters of 2005/06 to 2014/15 and compare it with the winters of 2090/91 to 2099/2100. Since aerosol emissions are reduced by approximately 50% in eastern China in the RCP8.5 scenario, a fixed threshold used in observations defining haze events is not practical in climate models. An objective definition method is used here by considering three aspects of haze events together (strength, regional
impact, and duration) based on the concentration of surface sulfate aerosol, similar to the approach used for defining extreme precipitation events (e.g. Zhao, Deng, and Black 2017). First, for each grid point over eastern China (10°–40°N, 110°–130°E), the 90 daily surface aerosol concentrations for a certain winter are sorted and the top 50% of days are classified as anomalous high-concentration days for this grid point. Second, we require that for any single day the number of grid points with anomalously high sulfate concentrations must exceed 50% of the total number of grid points in eastern China, and only the days fulfilling the above two criteria are defined as haze days. Third, the duration of a haze event is defined as the number of consecutive haze days, and events whose duration is more than 2 days are defined as extreme haze events in CESM. The combination of these three criteria yields 5–6 extreme haze events and around 30 haze days per winter, which is similar to those from observation during 1993 to 2012 with an average of about 30 haze days per winter (Yin, Wang, and Chen 2017). The day 0 of haze events is chosen as the day that has the most extensive areal coverage of haze grid points. The frequency of haze events in the three scenarios is shown in Table S1. The daily anomaly of wind and geopotential height is extracted by removing the annual cycle from the raw data. Based on the haze events defined above, composite anomalies associated with haze events are obtained by averaging daily meteorological anomalies from all identified haze events at individual lead days (from –6 to 6 days), similar to the composite analysis adopted in Zhong, Yin, and Wang (2019).

2.3. Diagnostic tools

A 3D wave-activity flux (WAF, represented by $W$ in the equation below), which can represent the wave energy propagation in accordance with the background flow, is used here to analyze the propagation of Rossby wave trains conducive to haze events:

$$
W = \frac{P \cos \phi}{2|V_H|} \left\{ \frac{u}{\partial \cos \phi / \partial \hat{z}} \left( \left( \frac{\partial u}{\partial \hat{z}} \right)^2 - \psi' \frac{\partial \psi'}{\partial \hat{z}} \right) + \frac{v}{\partial \cos \phi / \partial \hat{z}} \left( \left( \frac{\partial v}{\partial \hat{z}} \right)^2 - \psi' \frac{\partial \psi'}{\partial \hat{z}} \right) \right\}
$$

$$
= \frac{u}{\partial \cos \phi / \partial \hat{z}} \left( \left( \frac{\partial u}{\partial \hat{z}} \right)^2 - \psi' \frac{\partial \psi'}{\partial \hat{z}} \right) + \frac{v}{\partial \cos \phi / \partial \hat{z}} \left( \left( \frac{\partial v}{\partial \hat{z}} \right)^2 - \psi' \frac{\partial \psi'}{\partial \hat{z}} \right)
$$

(1)

where $P$ is pressure coefficient (pressure/1,000 hPa), $a$ is Earth’s radius, and $(\phi, \lambda, z)$ are latitude, longitude, and height, respectively. $V_H = (U, V)$ are the ten-year climatological winter-mean horizontal wind velocity averaged over 2006–15 (before the warming) and 2090–99 (after the warming) for WAF comparison; and $\psi'$ represents small-amplitude geostrophic perturbations on a steady zonally inhomogeneous basic flow (Takaya and Nakamura 2002).

The potential energy conversion rate from mean flow ($P_{\text{bar}}$) to low-frequency eddy ($P_{\text{transient}}$) is defined below to analyze the baroclinic process in the formation of an anticyclone conducive to haze events (Cai et al. 2007):

$$
C(P_{\text{bar}} \to P_{\text{transient}}) = \frac{C_{V/C_{\rho R}}}{2 \left( \frac{-\partial \theta}{\partial \phi} \right) g} (\bar{T} - C_2 \left( \frac{\hat{u} T_{/x} + \hat{v} T_{/y}}{\partial T} \right)).
$$

(2)

Here, $T$ is the winter climatological mean temperature field at 500 hPa and $(\hat{u}, \hat{v}, \hat{T})$ are the low-frequency eddy part of the 500-hPa geostrophic winds and temperature. $i$ and $j$ are unit vectors along $x$- and $y$-axis, respectively. The constant $C_2$ is defined as:

$$
C_2 = \frac{2R/c_p}{g} \frac{C_{V/C_{\rho R}}}{\left( \frac{-\partial \theta}{\partial \phi} \right)}
$$

(3)

Here $R$ is the gas constant, $c_p$ and $C_V$ are the specific heat capacity at the constant pressure and volume and $-\partial \theta / \partial \phi$ is the mean atmospheric static stability in extratropics which has been set to be 3.5 K/100 hPa.

3. Results

Since aerosol emissions decline during the twenty-first century under the RCP scenarios, we assess the effects of climate change on haze events by comparing the percentage strength of haze events between 2006–15 and 2090–99 in three sensitivity experiments. The percentage strength is defined as the ratio of the regional mean (30°– 40°N, 110°–120°E) surface sulfate concentration on extreme haze days to the climatological sulfate concentration over the 10-year periods examined to exclude effects of background emission changes. Sulfate is chosen to represent haze event strength as it accounts for most of the PM$_{2.5}$ surface concentration in the model (Chen et al. 2019). The frequencies of haze events in the RCP8.5 experiment and the other two experiments are shown in Table S1. The frequency increases from 380 events during 2006–15 to 421 events during 2090–99 for seven simulations under RCP8.5. The average relative strength of extreme haze events under RCP8.5 increases from 150% during 2006–15 to 190% during 2090–99, which is statistically significant at the 95% confidence level based on the Student’s $t$-test. A systematic shift towards higher values in the relative strength is evident in Figure 1(a). This shift mainly comes from aerosol reduction (Figure 1(c)) and not from increased GHGs, as a similar shift from 150% to 190% is found in
RCP8.5 fixedGHG and the change is insignificant in RCP8.5 fixedaerosol (Figure 1(b)).

The composite patterns of the circulation anomaly and sulfate concentration of haze events from a lead of −6 to +6 days are shown in Figure 2. The results indicate that the emergence or dissipation of severe haze is associated with the advance or retreat of the anticyclone anomaly, and here the anomaly fields are calculated by removing the annual cycle from the raw data as described in Section 2.2. When severe haze events occur, the EAWM begins to weaken, as indicated by the change from the cyclone to anticyclone anomaly in the East Asian trough from a lead of −6 to 0 days (Figure 2(a–d)). Accompanying the anticyclone anomaly at 500 hPa, the zonal pressure gradients over the southern part of East Asia increase, the southerly wind anomaly at 850 hPa strengthens, the meridional circulation is weakened, and it is harder for cold air to reach the midlatitudes of East Asia. Here, the diffusion conditions are deteriorated by the anticyclone anomaly and the sulfate aerosol center is moving from the Sichuan Basin to the North China Plain (30°–40°N, 110°–120°E) (Figure 2(d,e)).

The circulation pattern is also compared with observation, and a similarly strong anticyclone anomaly is found for haze events, defined using similar criteria as mentioned above, over eastern China, based on observation (PM$_{2.5}$ concentration is used in the observation) (data not shown). A similarly strong anticyclone anomaly also exists for haze events over the Beijing–Tianjin–Hebei region (Zhong, Yin, and Wang 2019) and North China Plain (Yin, Wang, and Chen 2017).

The change in the strength and duration of the anticyclone anomaly under RCP8.5 (Figure 3) is consistent with changes in haze events (Figure 1). By analyzing the intraseasonal 9–29-day anticyclone anomaly change in Figure 3 (d–f), we find the original change is largely contributed by 9–29-day low-frequency eddy change (Jiao, Wu, and Song 2019). The strength of the anticyclone anomaly on the severest haze day is 13 gpm for haze events in 2006–15 but 18 gpm in 2090–99, which is an increase of 40% (Figure 3(d)). The duration of anticyclone anomalies larger than 6 gpm is around 5 days for both 2006–15 and 2090–99, while for those larger than 12 gpm the duration is 3 days for 2006–15 but 4 days for 2090–99 (Figure 3(d)). So, not only does the intensity of the anticyclone anomaly increase, but also the residence time for extreme conditions. All the above changes can only be detected in RCP8.5 fixedGHG, and not in RCP8.5 fixedaerosol, which shows the change is mainly induced by aerosol reduction (Figure 3(c,f)).

The anticyclone anomaly is often linked to upstream atmospheric wave activities (Yin and Wang 2017). The intraseasonal low-frequency (9–29 days) WAF shows that two intraseasonal wave trains propagate energy eastwards and converge to the anticyclone, which is conducive to haze events (Figure 4(a–c)). Results from the original fields are almost the same as those based on the filtered low-frequency fields (not shown), which means the 9–29-day timescale accounts for most of the variation of haze events, consistent with the discussion in the previous paragraph. The northern wave train emanates from the North Atlantic, propagates south-eastward, and finally converges into eastern China along the polar jet, which serves as a waveguide. The southern wave train first converges to the North Indian
Ocean and then it gradually acts as a new Rossby wave source to diverge energy to eastern China along the subtropical Asian jet. After a lead of −4 days for composite haze events, the northern wave train is stronger than the southern wave train and the relative intensity of the northern and southern wave train is related to the relative intensity between the subtropical jet and polar jet.

The positive vertical WAF in eastern China indicates baroclinic energy conversion from the basic state to the anticyclone anomaly, which can maintain its persistence there (Figure 4(d–f)). The temporal features of the available potential energy conversion rate over (30°–50°N, 120°–150°E) indicates that anticyclone anomalies at 500 hPa in East Asia develop ahead of the energy conversion peak. This implies the anticyclone is first induced by an upstream Rossby wave convergence and then the anticyclone anomaly–induced perturbation heat fluxes extract potential energy from the basic state, which makes the anticyclone intensify further and remain in place. The zonal-mean meridional temperature contrast contributes half of the potential energy conversion, followed by the stationary wave–induced meridional contrast, and the

Figure 2. Composite anomalies of the 500-hPa geopotential height (contours), 850-hPa wind fields (vectors), and surface sulfate aerosol concentration (shading; units: µg kg$^{-1}$) at a lead of (a) −6, (b) −4, (c) −2, (d) 0, (e) +2, (f) +4, and (g) +6 days of composite haze events during 2006–15 under RCP8.5. Contour intervals are 8 gpm and negative contour values are dashed. The red rectangle in (a) (30°–50°N, 120°–150°E) is the region averaged for the anticyclone anomaly.
zonal contrast contributes little to the conversion term. In addition, as depicted by the composite jet stream in Figure 4(h) and Eady growth rate in Figure 4(i), the background baroclinicity south of 40°N has been weakened while north of it has been intensified, which manifests as decreased vertical wind shear and more stable atmospheric stratification south of 40°N.

For the future projections, the anticyclone anomaly (30°–50°N, 120°–150°E) in eastern China is intensified by a stronger northern wave train convergence at a lead of −2 days and after (Figure S1(a–c)). The intensified northern Rossby wave train originated from the increased anticyclone anomaly near the Barents–Kara Sea converges more wave energy to eastern China, thus contributing to
the stronger anticyclone and worse diffusion conditions responsible for the larger percentage strength change for the future haze events (Figure 1(a)). The local feedback process is also strengthened by the stronger upstream wave train (Figure S1(d–f)). The upstream stronger northern wave train induces stronger perturbation in heat fluxes and available potential energy conversion in eastern China, which is depicted by the increased upward WAF in that region. The increase of baroclinic energy conversion in Figure S3(a) is induced by the stronger upstream wave train but not by the climatological change of the background horizontal temperature gradient, since the energy conversion rate change calculated from first 10 years or last 10 years of climatological horizontal temperature gradient remain almost the same (Figure not shown). Besides, the intensified jet stream and Eady growth rate anomaly around 40°N (Figure S3(b,c)) also indicates a stronger feedback process there.

The percentage strength changes of haze events and anticyclone anomaly change both indicate the change is

**Figure 4.** Composite anomaly of the 9–29-day filtered 500-hPa geopotential height (shading) with 200-hPa wave activity flux (vector) at a lead of (a) −4, (b) −2, and (c) 0 days of haze events during 2006–15 under RCP8.5. Dotted areas are statistically significant at the 0.05 level based on 1000-times bootstrap tests. (d–f) As in (a–c) but for 850-hPa wave activity flux (shading). (g) Lead and lag change of the 500-hPa potential energy conversion term in the East China Sea (30°–45°N, 120°–150°E). (h) Composite anomalies of 200-hPa zonal wind (shading) at a lead of −1 day and the climatological jet stream (contours). (i) Composite maximum Eady rate (shading) at a lead of −1 day and its climatology (contours).
mainly attributable to the climate effects of aerosol reduction (Figures 1 and 3). So, here, we analyze the 3D WAF changes in the other two scenarios (Figures 5 and S2). For the decreased aerosol emissions scenario, the 9–29-day fields show that the 500-hPa geopotential height increases near the Barents–Kara Sea, and the horizontal wave activity shows an intensified southward wave train propagation for leads of −4, −2, and 0 days of haze events (Figure 5(a–c)). The 850-hPa vertical WAF increases both near the Barents–Kara Sea and eastern China. The anticyclone located in eastern China increases in this scenario, especially at a lead of 0 days. The same change can also be detected in the original fields (not shown). Our results show that the wave train conducive to haze events can be intensified by aerosol reduction, while the effect of increased GHGs is small.

4. Discussion and conclusions

In this study, the formation process and the future change of the anticyclone anomaly conducive to extreme haze events are investigated. The average relative strength of extreme haze events (30°–40°N, 110°–120°E) changes from 150% during 2006–15 to 190% during 2090–99. Consistent with changes in haze events, the strength and duration of the anticyclone anomaly in eastern China also increases and is mainly induced by the climate effects of aerosol reduction. The formation and future change of the anticyclone anomaly, contributed mainly by intraseasonal 9–29-day low-frequency waves, can be understood by examining the upstream wave train and local feedback process. For the future projection of extreme haze events, intensified northern wave train convergence originated from the increased anticyclone anomaly near the Barents–Kara Sea induces enhanced local feedback in eastern China. Combined with the other two scenarios, we find the intensified upward WAF is mainly induced by the climate effects of reduced aerosol emissions, possibly associated with the increased background baroclinicity induced by an aerosol reduction in eastern Europe.

In previous studies, the North Atlantic Ocean appears to be a source region in the formation of the 9–29-day Eurasian-like low-frequency wave train. It is shown to be related to the combination of anomalous divergence and convergence in different regions and is generated by internal atmospheric dynamics (Jiao, Wu, and Song 2019; Wang et al. 2019). In addition, the feedback of the transient eddies onto the mean flow is often of equal importance to the direct divergence forcing of the
Rossby wave, and it would be interesting to further explore and quantify the role transient eddies play in the anticyclonic formation conducive to haze events (McIntosh and Hendon 2018).

Here, we find that the climate effects of decreased aerosol can intensify the upcoming intraseasonal Rossby wave and increase the strength and duration of the anticyclone anomaly in eastern China. This induces deteriorating diffusion conditions that are more favorable for extreme haze formation in the future, and therefore more stringent regulations on aerosol emissions in China are needed to meet air quality standards. It is still unclear as to which type of aerosol reduction, such as absorption or scattering aerosol, intensifies the background baroclinicity in eastern Europe and the upstream wave train near the Barents–Kara Sea, and so this too merits further investigation.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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