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Factors Affecting Relative Humidity and Its Relationship with the Long-Term Variation of Fog-Haze Events in the Yangtze River Delta

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Abstract: Relative humidity (*RH*) is one of the most important parameters in the study of fog-haze. This paper first estimates the contributions of the key quantities (temperature, water vapor, air pollutant) and their combinations to the relative change of *RH*, and then investigates the relationships of *RH* with the long-term variation of haze and fog days based on the meteorological data in the Yangtze River Delta over the period 1970-2010.The main conclusions are as follows. (1) Temperature is the foremost factor influencing *RH*, with the effect of specific humidity and direct contribution of air pollutant being second and third. (2) *RH* shows a prominent descending trend in the Yangtze River Delta, due to global warming and the ‘analogous heat island effect’ (AHIE). (3) Decreasing *RH* was responsible for the reduction of fog days. The AHIE can explain the phenomenon in which fog days in metropolises are less than in small cities. (4) Granger causality analysis of inter-annual variation further shows that increasing aerosol loading Ganger-causes the increasing haze days, rather than meteorological parameters such as RH. High *RH* would enhance hygroscopic growth of particle and then suppress the planet boundary layer height (*PBLH*) and lead to more haze days, lower *PBLH* further increases aerosols and *RH*, this positive feedback mechanism can be established under the condition of aerosol loading being relatively stable and beyond the threshold for haze formation. (5) If aerosol emissions maintain the status quo, climate cooling would result in serious fog-haze events occurring more frequently in Yangtze River Delta. The research provide a scientific basis for understanding the influence of meteorological factors on *RH* and the connection between the variation of long term fog-haze days and *RH* under the background of climate change.

Keywords: relative humidity; contribution efficiency; impact factors; fog and haze days; climate change.

1 Introduction

Under the background of global warming, emissions of anthropogenic aerosols have increased substantially in China over the last two decades due to rapid economic development and urbanization. (Richter et al., 2005; Yoon et al., 2011).Over the same period, an increase in haze days (Che et al., 2009; Zhao et al., 2011; Ding and Liu, 2014; Zhang et al., 2015) and a reduction in fog days (Chen et al., 2006; Ding and Liu, 2014; Yin et al., 2015) have become progressively more common in China, especially in the densely populated economic zones of the Pearl River Delta, Yangtze River Delta, and Beijing-Tianjin-Hebe region. Many studies have been conducted to determine the causes and formation mechanisms of fog and haze, with the results showing that they are closely connected to pollutant concentrations, surface meteorological conditions, and climate change. On one hand, it has been found that fog formation can be enhanced by abundant water vapor (Niu et al., 2010), radiative cooling (Brown and Roach, 1976; Bergot and Guedalia, 1994), temperature inversions and stable weather (Zhang et al., 2005), adequate supply of aerosols as condensation nuclei (Sachweh and Koepke, 1995; Ming and Russel, 2004; Mohan et al., 2009), as well as climate change (Chen et al., 2006; Klemm and Lin, 2016). On the other hand, some conditions can lead to the occurrence of haze, such as the increase of primary and secondary aerosols (Li et al., 2010; Guo et al., 2014; Huang et al., 2014), aerosol hygroscopic growth (Kim et al., 2006; Pan et al., 2009; Fu et al., 2014) and gas-particle conversion (Yue et al., 2015; Shen et al., 2010). Climate change has also been found to affect haze occurrence (Jacob and Winner, 2009; Wang and Chen, 2016; Cai et al., 2017) including the maintenance of stable weather and atmospheric circulation (Xu et al., 2011; Zhao et al., 2013), and a weakened East Asia monsoon (Cao et al., 2015; Li et al., 2016). Despite the progress in identifying the potential factors affecting fog-haze events, determining the relative contributions of these factors to the development and evolution of fog-haze events remain a challenge.

Relative humidity (*RH*), defined as the ratio of actual vapor pressure to the saturated vapor pressure (Wallace and Hobbs, 2006), is one of the important factors in distinguishing fog and haze. Based on the criterion given by the China Meteorological Administration (CMA, 2003), when visibility is <10 km and *RH* <80%, the air condition is recognized as haze. When the visibility is <1 km and *RH* >90%, the air condition is defined as fog. *RH* is not only a key factor affecting fog and haze process but also plays an important role in the interactive transformation mechanism between them. The increase in anthropogenic aerosol emissions, global warming, urban heat island effect, and the variation of atmospheric water vapor all have an impact on *RH*. However, few studies have quantified the contribution of each factor, and their combinations, to the changes in *RH*. If the theoretical formula for the relationships between *RH* and single or multiple meteorological parameters can be deduced, a variety of sensitivity experiments could be conducted with different parameter combinations, and the results could be applied to investigate actual fog and haze historical trends, it would improve our understanding of the long-term changes in *RH* and fog-haze events.

In this study, we investigate the influences on *RH* of temperature, water vapor, aerosols, and their combinations in terms of their theoretical relationships. Taking the Yangtze River Delta as an example, a long-term dataset on temperature, humidity, fog days, and haze days are used to calculate the contributions of each parameter and their combinations to the relative change of *RH*. We identify the real causes of historical trends of fog and haze days based on the results. On this basis, the relationship between fog-haze events and *RH* under the background of climate change are determined, and possible further developments in climate cooling circumstances are predicted from the perspective of changes in *RH*.

2. Data sources, study area, and methodology

The study focus on the area of Yangtze River Delta during the period of 1970 to 2010. The dataset of temperature, visibility and *RH* are obtained from the National Meteorological Information Center (http://www.nmic.cn/web/channel-8.htm). The data they provided has been quality controlled to ensure the accuracy and reliability based on the international universal methods (Feng et al., 2004). According to the criterion issued by the CMA (2003) when the visual range is <10 km and *RH* <80%, conditions are recognized as haze, whereas when the visual range is <1 km and *RH* >90%, fog occurs. When *RH* is between 80% and 90%, the deterioration of visibility is caused by the interaction of haze and fog, but the main component is haze (Wu, 2006). Therefore, *RH* = 90% is used as the criterion to discriminate between fog and haze. The daily 08:00 sounding data in Nanjing from 1973 to 2010 comes from http://weather.uwyo.edu/upperair/sounding.html, daily maximum planet boundary layer height is estimated by Holzworth (1964) methods. The aerosol optical depth (AOD) data in Yangtze River Region are from Zhang et al. (2017) for the period of 1973 to 2010.

Granger causality analysis (Granger, 1969) is a statistical hypothesis test for determining whether one time series is useful in forecasting another. A variable X that evolves over time Granger-causes another evolving variable Y if predictions of the value of Y based on its own past values and on the past values of X are better than predictions of Y based only on its own past values. If X and Y are stationary time series determined by Augmented Dickey-Fuller (ADF) Test (Dickey and Fuller, 1979), Granger causality analysis can test whether the high correlation between X and Y has causality, which will help to reveal the interactive connections. Detailed information can be found in relevant literature (Granger, 1969).

The Yangtze River Delta is an economically developed area with a dense population, covering an area of 2. 107 x 10^5 km^2 including Shanghai, Jiangsu Province, Zhejiang Province, and Anhui Province. According to Guo et al. (2016), the criterion used to distinguish between a metropolis and a small city is based on the urban GDP and population in 2010. A metropolis has GDP over 100 billion RMB or a total population of more than 5 million. Based on these criteria, the cities of Shanghai, Nanjing, Hangzhou, Suzhou, Wuxi, Hefei and other 18 cities in the Yangtze River Delta can be classed as metropolises, the remaining urban areas are classified as small cities.

To calculate the relative change of *RH*, we selected the *RH* of a specific year as a reference and assumed that only one meteorological factor changes, while the others remains constant over the historical period. On the basis of the definition of *RH* (Wallace and Hobbs, 2006), the relative change can be defined as (calculated value-reference value)/reference value × 100%. For example, if *RH* for the reference year is 79%, the calculated *RH* is 80%, then the relative change is (80%-79%)/79% × 100% = 1.3%.

3. Theoretical analysis

Sensitivity experiments are designed to investigate the variation of *RH* with temperature, water vapor, and levels of gaseous pollutants. As shown in Fig. 1, two ideal boxes (Case 1 and Case 2) are considered. Case 1 represents the ideal atmosphere with temperature (T), water vapor mass (m_v) and dry air mass (m_d); Case 2 represents a polluted atmosphere that experienced an increase in temperature (ΔT), water vapor (Δm_v), and gaseous pollutants (Δm) compared to the ideal atmosphere in Case 1. The pressure P is assumed to be the same for both cases.

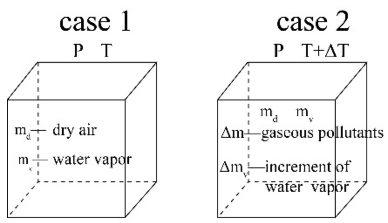

Fig. 1 Illustration of the two cases in the sensitivity experiment

Thus, *RH* in Case 1 (RH_1) is given by

$$RH_1 = [\frac{e}{\mathrm{k}e_s(T)}]_{p,t} \times 100\% \tag{1}$$

e and $e_s(T)$ are the actual and saturated vapor pressure, respectively. K is an enhancement factor that is used to indicate the deviation of the actual atmosphere from the ideal atmosphere (k ≈ 1.004, normal temperature and pressure). The water vapor mixing ratio r defined as the ratio of the water vapor mass (m_v) to dry air mass (m_d) is related to e with

$$r = \frac{m_v}{m_d} \approx \frac{\varepsilon e}{p} \tag{2}$$

where $\varepsilon = 0.622$. Combination of equations (1) and (2) yields

$$RH_1 = [\frac{e}{e_s(T)}]_{p,t} \approx \frac{rp}{\varepsilon e_s(T)} = \frac{m_v}{m_d} \times \frac{p}{\mathrm{k}\varepsilon e_s(T)} \tag{3}$$

Similarly, *RH* in Case 2 (RH_2) satisfies the expression:

$$RH_2 = \frac{m_v + \Delta m_v}{m_d + \Delta m} \times \frac{p}{\mathrm{k}\varepsilon e_s(T + \Delta T)} \tag{4}$$

Thus, the relative change (*RC*) of RH_2 relative to RH_1 is given by

$$RC = \frac{RH_2 - RH_1}{RH_1} = \frac{e_s(T)}{e_s(T + \Delta T)} \times \frac{m_d}{m_d + \Delta m} \times \frac{m_v + \Delta m_v}{m_v} - 1 \tag{5}$$

Equation (5) indicates that given RH_1, *RC* is linearly related to RH_2 and can be used to identify the response of each factor to RH_2.

To verify the effect of the individual factor on *RC*, we conducts a series of sensitivity tests whereby only one factor (Δm, Δm_v, or ΔT) varies. Equation (5) can be simplified as follows accordingly:

$$RC = \frac{m_d}{m_d + \Delta m} - 1 = -\frac{1}{1 + \frac{\Delta m}{m_d}} \tag{6a}$$

when only Δm changes ($\Delta T = 0$, $\Delta m_v = 0$) .

$$RC = \frac{m_v + \Delta m_v}{m_v} - 1 = \frac{\Delta m_v}{m_v} \tag{6b}$$

when only Δm_v changes.

$$RC = \frac{e_s(T)}{e_s(T + \Delta T)} - 1 \tag{6c}$$

when only ΔT changes.

The saturation water vapor pressure is given by use of Teten's empirical expression (Coulson, 1959):

$$e_s(T) = 6.1078\exp[\frac{17.2693882(T - 273.16)}{T - 35.86}] \quad (7)$$

Figure 2 compares the contributions showing *RC* as a function of the relative changes in the individual variables generically denoted by$\Delta X_i/X_i$,.

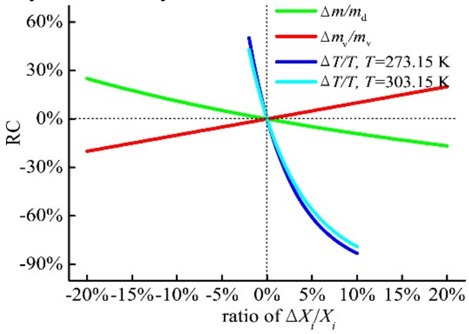

Fig. 2 The influence temperature (blue and light blue), water vapor (red), gaseous pollutants (green) on the relative change of relative humidity(*RC*), X_i stands for three factors.

Several points are evident from Fig 2. First, with an increase in gaseous pollutants, $\Delta m/m_d$ increases, which leads to a decrease of *RC*, it reflects the fact that the introduction of gaseous pollutants reduces *RH*. The lower the $\Delta m/m_d$, the faster *RC* declines. For example, when $\Delta m/m_d$ increases from 0 to 10%, *RC* fall by 9%, while it decreases by only 7% when $\Delta m/m_d$ increases from 20% to 30%. Second, *RC* exhibits a linear upward increase as the amount of water vapor increases, suggesting that water vapor only influences the value of RH_2, while not affecting the change rate of RH_2 as it varies. Third, *RC* declines nonlinearly with increasing $\Delta T/T$, with a faster decrease at a smaller change of temperature. For example, if the initial temperature is 273.15K, $\Delta T/T$ increases from 0 to 2%, *RC* decreases by 32%, while $\Delta T/T$ ranges from 8 to 10%, which only causes a maximum amplitude reduction of 7% for *RC*. Note that the effect of temperature change depends slightly on the reference temperature *T*. For the same variation of $\Delta T/T$, a lower initial temperature (*T*) corresponds to a greater *RC*. For example, if $\Delta T/T$ increases from 0 to 2%, RC declines by 32% for T=273.15 K, and 29% for 303.15 K, respectively. Forth, comparative analysis shows that the influences of water vapor and gaseous pollutants on *RC* are similar in magnitude for the same relative changes, and are far less than that of the temperature change. For example, at T = 273.15 K, $\Delta T/T$ increases from 0 to 2%, *RC* decreases by 32%, while under the same amplitude, the *RH* changes caused by $\Delta m_v/m_v$ or $\Delta m/m_d$ are only about 2%. This suggests that temperature is the most significant factor affecting *RH*, because the exponential dependence of saturation pressure on temperature.

Generally speaking, the air mass is about 1.29 kg per cubic meter, the gaseous contaminant mass is usually in the range of 10^2 to 10^3 μg per cubic meter (Zhao et al., 2013; Guo et al., 2014). Thus, the mass of air taken up by a gaseous pollutants is only about 10^{-7}. However, in addition to gaseous pollutant there are aerosols particles present in the air with greater mass magnitude. For example, according to monitoring records, the pollutant mass of 1016 mg in a cubic meter occurred during a sandstorm in 1993 in Jinchang City, Gansu Province (Weather China, 2014). But even if all these particles are regarded as gaseous pollutants, there is still a huge difference in magnitude between its mass and air mass, and therefore $\Delta m/\Delta m_d$ would not exceed 1‰. Although treating gaseous pollutants as aerosols is an ideal assumption, the mass ratio is still far less than 1. Hence, we could roughly infer that the direct contribution pollutants including aerosols to *RH* is nearly negligible. Thus, we focus hereafter on the contributions of temperature and water vapor only.

When $\Delta m/m_d \approx 0$, equation (5) become:

$$RC = \frac{RH_2 - RH_1}{RH_1} = \frac{e_s(T)}{e_s(T+\Delta t)} \times \frac{m_v + \Delta m_v}{m_v} - 1 \quad (8)$$

Because of the opposite effects of temperature and water vapor on *RH*, to maintain a constant *RC*, relative changes in temperature and *RH* are positively related to each other, as shown in Fig. 3. Also shown in Fig.3 is that as a result of the greater impact of temperature on *RH*, for the same *RC*, $\Delta m_v/m_v$ increases exponentially with $\Delta T/T$ ($\Delta m_v/m_v = 0.0693\times\exp(0.081\times\Delta T/T)$ for $RC=0$). In other words, for *RH* to remain unchanged, a larger increase of water vapor is needed to compensate for the effect of a smaller increase of *T*. For example, at $RC = 0$ and T = 273.15K, $\Delta T/T$ varies from 0 to 4% whereas $\Delta m_v/m_v$ ranges from 0 to 113%. In addition, under the same $\Delta T/T$, the greater the *RC* is, the more water vapor is needed. For example, at RC= -10%, $RC = 0$, and $RC = 10\%$, $\Delta T/T$ increases from 0 to 4%, and $\Delta m_v/m_v$ needs to increase from 0 to 92%, 113%, and 134%, respectively.

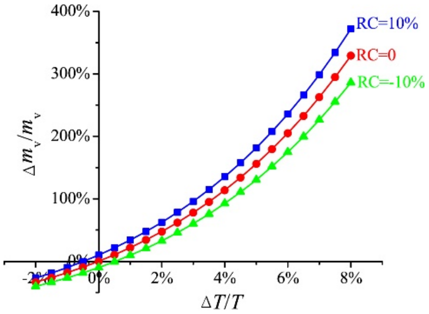

Fig. 3 The relationship between $\Delta m_v/m_v$ and $\Delta T/T$ at constant relative change of *RH* (*RC*s).

The above theoretical analysis reveals that temperature is the most significant meteorological factor affecting *RH*, and the larger amplitude of variation in *RH* can be witnessed under low temperature conditions. The contribution of water vapor is less significant than that of temperature. For *RH* to remain unchanged, a larger increase of water vapor is needed to compensate for the effect of a smaller increase of *T*. Moreover, the direct contribution of pollutants including aerosols to *RH* can be negligible in the actual atmosphere. Next we further examine the observational data to quantify the empirical range of variations of these variables and their relationships.

4. Observational analysis

4.1 Climate change characteristics

Figure 4 compares the annual changes of temperature, water vapor pressure, and *RH* during the period of 1970 to 2010 in the Yangtze River Delta. Temperature displayed a fluctuating upward tendency, with the metropolitan temperature being always higher than that of the small cities. The temperature difference between the two city types increased slightly over time. The difference in the water vapor pressure between big and small cities was extremely small (< 2%), which indicates that the scale of the urban area has little effect on the actual water vapor distribution. *RH* decreased with time, with *RH* in metropolises being lower than in small cities. The correlation coefficients between metropolises and small cities are 0.99, 0.99, 0.97, for temperature (a), water vapor (b)and *RH* (c) respectively, with significance level of $\alpha = 0.001$, indicating consistent changes in these meteorological variables between large and small cities in the Yangtze River Delta.

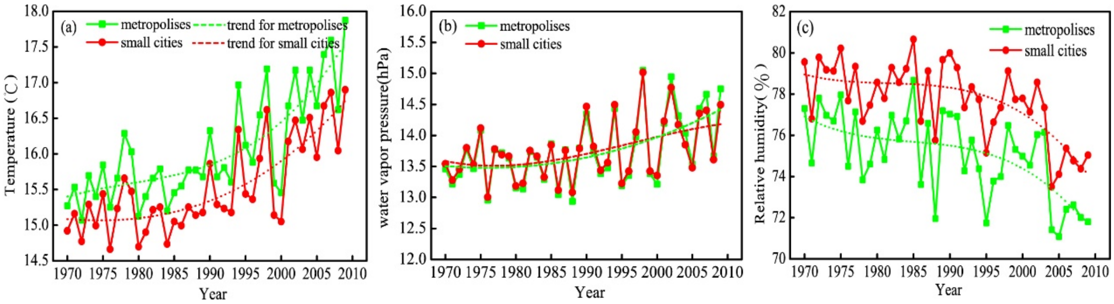

Fig. 4. Changes of temperature (a), water vapor pressure (b), and *RH* (c) in metropolises (green) and small cities (red) from 1970 to 2010 in the Yangtze River Delta.

The temperature trends in large and small cities were roughly consistent with the tendency of global warming (Xie et al., 2010), but there was a certain temperature difference (ΔT) between them. The heat island effect defined as the temperature difference between urban and rural areas is another factor affecting the temperature variation. Similarly, here the temperature difference between the metropolises and small cities is used to represent the 'analogous heat island effect' (AHIE), and the value of ΔT measures the AHIE intensity. If the temperature trend for small cities T_s is a consequence of global warming, then according to Figure 4a, the temperature trend for metropolises T_m can be expressed as $T_m = T_s + \Delta T$, i.e., the metropolitan temperature is influenced by global warming and the AHIE. In addition, the water vapor pressure varied by 1.3 hPa with a slight increase over the past 40 years, which is consistent with the other researches (Ding et al., 2008; Hu et al., 2016). The weakening of the East Asian summer monsoon has been identified as one of the most important reasons for the increase in water vapor in this area. It causes the monsoon rain belt to retreat southward, bringing more abundant water vapor to south China (Ding et al, 2008; 2010). The increase of water vapor may be related to the pattern of "flooding in the south and drought in the north".

4.2 Impacts of water vapor and temperature on the relative change of *RH*

The effects of individual factors (temperature and water vapor) on *RH* are determined theoretically in the previous section. This section examines their influences on the actual variation of water vapor and temperature in the Yangtze River Delta. To eliminate short-term fluctuations in temperature and water vapor, trends are used to represent the long-term characteristics of meteorological parameters in metropolises and small cities. The trends of meteorological factors are determined by the annual value in 1970 as the reference point when *RH* = 79%, T= 14.9 ℃, and e=13.5 hPa. The humidity relative change comparing to 1970 are defined as *RCF*, with independent contributions from temperature and water vapor denoted by RCF_t and RCF_v, respectively. In addition, RCF_{tv} denotes the relative change under the conditions when temperature and water vapor pressure vary simultaneously. The method used for the calculation is provided in section 2.

It can be seen from Fig. 5a, that RCF_v was negative from 1970 to 1985 (<-1%) but gradually became positive over time and increased significantly to be larger than 4% by 2010. As expected, RCF_v had a similar trend to that of water vapor (Fig. 4b). When the vapor pressure was lower than the reference point, RCF_v was negative; but RCF_v became positive when vapor pressure was greater than the reference point. As can be seen from Fig. 4b, over the past four decades, water vapor pressure reached a minimum around 1978 in the Yangtze River Delta, and due to the slight increase, RCF_v was less than 5%.

The metropolitan temperature trend is affected by both global warming and AHIE. The individual contributions of these two factors to RCF_t are shown in Fig. 5b. Relative humidity significantly reduced as the temperature increased, and as a result RCF_t was always negative. The intensity of the AHIE (ΔT) increased slowly over the past 40 years with a very small change (<0.5°C), which led to RCF_t ranging from -2.0 to -4.5%. The increase in temperature due to global warming was less than 0.25°C before 1990, thus RCF_t could not reach -1.6% by that. However, the temperature trend exhibited a significant upward tendency and increased by over 1°C from 1991 to 2010 due to the rapid global warming. As a result, RCF_t exceeded -10.1% by 2010. When comparing the contributions of global warming with the AHIE to RCF_t in metropolises, the contribution of AHIE is greater (less) than global warming before (after) 1995.

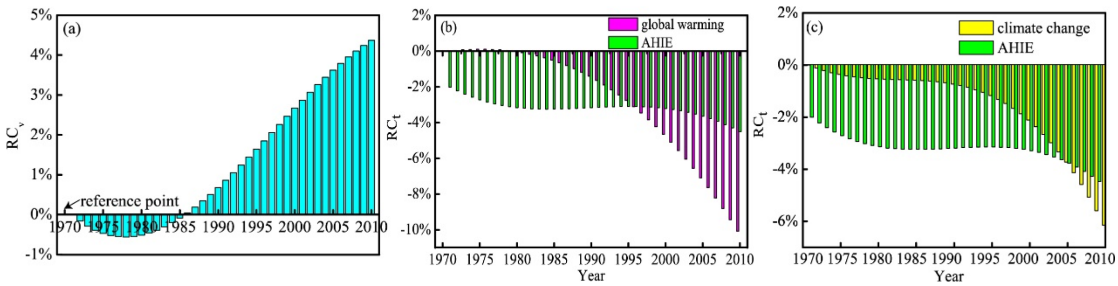

Fig. 5 The relative change of *RH* caused by water vapor, temperature, and climate change

Climate change manifests itself in changes of both temperature and water vapor. According to Fig. 4, the climate had gradually become warmer and moister with time in the Yangtze River Delta, suggesting the opposite impacts on *RH* of changes in temperature and water vapor. After taking the effects of actual water vapor into consideration, the RCF_{tv} caused by climate change was found to have significantly reduced (Figs. 5b, 5c) compared to that induced only by global warming. For example, in 2010, the RCF_t was -10.1% without the influence of water vapor, but the RCF_{tv} was only -6.1%. It can be inferred that the increase of water vapor pressure negated the *RH* reduction due to global warming to some extent. Furthermore, the increasing water vapor postponed the time at which the contribution of climate change became greater than that of AHIE from 1995 to 2004.

4.3 The relationship between fog/haze and *RH* in the Yangtze River Delta

This section investigates the variations of fog/haze days and their relationships to *RH*. Figure 6a shows the annual change of the number of fog days for metropolises and small cities in the Yangtze River Delta from 1970 to 2010. It can be seen that fog days have exhibited significant descending trends both in metropolises and small cities since 1980, and the fog days were less in metropolitan than in small cities. Figure 6b further shows the relationships between the number of fog days and *RH*. It is evident that fog days was significantly positively correlated with the annual *RH* ($R = 0.76$ and $\alpha = 0.001$). This result agrees with Yin et al. (2015). Although the water vapor pressure was almost same for metropolises and small cities (Fig. 4b), because of the AHIE, the metropolitan *RH* was less small cities, which resulted in a lower occurrence of fog and fewer fog days in metropolises. Namely, the AHIE can explain the phenomenon in which fog days in metropolises were less than in small cities. Due to the average effect of *RH* for both non-fog and fog days, annual mean relative humidity was maintained in a range from 70% to 81%.

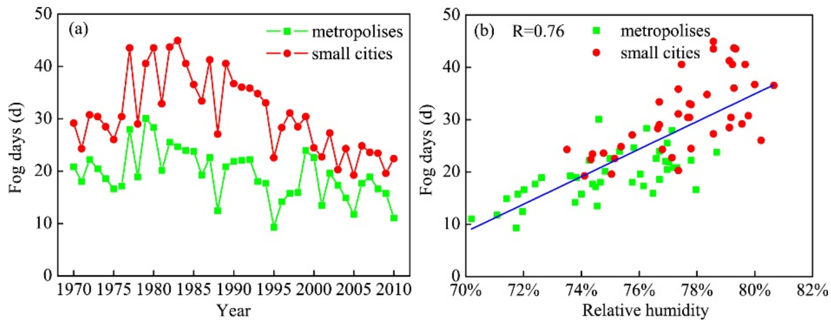

Figure 6. (a) Temporal change of the annual number of fog days; (b) relationship between the annual number of fog days and annual mean relative humidity.

Figure 7 shows the temporal change of the annual haze days (a) and the relationship between haze days and *RH* for the metropolises (green) and small city (red) in the Yangtze River Delta from 1970 to 2010. Haze days were significantly negatively correlated with *RH* (R = -0.70 and α = 0.001), which was also found in the Jiang-Huai Basin and southern China (Wu et al., 2016). Meanwhile, the annual number of haze days had a significant positive correlation with temperature (R = 0.76 and α = 0.001). However, Granger causality analysis shows that neither temperature nor relative humidity Ganger-caused haze days (Table 1). China has begun large-scale industrialization and urbanization expansion since 1978, resulting in the surge of aerosol emissions, this is confirmed by an obvious uptrend in aerosol optical depth (AOD) with amplitude of 17% in 2010 when compared with 1970 (Fig. 8a). Granger causality reveals the increasing AOD accounts for the increasing haze days (Table 1).

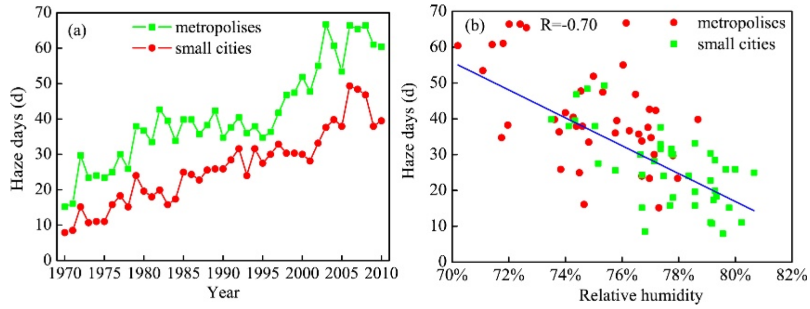

Figure 7. (a) Temporal change of the annual number of haze days; (b) relationship between the annual number of haze days and annual mean relative humidity.

Table 1 Granger causality analysis results

	Temperature	*RH*	AOD	*PBLH* (Nanjing)
Haze days	P>0.05	P>0.05	P=0.040（lag=9, 10）	P>0.05

All the time series are stationary after ADF Test examination. Null hypothesis: temperature, *RH*, AOD doesn't Granger-cause haze days, significant level: 5%, P>0.05, accept null hypothesis, P<0.05, reject null hypothesis.

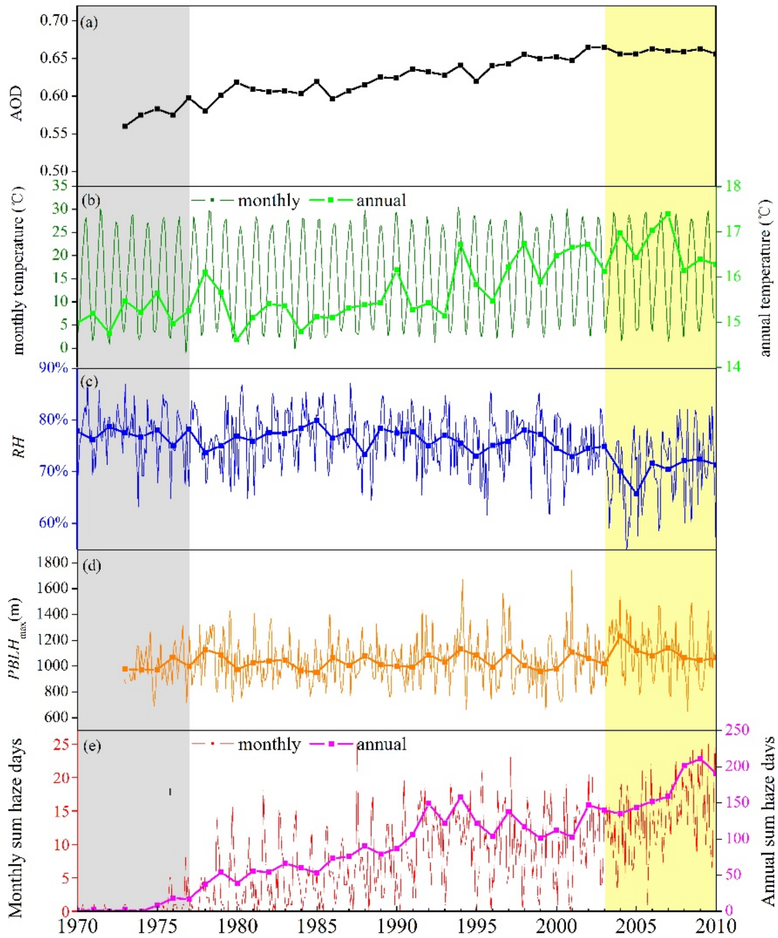

Fig. 8 Annual average of AOD(a), monthly and annual average of temperature (b), *RH* (c), daily maximum planet boundary layer height ($PBLH_{max}$) (d), monthly and annual sum of haze days(e) in Nanjing

The relationships between meteorological factors and AOD with haze days are further analyzed based on the monthly and annual average variations in Nanjing (a representative station in Yangtze River Delta). Before 1978 (light gray shadow), the variation of temperature, *RH* and daily maximum planet boundary layer height ($PBLH_{max}$) had little impacts on haze days (R≈0), because of the extremely low aerosol emissions as manifested by low AOD, which was largely below the threshold for haze formation. Under such low aerosol loading conditions, no matter how meteorological parameters changed, they would not affect the haze days. After aerosol loading increased beyond the haze threshold in 2000s, haze events would frequently occur under appropriate meteorological conditions (e.g., low wind, high RH etc). For the inter-annual variation, although Granger causality analysis shows that increasing aerosol loading was responsible for increasing haze days rather than *RH* or $PBLH_{max}$ (Table 1), when aerosol emission was high and relatively constant (yellow shadow), *RH* and $PBLH_{max}$ would influence the haze formation and development. For example, during the period of 2003 to 2010, AOD remained stable at the level of 0.65, each haze day and the corresponding daily *RH* and $PBLH_{max}$ for this episode in Nanjing were counted (1222 samples), the frequency of haze days and average $PBLH_{max}$ on different ranges of *RH* are shown Fig.9. An increase in frequency of haze days and a decrease in $PBLH_{max}$ were associated with the ascending *RH*. Similar variations can be also seen in each season (Fig. 10), with only a slight decrease of the frequency of haze days in the range of 71%~80% in spring (Fig.10a). More gales in spring may affect its distribution (Wang et al., 2008). This result supported the positive feedback between meteorology (including *RH* and PBLH) and haze (Tie et al., 2017). High *RH* induces an increase of aerosol particle size and mass by enhanced hygroscopic growth, multiphase reactions (Asmi et al., 2010) and secondary aerosol formation (Quan et al., 2018), which would further reduce the radiation and thus suppress *PBLH* (Quan et al., 2018), the decreased *PBLH* weakens the diffusion of water vapor leading to an increase in *RH*, the increased *RH* would in turn trigger the positive feedback mechanism of *RH*-aerosol-*PBLH* again.

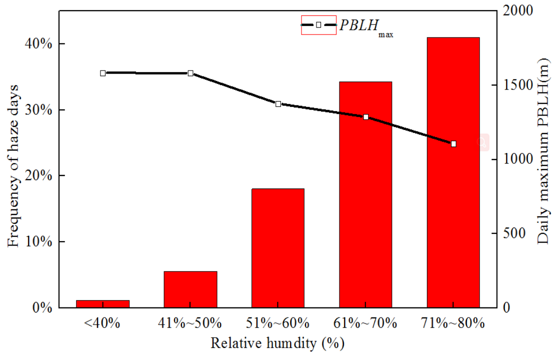

Fig.9 The frequency of haze days and the corresponding average $PBLH_{max}$ on different ranges of *RH* during 2003~2010 in Nanjing.

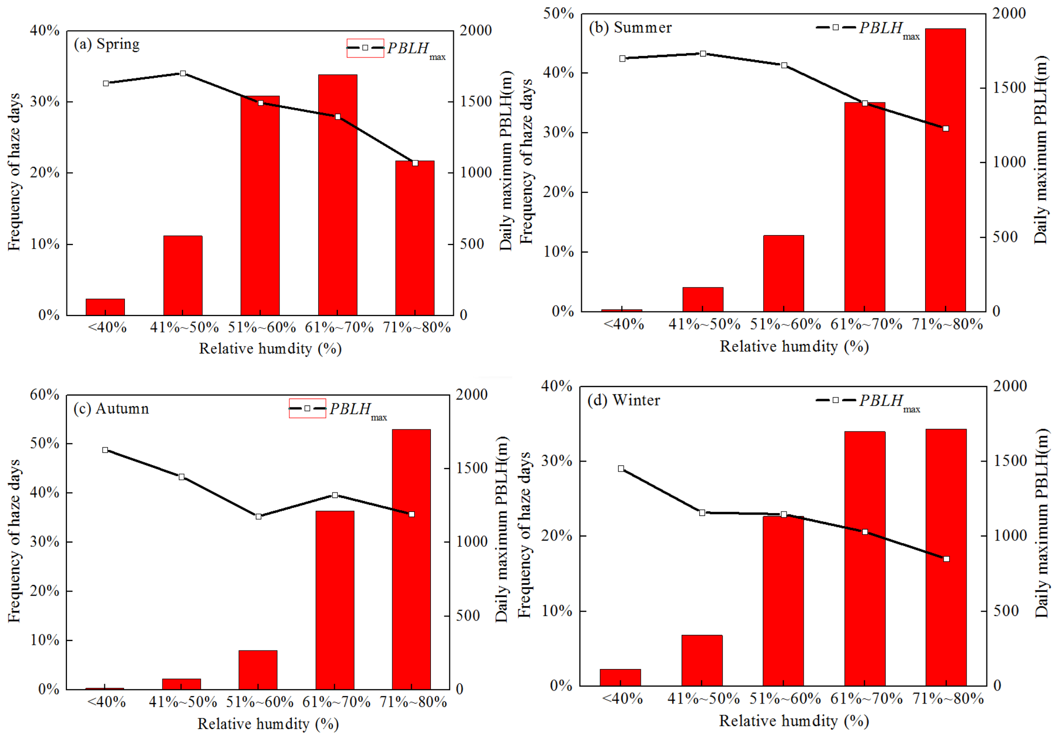

Fig.10 The frequency of haze days and the corresponding average $PBLH_{max}$ on different ranges of *RH* in spring(a), summer(b), autumn(c), winter(d) during 2003~2010 in Nanjing.

Overall, for inter-annual variation, increasing aerosol loading is responsible for ascending haze days rather than temperature, *RH* or $PBLH_{max}$, which has been confirmed by Granger causality analysis. When aerosol loading stays relatively stable and beyond the threshold for haze formation, high *RH* will enhance haze development.

4.4 Further discussion

The existence of hygroscopic aerosols will promote the activation of nuclear condensation and trigger fog episodes under a high *RH* condition even when humidity does not reach the saturation state (Mohan et al., 2009), the present fog event actually evolves into a "fog-haze" weather under high *RH* condition. For example, Sun et al. (2015) collected droplet samples during a fog event when *RH* was higher than 90% in Nanjing, finding the aerosol concentration is much higher than clean areas. Based on the analysis above, we suggests that with the enhancement of global warming and the AHIE, increasing temperature is the key reason for the reduction of fog days, and the fact that the aerosol loading beyond the threshold for haze formation is responsible for the increasing haze days. On the basis of these results, we may infer that if aerosol emissions maintained status quo, when climate became cooling, the ascending *RH* would result in more fog and haze days, which is likely to cause serious "fog-haze" events occurring more frequently. This conclusion is contrary to the predictions from Cai et al. (2017), who argues that the occurrence of stable weather in the North China Plain region would increase if climate continued to warm in the future, which would intensify the frequency and duration of winter haze events. Based on this premise, the frequency of haze episodes would increase by 50% relative to historical climatic conditions. Although our prediction and that of Cai et al. (2017) clearly differ, it indicates climate change has a complex and multifaceted impacts on severe haze event and more efforts are required to determine the overall influence of climate change on it. Regardless of the climate changes, aerosol loading will determine the frequency of haze events from a long-term point of view. Hence, controlling the aerosol emissions should be the major starting point for the management of haze problems in the next few decades.

5. Conclusion

Based on the definition of *RH*, an ideal box considering air pollutant is built to theoretically discuss the influences of temperature, water vapor, air pollutant and their combinations on *RH*. Taking the Yangtze River Delta as an example, the meteorological data from 1971 to 2010 are applied to analyze the contributions of each factor, as well as their combined effects on the relative change of *RH*. The long-term variations of haze and fog days and their connections with *RH* are revealed and future development of fog-haze events are also predicted from the perspective of *RH* and climate change. The main conclusions are as follows:

1) Temperature is the foremost factor influencing *RH* followed by water vapor pressure. A larger increase of water vapor is needed to compensate for the effect of a smaller increase of T. The direct contribution of aerosols to humidity can be ignored in the actual atmosphere.

2) The increase in temperature due to global warming and AHIE is responsible for the prominent descending trend of *RH* in the Yangtze River Delta. *RH* in metropolis is mainly dominated by AHIE before 1995, and global warming becomes more significant after that. But the increase in water vapor leads to the time when the contribution of climate change firstly exceeds that of the AHIE being postponed until 2004.

3) Fog days decrease after 1980 in the Yangtze River Delta and positively correlate with *RH*, increasing temperature is responsible for the deduction of fog days. The AHIE can explain the phenomenon whereby the fog days in metropolises are less than in small cities.

4) Granger causality analysis show the increasing aerosol loading results in the increasing haze days rather than meteorological parameters in inter-annual variation. Under the condition of aerosol

loading being relatively stable and beyond the threshold for haze formation, high *RH* would lead to the deceasing *PBLH* and more haze days.

5) If pollutant emissions maintained status quo, climate cooling would cause more fog days in the Yangtze River Delta region, the superposition of the growing number of fog days and haze days is likely to lead to serious fog-haze events occurring more frequently.

Based on the *RH* definition, although the ideal box model containing air pollutant is built, it still has limitations. Such as, gaseous pollutant cannot be completely regarded as aerosols, but due to the concentration only comprises a tiny share in atmosphere, the direct contribution to RH can be negligible. However, its indirect effects on *RH* and haze events cannot be ignored. For example, aerosols can indirectly affect *RH* and have major impacts on atmospheric stabilization during severe haze events in China by scattering and absorption (Wang et al., 2013, Peng et al., 2016). At the same time, aerosols can also impact water vapor distribution and precipitation pattern which would influence the regional *RH*. In addition, aqueous chemistry (zhang et al., 2015), urban emissions of water vapor from combustion sources and electricity generation facility cooling towers (Salmon et al., 2017) can also be the factor affecting *RH*. However, due to the uncertainties associated with the effects of aerosols (IPCC, 2013) on climate, it is difficult to estimate its indirect effect on *RH*. Moreover, the latest study shows that *RH* over land could be also adjusted by *RH* over ocean at global-scale trends (Byrne and Gorman, 2018), but more investigations are needed on how ocean humidity affects the continental humidity at regional scale.

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