https://doi.org/10.5194/acp-2020-1140
Preprint. Discussion started: 5 January 2021

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'Warm Cover'- Precursory 'Strong Signals' hidden in the Middle Troposphere for Haze Pollution

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Abstract. Eastern China (EC), located on the downstream region of Tibetan Plateau (TP), is a large area that has become vulnerable to frequent haze. In addition of air pollutant emissions, meteorological conditions were a key 'inducement' for air pollution episodes. Based on the study of the Great Smog of London in 1952 and haze pollution in EC over recent decades, it is found that the abnormal 'warm cover' in the middle troposphere, as a precursory 'strong signal' hidden, could connect to severe air pollution events. The convection and diffusion in the atmospheric boundary layer (ABL) were suppressed by a relatively stable structure of 'warm cover' in the middle troposphere, leading to the ABL height decreases, which were favourable for the accumulation of air pollutants in the ambient atmosphere. The warming TP built the 'warm cover' in the middle troposphere from the plateau to the downstream EC region and even the entire East Asian region. The frequent haze events in EC is connected with a significantly strong 'warm cover' in the interdecadal variability. It is also revealed that a close relationship existed between interannual variations of the TP's heat source and the 'warm cover' hidden in the middle troposphere over EC.

1 Introduction

In China, mainly over the region east of 100 °E and south of 40 °N (Tie et al., 2009), $PM_{2.5}$ (particulate matter with an aerodynamic diameter equal to or less than 2.5 μm) has become the primary air pollutant (Wang, et al., 2017). Anthropogenic pollutant emissions and unfavorable meteorological conditions are commonly regarded as two key factors for haze pollution with excessive concentrations of $PM_{2.5}$ (Ding and Liu, 2014; Yim et al., 2014; Zhang et al., 2015). The thermodynamic structures in atmospheric boundary layer and the free troposphere are the key meteorological conditions influencing the formation and maintenance of heavy pollution episodes (Wang et al., 2015; Cheng et al., 2016; Wang et al., 2016; Tang et al., 2016; Wang et al., 2019).

This study investigated whether the structure of atmospheric thermodynamics in the troposphere and its intensity variation could act as a 'strong forewarning signal' for surface $PM_{2.5}$ concentration variations in heavy air pollution. In order to explore the interaction between the free troposphere and the atmospheric boundary layer and the impact on the heavy air pollution in Eastern China, this study extended the meteorological conditions for heavy air pollution from the boundary layer to the middle troposphere. We

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identify a precursory 'strong signals' hidden in the free troposphere for frequent haze pollution in winter in Eastern China.

2 Data and methods

The data used in this study included NCEP/NCAR and ERA-Interim reanalysis data of meteorology, as well as data of surface $PM_{2.5}$ concentration measurement, air temperature observation and L-band sounding, as briefly described as follows:

The monthly NCEP/NCAR reanalysis data of meteorology with horizontal resolution of 2.5° for 1960-2019 were obtained from the U.S. National Center for Environmental Protection (NCEP, https://www.esrl.noaa.gov/).

The daily and monthly ERA-Interim reanalysis data of meteorology with horizontal resolution of 0.75° were derived from the European Center for Medium-range Weather Forecasts (ECMWF, https://www.ecmwf.int/), including air temperature, geopotential height, humidity, wind field and vertical velocity, etc.

The hourly $PM_{2.5}$ concentration data during 2013-2019 were collected from the national air quality monitoring network operated by the Ministry of Ecology and Environment of China (http://www.mee.gov.cn/). In addition, we categorized air pollution levels with the surface $PM_{2.5}$ concentrations based on the National Ambient Air Quality Standards of China (HJ633-2012) released by the Ministry of Ecology and Environment in 2012 as shown in Table 1.

We also used the monthly air temperature of surface observation data during 1960-2014 from 58 meteorological observation stations in the plateau area with an altitude above 3000 meters, which were archived from the China Meteorological Information Center (http://cdc.cma.gov.cn/).

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Furthermore, the L-band sounding 'seconds-level' data of the site Beijing from 2010 to 2019 to were used to calculate the height of atmospheric boundary layer (ABL, Liu and Liang, 2010). The height of ABL top is characterized by the L-band sounding observations at 20:00 (local time is used for this paper). The L-band sounding 'seconds-level' data has been undergone the quality control before analysis, and interpolation was implemented in a vertical direction at an interval of 5-hPa (Zhu et al., 2018). The L-band detection data provided by the Meteorological Observation Network (http://cdc.cma.gov.cn/) contains several automatic observation meteorological elements with time resolution of 1.2 s and vertical resolution of 8 m. More detail information can be found in Li et al. (2009) and Cai et al. (2014).

Table 1. Air pollution degrees categorized with surface $PM_{2.5}$ concentrations

Air pollution degrees	$PM_{2.5}$ concentrations
'less-serious' pollution	$75 \mu g \cdot m^{-3} < PM_{2.5} \leqslant 115\ \mu g \cdot m^{-3}$
'serious' pollution	$115\ \mu g \cdot m^{-3} < PM_{2.5} \leqslant 150\ \mu g \cdot m^{-3}$
'more-serious' pollution	$150\ \mu g \cdot m^{-3} < PM_{2.5} \leqslant 250\ \mu g \cdot m^{-3}$
'most-serious' pollution	$PM_{2.5} > 250\ \mu g \cdot m^{-3}$

3 Results

3.1 A precursory 'strong signal' of 'warm cover' in the middle troposphere

In the Beijing area and surroundings over North China Plain during February 18–27, 2014, the regional average $PM_{2.5}$ concentrations reached up to 250 $\mu g\ m^{-3}$ for the prolong heavy air pollution. The Great Smog of London in 1952 was attributed to the accumulation of low-level smoke and sulfur-dioxide pollutants under the influence of certain weather systems (Whittaker et al., 2004). To find the precursory 'strong signals' hidden of the meteorology for both heavy air pollution events, we retrieved the three-dimensional atmospheric dynamics and thermal structure during December in 1952 as well as February in 2014 by analyzing vertical anomalies of meteorology. There were high-pressure systems moved to London as well as Beijing and stagnated over the area at 500-hPa geopotential height anomalies, as shown in Fig. 1a–

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b. Prior to the heavy-pollution events, a high-pressure system over London as well as Beijing gradually strengthened (Fig. 1c–d), and the middle troposphere was characterized by a 'warm cover', i.e. a 'upper warming and bottom cooling' vertical structure (Fig. 1e–f).

By comparing Fig. 1a and Fig. 1b, we found that two long heavy air pollution events occurred during the maintenance stage of stable high pressure system. It can be seen that the 3D dynamical and thermodynamical structures were significantly modulated by the persistent large-scale anomalous circulation. The air temperature inversion effect of the high pressure system continuously strengthened the 'warm cover' structure in the middle troposphere (Cai et al., 2020). Moreover, it was obvious that 'strong signals' arising from the thick 'warm cover' persisted during the abnormal air-pollution episode during December 5–9, 1952 in London as well as February 21-26, 2014 in Beijing. It is worth pointing out that the bottom edge of 'warm cover' in the free troposphere declined day-by-day. During the heavy pollution incident, the 'warm cover' dropped to 900-hPa (Fig. 1g, h). The above analysis shows that in the upper air over London during December 5–9, 1952 and Beijing during February 21–26, 2014, the 'subsidence inversion of air temperature in the high pressure system and the inversion layer decreased, which made the atmospheric structure stable for accumulation of aerosols. The deep 'warm cover' structures in the middle troposphere acted as a precursory 'strong signal' of the Great Smog of London and Beijing's heavy air pollution.

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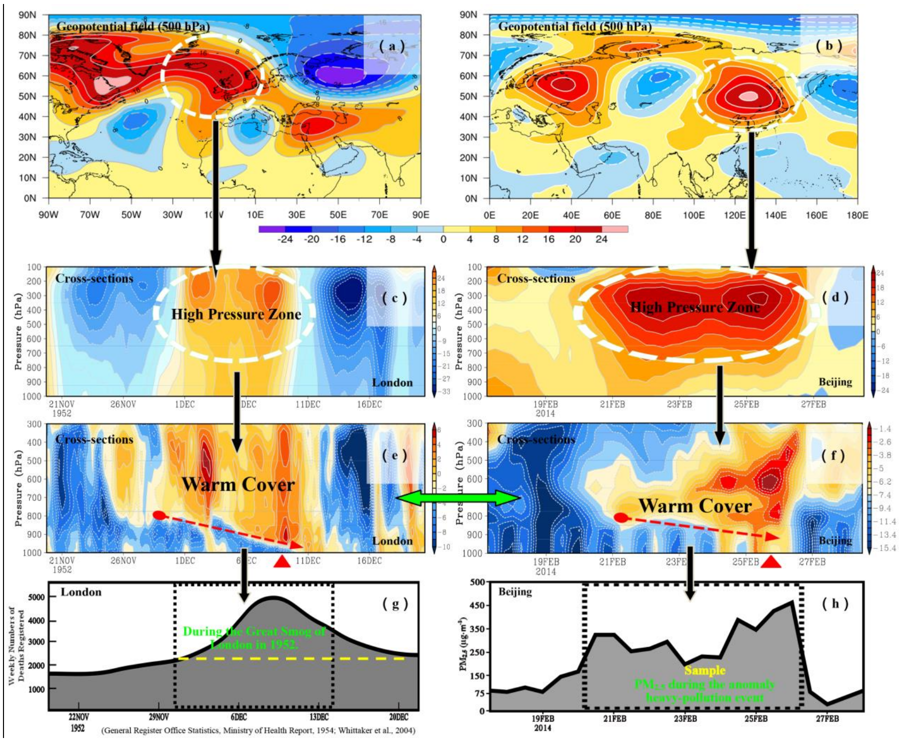

Figure 1. 3D dynamical and thermodynamical structures and air-pollution variations. (a) Geopotential height anomalies at 500-hpa during December 5 to 9, 1952 (during the Great Smog of London; unit: dagpm), (b) the same as (a) but during February 21 to 26, 2014. Time-vertical cross-sections of (c) the geopotential height anomalies (unit: dagpm) in the high pressure zone (50-70 °N; -20-10 °E) during November 20 to December 20, 1952, (d) the same as (c) but in the high pressure zone (40-63 °N; 115-138 °E) during February 17-28, 2014. (e) Time-vertical cross-sections of air temperature anomalies over London (unit: ℃, here the Red dotted arrow shows the bottom edge of the 'warm cover' during the Great Smog in London.) during November 20 to December 20, 1952, (f) the same as (e) but during the heavy pollution in February 2014 over Beijing. (g) Weekly death rate in London prior to, during and after the 1952 pollution episode (General Register Office Statistics, Ministry of Health Report, 1954; Whittaker et al., 2004). (h) The variation of surface $PM_{2.5}$ concentrations (units: $\mu g \cdot m^{-3}$) during the heavy pollution in February 2014 over Beijing.

3.2 'Warm Cover' in the free troposphere and boundary layer with aerosol variations

During five heavy air pollution episodes over Beijing in December 2015 and 2016 the vertical structures of air temperature anomalies presented the 'warm cover' structure in the free troposphere (see Fig. S1).

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During winter 2014-2017, Fig. 2a and Fig. 2b demonstrated the significant negative correlations passing 0.001 confidence degree between the height of the atmospheric boundary layer (ABL) and air temperature anomalies over same period and 24 hours ahead in Beijing, reflecting that the 'warm cover' structure hidden in the middle troposphere with significant 'strong-signal' features is of persistent premonitory significance for the heavy pollution episodes. Fig. 2c–e presented the significant positive correlation passing 0.001 confidence degree between $PM_{2.5}$ concentrations and air temperature anomalies over same period and 24, 48 hours ahead in Beijing. Air temperature anomalies over 24 hours ahead reflected that 'warm cover' hidden in the middle troposphere could be regarded as the precursory 'strong-signal' for air pollution change. Furthermore, such a 'stable' structure also restricted the transport of moist air from the lower to the middle troposphere for forming secondary aerosols, which could dominate $PM_{2.5}$ concentrations in air pollution over China (Huang et al., 2014; Tan et al., 2015).

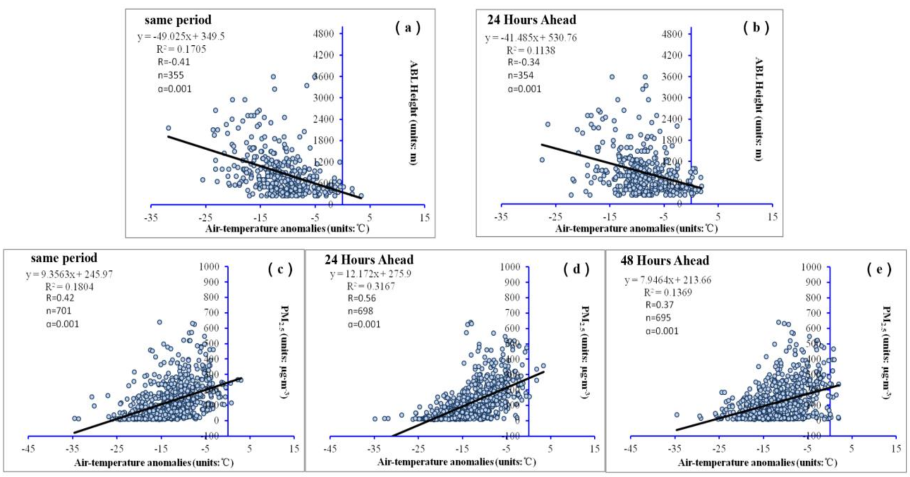

Figure 2. (a) The correlations between atmospheric boundary layer (ABL) height and air temperature anomalies, at 800-hPa. (b) 24 hours ahead at 650-hPa in Beijing during winter 2014–2017. The correlations between $PM_{2.5}$ concentration and air temperature anomalies, (c) at 850-hPa; (d) 24 hours ahead, at 800-hPa; (e) 48 hours ahead, at 724-hPa in Beijing during winter 2014–2017.

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3.3 Changes of the 'warm cover' structure

The 'warm cover' structure of air temperature anomalies in the troposphere indicated the intensification of heavy air pollution. The 'warm cover' structure is a precursory 'strong signal' for the frequent occurrence of regional haze events. The air pollution in Eastern China (EC) exhibited the significant seasonal variations. Our study revealed that seasonal differences in terms of the thermal structures in the atmosphere over EC. In spring (Fig. 3a, e) and summer (Fig. 3b, f), the middle troposphere was characterized by a 'upper cooling and bottom warming' vertical structure for less air pollution. When the autumn (Fig. 3c, g) or winter (Fig. 3d, h) arrived, the middle troposphere was characterized by a 'upper warming and bottom cooling' vertical structure, which intensified the air pollution. In autumn, atmospheric thermal structure over EC was marked with a transition between summer and winter (Fig. 3c). The atmosphere condition reversed in winter, a large-scale anomalous air temperature pattern of 'upper warming and bottom cooling' in the middle troposphere appeared from the plateau to downstream EC region and even the entire East Asian region (Fig. 3d). The structure of 'warm cover' in winter was much stronger than that in autumn, and its height of the former was much lower than that of the latter. Therefore, the intensity of air pollution over EC during winter is significantly higher than other seasons (Fig. 3h).

From the perspective of interdecadal variations, our study revealed a close relationship between the frequent occurrence of haze events in EC and the atmospheric thermal structure in the eastern TP. Furthermore, the thermal structures of the troposphere exhibited the distinct interdecadal variations (Fig. 4a-c). A cooling structure was identified in the wintertime air temperature anomalies over the east region of TP during 1961–1980 (Fig. 4a); the upper level of the eastern TP during 1981–2000 showed a 'upper cooling and bottom warming' vertical structure (Fig. 4b). The interdecadal changes of vertical structure reversed during 2001–2018 with a significant 'warm cover' (Fig. 4c). The years of 2001–2018 witnessed the highest frequency of haze days (Fig. 4f), and 1981–2000 saw a middle-level occurrence of haze days

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(Fig. 4e), while the lowest frequency of haze days occurred during 1961–1980 (Fig. 4d).

It is worth considering whether the variations of the plateau's heat structures could lead to the interdecadal variations of the 'warm cover' in the troposphere for the frequent occurrence of haze in EC since the 20th century (Fig. 4c, f). By analyzing TP's apparent heat source (Q1) and air temperature observed at meteorological stations over the TP in the winters during 1960-2014 (Fig. 5a, b), we found that the 'warm cover' changes in the middle troposphere over EC and even in East Asia was closely related to the surface temperature and TP's apparent heat.

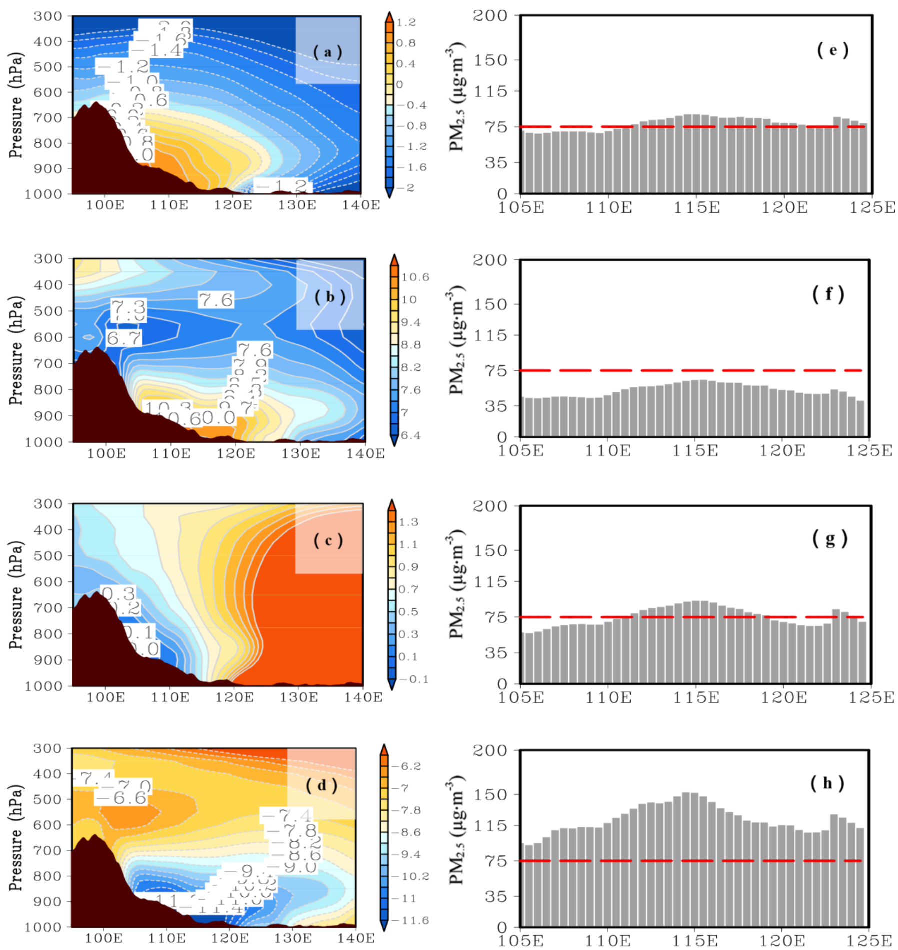

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Figure 3. Cross sections of (a-d) air temperature anomalies (unit: ℃) , and (e-h) the $PM_{2.5}$ concentrations (unit: μg·m^{-3}) averaged along 25-40 °N in spring (a, e), summer (b, f), autumn (c, g), winter (d, h) from 2013 to 2018.

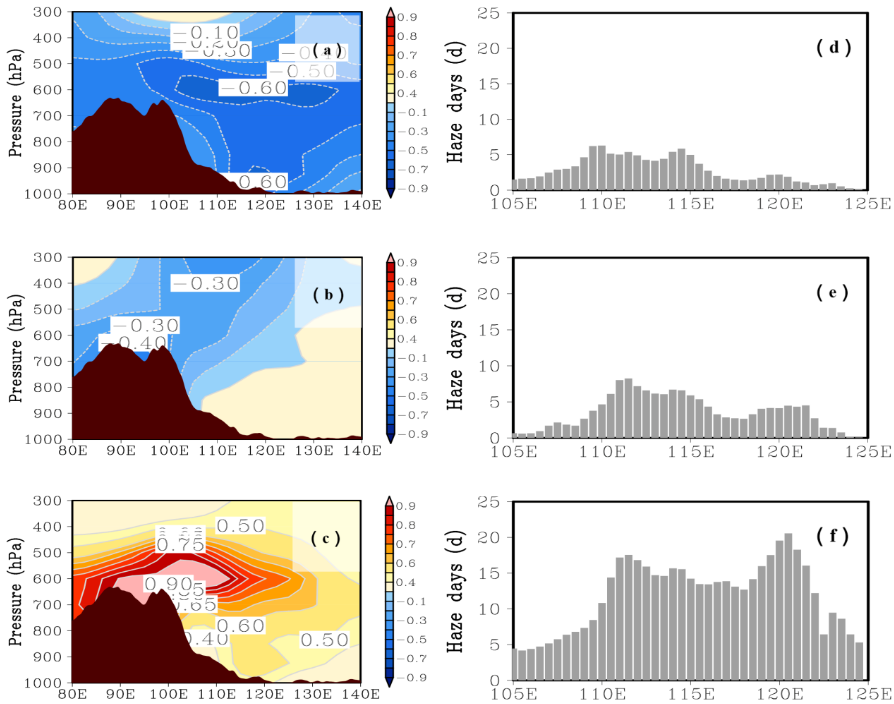

Figure 4. Cross sections of (a-c) air temperature anomalies (unit: ℃) and (d-f) the number of haze days averaged along 25-40 °N in winter during three decadal periods 1961-1980 (a, d), 1981-2000 (b, e) and 2001-2018 (c, f).

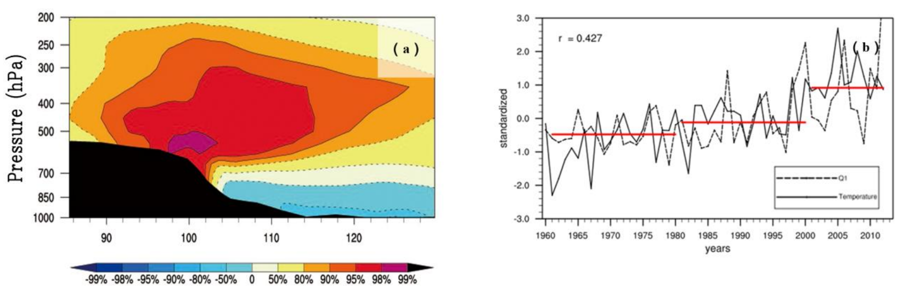

Figure 5. TP's apparent heat source (Q1) and air temperature variations. Cross sections of (a) the correlations between TP's apparent heat (Q_1) and air temperature latitude-averaged along 30-35 °N in the winters during 1960-2014; (b) interanual variations of TP's apparent heat source (Q_1) and air temperature of meteorological stations in the TP with the altitudes above

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3000 meters in the winters during 1960-2014.

4 Conclusions and discussion

Based on the study of the Great Smog of London in 1952 and Beijing's heavy air pollution in 2014, as well as $PM_{2.5}$ pollution over EC, the 'warm cover' in the free troposphere was identified as a precursory 'strong signal' for severe air pollution events, which could be attributed to climate change. A stable atmospheric thermal structure in the middle troposphere, i.e. a 'warm cover', suppressed the atmospheric boundary layer (ABL) development, which was a key 'inducement' for the accumulation of air pollutants in the ambient atmosphere.

A large-scale anomalous air temperature pattern of 'upper warming and bottom cooling' in the middle troposphere appeared from the plateau to the downstream EC region and even the entire East Asian region. The frequent haze pollution events in EC since the start of the 21st century happens to be within a significant positive phase in the interdecadal variations of 'warm cover' in the middle troposphere. A close relationship between the plateau's heat and the thermal structure in the atmosphere in EC and even the entire East Asian region reflected an important role of TP's thermal forcing in environment change over China.

Data availability. The monthly NCEP/NCAR reanalysis data of meteorology are collected from the U.S. National Center for Environmental Protection (NCEP, https://www.esrl.noaa.gov/); the daily and monthly ERA-Interim reanalysis data of meteorology are collected from the European Center for Medium-range Weather Forecasts (ECMWF, https://www.ecmwf.int/); the hourly $PM_{2.5}$ concentration data are collected from the national air quality monitoring network operated by the Ministry of Ecology and Environment of

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China (http://www.mee.gov.cn/); the air temperature of surface observation data and L-band sounding data are obtained from the China Meteorological Information Center (http://cdc.cma.gov.cn/). All data presented in this paper are available upon request to the corresponding author (Wenyue Cai, caiwy@cma.gov.cn).

Author contributions. XDX and WYC designed the study. XDX, WYC and TLZ performed the research. WYC performed the statistical analyses. XDX, WYC and TLZ wrote the initial paper. TLZ, XFQ, WHZ, CS, PY and CZW contributed to subsequent revisions.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This study is supported by the Atmospheric Pollution Control of the Prime Minister Fund (DQGG0104), the National Natural Science Foundation of China (91644223) and the Second Tibet Plateau Scientific Expedition and Research program (STEP, 2019QZKK0105).

Financial support. This research has been supported by the Atmospheric Pollution Control of the Prime Minister Fund (DQGG0104), the National Natural Science Foundation of China (91644223) and the Second Tibet Plateau Scientific Expedition and Research program (STEP, 2019QZKK0105).

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