



Roles of Climate Variability on the Rapid Increase of Winter Haze Pollution in North China after 2010

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10 Abstract. North China experiences severe haze pollution in early winter, resulting in many premature deaths and considerable

11 economic losses. The number of haze days in early winter in North China (HD_{NC}) increased rapidly after 2010 but declined

12 slowly before 2010, reflecting a trend reversal. Global warming and emissions were two fundamental drivers of the long-term

- 13 increasing trend of haze, but no studies have focused on this trend reversal. The autumn SST in the Pacific and Atlantic,
- 14 Eurasian snow cover and central Siberian soil moisture, which exhibited completely opposite trends before and after 2010,
- 15 were proven to stimulate identical trends of meteorological conditions related to haze pollution in North China. Numerical
- 16 experiments with a fixed emission level confirmed the physical relationships between the climate drivers and HD_{NC} during
- 17 both decreasing and increasing periods. These external drivers induced a larger decreasing trend of HD_{NC} than the observations,
- 18 and combined with the persistently increasing trend of anthropogenic emissions, resulted in a realistic slowly decreasing trend.
- 19 However, after 2010, the increasing trends driven by these climate divers and human emissions jointly led to a rapid increase
- 20 in HD_{NC}.
- 21 Keywords: haze, PM_{2.5}, trend reversal, anthropogenic emission, climate variability

22 1 Introduction

23 Haze pollution, characterized by low visibility and a high concentration of fine particulate matter (PM_{2.5}), has become a 24 serious environmental and social problem in China, as haze dramatically endangers human health, ecological sustainability 25 and economic development (Ding and Liu, 2014; Wang and Chen, 2016). Exposure to PM2.5 was estimated to cause 4.2 million 26 premature deaths worldwide in 2015 (Cohen et al., 2017), and in China, PM_{2.5} caused up to 0.96 million premature mortalities 27 in 2017 (Lu et al., 2019). Air pollution accounts for a loss of 1.2-3.8% of the gross national product (GNP) annually (Zhang 28 and Crooks, 2012). The most polluted areas in China are North China (NC; 34-42°N, 114-120°E), Fenwei Plain, Sichuan 29 Basin and Yangtze River Delta; among them, NC is the most polluted (Yin et al., 2015). Meteorological conditions 30 characterized by low surface wind speeds and a shallow boundary layer result in stagnant air, which limits the horizontal and





vertical dispersion of particles and induces the accumulation of pollutants (Niu et al., 2010; Wu et al., 2017; Shi et al., 2019).
High relative humidity favors the hygroscopic growth of pollutants (Ding and Liu, 2014; Yin et al., 2015), and anomalous
ascending motions weaken the downward invasion of cold and clear air from high altitudes (Zhong et al., 2019). The
forecasting of meteorological conditions is more accurate on the synoptic scale, but the predictions of interannual variations
are not good enough. Thus, the prediction of haze is a considerable challenge.

36 Previous studies proved that the interannual to decadal variations in winter haze have strong responses to external forcing 37 factors, such as the sea surface temperature (SST) in the Pacific and Atlantic, snow cover and soil moisture (Xiao et al., 2015; 38 Yin and Wang, 2016a, b; Zou et al., 2017). Anomalies of these factors exerted their impacts to modulate local dispersion 39 conditions by atmospheric teleconnections and greatly intensified haze pollution in NC. The eastern Atlantic/western Russia 40 (EA/WR), western Pacific (WP) and Eurasia (EU) patterns served as effective atmospheric bridges linking distant and 41 preceding external factors to the anomalous anticyclonic circulations over Northeast Asia (Yin and Wang, 2017; Yin et al., 42 2017). With enhanced anticyclonic anomalies, the haze pollution in NC was significantly aggravated by poor ventilation 43 conditions and high moisture.

44 The long-term trend of haze pollution has always been attributed to increasing human activities directly related to aerosol 45 emissions (Yang et al., 2016; Li et al., 2018). It is true that emissions are important in the formation of haze, but their role 46 varies from region (Mao et al., 2019). The trend of haze days in Yangtze River Delta and Pearl River Delta was 47 closely related to the trend of particle emissions (Fig. S1b, c), while a weak correlation existed in Fenwei Plain (Fig. S1d). A 48 surprising phenomenon can be seen in NC: the number of winter haze days and particle emissions showed similar trends before 49 early 1990s, but afterward, their close relationship disappeared (Fig. S1a). Many recent studies also showed that the long-term 50 trend in the haze problem has likely been driven by global warming (Horton et al., 2014; Cai et al., 2017). Weakening surface 51 winds have been reported over land in the last few decades while the global surface air temperature (SAT) has warmed 52 significantly (Mcvicar et al., 2012). In addition, enhanced vertical stability, which favors the accumulation of pollutants, has 53 been observed with global warming (Liu et al., 2013). However, none of the above studies focused on the change in the haze 54 trend. Over the past few decades, the global and Northern Hemispheric SAT averages generally displayed a continuous 55 warming trend, which was not exactly similar to the trend of haze days in NC (Fig. S2). It follows that haze pollution, especially 56 the change in its trend, is regulated by multiple drivers and that the long-term impacts of external climate forcings, which 57 efficiently modulate the interannual and decadal variations in haze, deserve further investigation.

58 2 Datasets and Methods

59 2.1 Data description





60 Monthly mean meteorological data from 1979 to 2018 were obtained from NCEP/NCAR reanalysis datasets (2.5°×2.5°), 61 including the geopotential height at 500 hPa (H500), vertical wind from the surface to 150 hPa, surface air temperatures (SAT), 62 wind speed, and special humidity at the surface (Kalnay et al., 1996). The boundary layer height (BLH, 1°×1°) values were 63 from Interim reanalysis data (ERA-Interim) obtained from the European Centre for Medium-Range Weather Forecasts 64 (ECMWF) (Dee et al., 2011). The number of haze days was calculated from the long-term meteorological data, mainly based 65 on observed visibility and relative humidity (Yin et al., 2017). The PM25 concentrations from 2009 to 2016 were acquired 66 from the US embassy, and those from 2014 to 2018 were from China National Environmental Monitoring Centre. Monthly 67 total emissions of BC, NH₃, NO_x, OC, SO₂, PM₁₀ and PM_{2.5} are obtained from the Peking University emission inventory. The 68 monthly mean extended reconstructed SST data $(2^{\circ} \times 2^{\circ})$ were obtained from the National Oceanic and Atmospheric 69 Administration (Smith et al., 2008). The monthly snow cover data were supported by the Rutgers University (Robinson et al., 70 1993). And the monthly soil moisture data $(0.5^{\circ} \times 0.5^{\circ})$ were downloaded from NOAA's Climate Prediction Center (Huug et 71 al., 2003)

72 2.2 Geos-Chem description and experimental design

We used the GEOS-Chem model to simulate PM_{2.5} concentrations (http://acmg.seas.harvard.edu/geos/). The GEOS-Chem model was driven by MERRA-2 assimilated meteorological data (Gelaro et al., 2017). The nested grid over Asia (11°S– 55°N, 60–150°E) had a horizontal resolution of 0.5° latitude by 0.625° longitude and 47 vertical layers up to 0.01 hPa. The GEOS-Chem model includes fully coupled O₃-NOx-hydrocarbon and aerosol chemical mechanisms with more than 80 species and 300 reactions (Bey et al., 2001; Park et al., 2004). The PM_{2.5} components simulated in GEOS-Chem include sulfate, nitrate, ammonium, black carbon and primary organic carbon, mineral dust, secondary organic aerosols and sea salt.

In this study, we designed two kinds of experiments. One was an experiment for simulating $PM_{2.5}$, and the other was a composite using simulated data. The simulation had changing meteorological fields in winter from 1980 to 2018 and the fixed emissions in 2010 representing a high emission level. The emissions data in 2010 were from MIX 2010 (Li et al., 2017). The numerical experiment was performed to examine the variation of $PM_{2.5}$ in the meteorological parameters during 1980–2018 under fixed-emission scenarios.

The composite was conducted to analyze the differences in the simulated HD_{NC} according to the years selected for the external forcing factors. Using the simulated dataset with the fixed-emission scenario was capable of eliminating the impacts of emissions and simply considering the effect of the four external forcing factors. The four (two) years with the largest (Favor Years) and smallest (Unfavor Years) four external forcing indices (i.e., SST_P , $-1 \times SST_A$, Snowc and $-1 \times Soilw$) were selected, and the differences in the simulated HD_{NC} under these four conditions in P1 (P2) were calculated. The simulated HD_{NC} in Favor Years minus the simulated HD_{NC} in Unfavor Years was calculated to analyze the effect of these four forced factors.





90 2.3 Statistical methods

91 In this study, the statistical model of fitted HD_{NC} was built based on MLR. This approach, a model-driven method, was 92 ultimately expressed as a linear combination of K predictors (x_i) that could generate the least error of prediction \tilde{y} (Wilks, 93 2011). With coefficients β_i , intercept β_0 , and residual ε , the MLR formula can be written in the following form: \tilde{y} 94 $=\beta_0+\Sigma\beta_i x_i+\varepsilon$.

95 The trends calculated in this study were obtained by linear regression after a 5-year running average. This method removed 96 the interannual variation and more prominent trend characteristics. Moreover, the stage trends were calculated according to 97 the inflection point, which passed the Mann-Kendall test.

98 **3** Trend change of early winter haze

99 Throughout the winter in North China, the haze pollution in early winter is the most serious (Yin et al., 2019). The number 100 of haze days in early winter in North China (HD_{NC}) reached a remarkable inflection point in 2010 (Fig. 1a), passing the Mann-101 Kendall Test. The trend of HD_{NC} was vastly different before and after 2010: slowly decreased during 1991–2010 (P1) with a 102 rate of 4.67 days/10 yr but rapidly increased after 2010 (P2, 2010-2018) with a rate of 25.43 days/10 yr. Recent studies 103 generally revealed that based on observations, the number of boreal winter haze days across NC had a slightly decreasing trend 104 after 1990 (Ding and Liu, 2014; He et al., 2019; Mao et al., 2019; Shi et al., 2019), which is consistent with the decreasing 105 trend presented by the dataset in our research. In addition, Dang and Liao (2019) confirmed the varying trend of HD_{NC} via 106 simulations of the global 3-D chemical transport (GEOS-Chem) model; using the well-simulated frequency of serious haze 107 days in winter, they also revealed the abovementioned changing trend of HD_{NC} , i.e., decreasing in the early stage and increasing 108 in the later stage. To further determine the reliability of the post-2010 upward trend of HD_{NC}, we used hourly PM_{2.5} 109 concentrations observed at the US embassy in Beijing from 2009 to 2017 and those monitored by China National 110 Environmental Monitoring Centre from 2014 to 2018 to count the number of days when the PM2.5 concentrations were >75 μ g m⁻³ and >100 μ g m⁻³ (Fig. 1a). These statistics also reflected the rising trend after 2010, as well as the improved air quality 111 112 in 2017 and a rebound in pollution in 2018. Although there was a certain gap between HD_{NC} (basing on visibility and humidity) 113 and these statistics, the two datasets revealed the same variations after 2010, and the statistics confirmed the robustness of the 114 observed HD_{NC}.

The above analysis substantiated the rapid aggravation of haze pollution in early winter after 2010. With regard to the increase in air pollution, there is no doubt that anthropogenic emissions were the fundamental cause of this long-term variation. Before the mid-2000s, the particle emissions throughout NC sustained stable growth but gradually began to decline afterward, which is inconsistent with the trend of HD_{NC} or even contrary in some subperiods. The previous decreasing trend of HD_{NC} hid the effects of the increased pollutant emissions; thus, people ignored the pollution problem and failed to control it in time. As





a consequence, the subsequent rise in HD_{NC} was extremely rapid and seriously harmed the biological environment and human health. The stark discrepancy between the trends of pollutant emissions and HD_{NC} strongly indicate that anthropogenic emissions were not the only factor leading to a sharp deterioration in air quality after 2010 (Wei et al., 2017; Wang 2018). Therefore, an important question must be asked: in addition to human activities, what factors caused the rapidly increasing trend of HD_{NC} after 2010?

125 As mentioned above, local meteorological factors could modulate the capacity to disperse and the formation of haze 126 particles, which have critical influences on the occurrence of severe haze pollution. To reveal the impacts of meteorological 127 conditions on the changing trend of HD_{NC} , the area-averaged linear trends of these meteorological factors in NC during P1 and 128 P2 were calculated, all of which exceeded the 95% confidence level (Fig. 2). In P1, the area-averaged linear trends of the 129 boundary layer height (BLH), wind speed and omega all showed significant positive trends, while specific humidity showed a 130 significant negative trend in NC; these conditions favored a superior air quality (Niu et al., 2010; Ding and Liu, 2014; Yin et 131 al., 2017; Shi et al., 2019; Zhong et al., 2019). However, the trends of these four meteorological factors completely reversed 132 in P2. Reductions in the BLH and wind speed, the enhancement of moisture, and an anomalous descending motion resisted 133 the vertical and horizontal dispersions of particles and helped more pollutants gather in relatively narrow spaces. These four 134 meteorological factors expressed an evident influence on the change trend of HD_{NC} and showed reversed trends between P1 135 and P2, similar to HD_{NC}. Furthermore, the magnitudes of the change rates of these factors were stronger in P2 than in P1 (Fig. 136 2), and HD_{NC} displayed this feature as well. The GEOS-Chem simulations with changing emissions and fixed meteorological 137 conditions failed to reproduce the change trend of haze (Dang and Liao, 2019). We designed an experiment driven by changing 138 meteorological conditions in winter from 1980 to 2018 and fixed emissions at the relatively high 2010 level. According to the 139 technical regulation on the ambient air quality index (Ministry of Ecology and Environment of the People's Republic of China, 140 2012), a haze day was defined as a day with daily mean $PM_{2.5}$ concentration exceeding 75 µg m⁻³. The simulations of the 141 frequency of haze days in NC by GEOS-Chem reproduced the trend reversal of haze pollution (Fig. 1b). The simulation results 142 were highly correlated with HD_{NC} and showed the feature that the trend in P2 was stronger than that in P1, indicating that 143 meteorological conditions drove the trend change of haze pollution.

144 4 Climate variability drove the trend reversal

According to many previous studies, the variabilities of the Pacific SST, Atlantic SST, Eurasian snow cover and Asian soil moisture played key roles in the interannual variations in haze pollution in NC (Xiao et al., 2015; Yin and Wang, 2016a, b; Zou et al., 2017), and the associated physical mechanisms were evidently revealed. Thus, the following question is raised here: did these four factors drive the trend reversal of HD_{NC}, and if so, how?





149 As shown in Figure S3a, the preceding autumn SST in the Pacific, associated with the detrended HD_{NC}, presented a 150 Pacific Decadal Oscillation (PDO)-like "triple pattern" with two significant positive regions and one nonsignificant negative 151 region (Yin and Wang, 2016a; Zhao et al., 2016). In the following research, the SST anomalies in the two positively correlated 152 regions located in the Gulf of Alaska (40-60°N, 125-165°W) and the central eastern Pacific (5-25°N, 160°E-110°W) were 153 used to represent the effects originating from the North Pacific. The area-averaged September-November SST of these two 154 regions was calculated as the SST_P index, and the correlation coefficients with HD_{NC} were 0.59 and 0.67 before and after 155 removing the linear trend during 1979–2018, respectively; both correlation coefficients were above the 99% confidence level. 156 The responses of the atmosphere to these positive SST_P anomalies were the positive phase of the EA/WR pattern and the 157 enhanced anomalous anticyclone center over NC (Yin et al., 2017; Fig. S4). Modulating by such large-scale atmospheric 158 anomalies, increased moisture, anomalous upward motion and reduced BLH and wind speed (Fig. S4) created a favorable 159 environment for the accumulation of fine particles (Niu et al., 2010; Ding and Liu, 2014; Shi et al., 2019; Zhong et al., 2019). 160 A numerical experiment based on the Community Atmosphere Model version 5 (CAM5) effectively reproduced the observed 161 enhanced anticyclonic anomalies over Mongolia and North China in response to positive PDO forcing, which resulted in an 162 increase in the number of wintertime haze days over central eastern China (Zhao et al., 2016). The trend changes of the North 163 Pacific SST were examined in P1 and P2. Consistent with the changing trend of HD_{NC}, reversed trends were also found in the 164 North Pacific, i.e., a significant negative trend during P1 and a positive trend during P2 over the two Pacific areas (Fig. 3a, b). 165 These similar trend changes suggest that the North Pacific SST might have been a major driver of the abrupt change in HD_{NC}. 166 It is clear that SST_P underwent a significant trend change around 2010 (Fig. 4a). Thus, the persistent decline in SST_P during P1 167 (at a significant rate of -0.2 °C/10 yr; Table 1) contributed to the slowly decreasing trend of HD_{NC} (Fig. 4a) via the modulations 168 of SST_P on the atmospheric circulation (Fig. S4). During P2, the larger increase in SST_P at a rate of 2.0 °C/10 yr dramatically 169 drove the rapid increase in HD_{NC} .

170 Besides the triple pattern in the Pacific, two areas exhibiting significant negative correlations with HD_{NC} were examined 171 in the Atlantic (Shi et al., 2015; Shi et al., 2015): one located over southern Greenland (50-68°N, 18-60°W) and another 172 located over the equatorial Atlantic (0-15°N, 30-60°W; Fig. S3a). The area-averaged September-November SST of the two 173 negatively correlated regions in Atlantic was defined as the SST_A index, whose correlation coefficients with HD_{NC} were -0.55 174 and -0.64 from 1979 to 2018 before and after detrending, respectively (above the 99% confidence level). The response of 175 atmospheric circulation to these negative SSTA anomalies culminated in a positive EA/WR pattern, and the stimulated easterly 176 weakened the intensity of East Asian jet stream (EAJS) in the high troposphere (Fig. S5). Influenced by the colder SST_A , there 177 was a very obvious abnormal upward movement above the boundary layer, reducing both the BLH and the surface wind speed; 178 thus, pollutants were prone to gather, causing haze pollution (Niu et al., 2010; Wu et al., 2017; Shi et al., 2019). With a linear 179 barotropic model, Chen confirmed the important role of subtropical northeastern Atlantic SST anomalies in contributing to the





anomalous anticyclone over Northeast Asia and anomalous southerly winds over NC, which enhanced the accumulation of pollutants (Chen et al., 2019). The spatial linear trend in the SST of both Atlantic areas changed from positive in P1 to negative in P2, which was completely contrary to the trend of HD_{NC} (Fig. 3a, b). The SST_A reached a infection point in 2010 (Fig. 4b) and contributed to the falling of HD_{NC} during P1 (change rate of SST_A = 0.55 °C/10 yr) and the rising of HD_{NC} during P2 (change rate of SST_A = -0.52 °C/10 yr).

185 The effect of Eurasian snow cover on the number of December haze days in NC intensified after the mid-1990s (Yin and 186 Wang, 2018). The roles of extensive boreal Eurasian snow cover were also revealed by numerical experiments via the 187 Community Earth System Model (CESM): positive snow cover anomalies enhanced the regional circulation mode of poor 188 ventilation in NC and provided conducive conditions for extreme haze (Zou et al., 2017). The correlation between the October-189 November snow cover and HD_{NC} was significantly positive in eastern Europe and western Siberia (46-62°N, 40-85°E, Fig. 190 S3b), where the spatial linear trend of snow cover was consistent with that of HD_{NC}. A significant negative trend in P1 and a 191 positive trend in P2 were discovered (Fig. 3c, d). The area-averaged October-November snow cover over eastern Europe and 192 western Siberia was defined as the Snowc index, and its correlation coefficients with HD_{NC} were 0.43 and 0.54 from 1979 to 193 2018 before and after detrending, respectively (above the 99% confidence level). The features of the weakened EAJS and 194 significant anomalous anticyclone could be found clearly in the induced atmospheric anomalies associated with the positive 195 Snowc anomalies (Fig. S6). The related abnormal upward motion restricted the momentum to the surface. In addition, the 196 corresponding lower BLH and weaker surface wind speed also reduced the dispersion capacity, resulting in the generation of 197 more haze pollution (Fig. S6). The Snowc index fell slowly until 2010 (with a rate of -1.8%/10 yr) and then rose rapidly (with 198 a rate of 28.3%/10 yr) and experienced a large trend reversal in 2010, in accordance with the behavior of HD_{NC} (Fig. 4c). 199 Therefore, relying on the revealed physical mechanisms, the strengthened relationship between Snowc and HD_{NC} and the 200 tremendous increase in Snowc during P2 substantially triggered the rapid enhancement of haze pollution in NC.

201 In addition to snow cover, soil moisture was another important factor affecting HD_{NC} (Yin and Wang, 2016b). The 202 September-November soil moisture and HD_{NC} were negatively correlated in central Siberia (54-70°N, 80-130°E; Fig. S3c). 203 The area-averaged September-November soil moisture over central Siberia was denoted as the Soilw index, whose correlation 204 coefficients with HD_{NC} were -0.57 and -0.60 from 1979 to 2018 before and after detrending, respectively (above the 99% 205 confidence level). Negative Soilw anomalies could induce a positive phase of EA/WR, and the associated anticyclonic 206 circulations occurred more frequently and more strongly (Fig. S7). Correspondingly, the local vertical and horizontal 207 dispersion conditions were limited. With increasing moisture, pollutants can more easily accumulate in a confined area. The 208 spatial linear trend of soil moisture also shifted from increasing to decreasing in 2010, opposite to the trend of HD_{NC} (Fig. 3e, 209 f). The change rate of Soilw was 38.8 mm/10 yr (opposite that of HD_{NC}) during P1, and the rate of change became more intense 210 (-51.8 mm/10 yr) during P2, physically driving a similar large change in HD_{NC} (Fig. 4d).





211 The varying trends of these four preceding external factors jointly drove the trend reversal of HD_{NC} based on their physical 212 relationships with the haze pollution in North China. To exclude the impacts of the stage trends of these variables on the 213 physical links between the climate drivers and HD_{NC}, the correlations between these factors and HD_{NC} were explored during 214 the decreasing stage (i.e., 1979-2010) and increasing stage (2010-2018), and all of these correlations were significant (Table 215 1). Thus, the physical relationships between HD_{NC} and these four factors were long-standing and did not disappear as the trend 216 changed. These four external factors had completely opposite trends in P1 and P2. Excluding SST_A, the amplitudes of the 217 change trends of the other three indices in P2 were obviously stronger than those in P1 and were identical to those of HD_{NC} 218 (Table 1). In our study, we composited the simulations based on the GEOS-Chem model to determine the impact on haze 219 pollution of each factor under the fixed-emissions level. The years in the top 20% and the bottom 20% of the four indices (i.e., 220 SST_{P_2} , $-1 \times SST_A$, Snowc and $-1 \times Soilw$) in P1 and P2 were selected, which could remove the effects of different trends. The 221 composite differences for the four external forcing factors were significant in the selected regions and passed the Student's t 222 test (Fig. S8). The responses of simulated HD_{NC} to the original (detrended) sequences of SST_P, SST_A, Snowc and Soilw were 223 all positive, which are consistent with the observational results (Fig. 5). Specifically, for the four original (detrended) drivers, 224 the resulting differences in simulated HD_{NC} were 3.94 (5.28), 5.97 (5.07), 1.86 (1.86) and 6.49 (6.49) days in P1 and 4.46 225 (4.46), 4.26 (4.26), 7.54 (7.54) and 7.35 (7.35) days in P2 (Fig. 5). These differences were distinct and further confirmed that 226 each factor played a role in the occurrence of haze pollution in NC.

227 These four indices were employed to linearly fit HD_{NC} based on a multiple linear regression (MLR) model (Wilks, 2011). 228 As shown in Figure 4e, the correlation coefficient between the fitted and observed HD_{NC} was 0.82. After a five-year running 229 average, the correlation coefficient reached 0.92. This model showed good ability to fit the infection point in 2010 and 230 highlighted the trend changes. Such a good fitting effect indicates that changes in the four external forcing factors could well 231 have influenced the variation in HD_{NC}. By exciting stronger responses in the atmosphere, such as the positive EA/WR phase 232 and the strengthened anomalous anticyclone over NC, the abovementioned climate drivers created stable and stagnant 233 environments in which the haze pollution in NC could rapidly exacerbate after 2010 (Table 1). Among the four indices, the 234 correlation coefficients between SST_P and Snowc (Pair 1) and between SST_A and Soilw (Pair 2) were high, while the rest were 235 insignificant. The variance inflation factors of the four factors in the MLR model were less than 2, showing that the collinearity 236 among them was weak. When selecting one factor from both Pair 1 and Pair 2 to refit HD_{NC}, the correlation coefficient between 237 the fitted and observed HD_{NC} and the trends of the fitted HD_{NC} in P2 worsened (Fig. S9). Therefore, these four external factors 238 were all indispensable to achieve a better fitting effect. The intercorrelated climate factors of Pair 1 and Pair 2 contributed 239 27.8% and 84.6%, respectively, to the trends of HD_{NC} in P1 and 54.8% and 20.4% to the trends in P2. Thus, the joint effect of 240 SSTA and Soilw played a more important role in the decreasing trend of HD_{NC} in P1; however, the impacts of SSTP and Snowc 241 were more than twice those of SST_A and Soilw in P2. More importantly, the fitted curve revealed a decreasing trend of HD_{NC}





 $\begin{array}{ll} 242 & (-5.24 \text{ days/10 yr}) \text{ that was larger than observed } (-4.67 \text{ days/10 yr}) \text{ during P1}. \text{ Many studies have noted that human activities} \\ 243 & \text{have led to persistently increasing trends of HD}_{\text{NC}} (\text{Yang et al., 2016; Li et al., 2018}). \text{ The combination of the exorbitant} \\ 244 & \text{decreased trend indicated by climate conditions and the long-term trend from anthropogenic emissions resulted in a realistic} \\ 245 & \text{slow decline (Table 2)}. \text{ This proportion of the trend explained by climate drivers } (72.3\%) decreased in P2 because the \\ 246 & \text{increasing trend driven by the climate divers and emissions jointly led to a rapid increase in HD}_{\text{NC}}. \end{array}$

247 5 Conclusions and discussions

Haze events in early winter in North China exhibited rapid growth after 2010, which was completely different from the slow decline observed before 2010, showing a trend reversal in the year 2010 (Fig. 1). The trend changes of associated meteorological conditions exhibited identical responses. After 2010, the lower BLH, weakened wind speed, sufficient moisture and anomalous ascending motion (all with larger tendencies than before 2010) limited the horizontal and vertical dispersion conditions and thus enhanced the occurrence of early winter haze pollution (Fig. 2). However, before 2010, the climate conditions showed the opposite characteristics and could create an environment with adequate ventilation for the dissipation of particles.

255 In this study, the external forcing factors that caused the significant growth of HD_{NC} after 2010 and the associated physical 256 mechanisms were investigated. These factors could stimulate and strengthen the anomalous anticyclone over NC via exciting 257 the EA/WR teleconnection pattern, thus regulating the meteorological conditions, weakening the dispersion conditions and 258 facilitating the accumulation of haze pollutants. The four climate drivers physically related to HD_{NC} showed exactly opposite 259 trend changes with an inflection point in 2010, which agrees with the behavior of HD_{NC} (Fig. 4). The factors of Pair 1 (SST_A 260 and Soilw) and Pair 2 (SST_P and Snowc) had joint effects and played more important roles in the increasing trend of HD_{NC} in 261 P2 and the decreasing trend of HD_{NC} in P1, respectively (Table 2). The fitting result of the four factors with the trend of HD_{NC} 262 showed a strongly decreasing trend in P1 and a weakly increasing trend in P2. Together with increasing emissions, these factors 263 jointly led to a relatively slow decreasing trend of HD_{NC} before 2010 and rapid growth afterward. Therefore, both the 264 decreasing trend in P1 and the increasing trend in P2 were caused by a combination of climate drivers and emissions.

Anthropogenic emissions have exceeded a high level since the 1990s, providing a sufficient foundation for the generation of severe haze pollution (Fig. 1). However, the effects of climate variability delayed warnings before 2010. Together with the local meteorological conditions, the trends of the climate drivers reversed in 2010, initiating a dramatically increase in HD_{NC} after 2010, which quickened the worsening of haze pollution in NC (Fig. 5; Table 1). The superimposed effect of high-level human emissions with strengthened climate anomalies loudly sounded the alarms through the extremely rapid rise of haze pollution.





271 Data availability. The monthly mean meteorological data are obtained from NCEP/NCAR reanalysis datasets 272 (https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.html). The boundary layer height data are available from the 273 Interim reanalysis dataset (http://www.ecmwf.int/en/research/climate-reanalysis/era-interim). The number of haze days can be 274 obtained from the authors. The PM2.5 concentrations from 2009 to 2016 can be downloaded from the US embassy 275 (http://www.stateair.net/web/historical/1/1.html), and those from 2014 to 2018 can be downloaded from China National 276 Environmental Monitoring Centre (http://beijingair.sinaapp.com/). The monthly total emissions of BC, NH₃, NO₅, OC, SO₂, 277 PM10 and PM25 are obtained from the Peking University emission inventory (http://inventory.pku.edu.cn/). SST data are 278 downloaded from http://www.esrl.noaa.gov/psd/data/gridded/data.noaa.ersst.v4.html. Soil moisture data are obtained from 279 https://www.esrl.noaa.gov/psd/data/gridded/data.cpcsoil.html. Snow cover data can be downloaded from Rutgers University: 280 http://climate.rutgers.edu/snowcover/. The emissions of 2010 can downloaded from be 281 http://geoschemdata.computecanada.ca/ExtData/HEMCO/MIX.

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287 Author contributions

- 288 Wang H. J. and Yin Z. C. designed the research. Yin Z. C. and Zhang Y. J. performed research. Zhang Y. J. prepared the
- 289 manuscript with contributions from all co-authors.

290 Competing interests

- 291 The authors declare no conflict of interest.
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419 Table and Figure legends

- 420 Table 1. Correlation coefficients (CCs) between HD_{NC} and the SST_P, SST_A, Snowc and Soilw indices after detrending and the 421 trends of the SST_P, SST_A, Snowc and Soilw indices for the periods 1991-2010 and 2010-2018. CC₁, CC₂, and CC₃ represent the correlation coefficients from 1979 to 2018, 1979 to 2010 and 2010 to 2018, respectively. "***" indicates that the CC was 422 423 above the 99% confidence level, "**" indicates that the CC was above the 95% confidence level, and "*" indicates that the 424 CC was above the 90% confidence level. 425 Table 2. The contribution rate of fitted HD_{NC} and each external forcing factor to the trend of HD_{NC} in P1 and P2, respectively. 426 Figure 1. (a) Variations in the December-January emissions (unit: Tg) of black carbon (BC), ammonia (NH₃), nitrogen oxide 427 (NO_x), organic carbon (OC), sulfur dioxide (SO₂), PM₁₀ and PM_{2.5} over North China from 1979 to 2013 and the variation in
- 428 HD_{NC} from 1979 to 2018 (black solid line). The red dashed line represents the total emissions of the seven pollutants. The blue
- 429 and green solid (dashed) lines indicate the number of days when the hourly $PM_{2.5}$ concentrations in a day exceeded 75 μ g m⁻³
- 430 and 100 μ g m⁻³, respectively, from 2009 to 2016 (2014 to 2018) using observed data from the US embassy (China National
- 431 Environmental Monitoring Centre). (b) Temporal evolutions of HD_{NC} (in black), simulated haze days (unit: days; red) and (c)
- 432 average $PM_{2.5}$ concentrations (unit: $\mu g m^{-3}$; blue) in NC. The dashed lines denote linear regressions for 1991–2010 (P1) and
- 433 2010–2018 (P2). Trend 1 and Trend 2 represent the linear trends of the simulations in P1 and P2, respectively.
- 434 Figure 2. Area-averaged linear trends of the BLH (unit: m/yr), specific humidity (unit: %/10 yr), surface wind speed (unit: m
- 435 $s^{-1}/10^2$ yr) and omega (unit: pascal $s^{-1}/10^3$ yr) over NC in early winter for the periods 1991–2010 (P1) and 2010–2018 (P2).
- 436 All datasets were 5-year running averages before calculating the trends.
- 437 Figure 3. Linear trends of the Pacific and Atlantic SST (unit: °C/yr; a, b), Eurasian snow cover (unit: %/yr; c, d), and central
- 438 Siberian soil moisture (unit: mm/yr; e, f) for the periods 1991–2010 (P1) and 2010–2018 (P2). All datasets were 5-year running
- 439 averages before calculating the trends. The green boxes represent the regions where the four indices are defined. Black dots
- 440 indicate that the trends were above the 95% confidence level.
- 441 Figure 4. Variations in HD_{NC} (in black) and the SST_P (unit: °C; a, red), SST_A (unit: °C; b, blue), Snowc (unit: %; c, yellow),
- 442 and Soilw (unit: mm; d, green) indices and the HD_{NC} values fitted by the MLR model by the above four factors (unit: days; e,
- 443 purple) from 1979 to 2018. Thick lines indicate 5-year running averaged time series. The rectangles and triangles indicate the
- 444 inflection points of HD_{NC} and the four indices, which were tested by the Mann-Kendall test.
- 445 Figure 5. Composite of the simulated HD_{NC} caused by the four external forcing factors (Favor Years minus Unfavor Years).
- 446 The circles and crosses represent the original and detrended sequences, respectively.
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450Table 1. Correlation coefficients (CCs) between HD_{NC} and the SST_P , SST_A , Snowc and Soilw indices after detrending and the451trends of the SST_P , SST_A , Snowc and Soilw indices for the periods 1991–2010 and 2010–2018. CC1, CC2, and CC3 represent452the correlation coefficients from 1979 to 2018, 1979 to 2010 and 2010 to 2018, respectively. "***" indicates that the CC was453above the 99% confidence level, "**" indicates that the CC was above the 95% confidence level, and "*" indicates that the454CC was above the 90% confidence level.455

	CC with UD	Trend / 10yr		
	CC with HD _{NC}	1991–2010	2010-2018	
SST _P	$\begin{array}{c} CC_1 = 0.67 *** \\ \hline CC_2 = 0.39 ** \\ \hline CC_3 = 0.66 *** \end{array}$	-0.20 °C***	1.99 °C***	
SST _A	$\begin{array}{c} CC_1 = -0.64 & *** \\ CC_2 = -0.54 & *** \\ CC_3 = -0.61 & *** \end{array}$	0.55 °C***	-0.52 °C***	
Snowc	$\begin{array}{c} CC_1 = 0.54 *** \\ \hline CC_2 = 0.46 *** \\ \hline CC_3 = 0.53 *** \end{array}$	-1.79%**	28.35%***	
Soilw	$CC_1 = -0.60 ***$ $CC_2 = -0.30 *$ $CC_3 = -0.66 ***$	38.78mm***	-51.81mm***	

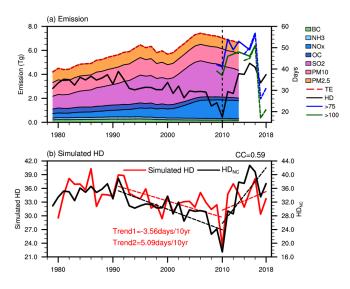
Table 2. The contribution rate of fitted HD_{NC} and each external forcing factor to the trend of HD_{NC} in P1 and P2,

458 respectively.

	Fitted HD _{NC}	SST _P	SST _A	Snowc	Soilw
P1	112.2%	23.3%	43.9%	4.5%	40.7%
P2	72.3%	41.9%	7.5%	12.9%	10.0%

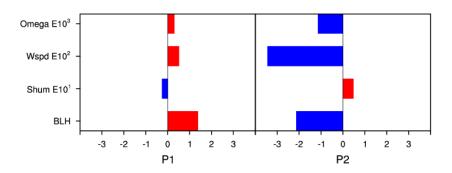








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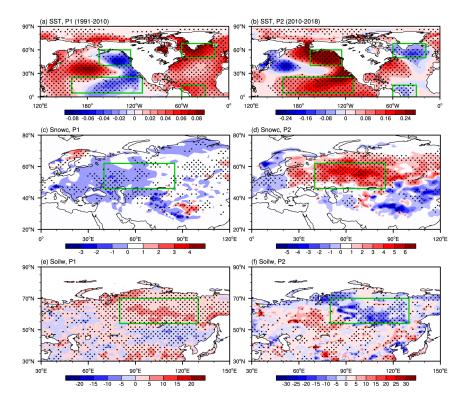
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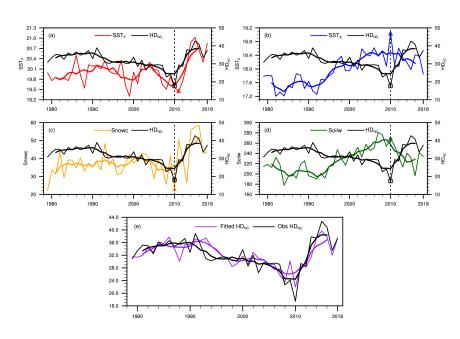
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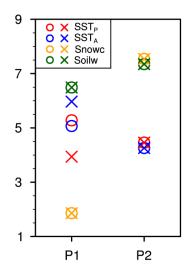
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