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 followings we quoted each review question in the square brackets and presented our response after each paragraph.

[Review Comment: This work proposed that abnormal 'warm cover' in the middle troposphere could suppress the convection and diffusion in the boundary layer, leading to haze pollution in Eastern China. It is also indicated that such 'warm cover' is attributed to the warming of the Tibetan Plateau. I think this work well fits the scope of this journal. Overall, this manuscript is well structured but needs more in-depth analysis to further improve this article. Besides, the writing needs to be polished. It is worth being published after addressing the following issues.]

Reply: Thank you for the encouraging comments.

Major comments:

[1. The introduction is too simple and is not sufficient to clearly demonstrate the background and scientific significance of this work. Pre-existing literature on this subject is suggested to be fully reviewed, and a comprehensive introduction ought to be provided in this part.]

Reply: Many thanks for the referee's discussion. For the introduction, we have adjusted it as required and added new content.

"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 in winter (Wang, et al., 2017). Therefore，in September 2013, the Chinese government launched the China's first air pollution control action plan-'The Airborne Pollution Prevention and Control Action Plan (2013-2017)' (State Council of the People's Republic of China, 2013). By 2017, about 64 % of China's cities are still suffering from air pollution, especially Beijing-Tianjin-Hebei region and surrounding areas (Wang et al., 2019; Miao et al., 2019). Then, in July 2018, the Chinese government launched the second three-year action plan for air pollution control, 'the blue sky defense plan', which demonstrates China's firm determination and new measures for air pollution control (State Council of the People's Republic of China, 2018). After the implementation of air pollution control action plans, air quality in many regions in China has been significantly improved.

Anthropogenic pollutant emissions and unfavorable meteorological conditions are commonly regarded as two key factors for air pollution (Ding and Liu, 2014; Yim et al., 2014; Zhang et al., 2015). Air pollutants mainly come from surface emission sources, and most of air pollutants are injected from the surface to the atmosphere through the atmospheric boundary layer (ABL) (Quan et al., 2020). The ABL structures are the key meteorological conditions which influences the formation and maintenance of heavy air pollution episodes (Wang et al., 2015; Cheng et al., 2016; Wang et al., 2016; Tang et al., 2016; Wang et al., 2019).

Most of the previous studies focused on exploring the impact on the heavy air pollution in Eastern China (EC) from the meteorological conditions in ABL. However, the thermodynamic and dynamic structures of free troposphere can affect the meteorological conditions in ABL (Cai et al., 2020). The convection and diffusion in the ABL are suppressed by a relatively stable structure in the middle troposphere, leading to the ABL height decreases, which was favourable for the formation and persistence of heavy air pollution (Quan et al., 2013; Wang et al., 2015; Cai et al., 2020).

This study investigated whether the thermodynamic structure of the troposphere and its intensity changes can be used as a ‘strong warning signal’ for the changes of $PM_{2.5}$ concentrations in heavy air pollution, and whether this strong signal exists in the time scales of seasonal, interannual and interdecadal changes. In order to explore the interaction between the free troposphere and the ABL and the impact on the heavy air pollution in EC, this study extended the meteorological conditions for heavy air pollution from the boundary layer to the middle troposphere. We identify a precursory ‘strong signals’ hidden in the free troposphere for frequent haze pollution in winter in EC.”

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

Miao, Y. C., Li, J., Miao, S. G., Che, H. Z., Wang, Y. Q., Zhang, X. Y., Zhu, R., and Liu, S. H.: Interaction Between Planetary Boundary Layer and $PM_{2.5}$ Pollution in Megacities in China: a Review. Curr. Pollut. Rep., 5, 261–271, https://doi.org/10.1007/s40726-019-00124-5, 2019.

Quan, J. N., Gao, Y., Zhang, Q., Tie, X. X., Cao, J. J., Han, S. Q., Meng, J. W., Chen, P. F., and Zhao, D. L.: Evolution of planetary boundary layer under different weather conditions, and its impact on aerosol concentrations, Particuology, 11(1), 34-40, https://doi.org/10.1016/j.partic.2012.04.005, 2013.

Quan, J. N., Xu, X. D., Jia, X. C., Liu, S. H., Miao, S. G., Xin, J. Y., Hu, F., Wang, Z. F., Fan, S. J., Zhang, H. S., Mu, Y. J., Dou, Y. W., and Cheng, Z.: Multi-scale processes in severe haze events in China and their interactions with aerosols: Mechanisms and progresses (in Chinese). Chin. Sci. Bull., 65, 810–824, https://doi.org/10.1360/TB-2019-0197, 2020.

State Council of the People’s Republic of China: Notice of the General Office of the State Council on Issuing the Air Pollution Prevention and Control Action Plan, State Council of the People’s Republic of China website. Available at: http://www.gov.cn/zwgk/2013-09/12/content_2486773.htm, 2013.

State Council of the People’s Republic of China: Notice of the General Office of the State Council on

Issuing the Air Pollution Prevention and Control Action Plan, State Council of the People's Republic of China website. Available at: http://www. gov.cn/zhengce/content/2018-07/03/content_5303158.htm. 2018.

Wang, Y. S., Li, W. J., Gao, W. K., Liu, Z. R., Tian, S. L., Shen, R. R., Ji, D. S., Wang, S., Wang, L. L., Tang, G. Q, Song, T., Cheng, M. T., Wang, G. H., Gong, Z. Y., Hao, J. M., and Zhang, Y. H.: Trends in particulate matter and its chemical compositions in China from 2013–2017. Sci. China Earth Sci., 62: 1857–1871, https://doi.org/10.1007/s11430-018-9373-1, 2019.

[2. The Great Smog of London in 1952 is one of the most well-known air pollution events across the world. Comparatively speaking, the haze in the North China Plain in February 2014 is not that "eye-catching". Why chose this pollution episode for comparison? 2013 Beijing Haze has drawn more attention from both scientific research and public concern.]

Reply: Meteorological conditions in February 2014 were worse than that in January 2013. In February 2014, a rarely persistent air pollution weather process occurred in central and eastern China, this process had caused severe air pollution in more than 50 cities, with an impact area of 2.07 million km^2. In the Beijing area during February 20–26, 2014, the regional average $PM_{2.5}$ concentration exceed the 'most-serious' air pollution level, and with a peak value of up to 456 $\mu g\ m^{-3}$.

[3. It is plausible that 'Warm Cover' may intensify the haze pollution in Eastern China, theoretically. However, as mentioned by the authors, the thermodynamical structure is closely related to circulation, which can significantly influence the regional transport/ventilation of air pollutants. It needs to be clarified whether the anomalous circulation or thermodynamical structure (ABL height decrease) is the main cause of haze pollution. This work only provides correlation and cross-sections of temperature anomalies and $PM_{2.5}$ concentration, both of which are a little too descriptive. More in-depth discussion and some quantitative analysis are suggested to be provided.]

Reply: Your constructive suggestions are greatly appreciated and very helpful for our further study. Due to the limited space of the article, some quantitative analysis will be given in the future, such as the contribution of each meteorological element to polluted weather. This study focused on exploring whether the thermodynamic structure of the troposphere and its intensity changes can be used as a ‘strong warning signal’ for the changes of $PM_{2.5}$ concentrations in heavy air pollution, and whether this strong signal exists in the time scales of seasonal, interannual and interdecadal changes.

Minor comments:

[1. Line 26: "In addition to"]

Reply: Following this comment, we have adjusted it as required.

[2. Line 43: Delete "with excessive concentrations of PM2.5"]

Reply: It has been deleted in the revised manuscript.

[3. Line 87: the North China Plain. Please check it throughout the manuscript.]

Reply: Following this comment, we have checked it throughout the manuscript.

[4. Line 88-89: Change to "for the long-lasting and heavy haze pollution". This statement needs to be rephrased. "sulfur-dioxide pollutants” is not appropriate.]

Reply: It has been done in the revised manuscript.

[5. Line 98: What do you mean by "long heavy air pollution "?]

Reply: Following this comment, we have adjusted it as required.

"persistent air pollution".

[6. The labels in the contour plot in Fig.3-4 are overlaid and need to be optimized.]

Reply: Following this comment, we have adjusted it as required.

[7. All the abbreviations should be defined for the first time. Please check throughout the article.]

Reply: Following this comment, we have adjusted it as required.

Reply to Referee 2

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

[Review Comment: Anthropogenic pollutant emissions and unfavorable meteorological conditions are commonly regarded as two key factors for haze pollution. 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. It is a very interesting topic and significant for air pollution control. However, I think the current analysis is not sufficient to support the conclusion. Thus, some quantitative estimation and mechanisms illustration is suggested before publication. The*

detailed reason and suggestions are listed below.]

Reply: Thank you for the encouraging comments.

[1. Fig 1 and Fig 2 demonstrate the key role of "warm cover" in the haze process. However, the illustration of the connection of "warm cover" with the Tibetan Plateau has lacked. The "warm cover" shown in Figure S1 is below 900 hPa, which is similar to the height of the PBL top. It results in a very stable ABL and further improves the surface $PM_{2.5}$ *concentration. However, the "warm cover" induced by Tibetan Plateau is about 600 hPa, which is 4 km. The mechanisms of the impact of "warm cover" in such altitude on PBL is needed to be illustrated in the manuscript.]*

Reply: Many thanks for the referee's discussion. We agree with the suggestion. Following this comment, the content of Section 3.3 have adjusted (lines 168-173 and Figure 5) with following sentences:

"The concept of variations of the tropospheric 'warm cover' has been proposed in this work. Under the background of climate change, it is worth considering whether the variational tendency of the structure of the plateau's heat source induces variations of the tropospheric thermal structure in downstream areas of the Plateau, leading to the interdecadal variations of the frequency of haze events seen in Eastern China since the 21th century. Thermal anomalies of the TP also play an important role in the variations of the frequency of haze events in EC apart from the anthropogenic pollutant emission related to the rapid industrialization of China. The observational and modeling studies have demonstrated that the interannual variations in the thermal forcing of TP are positively correlated with the incidences of wintertime haze over EC (Xu et al., 2016). The TP induced changes in atmospheric circulation, increasing atmospheric stability and driving frequent haze events in EC (Xu et al., 2016). In this study, the data analysis concerning the interannual variations of the TP's apparent heat source and air temperature in wintertime at the TP with the altitudes above 3000 meters showed that since the 1960s the heat source in areas vulnerable to TP climate change strengthen continuously as the surface temperature increased (Fig. 5a). Furthermore, the TP's apparent heat and air temperature of the middle troposphere over EC presented the significant positive correlation passing (90 % confidence level), which is similar to 'warm cover' structures (Fig. 5b). Therefore, we considered that the 'warm cover' change in the middle troposphere over EC was closely related to TP's apparent heat and the surface temperature. The TP induced changes in thermodynamic structure of atmospheric provided favorable climatic backgrounds driving air pollution events in EC."

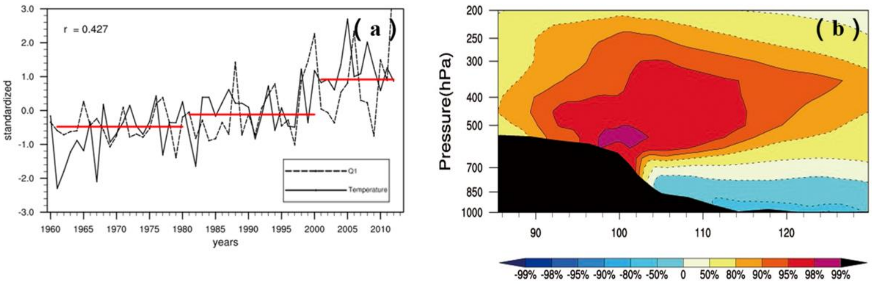

Figure 5. (a) Interanual variations of TP's apparent heat source (Q_1) and air temperature of meteorological stations in the TP with the altitudes above 3000 meters in the winters during 1960-2014; (b) Vertical cross sections of the correlations between TP's apparent heat (Q_1) and air temperature latitude-averaged along 30-35 °N in the winters during 1960-2014.

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

Xu, X. D., Zhao, T. L., Liu, F., Gong, S. L., Kristovich, D., Lu, C., Guo, Y., Cheng, X. H, Wang, Y. J., and Ding, G.: Climate modulation of the Tibetan Plateau on haze in China, Atmos. Chem. Phys., 16, 1365–1375, https://doi.org/10.5194/acp-16-1365-2016, 2016.

[2. Fig 3 shows that the "upper warming and bottom cooling" vertical structure in Autumn and Winter favors haze formation. It is interesting. However, the analysis is on the seasonal scale and did not directly support the haze formation on the daily scale.]

Reply: Based on the study of the Great Smog of London in 1952 and the heavy pollution of Beijing in February 2014, it is found that the abnormal 'warm cover' in the middle troposphere connected to both severe air pollution events (Sect. 3.1 and in Sect. 3.2). This study attempts to explore that whether such the similar structural characteristic of thermodynamic structure, i.e. the abnormal 'warm cover' in the middle troposphere, also exist from the perspective of different time scales, we have further analyzed the $PM_{2.5}$ concentrations and the number of haze days with seasonal and interdecadal variations of the thermodynamic structures in the atmosphere. We found that the thermal vertical structure of atmospheric showed a 'upper warming and bottom cooling' vertical structure under heavy pollution conditions. The concept of the tropospheric 'warm cover' has been confirmed on the seasonal and climatic scale.

Following this comment, we have added these in the revised Abstract (line 32) as follows:

"The anomalous structure of the troposphere's 'warm cover' not only exist in heavy haze pollution on the daily scale, but also provide seasonal, interannual and interdecadal 'strong signals' for frequently occurring regional haze pollution."

[3. Fig 4 compares the interdecadal change of thermal structure in EC and eastern TP with haze days. However, the anthropogenic emissions in EC have increased several times from 1961 to 2018. It is hard to attribute the increase of haze days to the change of TP thermal structure.]

Reply: The pollutant emission with high intensity was the internal cause of frequent air pollution in EC, and the adverse weather conditions were often the key 'inducement' for the accumulation of air pollutants in the atmosphere. Although the variation trends of air pollution in EC depend on the air pollutant emissions, the air pollution, including its intensity and duration, are closely related to meteorological conditions. Thus, we analyzed the anomalous thermodynamic structure (air temperature anomalies) from the perspective of meteorological conditions, in order to reveal the influence difference of the background field of meteorological conditions on regional atmospheric dispersion conditions. Furthermore, we found that the TP induced changes in thermodynamic structure of atmospheric provided favorable climatic backgrounds driving air pollution events in EC.

[4. I guess the impact of TP thermal structure on air pollution may cover a large part of EC. Maybe large-scale haze processes could be tried.]

Reply: Yes, the observational and modeling studies have demonstrated that the interannual variations in the thermal forcing of TP are positively correlated with the incidences of wintertime haze over EC (Xu et al., 2016). The TP induced changes in atmospheric circulation, increasing atmospheric stability and driving frequent haze events in EC (Xu et al., 2016).

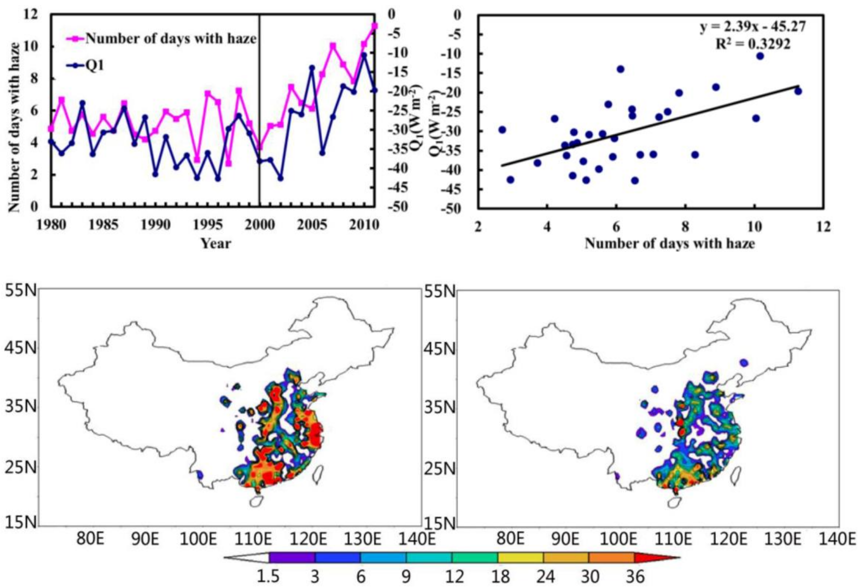

Figure 4. Interannual variability in the apparent heat source Q1 (the negative values denote cooling) integrated vertically over the TP and haze event frequency averaged in the CEC in winter (December, January and February) over 1980–2012 and their correlation (upper panel). The haze frequencies (days)

averaged in five winters with most positive (lower left panel) and most negative Q1 anomalies (lower right panel) on the TP relative to the mean haze frequency from 1980 to 2012 (Xu et al., 2016).

References:

Xu, X. D., Zhao, T. L., Liu, F., Gong, S. L., Kristovich, D., Lu, C., Guo, Y., Cheng, X. H, Wang, Y. J., and Ding, G.: Climate modulation of the Tibetan Plateau on haze in China, Atmos. Chem. Phys., 16, 1365–1375, https://doi.org/10.5194/acp-16-1365-2016, 2016.

Reply to the Peer-Reviewer

[Supplementary suggestions for revision: As pointed out by RC2, I still think this manuscript is too descriptive and the majority of the main text is just describing the "warm cover" phenomenon. It does need more quantitative estimation before publication. Or some additional discussion on the implication of this finding can add more scientific significance to this work. In addition, the tense and some English expressions in this manuscript are too confusing and need to be double-checked. English language editing is suggested.]

Reply: Thank you very much to the reviewers. We agree with the comments and suggestions of the reviewers. We have proofread and revised the language of the manuscript, and revised the content of Section 4 of the manuscript:

"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 anomalous 'warm cover' in the middle troposphere was identified as a precursory 'strong signal' for severe air pollution events, which could be attributed to climate change. A stable thermal structure in the middle troposphere, i.e. a 'warm cover', suppressed the ABL development, which was a key 'inducement' for the accumulation of air pollutants in the ambient atmosphere.

From the perspective of the thermal vertical structure in the troposphere, the abnormal vertical structure in the troposphere during heavy air pollution were understood in this study. The thermal structure formed by the conventional decline rate of atmospheric air temperature often 'covers up' the anomalous 'strong signal' of the troposphere in air pollution process, such as the abnormal stable structure with the middle warm and bottom cold in the troposphere with air temperature anomalies. The 'strong signal' of the 'warm cover' of air temperature anomalies in the middle troposphere during heavy air pollution can be described by the method of statistical comprehensive diagnosis analysis.

A large-scale anomalous air temperature pattern of 'upper warming and bottom cooling' in the troposphere appeared from the TP 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 TP'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."

Manuscript Revised edition Mark:

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

Xiangde Xu[1], Wenyue Cai[1, 2, 3], Tianliang Zhao[4], Xinfa Qiu[5], Wenhui Zhu[6], Chan Sun[1], Peng Yan[7], Chunzhu Wang[8], and Fei Ge[9]

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[8]Training Center, China Meteorological Administration, Beijing, China.

[9]School of Atmospheric Sciences/Plateau Atmosphere and Environment Key Laboratory of Sichuan Province/Joint Laboratory of Climate and Environment Change, Chengdu University of Information Technology, Chengdu, Sichuan Province, China.

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Correspondence: Wenyue Cai (caiwy@cma.gov.cn) and Tianliang Zhao (tlzhao@nuist.edu.cn)

Abstract. Eastern China (EC), located in the downstream region of Tibetan Plateau (TP), is a large area with frequent haze pollution. In addition to 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' (air temperature warm anomalies) in the middle troposphere, as a precursory 'strong signal', could connect to severe air pollution events. The convection and vertical 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 was favourable for the accumulation of air pollutants in the ambient atmosphere. The anomalous structure of the troposphere's 'warm cover' not only exist in heavy haze pollution on the daily scale, but also provide seasonal, interannual and interdecadal 'strong signals' for frequently occurring regional haze pollution. It is revealed that a close relationship existed between interannual variations of the TP's heat source and the 'warm cover' strong-signal in the middle troposphere over EC. The warming TP could lead to the anomalous 'warm cover' in the middle troposphere from the plateau to the downstream EC region and even the entire East Asian region for air pollution.

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 in winter (Wang, et al., 2017). Therefore, in September 2013, the Chinese government launched the China's first air pollution control action plan-'The Airborne Pollution Prevention and Control Action Plan (2013-2017)' (State Council of the People's Republic of China, 2013). By 2017, about 64 % of China's cities are still suffering from air pollution, especially Beijing-Tianjin-Hebei region and surrounding areas (Wang et al., 2019; Miao et al., 2019). Then, in July 2018, the Chinese government launched the second three-year action plan for air pollution control, 'the blue sky defense plan', which demonstrates China's firm determination and new measures for air pollution control (State Council of the People's Republic of China, 2018). After the implementation of air pollution control action plans, air quality in many regions in China has been significantly improved.

Anthropogenic pollutant emissions and unfavorable meteorological conditions are commonly regarded

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as two key factors for air pollution (Ding and Liu, 2014; Yim et al., 2014; Zhang et al., 2015). Air pollutants mainly come from surface emission sources, and most of air pollutants are injected from the surface to the atmosphere through the atmospheric boundary layer (ABL) (Quan et al., 2020). The ABL structures are the key meteorological conditions which influences the formation and maintenance of heavy air pollution episodes (Wang et al., 2015; Cheng et al., 2016; Wang et al., 2016; Tang et al., 2016; Wang et al., 2019).

Most of the previous studies focused on exploring the impact on the heavy air pollution in Eastern China (EC) from the meteorological conditions in ABL. However, the thermodynamic and dynamic structures of free troposphere can affect the meteorological conditions in ABL (Cai et al., 2020). The convection and diffusion in the ABL are suppressed by a relatively stable structure in the middle troposphere, leading to the ABL height decreases, which was favourable for the formation and persistence of heavy air pollution (Quan et al., 2013; Wang et al., 2015; Cai et al., 2020).

This study investigated whether the thermodynamic structure of the troposphere and its intensity changes can be used as a ‘strong warning signal’ for the changes of $PM_{2.5}$ concentrations in heavy air pollution, and whether this strong signal exists in the time scales of seasonal, interannual and interdecadal changes. In order to explore the interaction between the free troposphere and the ABL and the impact on the heavy air pollution in EC, this study extended the meteorological conditions for heavy air pollution from the boundary layer to the middle troposphere. We identify a precursory ‘strong signals’ hidden in the free troposphere for frequent haze pollution in winter in EC.

2 Data and methods

The data used in this study included NCEP/NCAR and ERA-Interim reanalysis data of meteorology, as

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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° of 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.

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 the People's Republic 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://data.cma.cn/).

Furthermore, the L-band sounding 'seconds-level' data of Beijing from 2010 to 2019 to were used to calculate the height of 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 (Zhu et al., 2018), and interpolation was implemented in a vertical direction at an interval of 2 hPa. The L-band detection data provided by the

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China Meteorological Information Center (http://data.cma.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	$PM_{2.5}$ concentration ranges
‘less-serious’ pollution	75 μg·m^{-3}< $PM_{2.5}$≤115 μg·m^{-3}
‘serious’ pollution	115 μg·m^{-3}< $PM_{2.5}$≤150 μg·m^{-3}
‘more-serious’ pollution	150 μg·m^{-3}< $PM_{2.5}$≤250 μg·m^{-3}
‘most-serious’ pollution	$PM_{2.5}$>250 μg·m^{-3}

3 Results

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

In February 2014, a rarely persistent air pollution weather process occurred in EC with severe air pollution in more than 50 cities, with an impact area of 2.07 million km^2. In the Beijing area during February 20–26, 2014 the regional average $PM_{2.5}$ concentration exceed the ‘most-serious’ air pollution level, and with a peak value of up to 456 μg·m^{-3}. In addition, the Great Smog of London in 1952 was attributed to the long-lasting and heavy haze pollution under the influence of certain weather systems (Whittaker et al., 2004). To find the precursory ‘strong signals’ hidden in meteorology for heavy air pollution events, we retrieved the three-dimensional atmospheric dynamic and thermal structures 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 both areas at 500 hPa geopotential height anomalies, as shown in Figs. 1a and 1b. During the heavy air pollution events, a high-pressure system over London as well as Beijing gradually strengthened (Figs. 1c and 1d), and the middle troposphere was characterized by a ‘warm cover’ with ‘upper warming and bottom cooling’ anomalies in vertical structure of air temperature (Figs. 1e and 1f).

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By comparing Figs. 1a and 1b, we found that two persistent heavy air pollution events occurred during the maintenance stage of stable high pressure system. During stagnation of the blocking high pressure system, the strength of the center of the geopotential height anomalies in the stable maintenance region of the blocking exhibited a synchronous response to the ‘warm cover’ above areas (Figs. 1c–1f). It can be seen that the local atmospheric thermal structure is significantly modulated by the persistent large-scale anomalous circulation. The ‘subsidence-induced air temperature inversion’ effect of the blocking high pressure system continuously strengthened the ‘warm cover’ structure in the middle troposphere, which suppressed the vertical diffusion capacity in the atmosphere (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 (Figs. 1g and 1h). The above analysis shows that in the ABL over London during December 5–9, 1952 and Beijing during February 21–26, 2014, the inversion layer height decreased, which made the ABL structure stable for accumulation of air pollutants. 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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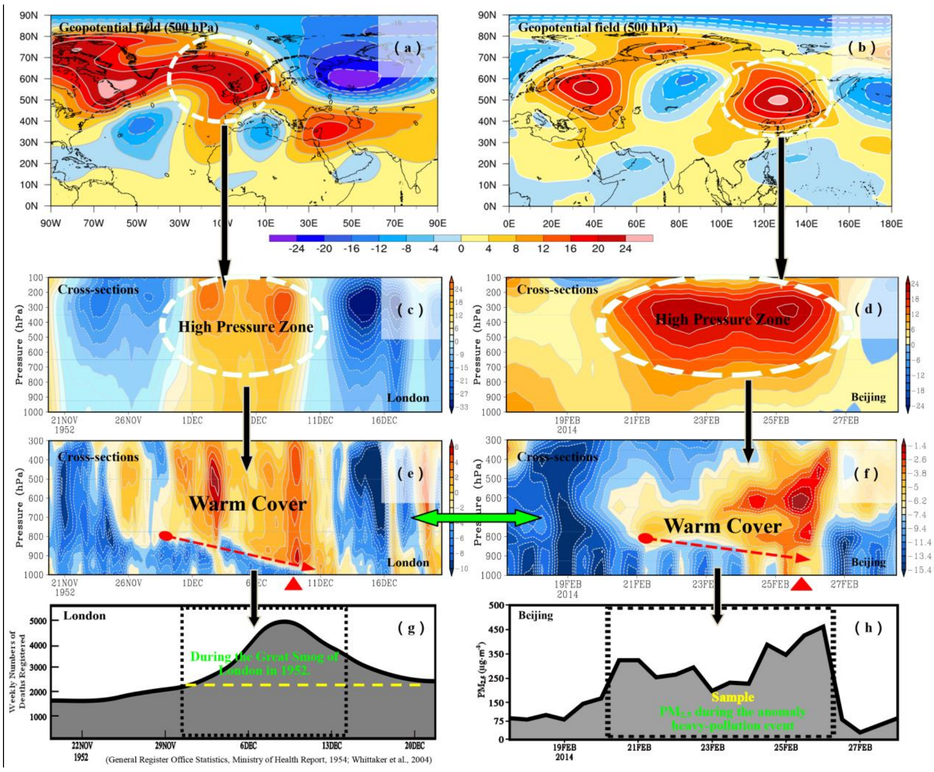

Figure 1. Dynamical and thermodynamical structures and air pollution variations: (a) geopotential height anomalies (unit: dagpm) at 500 hPa during December 5–9, 1952 for the Great Smog of London, (b) the same as (a) but during February 21–26, 2014. (c) Time-vertical cross-sections of the geopotential height anomalies (unit: dagpm) in the high pressure area (50-70 °N; 20 ° W-10 ° E) during November 20 to December 20, 1952，(d) the same as (c) but in the high pressure area (40-63 ° N; 115-138 ° E) during February 17–28, 2014. (e) Time-vertical cross-sections of air temperature anomalies (unit: ℃) over London (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 Effect of 'Warm Cover' in the free troposphere on ABL and surface $PM_{2.5}$ 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, Figs. 2a and 2b demonstrated the significant negative correlations between the height of the ABL and air temperature anomalies over same period and 24 hours ahead in Beijing, and the correlation coefficients were 0.41 and 0.34 (99.9 % confidence level), 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. Figures 2c–2e presented the significant positive correlations between $PM_{2.5}$ concentrations and air temperature anomalies over same period and 24, 48 hours ahead in Beijing, and the correlation coefficients were 0.42, 0.56 and 0.37 (99.9 % confidence level). Based on the above mentioned results, air temperature anomalies over 24 and 48 hours ahead could also be 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 vertical 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).

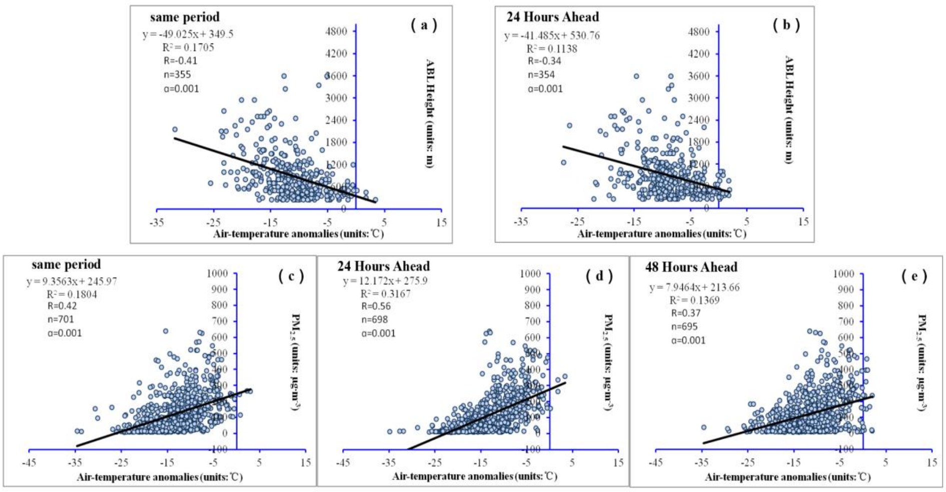

Figure 2. The correlations between ABL height and air temperature anomalies in Beijing during winter 2014–2017, (a) same period, at 800 hPa; (b) 24 hours ahead, at 650 hPa. The correlations between $PM_{2.5}$ concentration and air temperature

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anomalies in Beijing during winter 2014–2017, (c) same period, at 850 hPa; (d) 24 hours ahead, at 800 hPa; (e) 48 hours ahead, at 724 hPa

3.3 Changes of the 'warm cover' structure in the middle troposphere

The 'warm cover' structure of air temperature anomalies in the middle 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 EC exhibited the significant seasonal variations. Our study revealed that existed seasonal differences of the thermal structures in the atmosphere over EC. In spring (Figs. 3a and 3e) and summer (Figs. 3b and 3f), the middle troposphere was characterized by a 'upper cooling and bottom warming' vertical structure for less air pollution. When the autumn (Figs. 3c and 3g) and winter (Figs. 3d and 3h) 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 Tibetan Plateau (TP). Furthermore, the thermal structures of the troposphere exhibited the distinct interdecadal variations (Figs. 4a-4c). 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

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

The concept of variations of the tropospheric 'warm cover' has been proposed in this work. Under the background of climate change, it is worth considering whether the variational tendency of the structure of the plateau's heat source induces variations of the tropospheric thermal structure in downstream areas of the Plateau, leading to the interdecadal variations of the frequency of haze events seen in Eastern China since the 21th century. Thermal anomalies of the TP also play an important role in the variations of the frequency of haze events in EC apart from the anthropogenic pollutant emission related to the rapid industrialization of China. The observational and modeling studies have demonstrated that the interannual variations in the thermal forcing of TP are positively correlated with the incidences of wintertime haze over EC (Xu et al., 2016). The TP induced changes in atmospheric circulation, increasing atmospheric stability and driving frequent haze events in EC (Xu et al., 2016). In this study, the data analysis concerning the interannual variations of the TP's apparent heat source and air temperature in wintertime at the TP with the altitudes above 3000 meters showed that since the 1960s the heat source in areas vulnerable to TP climate change strengthen continuously as the surface temperature increased (Fig. 5a). Furthermore, the TP's apparent heat and air temperature of the middle troposphere over EC presented the significant positive correlation passing (90 % confidence level), which is similar to 'warm cover' structures (Fig. 5b). Therefore, we considered that the 'warm cover' change in the middle troposphere over EC was closely related to TP's apparent heat and the surface temperature. The TP induced changes in thermodynamic structure of atmospheric provided favorable climatic backgrounds driving air pollution events in EC.

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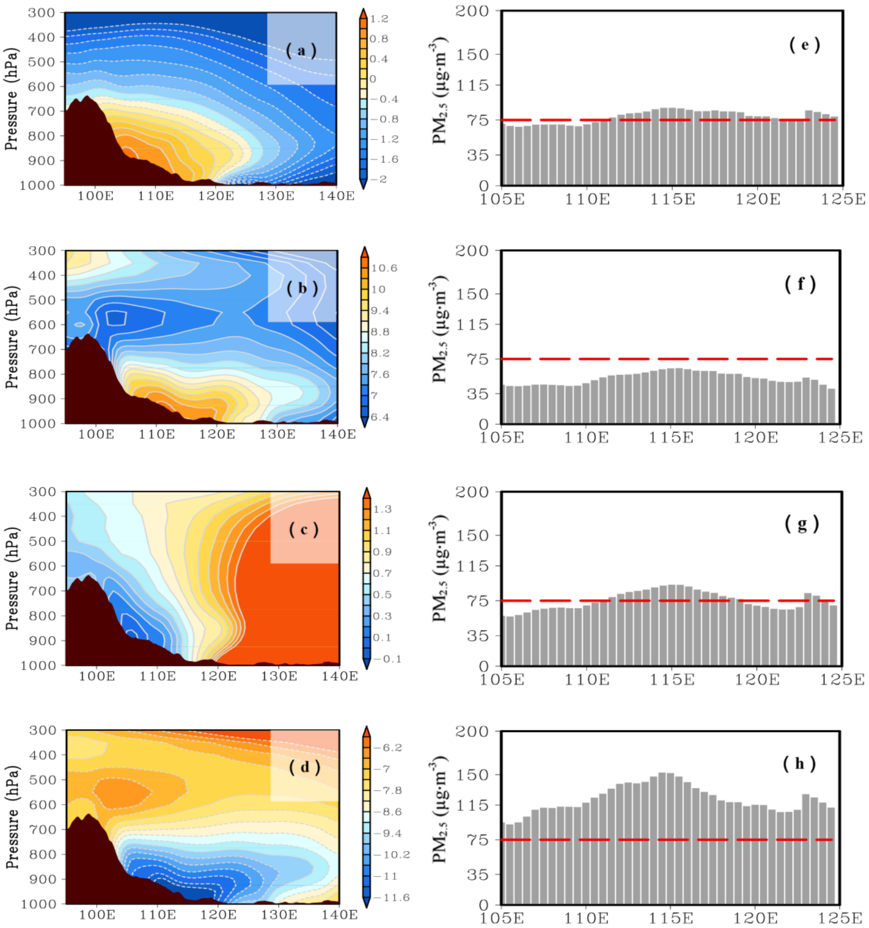

Figure 3. Vertical cross sections of (a-d) air temperature anomalies (unit: ℃) and (e-h) the $PM_{2.5}$ concentrations (unit: μg·m^{-3}) averaged along 25-40 °N in spring (a, e), summer (b, f), autumn (c, g), winter (d, h) from 2013 to 2018.

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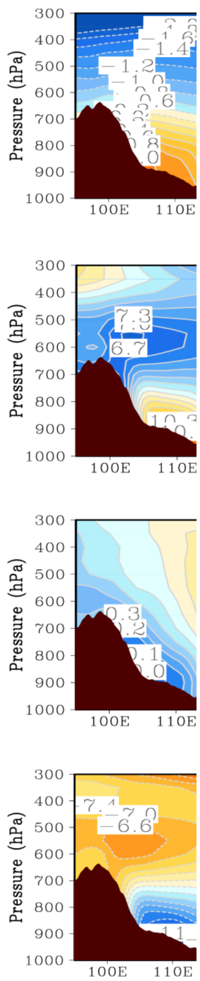

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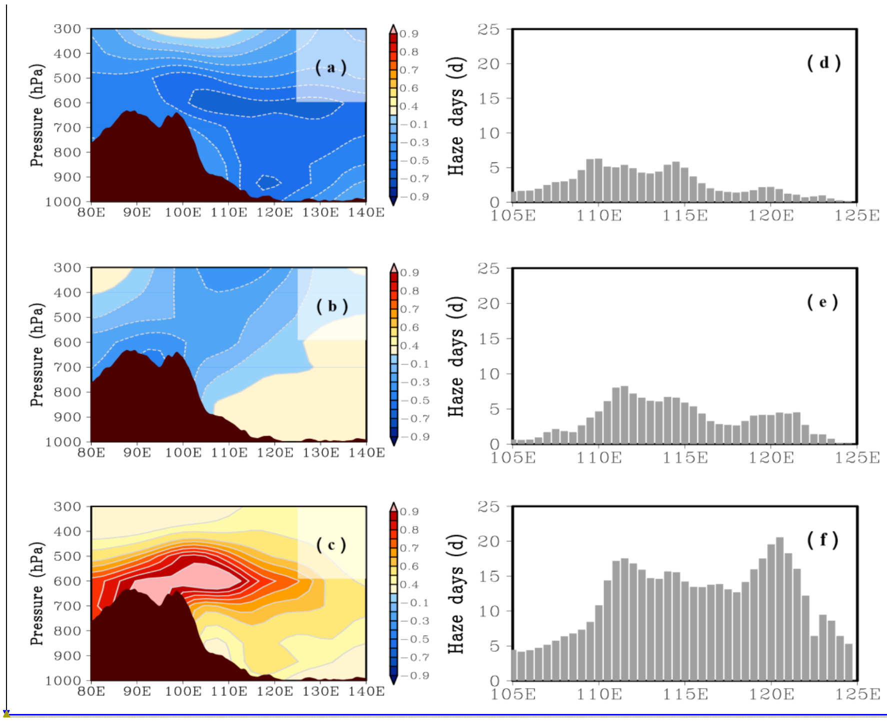


Figure 4. Vertical cross sections of (a-c) air temperature anomalies (unit: ℃) and (d-f) the number of haze days averaged along 25-40 °N in winter during 1961-1980 (a, d), 1981-2000 (b, e) and 2001-2018 (c, f).

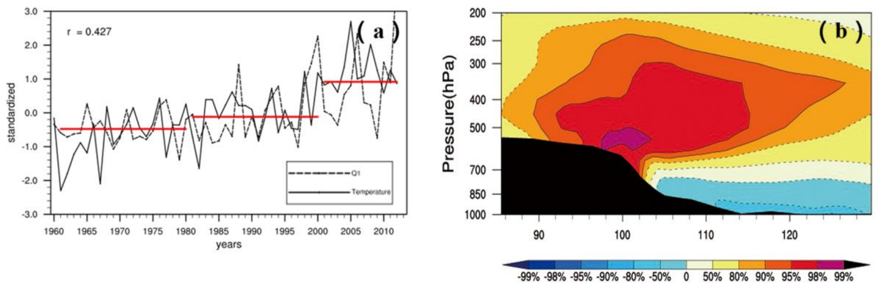
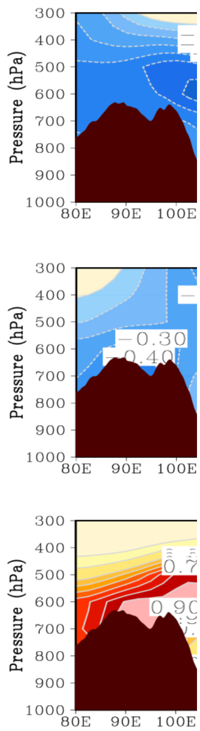


Figure 5. (a) Interanual variations of TP's apparent heat source (Q_1) and air temperature of meteorological stations in the TP with the altitudes above 3000 meters in the winters during 1960-2014; (b) Vertical cross sections of the correlations between TP's apparent heat (Q_1) and air temperature latitude-averaged along 30-35 °N 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



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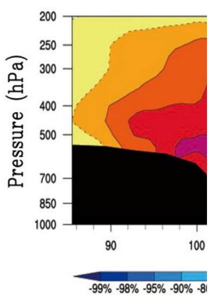

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as $PM_{2.5}$ pollution over EC, the anomalous ‘warm cover’ in the middle troposphere was identified as a precursory ‘strong signal’ for severe air pollution events, which could be attributed to climate change. A stable thermal structure in the middle troposphere, i.e. a ‘warm cover’, suppressed the ABL development, which was a key ‘inducement’ for the accumulation of air pollutants in the ambient atmosphere.

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From the perspective of the thermal vertical structure in the troposphere, the abnormal vertical structure in the troposphere during heavy air pollution were understood in this study. The thermal structure formed by the conventional decline rate of atmospheric air temperature often ‘covers up’ the anomalous ‘strong signal’ of the troposphere in air pollution process, such as the abnormal stable structure with the middle warm and bottom cold in the troposphere with air temperature anomalies. The ‘strong signal’ of the ‘warm cover’ of air temperature anomalies in the middle troposphere during heavy air pollution can be described by the method of statistical comprehensive diagnosis analysis.

A large-scale anomalous air temperature pattern of ‘upper warming and bottom cooling’ in the troposphere appeared from the TP 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 TP’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.

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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 the People's Republic of 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://data.cma.cn/). All data presented in this paper are available upon request to the corresponding author (Wenyue Cai, caiwy@cma.gov.cn).

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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, CZW and FG contributed to subsequent revisions.

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

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