Reply to Referee 1

We are grateful to the referee for the encouraging comments and careful reviews which helped to improve the quality of our paper. In the following we quoted each review question in the square brackets and added our response after each paragraph.

[Review Comment: In this paper, the observational and modeling studies show a relationship between the haze events over CEC and Tibetan Plateau (TP). I think it is a very interesting paper. It provides a new perspective to investigate the causes of the haze pollution in China. I believe this manuscript is appropriate for publication in Atmospheric Chemistry and Physics and would recommend publication subject to primarily minor revisions outlined below.]

Reply 1: Thank you for the encouraging comments.

[1. In this paper, which are the simulation results? Which are the observed results? Please comment on these.]

Reply 2: Following this comment, we have added a paragraph at the beginning of the revised Section 3 (Results and discussion) with following sentences:

"In this Section, we identify the contributions of pollutant emissions and climate change to interannual haze variations（in Sect. 3.1）, reveal a climatological "susceptible region" for haze formation in China (in Sect, 3.2), analyze the relationships between TP's thermal forcing and haze over CEC (in Sect, 3.3) and investigate the TP-warming inducing favorable meteorology for CEC's haze (in Sect. 3.4) based on the meteorological observations. In order to more convincingly demonstrate the observed results, Section 3.5 presents the results of a sensitivity simulation experiment about impacts of the TP's thermal forcing on CEC's aerosol variations."

[2. How to separate the time series into three phases in Fig.1? How to define the haze event in the figure? Please comment on these.]

Reply 3: In response to the referee's questions, we have clarified the statements in the revised manuscript as follows:

"In accompany with an unceasing increase in the Chinese pollutant emissions during recent decades, the significant interannual variations of haze occurrences in CEC over recent decades could be separated into three interdecadal phases with the trends of slow ascending (4.6d/10a) from the 1960s to 70s, less changing (1.7 d/10a) during the 1980s–1990s and sharply rising with a trend reaching 13.0d/10a going into the 21st century (upper panel of Fig. 1). Although of a continuous increasing trend in the pollutant emissions over the recent decades, the haze variations in CEC have evolved with the different trends of slow, less and sharply ascending over three interdecadal periods," (Please see the second paragraph of Sect. 3.1).

"This study adopts a widely-used comprehensive haze definition using surface in-situ observations of visibility, relative humidity and weather phenomenon. The observed relative humidity of less than 90% is used to distinguish haze from fog under the visibility <10km, (Schichtel et al., 2001; Doyle and Dorling, 2002; Ding and Liu, 2014)." (Please see the first paragraph of Sect. 2).

References:

Schichtel, B. A., Husar, R. B., Falke, S. R. and Wilson, W. E.: Haze trends over the United States 1980–1995, Atmos Environ, 35,5205–5210, 2001.

Doyle. M. and Dorling, S.: Visibility trends in the UK 1950–1997, Atmos Environ, 36,3161–3172, 2002.

[3.Please give us the more detailed information about CO2 emission.]

Reply 3: In the revised manuscript, we have added "The Chinese CO2 emission data over 1961-2012 are downloaded online from the website (http://cdiac.ornl.gov/CO2_Emission/timeseries/national)" in the first paragraph of Section 2, and "the Chinese CO_2 emission data source: http://cdiac.ornl.gov/CO2_Emission/timeseries/national" in the caption of Fig. 1.

[4. "In accompany with an unceasing increase in the Chinese pollutant emissions in recent decades, the significant interannual variations of haze occurrences in CEC have evolved with the trends of slow ascending from the 1960s to 70s, less changing during the 1980s–1990s and sharply rising with a trend reaching 13.0d/10a going into the 21st century (upper panel of Fig. 1), implying that climate change could also play an important role in the variations of haze events in CEC apart from the anthropogenic dimension of pollutant emission sources related to the rapid industrialization of China." I think this conclusion looks a little messy. You need to give us more evidence to prove this point.]

Reply 4: Following the suggestions, we have clarified that conclusion in the revised manuscript as follows (in Sect. 3.1):

"In accompany with an unceasing increase in the Chinese pollutant emissions during recent decades, the significant interannual variations of haze occurrences in CEC over recent decades could be separated into three interdecadal phases with the trends of slow ascending (4.6d/10a) from the 1960s to 70s, less changing (1.7 d/10a) during the 1980s–1990s and sharply rising with a trend reaching 13.0d/10a going into the 21st century (upper panel of Fig. 1). Although of a continuous increasing trend in the pollutant emissions over the recent decades, the haze variations in CEC have evolved with the different trends of slow, less and sharply ascending over three interdecadal periods, implying that climate change could also play an important role in the variations of haze events in CEC apart from the anthropogenic dimension of pollutant emission sources related to the rapid industrialization of China. A steady decline of East Asian monsoon winds is negatively correlated to haze occurrences in the CEC with the coefficient of determination, R2=0.6419 passing the confidence level of 99.9% (lower panel of Fig.1), indicating a consequence of East Asian monsoon climate change to CEC haze pollution."

[5. Fig.3: Surface wind speed or wind speed at 10 m? Please check it.]

Reply: Yes, it is wind speed at 10 m. It has been corrected in the caption of Fig.3.

[6. Could you give us the information about pollutant emission in lower TP's Q1 (1996,2002) and higher TP's Q1 (1998,2003)?]

Reply: The haze variations are generally controlled by pollutant emissions and meteorological conditions. In order to more convincingly demonstrate the contribution of the TP-warming to the haze frequency over CEC, a sensitivity simulation by employing the global air quality model GEM-AQ/EC is designed to isolate the emission influence on interannual variations of aerosols, where the monthly data of anthropogenic emissions by fossil fuel and biomass burning as well as the sulfate emissions compiled using EDGAR2.0 (Gong et al., 2012) are introduced without any interannual changes from 1995 to 2004. As designed in the sensitivity simulation, the pollutant emissions in lower TP's Q_1 (1996,2002) and higher TP's Q_1 (1998,2003) are same used in the simulation with the emission inventory dataset EDGAR2.0.

In the revised manuscript, we have added the above-discussions in Sect. 2 (the last paragraph) and in Sect. 3.5 (lines 348-349; lines 360-362) .

[7. As we known, dust is one of the absorbing aerosols in the atmosphere, which can influence the climate directly by modulating the radiation budget, affect the microphysical properties of clouds, and alter the surface albedo of the ground covered by snow or glacier. TP dust could impact on regional and global climate (e.g., Lau et al., 2006,2010; Huang et al., 2007; Chen et al., 2013). Could you consider the climatic

effects of the TP dust in this paper? I am wondering whether there is a relationship between TP dust and haze over CEC.

References:

1. Lau, K. M., M. K. Kim, and K. M. Kim (2006), Asian summer monsoon anomalies induced by aerosol direct forcing: The role of the Tibetan Plateau, Clim. Dyn., 26(7-8), 855–864, doi:10.1007/s00382-006-0114-z.

2. Lau, W. K. M., M. K. Kim, K. M. Kim,and W. S. Lee (2010), Enhanced surface warming and accelerated snow melt in the Himalayas and Tibetan Plateau induced by absorbing aerosols, Environ. Res. Lett., 5, 025204, doi:doi:10.1088/1748 9326/5/2/025204.

3. Huang, J., P. Minnis, Y. Yi, Q. Tang, X. Wang, Y. Hu, Z. Liu, K. Ayers,C. Trepte, and D. Winker (2007), Summer dust aerosols detected from CALIPSO over the Tibetan Plateau, Geophys. Res. Lett., 34, L18805,doi:10.1029/2007GL029938, 2007.

4. Chen, S., J. Huang, C. Zhao, Y. Qian, L. R. Leung, and B. Yang, Modeling the transport and radiative forcing of Taklimakan dust over the Tibetan Plateau: A case study in the summer of 2006, J. Geophys. Res. Atmos., 118,2013.]

Reply: Many thanks for the referee’s discussion. We agree with all the suggestions. We have added these in Section 1: Introduction (lines 120-130) as follows:

“Aerosol transport and deposition have been increasingly dirtying and even melting the snow- and ice-dominated wintertime TP (Ramanathan and Carmichael, 2008;Xu et al., 2009). This process leads to decreases in the snow and ice albedos, which could be largely responsible for climate change in the TP region (Hansen and Nazarenko, 2004). As one of the absorbing aerosols in the atmosphere, dust can influence the climate directly by modulating the radiation budget, affect the microphysical properties of clouds, and alter the surface albedo of the ground covered by snow or glacier TP dust could impact on regional and global climate (Lau et al., 2006,2010; Huang et al., 2007; Chen et al., 2013; Liu et al. ,2008). “

We have accordingly cited the following articles in the revised manuscript:

Chen, S., Huang, J., Zhao, C., Qian, Y., Leung, L. R., and Yang, B.:Modeling the transport and radiative forcing of Taklimakan dust over the tibetan plateau: a case study in the summer of 2006, J.Geophys. Res.-Atmos., 118, 797–812, doi:10.1002/jgrd.50122, 2013.

Lau, K. M., Kim, M. K., and Kim, K. M.: Asian summer monsoon anomalies induced by aerosol direct forcing: the role of the Tibetan Plateau, Clim. Dynam., 26, 855–864, doi:10.1007/s00382-006-0114-z, 2006.

Lau, W. K. M., Kim, M. K., Kim, K. M. and Lee, W. S.: Enhanced surface warming and accelerated snow melt in the Himalayas and Tibetan Plateau induced by absorbing aerosols, Environ. Res. Lett., 5, 025204, doi:10.1088/1748 9326/5/2/025204, 2010.

Liu, Z., Liu, D., Huang, J., Vaughan, M., Uno, I., Sugimoto, N., Kittaka, C., Trepte, C., Wang, Z., Hostetler, C., and Winker, D.: Airborne dust distributions over the Tibetan Plateau and surrounding areas derived from the first year of CALIPSO lidar observations, Atmos. Chem. Phys., 8, 5045-5060, doi:10.5194/acp-8-5045-2008, 2008.

Huang, J., Minnis, P., Chen, B., Huang, Z., Liu, Z., Zhao, Q., Yi, Y., and Ayers, J. K.: Long-range transport and vertical structure of Asian dust from CALIPSO and surface measurements during PACDEX, J. Geophys. Res., 113, D23212, doi:10.1029/2008JD010620, 2008.

Wang, H.-J., Chen, H.-P. and Liu J.-P.: Arctic sea ice decline intensified haze pollution in Eastern China, Atmos. Oceanic Sci. Lett., 8(1), 1-9, doi: 10.3878/AOSL20140081, 2015.

Zhang, R. H., Li, Q. and Zhang, R. N.:Meteorological conditions for the persistent severe fog and haze event over eastern China in January 2013, Science China: Earth Sciences, 57, 26–35, doi: 10.1007/s11430-013-4774-3, 2014.

Yan, L. and Liu, X..: Has climatic warming over the Tibetan Plateau paused or continued in recent years?, Journal of Earth, Ocean and Atmospheric Sciences, 1, 13-28, 2014.

Duan, A. , Wu, G. X., Zhang, Q., Liu, Y,: New proofs of the recent climate warming over the Tibetan Plateau as a result of the increasing greenhouse gases emissions, Chinese Science Bulletin,51(11), 1396-1400, 2006.

Liu, X. and Chen, B.: Climatic warming in the Tibetan Plateau during recent decades, Int. J. Climatol. 20, 1729-1742, 2000.

Reply to Referee 2 （Prof. Dr. Xuhui Lee）

We are grateful to Prof. Dr. Xuhui Lee for the encouraging comments and careful reviews which helped to improve the quality of our paper. In the following we quoted each review question in the square brackets and added our response after each paragraph.

[The authors of this paper propose that meteorological conditions on the Tibetan Plateau are partly responsible for the deteriorating air quality in eastern China. They found a robust correlation between wintertime visibility in eastern China with the heating rate (Q1) on the plateau derived from a reanalysis model. An air quality model was used to attribute changes in air quality to emission changes and to changes in meteorology. They found that during years of negative Q1 anomalies, the surface wind was weakened, the air in the boundary layer became more stable, and more moisture was transported from the ocean. All these factors contributed to unfavorable dispersion conditions. I support publication of the paper in ACP.]

Reply 1: Thank you for the encouraging comments

[The current manuscript style is not suited for the readership of ACP. Because literature review is mixed with methods, results and discussion, it is difficult to determine which statements describe past research by other people and which describe new results from this study. They should restructure the manuscript according to the standard template used by a technical journal, by separating the information clearly into different sections (introduction /literature review, methods, results, discussion, conclusions). Data and sources should be clearly explained. Model scenarios and their rationale should also be explained in the methods section.]

Reply 2: Following the comments, the revised manuscript has been restructured based on the original manuscript with the following outline:

1 Introduction

In this Sect., we present all the literature review with moving the descriptions of past research from the original Sect. 4 (please see lines 120-130 in the revised manuscript with track changes) and removed the new results and analysis from this study, as a part of results and discussion, to Sect. 3.1 (Contributions of pollutant emissions and climate change to interannual haze variations in China).

2 Data and methods

In this Sect., the design of sensitivity simulation is moved from the original Sect. 6 (please see lines 193-202 in the revised manuscript with track changes).

3 Results and discussion

3.1 Contributions of pollutant emissions and climate change to interannual haze variations in China

This Sect. is built with the sentences of the original introduction .

3.2 A climatological “susceptible region" for haze formation in China

3.3 Relationships between TP’s thermal forcing and haze over CEC

we removed the literature review to lines 120-130 (in the revised manuscript with track changes) in Sect. 1 (Introduction).

3.4 The TP-warming inducing favorable meteorology for CEC’s haze

3.5 A sensitivity simulation experiment on effect of the TP-warming

The design of sensitivity simulation is moved to lines 193-202 (in the revised manuscript with track changes) in the Sect. 2(Data and methods).

4 Conclusions

[The first name of all the authors should be spelled out completely. It is very difficult to distinguish Chinese names, and using initials only makes the job even more difficult.]

Reply 3: We have checked that the first names of all the authors were spelled out completely in our submitted PDF-file of manuscript, and after the submission the ACP changed them to the initials for the ACP discussion.

[Quality of the graphs should be improved. Some legends and axis labels are too small or too faint.]

Reply 4: We are sorry for the poor figure quality. All the Figures have been redrawn with high quality for ACP-publication (Please see the new Figs. In the revised manuscript).

[I understand that the authors have plan to continue to elucidate mechanisms underlying the Q1-air quality "teleconnection". Here I present several questions that arose from my reading of the manuscript. They do not need a response at this time but may be helpful to the authors in their future research:

1) Q1 is a quantity derived from atmospheric reanalysis. Given that meteorological observations are spare on the Tibetan Plateau, I wonder how robust this quantity is. The current research relies solely on one reanalysis product (NCEP?). Do other reanalyses show similar Q1 variabilities (both inter-annual and long-term trend)?

2) It has been known for some time that the eastern Asian monsoon has been weakening in the past three decades. As far as I know, there is no accepted theory for explaining why this is occurring. This study presents empirical evidence suggesting that the weakening trend is tied to changes in land cover on the plateau, as implicated by the strong correlation between Q1 and wind. In this regard, two questions are worth pursuing: a) what types of change on the surface (snow albedo change, glacier melt, desertification, shrinking of shrub land, etc) are responsible for the changing Q1 signal in the atmosphere? d) How do you test the TP hypothesis versus other competing hypotheses regarding the monsoon trend?

3) Related to my point 2 above, the author suggests that higher Q1 values are caused by contamination of snow cover by pollutants transported to the plateau, due to the fact that dirtier snow absorbs more solar energy. Can you find direct evidence for a declining trend in the snow albedo? If this proves true, the snow albedo effect constitutes a positive feedback on air quality at the regional scale, which may have contributed to the observed decline in visibility. However, the feedback mechanism cannot explain the large year-to-year variabilities in Q1. It is intriguing *that Q1 and air quality are correlated both in terms of the decadal trend and in terms of interannual variabilities. Why does Q1 change rapidly from one year to the next?]*

Reply 4: Your constructive suggestions are greatly appreciated and very helpful for our further study. In this preliminary study based on long-term observational data and a sensitivity simulation experiment, we investigate a relationship between the haze pollution in China and TP's environment and climate changes. It should be emphasized that considering the quality of reanalysis data over and around the TP, a comparison between NCEP/NCAR and other reanalysis data sets such as JRA-25, ERA-Interim, or MERRA is necessary in further work. The understandings of TP's thermal forcing changes and East Asian monsoon declines are challenging topics. The impacts of TP's climate change on air quality in China could be further studied on the shifts in weather patterns, pollutant emissions, depositions and chemical reactions in the atmosphere to comprehensively understand the meteorological drivers of air quality in a changing climate and also to consider the ocean-related impacts of climate change.

We have accordingly added the above discussions at the end of Sect. 4 (lines 402-410 of Conclusions in revised manuscript with track changes).

Climate modulation of the Tibetan Plateau on haze in China

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Key Points

1) A large-scale "susceptible region" for haze occurrences is climatologically identified over central-eastern China (CEC) harbored by the Tibetan Plateau (TP).

2) Thermal anomalies of the TP induce the change in meteorological drivers downstream for frequent haze events in CEC.

3) Implications of the TP-topography for atmospheric environment will be having potential utility for development planning in China.

Abstract

Rapid increases in pollutant emissions in conjunction with stagnant meteorological conditions result in haze pollution in China. Recent frequent haze in China has attracted worldwide attention. Here we show a relationship between the haze events and Tibetan Plateau (TP)'s environment and climate changes. Based on observational data taken over recent decades, we identify central-eastern China (CEC) as a climatological large-scale "susceptible region" of frequent haze, which is harbored by the TP with its impact on mid-latitude westerly winds. The observational and modeling studies demonstrate that the interannual variations in the thermal forcing of TP are positively correlated with the incidences of wintertime haze over CEC. Further analysis indicates that the TP-climate warming induced changes in atmospheric circulation driving frequent haze events in CEC. The frequent haze occurrences in CEC are consistent with decreasing winter monsoon winds, intensifying downward air flows and increasing atmospheric stability in the lower troposphere over the CEC in association with upstream plateau's thermal anomalies. Therefore, variations of haze in China are related to mechanical and thermal forcing by the TP. Our results also suggest that implications of the large TP-topography for environment and climate changes should be taken into account for air pollution mitigation policies in China.

1 Introduction

Haze in poor visibility with high particulate matter (PM) or aerosol levels is a pervasive air quality problem facing China, posing a major challenge for public health (Huang et al., 2014;Zhang et al., 2015). The frequent haze pollution has been notable for hitting record high levels of PM pollution over central-eastern China (CEC) in history since 1961 (Ding and Liu, 2014). In January 2013, extremely severe and persistent haze events swept over much of CEC-region. A large area of CEC from the North China Plain, including Beijing, across the Lower Yangtze Valley Plain to the Sichuan Basin, was blanketed in thick haze and smog for almost one month. It is estimated by the Chinese government that this wintertime haze covered a quarter of the total land area in China with 600 million people， half of the Chinese population, exposed to the haze air pollution (NDRC, 2013;Gu, 2013). China’s National Meteorological Center released its first ever “haze” orange alert (CMA, 2010) in response to the air quality index frequently reaching hazardous levels for this regional haze event. The $PM_{2.5}$ (PM with an aerodynamic diameter less than 2.5 micrometers) concentrations at 33 cities in the CEC region were more than 300 μg m^{-3} for longer than half a month, and some monitors reported hourly peak $PM_{2.5}$ levels of 900 μg m^{-3}, which is classified as “Beyond Index” (NDRC, 2013;Gu, 2013). The suffering of those in China from haze and poor air quality has attracted worldwide attention(Wang et al., 2014;Arden Pope III and Dockery, 2013;Chen et al., 2013b;Kan et al., 2011;Park et al., 2013;Zhao et al., 2013).

China has been experiencing the increased air pollution, commonly attributed to the large increases in pollutant emissions associated with the rapid economic development. However, air quality is modulated by changes in meteorology and climate (Tagaris et al., 2009;Zhang et al., 2014;Wang et al., 2015). In accompany with an unceasing increase in the Chinese pollutant emissions in recent decades, the CEC region had observed the significant interannual variations of haze occurrences (Ding and Liu, 2014). The changing East Asian monsoon climate could also play an important role in the variations of haze events in CEC apart from the anthropogenic dimension of pollutant emission sources related to the rapid industrialization of China. The surface wind speed associated with East Asian monsoons has significantly weakened in both winter and summer in the recent three decades (Xu et al., 2006;Oey et al., 2013). The weakening of the East Asian monsoons could increase air pollutants mainly by the changes in atmospheric circulation and weather conditions (Zhu et al., 2012;Niu et al., 2010). Weak advection of cold air, in conjunction with strong subsidence and stable atmospheric stratification, can easily produce a stagnation area in the lower troposphere resulting in regional pollutant accumulations, which are favorable for the development of CEC haze events (Zhao et al., 2013). In addition, in the presence of high soil moisture, strong surface evaporation results in increases in the near-surface relative humidity, which is also conducive to haze formation(Xiao et al., 2011). The contribution of the meteorological factors to the variance of the daily haze evolution was estimated to reach 0.68, which could explain more than 2/3 of the variance for the persistent severe haze events over CEC in January 2013 (Zhang et al., 2014). By changing East Asian winter monsoon climate, the Arctic sea ice decline could intensify haze pollution in CEC (Wang et al., 2015).

It is generally accepted that meteorological conditions in China are closely connected to the large topography of Tibetan Plateau (TP) (Yanai et al., 1992;Xu et al., 2010;Wu et al., 2012;Ye and Wu, 1998). Precipitation, land surface temperature and surface air temperature have increased on the TP over the past decades(Zhong et al., 2011). The TP has exhibited the largest surface radiative flux changes induced by aerosols (e.g. black carbon and dust) contaminating snow and ice compared to any other snow- and ice-covered regions in the world(Qian et al., 2011). Aerosol transport and deposition have been increasingly dirtying and even melting the snow- and ice-dominated wintertime TP (Ramanathan and Carmichael, 2008;Xu et al., 2009). This process leads to decreases in the snow and ice albedos, which could be largely responsible for climate change in the TP region (Hansen and Nazarenko, 2004). As one of the absorbing aerosols in the atmosphere, dust can influence the climate directly by modulating the radiation budget, affect the microphysical properties of clouds, and alter the surface albedo of the ground covered by snow or glacier TP. Dust transport and depositions could impact on regional climate and environment over the TP (Lau et al., 2006;Lau et al., 2010;Huang et al., 2008;Chen et al., 2013a;Liu et al., 2008). The question remains whether the rapid changes in climate experienced by the TP could exert an influence on the haze variations in the downstream CEC region, the lower flatlands harbored by the large TP-topography. The consequent processes linking the TP-climate change with the CEC haze pollution should be highly possible and worth investigating, even though it is obvious that increasing anthropogenic pollutant emissions contribute to high haze frequency (Zhang et al., 2013). In this study, we attempt to determine the physical connection between climate change in the TP and haze occurrences in the CEC region to more comprehensively understand the large-area haze formation in China especially under the background of global warming with the TP's environment and climate changes.

2 Data and methods

In this study, we used the observational records of visibility, weather phenomenon, relative humidity and 10m wind from 1961 to 2012 archived at the China Meteorological Administration（CMA）, and the meteorological variables of air temperature, winds and relative humidity from the reanalysis data generated by the US National Center for Environmental Prediction–National Center for Atmospheric Research (NCEP/NCAR). This study adopts a widely-used comprehensive haze definition using surface in-situ observations of visibility, relative humidity and weather phenomenon. The observed relative humidity of less than 90% is used to distinguish haze from fog under the visibility <10km, (Schichtel et al., 2001;Doyle and Dorling, 2002;Ding and Liu, 2014). The Chinese CO_2 emission data during 1961-2012 are downloaded online from the website (http://cdiac.ornl.gov/CO2_Emission/timeseries/national).

Following the studies of Yanai (1961), Yanai and Johnson (1993) , Yanai and Tomita (1998), the apparent heat source (Q_1) and apparent moisture sink (Q_2) are calculated. Atmospheric heat sources and moisture sinks are respectively gauged with the Q_1 and Q_2. As Q_1 includes Q_2 and radiative heating, here we concentrate only on the collective effect of apparent heating (Q_1) over the TP. The heat source column (in units of w m^{-2}) over the TP is obtained with both horizontal and vertical integration of Q_1 over the TP-area of 78°E-103°E and 28°N-38°N covering the most region with the altitude of higher than 3000m (see the large TP-rectangle in upper panel of Fig. 1) to form a one-dimensional variable representing the TP-thermal forcing. The correlation coefficients between the TP-heat source column and the meteorological variables (U-, V- and W-components of wind and air temperature) are calculated to build their horizontal and vertical distributions of correlations. Zonal, meridional and vertical components of the correlation vector are respectively derived through the correlation coefficients of the TP-heat source column to U-, V- and W-components of vector of wind and air temperature, indicating the changes in wind and air temperature induced by the TP-thermal forcing.

In the modeling study, we used the global air quality model system GEM-AQ/EC, which is an integration of gas phase chemistry and aerosol modules in the meteorological model GEM (Global Environmental Multiscale weather prediction model of Environment Canada). Full details of the development and evaluation of GEM-AQ/EC are given by Gong et al. (2012). The validations of 10 year (1995-2004)

GEM-AQ/EC modeling prove that the model provided satisfactory simulations of the distribution and variation of global and regional aerosols (Gong et al., 2012;Zhao et al., 2012). Regional variations of aerosols in East Asia are reasonably captured by the GEM-AQ/EC modeling compared to the observed aerosol concentrations and aerosol optical depth.

Air quality change is generally driven by two factors: pollutant emissions and meteorology. In order to exclude the emission influence on interannual variations of aerosols, a sensitivity simulation with GEM-AQ/EC is designed without year-to-year changes in anthropogenic aerosol emissions from 1995 to 2004 for an assessment on the impact of the TP-warming on air quality change in China. The sensitivity simulation experiment of GEM-AQ/EC was configured with 28 hybrid vertical levels and the model top at 10 hPa as well as the horizontal model grids in a global uniform resolution of 1 °×1 °. The GEM-AQ/EC was run with the fully nudged variables of wind, temperature, pressure and water vapor of NCEP-reanalysis meteorology every 24 hours from 1995 to 2004.

3 Results and discussion

In this Section, we identify the contributions of pollutant emissions and climate change to interannual haze variations（in Sect. 3.1）, reveal a climatological “susceptible region" for haze formation in China (in Sect, 3.2), analyze the relationships between TP’s thermal forcing and haze over CEC (in Sect, 3.3) and investigate the TP-warming inducing favorable meteorology for CEC's haze (in Sect. 3.4) based on the meteorological observations. In order to more convincingly demonstrate the observed results, Section 3.5 presents the results of a sensitivity simulation experiment about impacts of the TP's thermal forcing on CEC's aerosol variations.

3.1 Contributions of pollutant emissions and climate change to interannual haze variations in China

China has experienced the huge increases in CO_2 emissions from fossil fuel combustion with the certain attendant pollutant emissions and aerosol loading over recent decades (upper panel of Fig. 1), which has a direct physical link to more frequent haze occurrences in situ in China. The regional emissions of air pollutants contribute largely to the haze pollution in CEC with a high coefficient of determination, $R^2=0.9025$ between interannual variations of haze frequency and CO_2 emission in China (upper panel of Fig. 1), reflecting that the frequent haze events are strongly associated with the large increases in anthropogenic pollutant emissions in recent decades.

In accompany with an unceasing increase in the Chinese pollutant emissions during recent decades, the significant interannual variations of haze occurrences in CEC over recent decades could be separated into three interdecadal phases with the trends of slow ascending (4.6d/10a) from the 1960s to 70s, less changing (1.7 d/10a) during the

1980s–1990s and sharply rising with a trend reaching 13.0d/10a going into the 21st century (upper panel of Fig. 1). Although of a continuous increasing trend in the pollutant emissions over the recent decades, the haze variations in CEC have evolved with the different trends of slow, less and sharply ascending over three interdecadal periods, implying that climate change could also play an important role in the variations of haze events in CEC apart from the anthropogenic dimension of pollutant emission sources related to the rapid industrialization of China. A steady decline of East Asian monsoon winds is negatively correlated to haze occurrences in the CEC with the coefficient of determination, $R^2=0.6419$ passing the confidence level of 99.9% (lower panel of Fig.1), indicating a consequence of East Asian monsoon climate change to CEC haze pollution.

3.2 A climatological “susceptible region" for haze formation in China

Examination of ground-based observations of the frequency of haze events from 1961 to 2012 (CMA, 2010) reveals that the haze air pollution in China typically has the highest levels in the CEC region covering a vast area from the eastern edges of the TP and the Loess Plateau to China’s Pacific coast, and haze occurrences in CEC oscillate seasonally between the peak in winter and the low in summer (Fig. 2). Based upon these climate data, we could climatologically regard the CEC, with the lowlands harboured by the upstream plateaus of western China, as a large-scale “susceptible region" of frequent haze events in China (left panel of Fig. 2). Upper panel of Figure 3 shows that low average wind speeds tend to be coincident with the centers of pollutant haze events over the CEC (left panel of Fig. 2), reflecting the climatological “susceptible region" of haze occurrences in connection with a stagnation area in the lower troposphere in China.

Due to the influence of the TP terrain on the typical westerly winds in this region, the air flowing from the windward plateaus descends in a north-south oriented zone between about 110^{o}E and 125^{o}E (middle panel of Fig. 3). Accompanying this strong downward current are weak winds in the near-surface layers that lie in the lee side of the plateaus. These air flow and wind condition lead to development of a “harbor” that accumulates air pollutants in the CEC region. The weak wind and downward current areas coincide well with the centers of frequent haze events in China (middle and lower panels of Fig. 3). The “susceptible region” of haze events over the CEC region from the eastern edge of the plateaus to the lower flatlands is associated with the “harbor” effect of the unique TP topography under specific meteorological conditions that trap air pollutants.

Because haze is climatologically mostly a winter phenomenon in the CEC (right panel of Fig. 2), the following analysis on the TP’s climate effect on haze pollution in CEC and the related mechanisms is focused on the winter season.

3.3 Relationships between TP’s thermal forcing and haze over CEC

As a vast elevated landmass, the TP acts thermodynamically as a synoptic-scale wintertime cooling source protruding into the free atmosphere(Qiu, 2008;Liu and Chen, 2000b;Ruddiman and Kutzbach, 1989;Yeh et al., 1957). The TP region, as a wintertime cooling source (negative values of apparent heat source Q_1), has been experiencing a warming trend over recent decades, especially since 2001 (upper left panel of Fig. 4). A striking climate warming over the TP during the last decades has been revealed by many studies (Liu and Chen, 2000a;Duan et al., 2006;Yan and Liu, 2014).

Against the backdrop of global climate change, the question may be posed: Does the warming of the TP region cause changes in the atmospheric environment in China resulting in more frequent haze events in the CEC? The historical data analysis indicates that a significant correlation exists between the wintertime cooling source represented by the apparent heat source column Q_1 integrated over the TP and the number of haze days averaged regionally in the CEC over recent decades (upper panel of Fig. 4). It is also found in upper panel of Fig. 4 that the changes of wintertime Q_1 over the TP were reversed from cooling to warming in the late 1990s, which could be connected with the trends in haze occurrences with less changing over the 1980s-90s and sharp increasing during the 21st century in China under the increases in pollutant emission levels (upper panel of Fig. 1). Based on the composite analysis on the haze frequencies in winter with positive and negative anomalies in wintertime cooling source of the TP, the haze increasing and decreasing incidences over the CEC are found in good agreement with the positively and negatively anomalous TP-cooling sources (lower panel of Fig. 4). The frequency of haze events over the CEC region is positively correlated with climatic warming over the TP.

3.4 The TP-warming inducing favorable meteorology for CEC's haze

Further analyses provide information on the mechanisms relating climatic warming of the TP and enhancement in haze occurrence in the CEC. The favorable meteorology for haze occurrences is well known to be lower wind speeds, weaker vertical mixing, stronger subsidence, higher air humidity and more stable low-level stratification. We are still pondering the question whether climatic warming of the TP could strengthen the aforementioned meteorological conditions downstream for frequent haze events in the CEC to reveal the mechanism how thermal anomalies of the TP in climate change influence the incidence of haze over the CEC

The East Asian winter monsoon, which climatologically prevails over the CEC, typically maintains near-surface northeastern winds(Ding, 1994). In upper panel of Figure 5, two horizontal components of the correlation vector are derived through two correlation coefficients of Q_1 to U- and V-surface wind components, respectively, where the arrow length denotes the combined correlation with a longer arrow implying a better correlation, and the arrow direction means the direction of anomalous wind induced by the TP-thermal effect. The correlation vector over the CEC in upper panel of Figure 5 indicates that the variations of thermal forcing over the TP could give rise to the weakening winter monsoon winds (southwest wind anomalies) induced by changes of Q_1 over the TP. Furthermore, the anomalous south wind components resulting from climate change in the TP (positive correlations of Q_1 to V-wind components in upper panel of Fig. 5) can enhance transport of water vapor from the oceans to the CEC(Niu et al., 2010). By increasing the moisture in the lower troposphere driven by the strong vapor transport(Hung and Kao, 2010) (lower panel of Fig. 5), in addition to decline in the East Asian winter monsoon with weak advection of cold air, haze formation can be enhanced(Zhao et al., 2013;Xiao et al., 2011).

Upper panels of Figure 6 present the results of composite analysis on vertical variations in air temperature in five winters respectively with the most positive and the most negative anomalies in the TP cooling source. The air temperature changes with upper warmer and lower cooler are found in the middle and lower troposphere over the CEC region in winter with positive Q_1 anomalies on the TP, and an inverse structure of the CEC air temperature changes in winter with negative Q_1 anomalies on the TP (upper panel of Fig. 6). The TP's warming and cooling anomalies could lead to a "warm shield" and a "cool shield" in the atmosphere over the CEC. The correlation analyses of observation data over 1981-2012 confirm that the vertical structure of anomalous air temperature similar to that induced the TP's positive thermal effect (upper-left panel of Fig.6) with a "warm shield" intensifying the subsidence in the lower troposphere is responsible for the frequent haze occurrences over the CEC (lower panel of Fig. 6). Associated with the warming TP, the vertical variations of air temperature with upper warmer and lower cooler could easily build an inversion layer in the atmosphere over the polluted CEC, which results in more stably stratified atmosphere in this region (Fig. 6). Heavy haze pollution processes in winter are highly related with the existence of atmospheric inversion layer(Xu et al., 2003).

The cumulative consequences of weakening winter monsoon winds, intensifying downward air flows, a more humid and more stable atmosphere as the favorable meteorological conditions for haze formation would be expected to strengthen the air pollutant "harbor" effect of the TP and increase the number and severity of haze events in the CEC. Therefore, the haze formation over CEC is significantly modulated by the TP's climate change under the increase and even without changes in the current levels of anthropogenic pollutant emissions.

3.5 A sensitivity simulation experiment on effect of the TP-warming

In order to more convincingly demonstrate the connection of the TP-warming to the haze frequency over CEC, a sensitivity simulation by employing the global air quality model GEM-AQ/EC is designed to isolate the emission influence on interannual variations of aerosols, where the monthly data of anthropogenic emissions by fossil fuel and biomass burning as well as the sulfate emissions compiled using EDGAR2.0 (Gong et al., 2012) are introduced without any interannual changes from 1995 to 2004. The results of this sensitivity simulation are used to assess the impact of climate change on interannual change of air quality over CEC in this study.

Haze and aerosol changes are determined by both pollutant emission and meteorology, and the effects of meteorology are difficult to separate from aerosol observations. The 10-year GEM-AQ/EC simulation without interannual changes in the anthropogenic emissions provides a possibility to identify the meteorological effect on the interannual variations of aerosols. To investigate the implications of TP's climate change for interannual aerosol variations in CEC's haze, a composite analysis of surface aerosol concentrations over CEC (Fig. 7) were performed for two winters with lower TP's Q_1 (1996, 2002) and two winters with higher TP's Q_1 (1998, 2003) during the simulation period of 1995-2004 according to the interannual Q_1 changes over the TP (Fig. 4). As designed in the sensitivity simulation, the pollutant emissions in lower TP's Q1 (1996,2002) and higher TP's Q1 (1998,2003) are same in the simulation with the emission inventory dataset EDGAR2.0 (Gong et al., 2012). Because the effect of emissions was singled out in the interannual aerosol variations modeled in the sensitivity simulation experiment, the simulated variations in aerosol concentrations over CEC could be purely attributed to the changes of meteorological drivers in the context of changing climate. The analysis results show that the TP heating anomalies could lead to enhancements of 30-45% in wintertime surface aerosol concentrations over the CEC-region compared to the winters with the TP cooling anomalies (Fig. 7). Because changes of aerosol levels in the surface atmosphere determine haze formation, this sensitivity simulation confirmed that the frequent haze in China with the significantly interannual variations is closely related to thermal forcing by the TP, and climate change of the TP could intensify pollutant haze in China even without increases in the current anthropogenic pollutant emissions.

4. Conclusions

Based on observational data over the recent decades, we identify the CEC region, the lower flatlands along the eastern plateau edges in China as a climatological large-scale “susceptible region” of pollutant haze in connected with downward currents and weak near-surface winds in consequence of the “harbor” impact of large TP topography on mid-latitude westerlies. The climate analysis reveals that the increasingly frequent haze in the CEC region is related with decreasing winter monsoon winds, intensifying descending air and increasing atmospheric stability in the lower-troposphere over the CEC in association with plateau’s thermal anomalies. Climate impact of the TP’s mechanical and thermal forcing driving changes in atmospheric circulation and meteorological conditions downstream is potentially contributing to the increasing trend in haze events in China. A sensitivity simulation also confirmed that the frequent haze in CEC with the significantly interannual variations is closely connected with thermal forcing by the TP.

The Chinese government has been making great strides in reducing emissions and mitigating air pollution. However, the interplay of China’s unique landform distribution with climate change and its associated more extreme weather events could impair the effectiveness of air pollution control measures in China. With the influence of the TP climate change, the CEC region is facing a bigger challenge to realize air quality maintenance plan. The TP "harbor effect" and climate change should be considered in making decisions on the locations of new industrial facilities for development planning in China in order to preferentially reduce anthropogenic emissions in the "susceptible region" of haze and in turn reduce the number and severity of haze events in the central-eastern region of China.

In this preliminary study based on long-term observational data and a sensitivity simulation experiment, we investigate a relationship between the haze pollution in China and TP's environment and climate changes. It should be emphasized that considering the quality of reanalysis data over and around the TP, a comparison between NCEP/NCAR and other reanalysis data sets such as JRA-25, ERA-Interim, or MERRA is necessary in further work. The understandings of TP's thermal forcing changes and East Asian monsoon declines are challenging topics. The impacts of TP's climate change on air quality in China could be further studied on the shifts in weather patterns, pollutant emissions, depositions and chemical reactions in the atmosphere to comprehensively understand the meteorological drivers of air quality in a changing climate and also to consider the ocean-related impacts of climate change.

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Figure Captions

Figure 1. Interannual variations in the total CO_2 emissions in mainland China and the haze event frequency averaged in the CEC region over 1961-2012 with their relationship in the inserted figure (upper panel). The Chinese CO_2 emission data source: http://cdiac.ornl.gov/CO2_Emission/timeseries/national. Two blue dotted lines separate the time series into three phases of the 1960s-70s, the 1980s-90s and the 21st century. Interannual variations in wind at 10m (blue line) and the number of days with haze (red line) in the CEC over 1961-2012 (lower-left panel) and their scatter plot (lower-right panel).

Figure 2. Distribution of annual haze event frequency (days per year) averaged over 1961-2012 in China, and Chinese topography of the TP and the Loess Plateau with altitudes is shown with yellow shades (left panel). Monthly variation of haze frequency averaged from 1961-2012 over the CEC region (right panel).

Figure 3. Near-surface wind speed distribution (m s^{-1}) averaged over 1961-2012 in China with the red rectangle marking the region for cross sections in the middle and lower panels (upper panel). Cross sections of horizontal wind speed (m s^{-1}; filled color contours) and vertical circulations illustrated by stream lines (middle panel) and zonal variations of annual haze event frequency (lower panel) at 27°N-41°N averaged over 1961-2012. Note that near-surface vertical and horizontal winds are not illustrated well here due to north-south variations in the terrain and approximation of the location of the TP in this figure. All fields are for the annual-averages.

Figure 4. Interannual variability in the apparent heat source Q_1 (the negative values denote cooling) integrated vertically over the TP and haze event frequency averaged in the CEC in winter (December, January and February) over 1980-2012 and their correlation (upper panel). The differences of haze frequencies (days) averaged in five winters with most positive (lower left panel) and most negative Q_1 anomalies (lower right panel) on the TP relative to the mean haze frequency from 1980 to 2012.

Figure 5. The distributions of the interannual correlations of the apparent heat source Q_1 over the TP to the local V-component of surface wind in winter over 1961- 2012 (color shading). Arrows denote correlation vectors (showing both correlation coefficients of Q_1 to U- and V-surface wind components) in China. The

correlation coefficients of 0.12 (-0.12)，0.14 (-0.14) and 0.19 (-0.19) respectively passing the significance levels of 90%, 95% and 99%. A vertical section of the trends in vapour content (g kg^{-1} per 10 years) in winter over 1961-2012 averaged along 27^{o}N-41^{o}N (lower panel).

Figure 6. Vertical sections of the anomalous air temperature (^{o}C) averaged along 27^{o}N-41^{o}N in five winters with most positive (upper left panel) and most negative Q_1 anomalies (upper right panel) on the TP from 1980 to 2012, and vertical sections of the correlations of the number of haze days with air temperature (lower left panel) and vertical circulations (lower right panel) in winter from 1980 to 2012.

Figure 7. The percentages (%; contour lines) of differences of surface aerosol concentrations between winters of 1998 and 2003 with positive and winters of 1996 and 2002 with negative Q_1 anomalies on the TP relative to the surface aeorosol levels averaged over winters of 1996,1998, 2002 and 2003 (μg m^{-3}; color contours) modeled by the sensitivity simulation experiment with GEM-AQ/EC.