1	Future projections of daily <u>hazyhaze conducive</u> and clear weather conditions over the
2	North China Plain using a Perturbed Parameter Ensemble
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16 Abstract

We examine past and future changes in both winter haze and clear weather conditions over the 17 North China Plain (NCP) using a Perturbed Parameter Ensemble (PPE) and elucidate the 18 influence of model physical parameterizations on these future projections for the first time. We 19 use a large-scale meteorology-based Haze Weather Index (HWI), which was developed to 20 21 examine the) with values >1 as a proxy for haze conducive weather and HWI <-1 for clear 22 weather conditions for Beijing. We find that the HWI can be used as an indicator of winter haze across the entire over the NCP due to the extended spatial coherence of the local 23 meteorological conditions. The PPE generated using the UK Met Office HadGEM-GC3 model 24 shows that under a high-emission (RCP8.5) scenario, the frequency of haze conducive weather 25 (HWI>1) is likely to increase whereas the frequency of clear weather (HWI<-1) is likely to 26 27 decrease in future. However, a change of opposite sign with lower magnitude in the frequencies, though less likely, is also possible. In future, the total number of hazy 28 daysfrequency of haze conducive weather for a given winter can be as much as ~3.5 times 29 higher than the numberfrequency of clear days weather over the NCP. We also examined the 30 changes in the interannual variability of the frequency of hazy and clear days and find no 31 32 marked changes in the variability for future periods. The future frequencies of haze conducive weather (HWI>1) during winter hazy and clear days in the PPE are largely driven by associated 33 34 with changes in zonal-mean mid-tropospheric winds and the vertical temperature gradient over 35 the NCP. We do not also examined the changes in the interannual variability of the haze conducive and clear weather, and find any discernible no marked changes in the variability of 36 future periods. We find a clear influence of model physical parametrizations on climatological 37 38 mean frequencies for both haze conducive and clear weather. For mid to late 21st century (2033-2086), parametric effect can explain up to ~80% variance in climatological mean frequencies 39 of PPE members. Therefore, model parameterizations on-adds uncertainty in the future 40

- 41 projections of trendshaze conducive weather in addition to the frequency of hazy or clear 42 days.internal variability. We also find a clear impactgrowing influence of anthropogenic 43 climate change on future trends for both hazy and clear days, however, it is only discernible 44 for specific periods due to the large underlying internal variability in the frequencies of 45 climate change on future trends for both hazy and clear days, however, it is only discernible 46 for specific periods due to the large underlying internal variability in the frequencies of 47 climate change on future trends for both hazy and clear days, however, it is only discernible 48 for specific periods due to the large underlying internal variability in the frequencies of 49 climate change on future trends for both hazy and clear days, however, it is only discernible 40 for specific periods due to the large underlying internal variability in the frequencies of 40 climate change on future trends for both hazy and clear days, however, it is only discernible 41 for specific periods due to the large underlying internal variability in the frequencies of 42 climate change on future trends for both hazy and clear days, however, it is only discernible 43 climate change on future trends for both hazy and clear days, however, it is only discernible 44 for specific periods due to the large underlying internal variability in the frequencies of 43 climate change on future trends for both hazy and clear days of the large days of th
- 45 <u>hazymean frequencies of haze conducive</u> and clear days.

- 46 weather over the 21st century suggesting climate change can exacerbate the haze conducive
 47 weather and reduce the clear weather conditions in future over the NCP.
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49 **1. Introduction**

Over the last decade, a number of severe haze episodes (several days or longer) were 50 reported over the North China Plain (NCP) during boreal winter (December-January-February, 51 DJF). In January 2013, unprecedented $PM_{2.5}$ levels exceeding 450 µg m⁻³ were observed over 52 the NCP (Wang et al., 2014a; Wang et al., 2014b; Zhang et al., 2018; Zhang et al., 2013). 53 Similar events were also observed in November-December 2015 when the PM_{2.5} concentrations 54 reached as high as 1000 µg m⁻³ in Beijing and caused the first-ever 'red alert' for severe air 55 pollution (Liu et al., 2017; Zhang et al., 2017). In December 2016, around 25% of the land area 56 of China was covered with severe haze for around one week (Yin and Wang, 2017). These 57 severe haze events adversely impacted public health including mortality, visibility, and 58 ultimately the economy of the country (Bai et al., 2007; Chen and Wang, 2015; Kan et al., 59 2012; Kan et al., 2007; Wang et al., 2006; Xu et al., 2013; Hong et al., 2019). 60

61 Previous research has shown that the persistence of severe haze for days during winters over the NCP occurred due to the combined effect of local and regional high pollutant 62 emissions and stagnant meteorological conditions (Li et al., 2018; He et al., 2016; Jia et al., 63 64 2015; Pei et al., 2018; Zhang et al., 2021). The normal winter meteorological conditions over the NCP are characterized by northwesterly flow near the surface through to the mid-65 troposphere associated with the East Asian winter monsoon (circulation (Fig. 1a and 1b; also 66 see An et al., 2019; Chen and Wang, 2015; Li et al., 2016; Renhe et al., 2014; Li et al., 2016; 67 Xu et al., 2006). The northwesterly winds support the intrusion of relatively clean air from the 68 high latitudes to the NCP and therefore ventilate this region (Xu et al., 2006). However, during 69

70 the severe haze episodes, the near-surface northwesterlies appear to be weaker than normal and the mid-tropospheric trough was reported to be shallower and shifted northwards – collectively 71 leading to a weaker than normal northwesterly flow and reduced horizontal transport of air 72 73 pollutants from the NCP (Chen and Wang, 2015). The weaker northwesterlies near the surface also reduces the intrusion of cold and clean air from the high-latitudes to the NCP (Xu et al., 74 75 2006). Fig. 2a-b). In addition to changes in horizontal winds, the vertical temperature gradient between the lower and upper troposphere over the NCP enhancescan influence the vertical 76 dispersion of the pollutants. A warmer than normal temperature near the surface, accompanied 77 78 with colder temperature in the upper troposphere, would enhance the thermal stability and reduces reduce the atmospheric mixing leading to the build-up of the atmospheric pollutants 79 80 over this region (Fig. 2; also see Hou and Wu, 2016; Sun et al., 2014; Wang et al., 2014a; 81 Zhang et al., 2018; Cai et al., 2018). The planetary boundary layer height is also found to be 82 suppressed during extreme haze events leading to accumulation of pollutants, notably PM_{2.5} concentrations (Liu et al., 2018; Petäjä et al., 2016), due to an increase in moisture, reduced 83 vertical mixing and dispersion which aids aerosol growth during high haze events over the 84 NCP (An et al., 2019; Tie et al., 2017). 85

86 In this paper, our focus is on On a daily scale, past studies have examined the 87 meteorological driven changes leading to daily hazy or clearin haze conducive weather 88 conditions over the NCP. On a daily scale, recent studies suggest an increase in the occurrence 89 of large-scale meteorological conditions favourable for winter haze over the NCPChina under 90 climate change- scenarios using large-scale meteorology-based indexes. For example, Cai et al. (2017) have used four key variables, i.e. meridional wind at 850 hPa (V₈₅₀), zonal wind at 91 92 500 hPa (U₅₀₀), temperatures at 850 hPa (T₈₅₀) and 250 hPa (T₂₅₀) pressure levels to calculate a meteorology-based daily Haze Weather Index (HWI) and). They have projected a ~50% 93 94 increase in the frequency of winter haze conducive weather conditions, similar to the January

95	2013 event, over Beijing in the future (2050-2099) as compared to the historical (1950-1999)
96	period under the RCP8.5 scenario using 15 CMIP5 models. Using the HWI, Liu et al. (2019)
97	projected a 6-9% increase in the winter haze frequency under 1.5° and 2° global warming,
98	respectively based on 20 CMIP5 models whereas Qiu et al. (2020) projected a relatively high
99	increase of 21% and 18% in severe winter haze episodes under 1.5° and 2° global warming,
100	respectively using an ensemble of climate simulations from the Community Earth System
101	Model 1 (CESM1) (Kay et al., 2015). RCP8.5 scenario using 15 CMIP5 models. Han et al.
102	(2017) also Callahan and Mankin (2020) also used specific humidity, V ₈₅₀ , T ₈₅₀ and
103	temperatures at 1000 hPa to examine the haze favourable meteorology for Beijing, and found
104	a 10-15% increase in winter haze conducive weather in CMIP5 multimodel and CESM large
105	ensemble under 3° warming. These authors have also emphasized a large influence of internal
106	variability in addition to anthropogenic forcing on future haze conducive weather over Beijing.
107	In addition to the large-scale meteorology based indexes, several other stagnation
108	indices based on regional or local meteorological variables have also been used to determine
109	the influence of anthropogenic climate change on haze conducive weather for China as well as
110	global regions. Using minimum monthly mean wind speeds averaged over northwestern
111	Europe, Vautard et al. (2018) suggested a potential increase in the frequency of stagnant
112	conditions conducive to air pollution over northwest Europe; however, their results were
113	sensitive to models used for the analysis. Horton et al. (2014) have used thresholds for the daily
114	mean near-surface (10-m) wind speeds, mid-tropospheric (500 hPa) temperatures and
115	accumulated precipitation to calculate the Air Stagnation Index (ASI) under RCP8.5 scenario
116	using 15 CMIP5 models. They found an increase in air stagnation occurrence events leading to
117	poor air quality by up to ~40 days per year over a majority of the tropics and sub-tropics. Han
118	et al. (2017) examined indicators of haze pollution potential (e.g. horizontal transport, wet-
119	deposition, ventilation conditions) using three regional climate simulations and projected a

120 higher probability of haze pollution risk over the Beijing-Tianjin-Hebei region under the RCP4.5 scenario. LiuGarrido-Perez et al. (2019) projected (2021) took a 6-9% increase in the 121 winter haze frequency under 1.5° different approach as compared to analysing probabilistic 122 123 projections and 2° global warming, respectively used the ASI to generate stagnation storylines, i.e. plausible and physically consistent scenarios of stagnation changes based on 20 CMIP5 124 models. Qiu et al. (2020) also projected an increase of 21% and 18% in severe winter haze 125 episodes under 1.5° and 2° global warming, respectively using an ensemblethe response of 126 remote drivers under climate simulations from the Community Earth System Model 1 127 128 (CESM1) for a low warming experiment change(Kay et al., 2015). Callahan and Mankin (2020) found 10-15% increase in winter hazy days in CMIP5 multimodel and CESM large 129 130 ensemble under 3° warming and emphasized a large influence of internal variability in addition to anthropogenic forcing on future, for Europe and the United States (US). 131

While most studies indicate an increase in the haze conducive weather over Beijing. AChina, a few studies also find little impact of climate change on future projections of haze (Shen et al., 2018; Pendergrass et al., 2019), which could partly arise due to the under-sampling of internal variability associated uncertainty in their projections (Callahan and Mankin, 2020), as well as model-to-model differences. Hence, there is a large uncertainty as to how haze conducive weather conditions may change in the future and these depend on haze metricmetrics or underlying processes considered for <u>future</u> projections.

In order to account for the uncertainty in the future projections (e.g. of large-scale circulation) particularly at the regional scale (Hawkins and Sutton, 2012; Deser et al., 2012; Deser et al., 2014), it is desirable to use an ensemble of climate change simulations. Whilst a multimodel ensemble, e.g. CMIP5 or CMIP6, is commonly used for climate change studies, several other studies have also emphasised the use of an initialised ensemble or Perturbed Parameter Ensemble (PPE) from a single model to assess the uncertainties and obtain a

145 comprehensive range of possible future climate realisations for the same emission scenario for a given model (Knutti et al., 2010). All three methodologies have different advantages. For 146 instance, using multiple models allows us to sample structural uncertainty in future projections, 147 which cannot be sampled using a single model. On the other hand, using an initialised ensemble 148 from a single model allows us to sample a broader range of internal variability, which is often 149 under-sampled in a multimodel ensemble. The advantage of using the PPE over the initialised 150 or multimodel ensemble is that it not only accounts for internal variability but also model 151 uncertainty arising due to the different settings of the physical parameterisations in a single 152 model. 153

- Both multimodel ensemble and initialised ensemble from a single model have been 154 used to assess the future winter haze conducive conditions over Beijing. In this paper, we use 155 156 a PPE generated using the UK's Met Office HadGEM-GC3 model to assess for the first time the impact of both model physical parameterisations and anthropogenic climate change on 157 future daily haze conducive weather conditions-using the HWI. We first determine the spatial 158 extent for which the HWI can be used as an indicator of air quality over China (Section 3). We 159 160 examine the changes in the frequency of hazy and clear days for historical and three future periods, i.e. near (2006-2032), mid (2033-2059) and far (2060-2086) future, over the NCP 161 (Section 4). We also analyse the changes in the interannual variance of the frequency of hazy 162 and clear days for the future periods as compared to the historical (Section 5). We investigate 163 164 the importance of the different meteorological variables used in the HWI in determining the 165 future changes in haze conducive conditions in the PPE (Section 6). Finally, we assess the model physical parametrisations and anthropogenic climate change on the frequency of future 166 167 hazy and clear weather conditions over the NCP (Section 7). More details on the data and methods used in this paper are provided in the next section.. 168

169 In this paper, our focus is on the daily haze conducive and clear weather conditions over the NCP under a fixed high-emission scenario (RCP8.5). For this purpose, we use the 170 HWI proposed by Cai et al. (2018) as past research studies have shown a robust correlation 171 between the HWI, which is a large-scale meteorology based index, and haze conducive weather 172 for Beijing in China. Whilst Cai et al. (2018) originally proposed the HWI for Beijing, the 173 index is based on changes in large-scale meteorology over the NCP and thus offers a good 174 175 potential as the indicator of haze conducive weather over the NCP. One potential advantage of using the HWI for future projections, as opposed to a regional or local air stagnation index, is 176 177 that the general circulation models generally simulate large-scale meteorology reasonably well as compared to local or regional meteorology. Therefore, we expect the future projections of 178 179 clear or haze conducive weather provided using the HWI to be less uncertain than projections 180 provided using regional stagnation indexes. 181 The HWI uses four meteorological variables as stated above, but Cai et al. (2018) have 182 also examined the impact of the inclusion of more weather variables, such as geopotential height, boundary layer thickness and local stratification instability, in the HWI and did not find 183 184 any significant differences in the performance of the HWI. Therefore, we use the same 185 variables and methodology as Cai et al (2018) to calculate the HWI and provide future 186 projections of haze conducive and clear weather using the HWI. However, our analysis is based 187 on an underlying assumption that the large-scale meteorological conditions, which are used as 188 a basis for the HWI, will have a similar influence on the air quality of the NCP in the future climate as for present-day climate. 189

In this paper, we first examine the application of the HWI as a proxy for haze conducive
 and clear weather over NCP for the current climate using a suite of observations (Section 3).
 We then provide the projections of the haze conducive (HWI >1) and clear weather (HWI <-
 frequency over NCP for the historical and future period. We assess the impact of model





Figure 1 Average wind speed at (a) 850 hPa and (b) 500 hPa pressure level. The red dot
 represents the location of Beijing and black rectangle shows the location of the NCP. This
 figure has been repeated for a longer average period, i.e. 1979-2019 (not shown) and the result
 is similar.

205 2. Data & Methods

206 2.1 Observations, Reanalysis Outputs and PPE Model Simulations

Hourly PM_{2.5} concentrations are used from the US embassy site for Beijing for DJF 207 from 2009-2017. Daily mean PM_{2.5} concentrations are constructed using hourly data to identify 208 209 hazy and clear days and evaluate the performance of the HWI as a representative of haze conducive and clear weather conditions for Beijing (see Section 3). We also used newly 210 211 released gridded daily PM_{2.5} concentrations for DJF from Chinese Air Quality Reanalysis Datasets (CAQRA) provided by China National Environment Monitoring Centre for 2013-212 2017 (Kong et al., 2021) to test the performance of the HWI across entire China. The CAQRA 213 data has been produced by assimilating surface air quality observations from over 1000 214

monitoring sites in China and is available at a high spatial resolution of around 15×15 km and 215 hourly temporal resolution over China. More details on the validation of the CAQRA dataset 216 217 against the independent station data is provided in (Kong et al., (2021). The visibility data for Beijing (homogenized data for 20 stations in Beijing) is provided by the National 218 219 Meteorological Information Center of China, **Chinese**China Meteorological AgencyAdministration (CMA), for DJF 1999-2018. 220

We used daily ERA-5 reanalysis data of four variables: meridional wind at 850 hPa pressure level (V_{850}), zonal wind at 500 hPa pressure level (U_{500}), temperatures at 850 hPa level (T_{850}) and 250 hPa (T_{250}) to calculate the HWI for DJF 1979-2019. The ERA-5 data used here is available at 0.25° x 0.25° horizontal resolution and hourly temporal resolution (Hersbach et al., 2020).

226 We used a PPE of climate simulations produced using the recent configuration of the UK Met Office's HadGEM3-GC3.05 coupled model (Sexton et al., 2021; Yamazaki et al., 227 2021). The base model used for PPE, HadGEM3-GC3.05, has a horizontal resolution of ~60 228 km with 85 vertical levels. A total of 47 model parameters from seven parameterization 229 schemes were simultaneously perturbed to obtain the PPE (the full list of perturbed parameters 230 is provided in Table 1 of (Sexton et al., 2021). Here, we used daily outputs of V₈₅₀, U₅₀₀, T₈₅₀ 231 and T₂₅₀ for DJF for the historical (1969-2005) and future (2006-2089) under the RCP8.5 232 scenario. In addition, we also assessed internal variability using 200-year control simulations 233 for each PPE member where 1900 boundary conditions were prescribed. Overall, 16 PPE 234 members are available for all the control, historical and RCP8.5 simulations-235

236 **2.2 Calculation of the HWI**-

237 <u>The winter HWI is calculated using the methodology given by Cai et al. (2017).</u> We
 238 analyse the composite differences in the U₅₀₀, V₈₅₀, T₈₅₀ and T₂₅₀ for hazy (PM_{2.5} concentrations)

> 150 µg m⁻³ for Beijing) and clear (PM_{2.5} concentrations < 35 µg m⁻³ for Beijing) days across
China for DJF 2009-2017 (Fig. <u>12</u>) (see <u>next</u>-section <u>3.1</u> for <u>an explanation on the PM_{2.5}
<u>concentration</u> cut-offs values used <u>for PM_{2.5} concentration</u>). <u>here</u>). We also provide the
<u>composite values for these meteorological variables for hazy and clear days separately in Fig.</u>
<u>2.</u>
</u>



245 Figure 1 shows the 2 Winter composites of u-wind at 500 hPa level (U₅₀₀) over China for all available days for which data is available from US embassy station for Beijing for DJF 2009-246 2017 for (a) high PM_{2.5} (>150 μ gm m⁻³), (b) low PM_{2.5} (<35 μ gm m⁻³) concentrations and (c) 247 difference in the zonal wind speed with a dipole pattern suggesting a northward shift in between 248 249 the composites in (a) and (b). (d-f) same as (a-c) but for v-wind at 850 hPa level (V_{850}), (g-i) same as (a-c) but for temperature at 850 hPa level (T₈₅₀), and (j-l) same as (a-c) but for 250 temperature at 250 hPa pressure level (T_{250}). Black rectangles (B1-B5) in the last column show 251 the regions for which spatial means were used for the calculation of the HWI. The blue dot in 252 these columns shows the location of Beijing. 253

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254 <u>During the hazy days</u>, the mid-tropospheric trough (Fig. 1a), weakenedwesterly flow 255 becomes weaker over the NCP as compared to the clear days (Fig. 2a-c). The mid-tropospheric

- trough also moves northwards as suggested by the dipole pattern in Fig 2c, which shows the
- 257 <u>differences in the U_{500} for hazy and clear days. The</u> northerly flow (Fig. 1b), higher
- 258 temperatures in the near the surface is weaker during hazy days as compared to clear days (Fig.

259 2d-f). The lower troposphere and lower temperatures in is relatively warmer during hazy days as compared to clear days (Fig. 2g-i) whereas the upper troposphere (Fig. 1c-d) is cooler over 260 the NCP during hazy days as compared to the clear days. These findings(Fig. 2j-1). The changes 261 262 in these variables are also consistent with the previous studies (e.g. Cai et al., 2017) that showed similar changes in these meteorological variables. Cai et al. (2018) have examined the use of 263 other variables such as geopotential height, boundary layer thickness and local stratification 264 instability and do not find any significant differences in the performance of HWI by inclusion 265 of more weather parameters.for this time period. Therefore, we also use only these four 266 variables for our analysis the calculation of the HWI, which is used as a proxy for haze 267 conducive and clear weather conditions under a future climate. 268

The winter HWI is calculated using the methodology given by Cai et al. (2017). For the 269 270 calculation of observational HWI, we use ERA-5 reanalysis data for the period 1979-2019. We first create a daily DJF time series of each variable for each reanalyses grid point over China. 271 The daily DJF time series is concatenated for the period 1979-2019. A daily standardised 272 anomaly time series is created for each meteorological variable by first removing the daily 273 mean climatology from each day of the time series and then normalising by the standard 274 275 deviation. Spatial averages are then obtained over the relevant boxes (B1 to B5) for each meteorological variable following Cai et al. (2017) (Fig. 1). The HWI time-series is calculated 276 277 by using the following equation:

HWI (t) =
$$U_{500}$$
 (t) + V_{850} (t) + $dT(t)$

279 where $U_{500} = U_{500,B1}(t) - U_{500,B2}(t)$, $V_{850} = V_{850,B3}(t)$, and $dT = T_{850,B4}(t) - T_{250,B5}(t)$. The HWI 280 (t) time series is then itself normalized by its own standard deviation.

For the PPE historical and RCP8.5 simulations, the daily HWI time series is calculated for each ensemble member for DJF for 1969-2089 using the same methodology as used for

283 ERA-5, with the difference being that the normalisation of the PPE time-series (1969-2089) is performed using the historical standard deviation (1969-2005), following Cai et al. (2017). 284 Similarly, the HWI time series is calculated for the PPE pre-industrial control simulations for 285 286 170 model years out of 200 model years (the first 30 years are discarded as model spin-up period). The normalisation of the pre-industrial control time series is performed using the 287 standard deviation for 170 years. The pre-industrial control simulations used here are initialised 288 with past forcings corresponding to the year 1900 and therefore are an approximate 289 representative representation of the internal variability of the current climate as this does not 290 291 take into account any temporal changes in the internal variability from 1900 to the historical and future periods used here. 292

3. Relationship between the Haze Weather Index <u>as an indicator for clear</u> and air quality indicatorshaze conducive weather conditions over the NCP

We determine As the relationship between HWI and PM_{2.5} concentration was originally 295 proposed for Beijing. As visibility is an optical by Cai et al. (2018), we first determine if the 296 297 HWI can be used as a representative of haze (Wang et al., 2006) conducive and available clear weather conditions for a relatively long period (1999-2018) as compared to the present climate 298 for Beijing using (a) PM_{2.5} concentrations, we also correlate the HWI with from the visibility 299 overUS embassy station in Beijing. We then test the relationship between HWI and (b) PM_{2.5} 300 concentrations over entire China to averaged over larger Beijing domain from CAQRA 301 reanalysis and (c) visibility data from the CMA stations in Beijing. We then determine the 302 spatial extent of the region for which HWI can be used as an indicator of air qualityhaze 303 conducive and clear weather conditions using PM2.5 concentrations for China using CAQRA 304 reanalysis data. We use the 25th and 75th percentile values of daily mean PM_{2.5} concentrations 305 to identify the clear and hazy days, respectively for each dataset. For visibility, we use the 306 opposite criterion, i.e. 25th percentile as a threshold for hazy days and 75th percentile as a 307

threshold of clear days, as lower visibility is associated with hazy days and higher visibility
with clear days. The days with daily PM_{2.5} concentration or visibility lying between the 25th
and 75th percentile values are identified as moderately polluted days.

311 3.1 PM_{2.5} concentrations for Beijing versus HWI

Figure 2 We examine the relationship between the daily HWI and PM_{2.5} 312 concentrations for the US embassy station for Beijing. Figure 3 (a) shows that the daily HWI 313 increases linearly with increasing $PM_{2.5}$ concentrations for up to ~150 µg m⁻³ and for $PM_{2.5}$ > 314 150 µg m⁻³, the HWI starts to level-off (note the log scaling in the y-axis). The time-series 315 correlation between the HWI and PM_{2.5} concentration is ~0.58, which is significant at the 1% 316 level. Callahan et al. (2019) have also obtained a correlation coefficient of 0.58 for daily PM_{2.5} 317 concentrations from the U.S. embassy in Beijing and the HWI calculated using NCAR R1 318 reanalysis. 319

The 25th and 75th percentile values of daily mean PM_{2.5} concentrations for the US 320 embassy Beijing station for DJF 2009-2017 are ~35 and ~150 µg m⁻³ respectively. We 321 determine the percentage of hazy days (with daily mean $PM_{2.5}$ concentrations >150 µg m⁻³) and 322 clear days (with daily mean $PM_{2.5}$ concentrations < 35 µg m⁻³) for different HWI ranges (Fig. 323 2b3e). Out of all days with HWI >1, 64% have daily mean $PM_{2.5}$ concentrations > 150 µg m⁻³ 324 and 98% with PM_{2.5} concentrations >35 μ g m⁻³. This suggests that for HWI >1, almost all days 325 are hazy or moderately polluted. Similarly, almost all days with HWI < -1 are clear or 326 moderately polluted. Using HWI thresholds of ± 1 demarcates between the clear and hazy days, 327 i.e. almost no clear days occur for HWI >1 and almost no hazy days occur for HWI <-1. 328

We have also examined the relationship between the individual variables in the HWI (section 2.2) and PM_{2.5} concentrations observed at the US embassy in Beijing/CAQRA and find that the individual components have correlation values that are similar to or less than that of those used in the combined HWI. Also, physically multiple favourable weather conditions,
 as represented by each of these variables, collectively provide a conducive setting for haze.
 Hence, we focus on the HWI as a combined index rather than its individual components.





Figure 3 HWI versus daily mean (a) PM_{2.5} concentrations for the US embassy Beijing station for DJF 348 2009-2017 (b) PM_{2.5} concentrations spatially averaged over the region around Beijing (116.15-116.65 349 350 °E, 39.65 - 40.15 °N) from CAQRA reanalysis for DJF 2013-2017 (c) visibility averaged over 20 stations from the CMA for DJF 1999-2018 and (d) PM_{2.5} concentrations spatially averaged over the 351 NCP (36-43.5 °N, 107-122 °E) from CAQRA reanalysis. Blue lines show the 25th and 75th percentile 352 thresholds used to define clear and hazy days for each dataset. Percentage of clear, moderately polluted 353 and hazy days for different HWI ranges for the (e) US embassy Beijing station for DJF 1999-2018 (f) 354 larger Beijing domain (116.15-116.65 °E, 39.65 - 40.15 °N) from CAQRA reanalysis for DJF 2013-355 2017 (g) Beijing for DJF 1999-2018 (h) NCP from the CAQRA reanalysis for DJF 2013-2017. 356

357 <u>3.2 Visibility for Beijing versus HWI</u>

As visibility is an optical representative of haze (Wang et al., 2006) and the data for visibility is available for a relatively long period (1999-2018) as compared to the PM_{2.5} concentrations, we also correlate the HWI with the visibility over Beijing. Figure 3**3.2** Visibility for Beijing versus HWI

Figure 2 (c) shows that the HWI is inversely related to the visibility for the Beijing station. The time-series correlation between the HWI and visibility is -0.63, which is significant at the 1% level. The days with visibility < 8.5 km are identified as hazy days, days with visibility > 23.8 km are identified as clear days. For days with HWI > 1, no clear days occur and similarly for days with HWI< -1, only 6% of days are hazy (Fig 2d3g). This further confirms that the correlation between the HWI and haze is significant for a longer period (1999-2018) using visibility as a metric for haze (alternative to the PM_{2.5} concentrations used above).

369 **3.3** PM_{2.5} concentrations over North China Plain versus HWI

We now determine the spatial extent for which HWI can be used as an indicator of <u>haze</u> clear or haze conducive conditions using $PM_{2.5}$ concentrations using data from CAQRA reanalysis. We correlate the daily time-series of $PM_{2.5}$ concentration at each grid point with the HWI for DJF 2013-2017 (Fig. <u>34</u>). Over the entire NCP (36-43.5 °N, 107-122 °E), the correlation coefficient between the daily HWI and gridded $PM_{2.5}$ concentration is ~0.7, significant at the 1% level. The correlation is considerably lower but still significant over other eastern China regions, e.g. north easternmost China and the Sichuan Basin (27-32 °N, 102-107

377 °E).



Figure 34 Spatial distribution of correlation between winter PM_{2.5} concentrations and HWI time series
 at each grid point. Blue dot shows the Beijing station (39.3 °N, 116.4 °E) and the black rectangle shows
 the North China Plain (36-43.5 °N, 107-122 °E).

Considering daily mean PM_{2.5} concentrations averaged over the NCP, we also find a 383 linear relationship with the daily HWI (r = 0.66; significant at the 1% level; Fig 2e). The values 384 of PM2.5 concentrations for NCP are lower as compared to the station values of PM2.5 385 concentrations at the US Embassy Beijing and the correlation coefficient is higher. This could 386 be due to the different time periods for the two dataset, i.e. 2009-2017 for the US embassy and 387 2013-2017 for the CAQRA reanalyses, and spatial averaging of PM2.5 concentrations over the 388 389 NCP region.2d). We also calculate the percentage of clear and hazy days for different HWI ranges for the larger domain of the NCP using the 25th and 75th percentile values, respectively. 390 391 The percentage of hazy and clear days for HWI > 1 and HWI < -1 for NCP in CAQRA 392 reanalyses are very similar to the values obtained for the US embassy Beijing station (Fig 2f3h).

393 Overall, our results confirm that the daily HWI has a robust relationship with daily 394 PM_{2.5} concentrations not only for the Beijing station but across the NCP for the given time periods. Therefore, we use HWI > 1 as a thresholdproxy for hazy dayshaze conducive weather and HWI < -1 as a threshold-ofproxy for clear daysweather across the NCP region. This threshold is also consistent with several other studies (e.g., Cai et al., 2017; Callahan and Mankin, 2020; Callahan et al., 2019), that have used HWI >1, as a cut-off for hazy dayshaze conducive weather for Beijing. We now use the HWI to calculate the frequency of hazyhaze conducive weather (HWI >1) and clear conditionsweather (HWI <-1) for the past and future using ERA-5 reanalysis and PPE members.

402 4. Historical and future changes in the frequency of hazyhaze conducive and clear 403 conditionsweather occurrence

404 The changes in the number of hazyfrequency of haze conducive weather (HWI>1) and clear days per winter, as defined by HWI thresholds, weather (HWI <-1) from the ERA-5 405 406 reanalyses and the PPE are shown in Fig. 45. For ERA-5, the frequency of hazy dayshaze conducive weather has increased, whereas the frequency of clear daysweather (HWI<-1) has 407 reduced for the period 1979-2018. The mean frequency of hazy dayshaze conducive weather 408 409 using 16 PPE members shows a relatively larger increase than ERA-5 for the same 1979-2018 410 time-period (Fig. 4a5a). In contrast, the mean frequency of clear daysweather from the PPE 411 for this period shows a similar reduction to that obtained using the ERA-5 reanalyses (Fig. 412 4b5b).

We examine the changes in the frequency of hazyhaze conducive weather (HWI>1) and clear daysweather (HWI<-1) for the historical (1979-2005) and three future periods, i.e. near (2006-2032), mid (2033-2059) and far (2060-2086) future. The mean frequency of hazy daysfor haze conducive weather is 14.7 days per winter obtained from the ERA-5 reanalysis and 15.0 days per winter from the PPE mean for the historical period. The corresponding values for clear daysweather are 15.0 days and 15.2 days per winter for ERA-5 and PPE, respectively. This shows a good agreement between the mean frequencies of hazyhaze conducive and clear
 days for the ERA-5 data and the PPE mean for the historical period.



Figure 45 Frequency of hazy (haze conducive weather (HWI>1, pink line) and clear days (weather (HWI<-1, blue line) per winter from ERA-5 reanalysis (1979 to 2018). Year 1979 represents period from 1 December 1979 to 28 February 1980 and so on. For each winter (DJF), we calculate the total number of hazy days with HWI >1 as proxy for haze conducive weather and clear days with HWI <-1 as proxy for clear weather conditions. Grey lines show frequencies from 16 individual PPE members and black line shows the mean of frequency using all 16 PPE members for 1969-2087 under the RCP8.5 scenario. Linear trend is calculated using the line of best fit.

- 429 The mean <u>frequency of haze conducive weather for near, mid and far future is 17.9</u>,
- 430 <u>18.6 and 19.9, respectively. The mean frequency for the same future periods for clear weather</u>
- 431 is 13.2, 12.2 and 10.8, respectively (Fig. 6a). The mean change in the frequency of hazy days

432 averaginghaze conducive weather averaged across all PPE members is 2620%, 24% and 33% for the near, mid and far future respectively as compared to the historical period, suggesting 433 that the frequency of hazy dayshaze conducive weather will likely increase for all future 434 435 periods (Fig. 5). However, there exists a very large range in the projected change for all three future periods suggesting internal variability or parametric effect could influence the 436 437 future projections of haze conducive weather. For the near and mid future, hazy-days with <u>HWI>1</u> are projected to change by -81% to 6541% and -12% to 65% across the 16 PPE 438 members, respectively, as compared to the frequency for the historical period. For the far 439 440 future, the range of projected change is even larger, and an increase of $\sim 87\%$ in the frequency of hazy dayshaze conducive weather is also possible. It should be is noted that, for all three 441 442 periods, only one of the sixteen ensemble members suggests a decrease(E16 shown in daily 443 haze Fig. 10) shows a reduction in the haze conducive weather frequency. whereas other ensemble members show an increase in frequency for all periods. For the historical period, E16 444 ensemble member has a mean frequency of 16.3, which reduces to 16.2, 14.4 and 15.2 for near, 445 446 mid and far future. While E16 ensemble member shows a consistent reduction in mean frequency in future, the reduction is specific to only this ensemble member and is not a general 447 448 feature across PPE members.



Figure 6 (a) Mean frequency of haze conducive weather (HWI>1, pink) and clear weather (HWI<-1, 450 blue) for the historical period (1979-2005), near (2006-2032), mid (2033-2059) and far (2060-2086) 451 future under the RCP8.5 scenario. Circles represent PPE members and triangles PPE mean. Grey box 452 and whiskers show the distribution of 10,000 values of mean frequencies sub-sampled from the control 453 simulation, (b) same as (a) but shows variance across 16 PPE members for each period. For box and 454 455 whiskers, we first randomly sampled 10,000 time series of length 27 years using 2704 years of pre-456 industrial control simulation and calculated 10,000 values of mean frequency. We then randomly subsample 16 mean values (corresponding to the number of ensemble members) from the 10,000 mean 457 values, calculated their mean for (a) and variance for (b). This is repeated 10,000 to obtain a distribution. 458 459 The boxes are at the 25th and 75th percentile and the whiskers at 2.5th and 97.5th percentile of mean and variance distribution. For panel (a), the box and whiskers are comparable only to the ensemble means 460 461 (triangles) and not ensemble members (circles).

462 For clear weather (HWI<-1), the mean change in the frequency averaging across all PPE members is -2113%, -20% and -29% for near, mid and far future, respectively (Fig 56a). 463 Considering the range across the 16 PPE members, the frequency of clear weather for near, 464 mid and far future is projected to change by -4029% to 925%, -36% to 10% and -57% to -9%, 465 respectively. Overall, most ensemble members show an increase in the frequency of hazy 466 dayshaze conducive weather and a reduction in the frequency of clear daysweather for all three 467 468 future periods however. However, negligible change or even the opposite change, though less likely, but possible for all periods. 469

470 Figure 6 Frequency of winter hazy days versus clear days We also determine the influence

471 <u>of anthropogenic climate change and the parametric effect on the frequencies of haze conducive</u>

472 weather (HWI>1) and clear weather (HWI<-1) for the historical as well as the three future

473 periods. As shown in later Section 5, the estimate of interannual variance from the control is

474 representative of all time periods and shows no discernible parametric effect. Therefore, we

475 pool the 16 PPE control simulations to sample the internal variability for box and whiskers

476 <u>shown in Fig. 6 (a) and 6 (b) (see captions for details on resampling).</u>

In Fig. 6 (a), we show the mean frequency of haze conducive weather and clear weather
 for 16 individual PPE members (circles) and PPE mean (triangles). The grey box and whiskers
 represent the range of ensemble mean frequencies that can be explained by the internal

variability. If the PPE mean (triangles) lies within the whiskers (i.e. 95 percentile of the control
distribution) we conclude no influence of anthropogenic climate change on mean frequency
however if the PPE mean lies outside the whiskers, it would represent a climate change signal
in the mean frequency. Figure 6 (a) suggest that the mean frequencies for haze conducive as
well as clear weather lies within the box-whiskers for the historical but lies outside the whiskers
for the three future periods, thereby showing a clear impact of anthropogenic climate change
on the frequencies of both haze conducive and clear weather conditions.

We now examine whether the differences in the mean frequency across different PPE 487 members (shown by circles in Fig. 6a) for a given period can be explained by the internal 488 variability or if the differences in PPE members partly arise due to the parametric effect. The 489 490 triangles in Fig. 6b shows the variance across 16 PPE members, i.e. variance across 16 circles shown in Fig. 6a, for each time period. The whiskers in Fig. 6b show the 95th confidence 491 interval from the control simulation and is representative of the internal variability. For any 492 493 time period, if the PPE member variance (triangle) lies within the whiskers, we conclude that 494 the differences in mean frequencies in Fig. 6a can be fully explained by the internal variability and there is no discernible impact of the parametric effect. However, if the triangles lie outside 495 the whiskers in Fig. 6b, we conclude an impact of the parametric effect on the mean frequency 496 for that period. For the points that lie outside the whiskers in Fig. 6b, we also quantify the 497 percentage of variance that can be explained by the internal variability and parametric effect. 498 For any time period, the variance in ensemble mean due to the parametric effect is simply 499 calculated as follow and the remaining variance is attributed to the internal variability. 500 Total variance in the ensemble mean – Mean variance from the control simulation $\times 100$ 501 Total variance in the ensemble mean Figure 6b shows that the variance in PPE mean frequency for historical and future 502 periods lies within the range sampled by the internal variability for both haze conducive 503



512 <u>respective variance in the frequencies arise due to the parametric effect.</u>



513

Figure 7 Frequency of haze conducive weather (HWI>1) versus clear weather (HWI<-1) averaged over
 the historical period (1979-2005) and the far-future (2060-2086) period under RCP8.5 using all PPE
 members. Circles denote individual PPE members whereas triangles denote the mean of the members.
 Grey triangle shows mean frequency from ERA-5 reanalysis for the historical period (1979-2005). The
 black solid line shows the 1:1 (identity) line.



520 changes in the frequency of hazy dayshaze conducive weather (HWI>1) versus clear days for

521 a given winter.weather (HWI<-1). The average frequency of hazy dayshaze conducive and 522 clear days-weather frequency over the historical period are almost equal for each PPE member 523 (Fig-6. 7). All PPE members show a higher frequency of hazy days for haze conducive weather 524 than clear daysweather under the far future (2060-2085), however, there exists a substantial range in this change. The frequency of winter haze daysconducive weather can be similar or 525 up to 3.5 times the frequency of clear daysweather conditions (Fig. 67). Similar results are also 526 obtained for the near and mid-future. Averaged across the PPE members, the number of 527 hazyhaze conducive days can increase by ~2 times as compared to the number of clear days in 528 529 future. As noted in Fig. 67, the spread in the haze conducive weather frequency of hazy days amongst individual ensemble members is also larger for the far future (2060-2086) compared 530 to the historical period. This suggests a larger uncertainty and a larger range of possible future 531 532 meteorological conditions affecting haze and air quality as compared to the historical period. 533 Other studies have (e.g., Cai et al., 2017; Callahan and Mankin, 2020) also found similar 534 increases in the frequency of hazy dayshaze conducive weather for the future. However, the 535 range of projected change differdiffers substantially across models as well as ensemble members. In our study, in addition to the frequency of hazy dayshaze conducive weather, we 536 also evaluate the changes in the frequency of clear daysweather across different future periods 537 and compared the relative changes in both the frequencies, which is not examined in the past 538 studies. 539

540 5. Role of individual meteorological variables

We now investigate the role of individual constituent meteorological variables in driving the changes in the distribution of the HWI as well as individual constituents of the HWI between the far-_future (2060-86) and the historical (1979-2005) period. The probability distribution of the HWI shows a shift in the distribution towards higher magnitudes for the farfuture as compared to the historical period (Fig. 8). This implies an increased frequency in 546 hazyof haze conducive weather, as the number of days, as values with HWI >1 increase. A similar shift is apparent in the zonal-mean wind (U_{500}) and the vertical temperature profiles 547 (dT), whereas no apparent shift is noted in V_{850} suggesting a relatively less important role of 548 V_{850} in driving the future changes in the HWL. We also find that the shift in the HWI, as well 549 as U₅₀₀ and dT distribution, is not due to the shift in one particular PPE member or time period. 550 It is consistent across the 16 PPE members and is continual over time from the historical to the 551 far-future period. Therefore, for the PPE analysed here, the changes in the haze conducive 552 weather (HWI>1) is largely associated with the changes in the U₅₀₀ and dT, and V₈₅₀ appear to 553 554 have a less important role. Despite using a multimodel ensemble and a different time-period than used here, a similar result with a relatively larger shift in the PDFs of U_{500} and dT as 555 compared to V_{850} can also be noted in the Cai et al. (2017). 556



Figure 78 Probability Distribution Functions (PDF) for the winter HWI, meridional winds at 850 hPa pressure level (V_{850}), zonal winds at 500 hPa pressure level (U_{500}) and temperature gradient between the lower and upper troposphere (dT). <u>The PDF for the HWI is created using</u> the daily DJF time series of all 16 PPE members. PDFs for V_{850} , U_{500} and dT are created using daily DJF values the normalized daily DJF time series of each variable calculated for the HWI

(see section 2.2 for details) and represents the constituent variables of the HWI. Blues bars
 show the PDFs for the historical (blue)period and red for the far-future (red)-under the RCP
 8.5 by pooling in all 16 PPE members.scenario. Blue and red solid lines show the mean values
 of the PDF for historical and far future, respectively.

567 <u>65</u>. Interannual variability in <u>the frequency of hazyhaze conducive</u> and clear weather 568 <u>conditions</u>frequency

Large interannual variability in the frequency of hazyhaze conducive (HWI>1) and clear daysweather (HWI<-1) is apparent in both individual PPE members and ERA-5 reanalysis (Section 4). Therefore, we examine the changes in the interannual variance of the frequencies for future periods as compared to the historical period. We also compare the variance in historical and future time-_periods with the variance in the control simulation to discern the influence of the model physical parameterisations, i.e. parametric effect, on the variance.

The interannual variance for ERA-5 data is 27 days² and 39 days² for hazy and clear 576 577 days, respectively, for the historical period (1979-2005) (triangles in Fig. 8a-b). The interannual variance in the frequency of hazy days derived from the PPE members for the 578 historical period is larger than that for the ERA-5, whereas for the clear days the variance for 579 ERA-5 lies within the range of the PPE members. No consistent change in the interannual 580 variance of hazy days is noted for any of the PPE members (note the changes in colour ranking) 581 from the historical to the future periods suggesting little influence of the parametric effect on 582 the interannual variance of hazy days. 583



Figure 89 Interannual variance in frequency of winter (a) hazyhaze conducive weather (HWI>1) and 586 (b) clear <u>daysweather (HWI<-1)</u> for the control simulation, historical (1979-2005), and near (2006-587 2032), mid (2033-2059) and far-future (2060-2086) under RCP8.5 for all 16 PPE members. Coloured 588 circles are for individual PPE members and triangles for ERA-5 reanalysis. (c-d) are same as (b) but 589 with \log_{10} and square root power transformations. For (c-d), we first calculate the \log_{10} of (1+frequency) 590 and square-root of the frequency of clear days for the control simulation and each time-period, and then 591 592 estimate variance for each respective period. The length of control simulation and all future periods is 593 the same as historical, i.e. 27 years. The 27 years used for control here are randomly selected from 170-594 year control simulation for each member.

595	The interannual variance for ERA-5 data is 27 days ² and 39 days ² for haze conducive
596	and clear weather, respectively, for the historical period (1979-2005) (triangles in Fig. 9a-b).
597	The interannual variance in haze conducive weather frequency derived from the PPE members
598	for the historical period is larger than that for the ERA-5, whereas for the clear weather the
599	variance for ERA-5 lies within the range of the PPE members. No consistent change in the

<u>interannual variance of haze conducive weather is noted for any of the PPE members (note the</u>
 <u>changes in colour ranking) from the historical to the future periods suggesting little influence</u>
 of the parametric effect on the interannual variance of haze conducive weather.

In contrast, the frequency of clear daysweather for most PPE members show a marked 603 reduction in the interannual variance from historical to near-future (Fig. 8b9b). However, as 604 605 the frequency of clear days weather show a decreasing trend in time (see Fig. 4b5b), the mean frequency would be expected to reduce for the three future periods. Also, the reduction in 606 607 variance could arise as the frequencies of clear daysweather approach their lower bound of zero. With count data, a power transformation is often applied to stabilize the variance across 608 all time periods. We tried applied two power transformations, i.e. $\log_{10}(1+x)$ and square-root 609 (x), where x is the count data (Fig. $\frac{8e9c}{c}$ -d). We find the spread in the variance in the control 610 611 simulation across the PPE members is comparable with the historical as well as future periods 612 (Fig. <u>8e9c</u>-d). Note that for control simulation we randomly selected 27 years (length same as historical and future periods) from 170 years of control simulation from each PPE member, 613 however, we note comparable variance for the other randomly selected samples. Figure 89 (c-614 d) also shows that the individual PPE members show inconsistent changes in the variance 615 616 (noting changes in the colour ranking) from control to historical and future periods. Therefore, 617 no robust changes in the interannual variance of hazy or log_{10} (1+ frequency of haze conducive 618 and clear days)weather can be detected from control to historical and future periods. This 619 means we can use the variance in the control simulation as a representative estimate of internal variability. This enables us to quantify the influence of the parametric effect and anthropogenic 620 climate change on the mean frequencies (see previous section) and trends in frequencies (see 621 622 next section) across the different periods (see next section).

623 7<u>6</u>. Influence of the parametric effect and anthropogenic climate change <u>and parametric</u> 624 effect on trends

625 We discern the influence of the parametric effect and anthropogenic climate change and parametric effect on the future projections of the trends in the frequency of hazyhaze conducive 626 weather (HWI >1) and clear days. Figure 9 shows the weather (HWI <-1). The time series of 627 628 the frequency of winter hazyhaze conducive and clear days-weather frequency from ERA-5 and the 16 PPE members for the historical and future periods- is shown in Fig. 11 (a) and 11 629 (b). The 95th percentile values (blue shaded region) and the range (blue dotted lines) in the 630 frequency of hazyhaze conducive and clear days weather frequency from the respective control 631 simulation for each PPE member are also shown. 632

633 For hazy days, haze conducive weather (HWI>1), the time series for selected PPE members (e.g. E3, E4) show increasing positive trends. In particular, towards the end of the 634 21st century (Fig. 9a10a), the lower half of the control range is seldom sampled and more than 635 the expected number of values lie above the 97.5th percentile of the control frequencies. In 636 contrast, for other PPE members (e.g. E8, E10), the full time series sample the control 637 distribution evenly throughout the full period. For clear days, weather (HWI<-1), some 638 members (e.g. E3, E4) show a clear reduction during the 21st century whilst others (e.g. E16) 639 show that no trend and explore the control distribution evenly (Fig 9b10b). 640

We first examine In Section 4, we examined the influence of the anthropogenic climate change and parametric effect on the mean frequencies. The analysis of mean frequencies provides an estimate of the accumulated influence of climate change on frequencies with respect to the control simulations whereas analysis of trends would provide a better estimate of changes within a selected time period. Therefore, we apply the same analysis on the trends in the frequencies (Fig. 11).



653 conducive and clear weather for the pre-industrial control simulation of 170-years.

654 We calculate the ensemble mean trend obtained from the 16 individual PPE member trends to determine the influence of climate change for the historical period (see captions of 655 Fig. 11 for details). We describe the evolution of the historical trend for three equal-length 656 future time periods (i.e. near, mid and far future) and examine if the historical trends are 657 sustained across the 21st century and if the trends are discernible outside the range described 658 659 by the internal variability (Fig. 11a-b). The grey whiskers in Fig. 11 (a) and (b) cover the range 660 of trends that can be noted in section 6, for hazy days and log₁₀ (1+explained by internal variability and any trend values lying outside the grey whiskers represent the influence of 661 662 anthropogenic climate change. The mean trend in the frequency of clear days), the estimate of interannual variance 663 from the control is representative of all time periods and shows no discernible both haze 664 conducive (HWI>1) and clear weather (HWI <-1) for the historical period (1979-2005) lie 665 outside the 95% confidence interval of the control simulations. This suggests that the trends 666 667 noted for the historical period cannot be explained by internal variability alone and there is a substantial impact of anthropogenic climate change on the historical trends. The trends in haze 668 conducive weather lie within the envelope of internal variability for the three future periods 669 analysed here implying that the historical trend is not sustained over the 21st century and 670 indistinguishable from the internal variability for the future. Figure 11 (a) also shows a positive 671 mean trend in haze conducive weather (HWI>1) for historical, near and mid future, but a weak 672 673 negative trend for far future. While the frequency of haze conducive weather increases for all three future periods with respect to the historical period as shown in Fig. 6a, the trends only 674 show an increment or reduction for that period as these are not referenced to the historical 675 period. Therefore, trends could still be negative within any selected period, as in the case of 676 the far future. In contrast, the mean trends in clear weather frequency for near (2006-2032) and 677 mid future (2033-2059) lie outside the 95% confidence interval of the control simulation. This 678

679 <u>shows that for clear weather frequency (HWI<-1), the historical trend is sustained over the first</u>
680 <u>half of the 21st century and then it levels off.</u>

681 We now examine the influence of the parametric effect. Therefore, we pool on the 16 PPE control simulations to sample trends in the internal variability. frequency of haze conducive 682 and clear weather. In Fig. 10 (all (c) and (bd), we show the variance in trends for the time 683 684 series resampled using the control simulation (see captions for details on resampling). The grey box and whiskers show the 95th confidence interval of the control variance used to represent 685 the internal variability. The variance in PPE trends calculated using 16 PPE members for 686 687 selected time periods is overlaid (circles). In Fig. 10 (a-b11 (c-d), if the variance for historical or future periods lies outside the whiskers, we conclude an impact of the parametric effect on 688 the trends. However, if the variance across the 16 PPE members lies within the whiskers, we 689 conclude no impact of the parametric effect on the trend. Note that the variance in trends for 690 clear daysweather is in log-transformed space. As can be seen in Fig. 10a11c and 10b11d, the 691 variance in PPE trends for historical and future periods lies within the envelope95th percentile 692 distribution of the internal variability for both hazyhaze conducive and clear daysweather. 693 Therefore, we do not find any discernible influence of the parametric effect on the trends in the 694 695 frequency of hazy and clear days frequencies.



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704 Figure 10 Variance in 11 Mean PPE trends for the frequency of winter (a) hazyhaze conducive 705 weather (HWI>1) and (b) clear days.weather (HWI<-1) for winter. Circles show the variance 706 immean trends from 16 PPE members for the historical (1979-2005) and near (2006-2032), mid 707 (2033-2059) and far (2060-2086) future under the RCP8.5 scenario. Grey box and whiskers show the distribution of 10,000 values of variance in trends sub-sampled from the control 708 709 simulation. (c-d) same as (a-b) but variancemean is replaced by meanvariance in trends. For 710 box and whiskers, we first randomly sampled 10,000 time series of length 27 years using 2704 years of pre-industrial control simulation and calculated 10,000 values of trends. We then 711 randomly sub-sample 16 trends values from the 10,000 trend values and calculate the variance 712 and mean of 16 trend values. The boxes are at the 25th and 75th percentile and the whiskers at 713 2.5th and 97.5th percentile of mean and variance distribution. For clear days, the frequencies 714 were transformed to log space by applying a power transformation of $\log_{10} (1 + \text{frequency})$ 715 before calculating trends. 716

717 We now examine the influence of anthropogenic climate change on the trends in the frequency of hazy and clear conditions. We calculate the mean trend obtained from the 16 718 individual PPE member trends (Fig. 10c-d), to determine the influence of climate change across 719 selected time periods. The grey whiskers in Fig. 10 (c) and (d) cover the range of trends that 720 can be explained by internal variability and any trend values lying outside the grey whiskers 721 represent the influence of anthropogenic climate change. 722 The mean trend in the frequency of both hazy and clear days for the historical period 723 (1979-2005) lie outside the 95% confidence interval of the control simulations, suggesting a 724

- substantial impact of anthropogenic climate change on the historical trends in the PPE.
- Similarly, the mean trends for clear days for near (2006-2032) and mid future (2033-2059) lie 726
- outside the 95% confidence interval of the control simulation. Thus, we find the impact of the 727

climate change on both hazy and clear days. However, it is only discernible for specific periods
 due to the underlying large internal variability in the frequency of hazy and clear days.

730 **87**. Conclusions

In this study, we elucidate for the first time the influence of model physical 731 parametrisations, in addition to internal variability and climate change, on the future hazyhaze 732 733 conducive and clear weather conditions over the North China Plain (NCP) using the Perturbed 734 Parameter Ensemble (PPE) from the Met Office HadGEM3-GC3.05 model. We use a meteorology-based daily Haze Weather Index (HWI), which has been previously used by a 735 736 number of studies to examine the changes in winter (December-February) haze conducive and clear weather conditions for past and future over Beijing the NCP using a large-scale 737 738 meteorology-based daily Haze Weather Index (HWI). We first identify the regional extent of the application of the HWI over China. We find that the HWI >1 can be used as an indicator 739 740 of hazy and haze conducive weather conditions and HWI<-1 as an indicator of clear weather 741 conditions for the entire NCP due to the spatial coherence of regional meteorological conditions over this region. 742

743 The PPE shows that under the RCP8.5 emission scenario, the mean frequency of hazy dayshaze conducive weather (HWI>1) can increase by up to ~65% in the near (2006-2032) and 744 mid (2033-2059) future and by ~87% in far- future (2060-2086) as compared to the historical 745 746 period (1979-2005). In contrast, the frequency of clear <u>daysweather (HWI<-1)</u> can reduce by up to ~40% in the near and mid-future and by ~57% in the far- future. However, the opposite 747 change of relatively lower magnitude or negligible change in frequency of hazyhaze conducive 748 749 and clear weather, though less likely, is possible. The absolute number of hazy days for with haze conducive weather in the far future can remain the same or up to \sim 3.5 times higher than 750 the clear days for any given winter. weather over the NCP. There also exist a large interannual 751

752 variability in the frequency of hazyhaze conducive and clear weather conditions. However, no 753 systematic change in the interannual variance of the frequency of hazy or clear days frequencies 754 is projected noted in future as compared to the historical period. We also find that the future 755 changes in hazy or clear the haze conducive weather conditions are largely influenced by(HWI>1) for the future is associated with the changes in the mid-tropospheric zonal wind 756 757 component and strong vertical temperature gradient between the lower to upper troposphere over the North China Plain.NCP. We do not find any discernible a consistently growing 758 influence of the anthropogenic climate change and parametric effect on the mean haze 759 conducive and clear weather frequencies across the 21st century. This suggests that in addition 760 to the internal variability, the parametric effect adds as an additional source of uncertainty in 761 762 future projections of trends in the frequency of hazy and clear days. However, we haze conducive and clear weather, particularly towards the end of the 21st century. We find that the 763 impact of anthropogenic climate change on the is discernible in trends for both hazy and clear 764 days for the historical and specific future period for haze conducive weather and up to mid of 765 the 21st century for clear weather. Beyond these periods, suggesting climate change can 766 exacerbate the increase in the number of hazy and the reduction in the number of clear days in 767 future. the historical trends are not sustained and not distinguishable from the internal 768 variability. 769

This study considers four atmospheric variables to examine the changes in future hazyhaze conducive and clear weather conditions, however, other atmospheric variables (e.g., boundary layer height) or processes may also influence the hazy or clear weather conditions.occurrence of haze. Furthermore, even though our study shows the potential for an increase in hazyhaze conducive weather conditions and a reduction in clear weather conditions for the future periods examined using, the HWI, theactual formation of haze also depends will depend on future emissions of air pollutants and their precursors. If the source emissions are cut-off or reduced in the future, the risk of haze formation would naturally reduce.
Nevertheless, the projections of changes in the frequency and interannual variance in haze
conducive weather conditions can be very useful for developing successful adaptation and
mitigation policies for the future that consider both emissions and climate change, and therefore
can be beneficial for near and long-term planning and decision-making in relation to improving
future PM_{2.5} air quality.

783 Data Availability

The Copernicus Climate Change Service (C3S) (2017): ERA5: Fifth generation of ECMWF atmospheric reanalyses of the global climate data are available through Copernicus Climate Change Service Climate Data Store (CDS) (https://cds.climate.copernicus.eu/). The PM2.5 concentrations for the US Embassy station in Beijing are archived at the following website (http://www.stateair.net/web/historical/1/1.html). The haze weather index time series for PPE and visibility data used in this paper can be obtained from the authors. The CAQRA dataset can be freely downloaded at https://doi.org/10.11922/sciencedb.00053.

791 Author Contribution

SJ and RMD conceived and designed the manuscript; DS conducted PPE simulations using Met Office HadGEM model; LP provided the visibility data; SJ performed data analysis, produced figures, wrote the first draft; all co-authors provided comments on the manuscript and contributed to writing.

796 Competing interests

797 The authors declare no financial or non-financial conflict of interest.

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