Dear Editors and Referees:

Thank you very much for your careful review and constructive comments on our manuscript acp-2021-709. We have accordingly made the substantial revisions. The revised portions are highlighted in the revised manuscript. In the following, we quoted each review question in the square brackets and added our response after each paragraph.

Responses to Referee #1

*[**1. General comments:** The manuscript by Sun et al. analyzes the impact of meteorological factors on the changes in $PM_{2.5}$ concentration in Twain-Hu Basin, China using a Kolmogorov-Zurbenko filter. They conclude that interannual and seasonal meteorology have the largest impacts on the changes in $PM_{2.5}$. However, the method used in this work is not validated using synthetic data or with other methods, so the accuracy of the results is doubtful. In addition, this work focuses on a very small region, and the results may not have broader implications for national - global air pollution issues and may not fit the scope of the general ACP readership.]*

Response 1.1: Thanks for the referee's comments and suggestions. Please find our response as follows and the subsequent **Response 1.2** to the referee's comments and suggestions:

According to the suggestions, we have conducted the simulation experiments with Weather Research and Forecasting model with Chemistry (WRF-Chem) to validate the accuracy of the results with KZ filter, which is added in the new *Sect. 3.6* as follows:

3.6 Meteorological contribution to $PM_{2.5}$ changes validated with WRF-Chem modeling

The above observational study investigated the meteorological influence on the changes in $PM_{2.5}$ concentrations in the THB using KZ filter, with concluding the large impact of meteorology on the $PM_{2.5}$ changes over 2015–2019. To validate this conclusion of analyses with KZ filter, we designed three sets of modeling experiments CTRL, SENS-MET and SENS-EMI (Table S6) for December of 2015–2019, respectively driven with the changing meteorology and anthropogenic emissions over 2015–2019, the fixed meteorological conditions and anthropogenic emissions of 2015 with atmospheric chemical model

WRF-Chem (Weather Research and Forecasting model with Chemistry). Air pollutant emission inventories, modeling configuration, experiment design and modeling verification were described in the supplement. The modeling verification of experiments CTRL indicated that $PM_{2.5}$ and meteorology were reasonably reproduced by the WRF-Chem simulation (Figs.S4–S5, Table S7), and the designed three sets of modeling experiments CTRL, SENS-MET and SENS-EMI could be used in the further analyses of emission and meteorological impact on $PM_{2.5}$ change over 2015–2019 to confirm the results of KZ filter.

We derived the effect of meteorology by comparing the simulated $PM_{2.5}$ concentrations in the three sets of experiments CTRL, SENS-MET and SENS-EMI (Table S6). The relative contribution of meteorology to the interannual changes of $PM_{2.5}$ concentrations was calculated with a linear additive relationship of contributions of meteorology and emission in the following equations:

$$Con_{MET} = \frac{k_{MET}}{k_{CTRL}} \tag{11}$$

$$Con_{EMI} = \frac{k_{EMI}}{k_{CTRL}} \tag{12}$$

$$RCon_{MET} = \frac{Con_{MET}}{Con_{MET} + Con_{EMI}} \times 100\% \tag{13}$$

k_{CTRL}, k_{MET} and k_{EMI} represent the trends in interannual changes of $PM_{2.5}$ concentrations simulated by the experiments CTRL, SENS-MET and SENS-EMI, respectively. Con_{MET} and Con_{EMI} are the contribution of meteorology and emission, and $RCon_{MET}$ is the contribution rate (%) of meteorology to interannual changes of $PM_{2.5}$ concentrations (Zhang et al., 2020).

Based on WRF-Chem modeling experiments, we assessed the impact of meteorological changes on interannual $PM_{2.5}$ variations from 2015 to 2019 with *Eqs. (11–13)*. The relative contribution of meteorology to interannual $PM_{2.5}$ variations displayed the regional pattern of northern positive and southern negative values over the THB (Fig. 10), confirming the impact of meteorological changes by accelerating and offsetting the effects of emission reductions on $PM_{2.5}$ declining trends in the northern and southern THB, respectively. The general spatial distribution of meteorological contribution rates to $PM_{2.5}$ declining trends from the WRF-Chem simulation was consistent with the results using KZ filter (Figs. 9 and 10), validating the results with KZ filter that meteorological drivers exerted a contrary impact of northern positive and southern negative contribution on long-term changes of $PM_{2.5}$ concentrations in the THB.

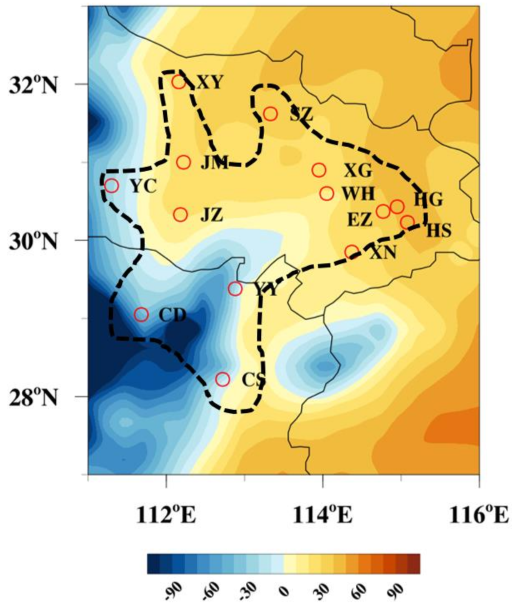

Figure 10 Spatial distribution of contribution rates of meteorological variations to $PM_{2.5}$ reductions based on WRF-Chem modeling experiments (contour, unit: %) in the THB outlined with black dashed line and surrounding regions for December of 2015–2019.

*[**1. General comments:** … … … … In addition, this work focuses on a very small region, and the results may not have broader implications for national - global air pollution issues and may not fit the scope of the general ACP readership]*

Response 1.2: In response to the above comments, we have clarified the highlights and implications in the revised *Abstract* and *Introduction* as follows:

The THB covering a large region of two provinces, Hubei and Hunan in central China, is surrounded by the high air pollutant emission regions in North China Plain (NCP) to the north, Yangtze River Delta (YRD) to the east, Pearl River Delta (PRD) to the south and Sichuan Basin (SB) to the west (Lin et al., 2018). Driven by East Asian monsoonal winds over Central Eastern China, THB is a major receptor region in regional transport of air pollutants over China (Shen et al., 2020). Governed by the multi-scale atmospheric circulations, air pollutants emitted from the upwind source regions can be transported easily to the downstream receptor region exacerbating the regional air quality, which can result in a complicated relation of source and receptor in regional transport of air pollutants (Hu et al., 2021). However, the previous studies mostly focused on the atmospheric environment change in the source regions with high anthropogenic emissions of air pollutants, and there have been few assessments on multi-scale changes of atmospheric environment over the receptor region in regional transport of air pollutants. In the present study of 5-year observations and modeling, we targeted the THB, a large region of heavy $PM_{2.5}$ pollutions over central China, to assess the meteorological effect on $PM_{2.5}$ changes over a receptor region in regional transport of air pollutants, and we assessed the contributions of air pollutant emissions and meteorological conditions to air quality change over this receptor region with the long-term observations over recent years. Our results highlight the effects of emission mitigation and meteorological changes on source-receptor relationship of region transport of air pollutants with the implication of long-range transport of air pollutants for regional and global environment changes. Therefore, the results in this paper have broader implications for regional - global air pollution issues and fit the scope of the general ACP readership.

References:

Hu, W. Y., Zhao, T. L., Bai, Y. Q., Kong, S. F., Xiong, J., Sun, X. Y., Yang, Q. J., Gu, Y., and Lu, H. C.: Importance of regional PM2. 5 transport and precipitation washout in heavy air pollution in the Twain-Hu Basin over Central China: Observational analysis and WRF-Chem simulation, Science of the Total Environment, 758, 143710, 2021.

Lin, C. Q., Liu, G. H., Lau, A. K. H., Li, Y., Li, C. C., Fung, J. C. H., and Lao, X. Q.: High-resolution satellite remote sensing of provincial PM2. 5 trends in China from 2001 to 2015, Atmospheric Environment, 180, 110-116, 2018.

Shen, L. J., Wang, H. L., Zhao, T. L., Liu, J., Bai, Y. Q., Kong, S. F., and Shu, Z. Z.: Characterizing regional aerosol pollution in central China based on 19 years of MODIS data: Spatiotemporal variation and aerosol type discrimination, Environmental Pollution, 263, 114556, 10.1016/j.envpol.2020.114556, 2020.

Zhang, W. J., Wang, H., Zhang, X. Y., Peng, Y., Zhong, J. T., Wang, Y. Q., and Zhao, Y. F.: Evaluating the contributions of changed meteorological conditions and emission to substantial reductions of PM2.5 concentration from winter 2016 to 2017 in Central and Eastern China, Science of The Total Environment, 716, 136892, 2020.

[2. Abstract, please clarify why THB is selected as the studied region in this work.]

Response 2: The THB covering a large region of two provinces, Hubei and Hunan in central China, is surrounded by the high air pollutant emission regions in North China Plain (NCP) to the north, Yangtze River Delta (YRD) to the east, Pearl River Delta (PRD) to the south and Sichuan Basin (SB) to the west (Lin et al., 2018). Driven by East Asian monsoonal winds over Central Eastern China, THB is a major receptor region in regional transport of air pollutants over China (Shen et al., 2020). Governed by the multi-scale atmospheric circulations, air pollutants emitted from the upwind source regions can be transported easily to the downstream receptor region exacerbating the regional air quality, which can result in a complicated relation of source and receptor in regional transport of air pollutants (Hu et al., 2021). However, the previous studies mostly focused on the atmospheric environment change in the source regions with high anthropogenic emissions of air pollutants, and there have been few assessments on multi-scale changes of atmospheric environment over the receptor region in regional transport of air pollutants. Thus, we assessed the contributions of air pollutant emissions and meteorological conditions to air quality change over this receptor region in central China with the long-term observations over recent years. Our results highlight the effects of emission mitigation and meteorological changes on source-receptor relationship of region transport of air pollutants with the implication of long-range transport of air pollutants for regional and global environment changes. In the revised *Abstract,* we have clarified why THB is selected as the studied region in this work as follows:

As an important issue in atmospheric environment, the contributions of anthropogenic emissions and meteorological conditions to air pollution have been few assessed over the receptor region in regional transport of air pollutants. In the present study of 5-year observations and modeling, we targeted the Twain-Hu Basin (THB), a large region of heavy $PM_{2.5}$ pollutions over central China, to assess the meteorological effects on $PM_{2.5}$ change over a receptor region in regional transport of air pollutants. … … … … Our results highlight the effects of emission mitigation and meteorological changes on source-receptor relationship of region transport of air pollutants with the implication of long-range transport of air pollutants for regional and global environment changes.

References:

Hu, W. Y., Zhao, T. L., Bai, Y. Q., Kong, S. F., Xiong, J., Sun, X. Y., Yang, Q. J., Gu, Y., and Lu, H. C.: Importance of regional PM2. 5 transport and precipitation washout in heavy air pollution in the Twain-Hu Basin over Central China: Observational analysis and WRF-Chem simulation, Science of the Total Environment, 758, 143710, 2021.

Lin, C. Q., Liu, G. H., Lau, A. K. H., Li, Y., Li, C. C., Fung, J. C. H., and Lao, X. Q.: High-resolution satellite remote sensing of provincial PM2. 5 trends in China from 2001 to 2015, Atmospheric Environment, 180, 110-116, 2018.

Shen, L. J., Wang, H. L., Zhao, T. L., Liu, J., Bai, Y. Q., Kong, S. F., and Shu, Z. Z.: Characterizing regional aerosol pollution in central China based on 19 years of MODIS data: Spatiotemporal variation and aerosol type discrimination, Environmental Pollution, 263, 114556, 10.1016/j.envpol.2020.114556, 2020.

[3. L99-100, please clarify what the numbers in the subscript of KZ stand for and why using 1.7 years here.]

Response 3: The KZ filter $KZ_{m,p}$ is a low-pass filter based on an iterative moving average to remove the high frequency variations from the daily observational data, *m* and *p* in the subscript of KZ are moving average (unit: day) and number of iterations (unit: time) respectively.

By comparing different sets of moving average *m* and number of iterations *p*, it was found that the decomposed time series using $KZ_{15,5}$ (15-day length with five iterations) filter exhibited no white noise (short-term component), and the trend of long-term component derived with $KZ_{365,3}$ (365-day length with three iterations) filter corresponded approximately to the interannual trend of the original data, so that $KZ_{15,5}$ and $KZ_{365,3}$ filters were used to decompose the short-term and long-term components from the daily observational data (Rao and Zurbenko, 1994; Eskridge et al., 1997).

Based on the spectral decompositions of the daily observational data and three components (Fig. R1), the power spectral of daily observational data in periods less than 33 days and longer than 632 days (1.7 years) have been well reproduced by short-term and long-term components, and seasonal component represents well the seasonal variations, i.e., periods between 33 days and 1.7 years. We also clarified why using 33 days and 1.7 years in the revised manuscript (Lines 106–113) as shown below:

By comparing different sets of moving average *m* and number of iterations *p*, it was found that the decomposed time series using $KZ_{15,5}$ filter exhibited no white noise (short-term component), and the trend of long-term component derived with $KZ_{365,3}$ filter corresponded approximately to the interannual trend of the original data (Rao and Zurbenko, 1994; Eskridge et al., 1997). Based on the spectral decompositions of the daily observational data and three components, the power spectral of daily observational data in periods less than 33 days and longer than 632 days (1.7 years) have been well reproduced by short-term and long-term components, and seasonal component represents well the seasonal variations, i.e., periods between 33 days and 1.7 years (Seo et al., 2018). Thus we applied $\mathrm{KZ}_{15,5}$ and $\mathrm{KZ}_{365,3}$ filters to remove the variations with the periods shorter than 33 days and 1.7 years in this study.

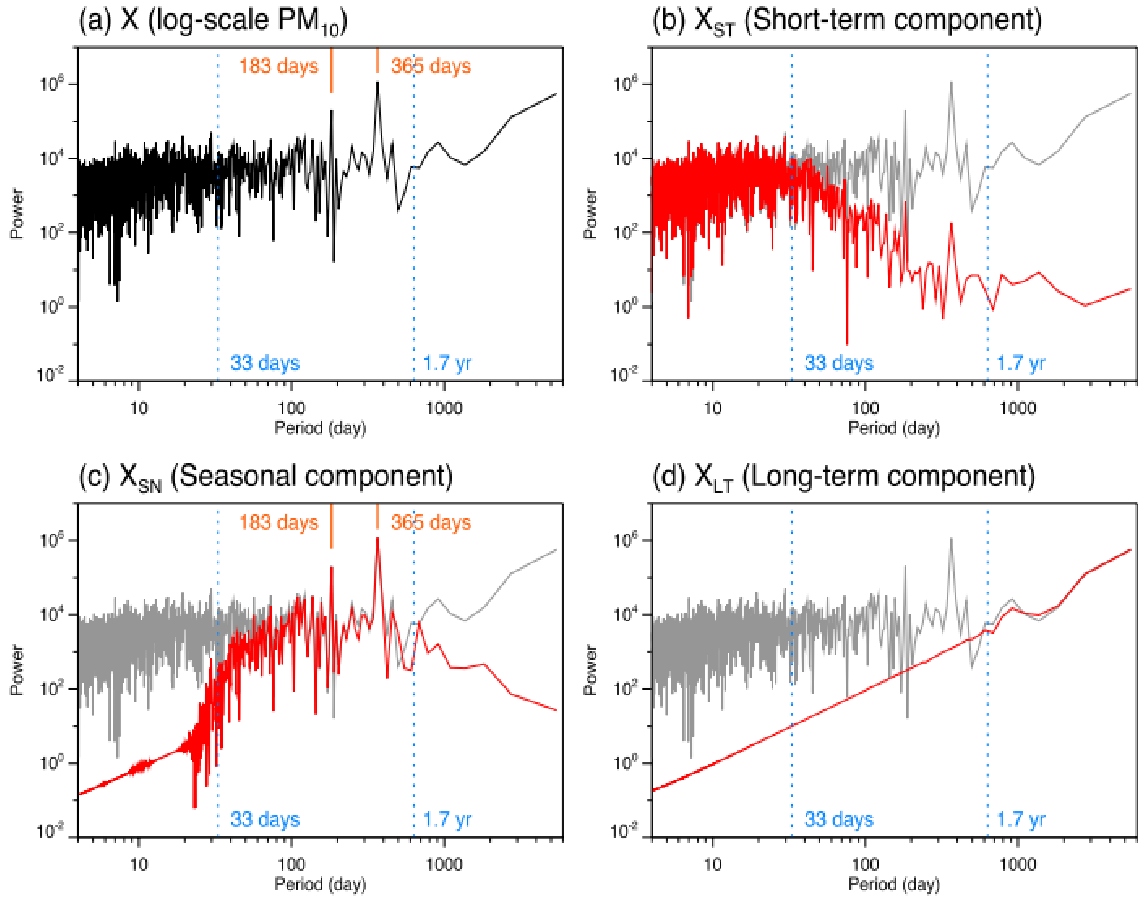

Figure R1 Power spectra of (a) log-transformed original time series X (black line) and (b) the short-term (less than 33 days), (c) seasonal (between 33 days and 632 days), and (d) long-term components (longer than 632 days) (red lines). Effective filter widths for $KZ_{15,5}$ filter (33 days) and $KZ_{365,3}$ filter (632 days) are marked with blue vertical dashed lines. The power spectrum of the original time series in (a) is represented with gray lines in (b-d) (Seo et al., 2018).

References:

Eskridge, R. E., Ku, J. Y., Rao, S. T., Porter, P. S., and Zurbenko, I. G.: Separating different scales of motion in time series of meteorological variables, Bulletin of the American Meteorological Society, 78, 1473-1484, 1997.

Rao, S. T., and Zurbenko, I. G.: Detecting and tracking changes in ozone air quality, Air & waste, 44, 1089-1092, 1994.

Seo, J., Park, D. S. R., Kim, J. Y., Youn, D., Lim, Y. B., and Kim, Y.: Effects of meteorology and emissions on urban air quality: a quantitative statistical approach to long-term records (1999–2016) in Seoul, South Korea, Atmospheric Chemistry and Physics, 18, 16121-16137, 2018.

[4. L148-154, it is not clear to me how the authors verified this approach. How are the results compared to analyses using other methods? Could synthetic data be generated to test this approach?]

Response 4.1: Many thanks for the referee's comments. Please find our response as follows and the subsequent parts in Responses 4.2 and 4.3 to the referee's comments and suggestions:

As presented in the response 3, the best moving average *m* and number of iterations *p* are chosen to separate the multi-scale components with the KZ filter in this study, as the correlation coefficients of 0.05, 0.01 and 0.04 among the decomposed short-term, seasonal and long-term components were near zero, indicating the orthogonal decomposition of multi-time scale components (Eskridge et al., 1997). Besides, the larger the total variance, the more independent the three components are of each other (Chen et al., 2019). The sum of the long-term, seasonal and short-term components contributed 91.4–94.4 % to the total variance with the regional averages of 92.7 % (Fig. 2), reflecting a satisfactory verification of the KZ filtering results. According to the decomposed long-term, seasonal and short-term components demonstrated in Fig. 3, the notable peaks of decomposed seasonal and short-term components were highly consistent with the peaks of $PM_{2.5}$ concentrations in the original observed data, which further proved a reasonable decomposition of the multi-scale components of $PM_{2.5}$ change over 2015–2019.

The verification of the decomposition using KZ filter have been added in Lines 164–167 and Lines 179-184 of the revised manuscript:

The larger the total variance, the more independent the three components are of each other (Chen et al., 2019). The sum of the long-term, seasonal and short-term components contributed 91.4–94.4 % to the total variance with the regional averages of 92.7 % (Fig. 2), reflecting a satisfactory verification of the KZ filtering results. (Lines 163–166)

… … … …

The correlation coefficients of 0.05, 0.01 and 0.04 among the decomposed short-term, seasonal and long-term components were near zero, indicating the orthogonal decomposition of multi-time scale components (Eskridge et al., 1997). According to the decomposed long-term, seasonal and short-term components demonstrated in Fig. 3, the notable peaks of decomposed seasonal and short-term components were highly consistent with the peaks of $PM_{2.5}$ concentrations in the original observed data, which further proved a reasonable decomposition of the multi-scale components of $PM_{2.5}$ change over 2015–2019. (Lines 178-183)

[4. L148-154: … … … … How are the results compared to analyses using other methods? … … … …]

Response 4.2: In response to this comment, we compared the decomposed long-term component using KZ filter with other studies and have revised the manuscript (Lines 196–198) as follows:

The change of long-term component of $PM_{2.5}$ exhibited a steadily declining trend over 2015–2019 (Fig. 3c), which was consistent with the interannual trend of observed regional $PM_{2.5}$ concentrations under the sustained impact of emission control (Zhang et al., 2019; Xu et al., 2020).

To further validate the accuracy of the meteorological contribution to $PM_{2.5}$ changes with KZ filter, we have conducted the simulation experiments with WRF-Chem, which is added in the new *Sect. 3.6*.

[4. L148-154: … … … … Could synthetic data be generated to test this approach?]

Response 4.3: There are various statistical methods using synthetic data to quantify the relative contribution of meteorology and emission on air pollution over China. Multiple linear regression model was constructed to quantify meteorological influences on the trends in $PM_{2.5}$ changes, with a novel focus on the contribution of the most influential meteorological factors to $PM_{2.5}$ trends for four seasons, contributing 2 %–29 % of the observed decreasing trend of $PM_{2.5}$ concentrations over China during recent years (Chen et al., 2020). The meteorology-driven anomalies contributed –3.9 % to 2.8 % of the annual mean $PM_{2.5}$ concentrations in China estimated from the generalized additive model driven by the satellite-based full-coverage daily $PM_{2.5}$ retrievals (Xiao et al., 2021). Based on the model-based environmental meteorology index, both meteorological variations and emission controls contributed to $PM_{2.5}$ decrease in the THB, with the meteorology contributing –45.5 % (Gong et al., 2021). These results emphasize the general accelerating effect of meteorology on $PM_{2.5}$ decline national wide and the offsetting effect for various regions. The comparison of the results using KZ filter with other studies using synthetic data have been added in Lines 356–359 of the revised manuscript:

Comparing with the statistical studies using synthetic data of meteorological influence on regional $PM_{2.5}$ changes in China with meteorological contribution from –45.5 % to 29.0 % over recent years (Chen et al., 2020; Xiao et al., 2021; Gong et al., 2021), the $PM_{2.5}$ pollution over the THB was affected contrarily by meteorological drivers with the northern positive and southern negative contribution from 2015 to 2019 (Fig. 9).

References:

Chen, L., Zhu, J., Liao, H., Yang, Y., and Yue, X.: Meteorological influences on PM2.5 and O3 trends and associated health burden since China's clean air actions, Sci Total Environ, 744, 140837, 10.1016/j.scitotenv.2020.140837, 2020.

Chen, Z. Y., Chen, D. L., Kwan, M. P., Chen, B., Gao, B. B., Zhuang, Y., Li, R. Y., and Xu, B.: The control of anthropogenic emissions contributed to 80 % of the decrease in PM 2.5 concentrations in Beijing from 2013 to 2017, Atmospheric Chemistry and Physics, 19, 13519-13533, 2019.

Eskridge, R. E., Ku, J. Y., Rao, S. T., Porter, P. S., and Zurbenko, I. G.: Separating different scales of motion in time series of meteorological variables, Bulletin of the American Meteorological Society, 78, 1473-1484, 1997.

Gong, S. L., Liu, H. L., Zhang, B. H., He, J. J., Zhang, H. D., Wang, Y. Q., Wang, S. X., Zhang, L., and Wang, P., Guo, H., Hu, J., Kota, S. H., Ying, Q., and Zhang, H.: Responses of PM2.5 and O3 concentrations to changes of meteorology and emissions in China, Sci Total Environ, 662, 297-306, 10.1016/j.scitotenv.2019.01.227, 2019.

Xiao, Q., Zheng, Y., Geng, G., Chen, C., Huang, X., Che, H., Zhang, X., He, K., and Zhang, Q.: Separating emission and meteorological contributions to long-term PM2.5 trends over eastern China during 2000–2018, Atmospheric Chemistry and Physics, 21, 9475-9496, 10.5194/acp-21-9475-2021, 2021.

Xu, Y., Xue, W., Lei, Y., Huang, Q., Zhao, Y., Cheng, S., Ren, Z., and Wang, J.: Spatiotemporal variation in the impact of meteorological conditions on $PM_{2.5}$ pollution in China from 2000 to 2017, Atmospheric Environment, 223, 117215, 10.1016/j.atmosenv.2019.117215, 2020.

Zhang, X. Y., Xu, X. D., Ding, Y. H., Liu, Y. J., Zhang, H. D., Wang, Y. Q., and Zhong, J. T.: The impact of meteorological changes from 2013 to 2017 on PM 2.5 mass reduction in key regions in China, Science China Earth Sciences, 62, 1885-1902, 2019.

[5. Section 3.1, how are the results compared to other studies?]

Response 5: In response to the referee’s comments, we clarified the verification of KZ filter in the revised manuscript from the following two aspects: (1) comparing the decomposited short-term, seasonal and long-term components; (2) the decomposition of emission- and meteorology-related long-term components.

(1) The comparison of decomposed multi-time scale components with other studies has been given in response 4.1 and have been added in Lines 164–167 and Lines 179-184 of the revised manuscript:

The larger the total variance, the more independent the three components are of each other (Chen et al., 2019). The sum of the long-term, seasonal and short-term components contributed 91.4–94.4 % to the total variance with the regional averages of 92.7 % (Fig. 2), reflecting a satisfactory verification of the KZ filtering results. (Lines 164–167)

… … … …

The correlation coefficients of 0.05, 0.01 and 0.04 among the decomposed short-term, seasonal and long-term components were near zero, indicating the orthogonal decomposition of multi-time scale components (Eskridge et al., 1997). According to the decomposed long-term, seasonal and short-term components demonstrated in Fig. 3, the notable peaks of decomposed seasonal and short-term components were highly consistent with the peaks of $PM_{2.5}$ concentrations in the original observed data, which further proved a reasonable decomposition of the multi-scale components of $PM_{2.5}$ change over 2015–2019. (Lines 179-184)

(2) The comparison of emission- and meteorology-related long-term components with other studies was clarified in the revised manuscript (Lines 196–198 and Lines 202–210) as follows:

The change of long-term component of $PM_{2.5}$ exhibited a steadily declining trend over 2015–2019 (Fig. 3c), which was consistent with the interannual trend of observed regional $PM_{2.5}$ concentrations under the sustained impact of emission control (Zhang et al., 2019; Xu et al., 2020). (Lines 196–198)

In previous studies, chemical transport models and statistical methods were both used to assess the changes in air pollution attributable to emissions and meteorology (Xiao et al., 2021). Significant declines in emission-related $PM_{2.5}$ concentrations occurred in central China (Wang et al., 2019; Chen et al., 2020), and the meteorology offset the impact of emission reduction in typical years of unfavorable meteorological conditions (Xu et al., 2020; Gong et al., 2021). The regional averaged emission- and meteorology-related long-term components as well as the long-term component over the THB are displayed in Fig. S1a, implying the steadily declining trend of $PM_{2.5}$ and the dominating impact of emission reduction on long-term $PM_{2.5}$ changes, which is consistent with the previous studies using multiple linear regression model for central China (Fig. S1b). The meteorology-related long-term component is positive value in certain periods, implying the significant modulation effect of meteorology on $PM_{2.5}$ decline in the THB. (Lines 202–210)

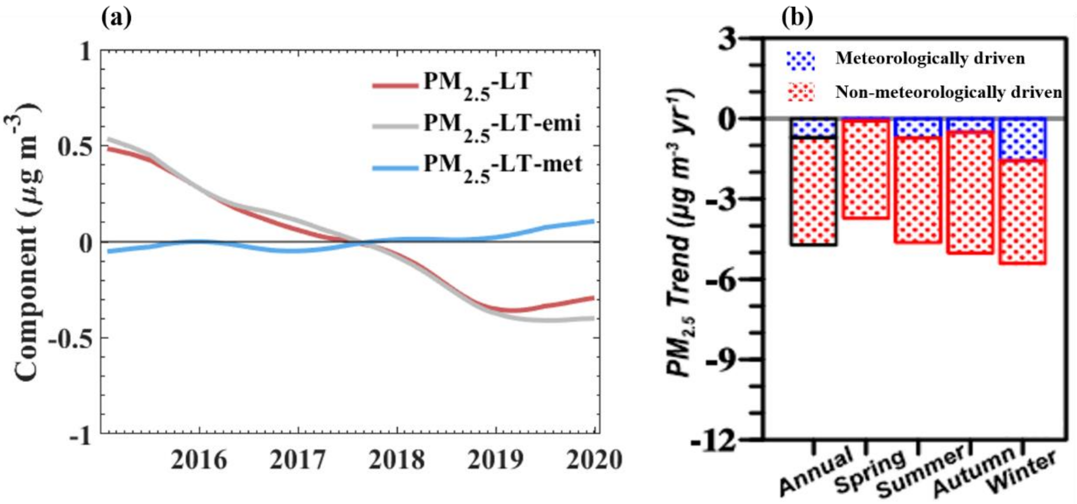

Figure S1 (a) The regional averaged long-term ($PM_{2.5}$-LT), emission-related long-term ($PM_{2.5}$-LT-emi) and meteorology-related long-term ($PM_{2.5}$-LT-met) components over the THB from 2015 to 2019. (b) Meteorologically driven, and non-meteorologically (emission) driven trends of annual and seasonal $PM_{2.5}$ concentrations during 2014–2018 for central China. Blue and red bars respectively represent meteorologically driven trends and non-meteorologically (emission) driven trends (reconstructed from Chen et al., 2020).

References:

Chen, L., Zhu, J., Liao, H., Yang, Y., and Yue, X.: Meteorological influences on PM2.5 and O3 trends and associated health burden since China's clean air actions, Sci Total Environ, 744, 140837, 10.1016/j.scitotenv.2020.140837, 2020.

Eskridge, R. E., Ku, J. Y., Rao, S. T., Porter, P. S., and Zurbenko, I. G.: Separating different scales of motion in time series of meteorological variables, Bulletin of the American Meteorological Society, 78, 1473-1484, 1997.

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Xiao, Q., Zheng, Y., Geng, G., Chen, C., Huang, X., Che, H., Zhang, X., He, K., and Zhang, Q.: Separating emission and meteorological contributions to long-term PM2.5 trends over eastern China during 2000–2018, Atmospheric Chemistry and Physics, 21, 9475-9496, 10.5194/acp-21-9475-2021, 2021.

Xu, Y., Xue, W., Lei, Y., Huang, Q., Zhao, Y., Cheng, S., Ren, Z., and Wang, J.: Spatiotemporal variation in the impact of meteorological conditions on PM2.5 pollution in China from 2000 to 2017, Atmospheric Environment, 223, 117215, 10.1016/j.atmosenv.2019.117215, 2020.

Zhang, X. Y., Xu, X. D., Ding, Y. H., Liu, Y. J., Zhang, H. D., Wang, Y. Q., and Zhong, J. T.: The impact of meteorological changes from 2013 to 2017 on PM 2.5 mass reduction in key regions in China, Science China Earth Sciences, 62, 1885-1902, 2019.

[6. L196, what are the relative contributions of emissions and meteorology to the long-term changes in $PM_{2.5}$ based on the analyses here?]

Response 6: We applied $KZ_{15,5}$ and $KZ_{365,3}$ filters to remove variabilities of periods shorter than 33 days and 1.7 years and decompose the daily environmental data into short-term, seasonal and long-term components. The long-term component can be further separated into emission-related and meteorology-related components by isolating the emission-related component using a multiple linear regression model with representative meteorological variables (Seo et al., 2018). The detailed methods about the separation of emission- and meteorology-related long-term components are displayed in Fig. R2 and *Sect. 2.3* of the revised manuscript.

The slope of the long-term component can reveal the long-term trend after short-term and seasonal variations are removed from the daily observational data. The difference between the slope of emission-related long-term and long-term components of $PM_{2.5}$ is caused by meteorological changes. The meteorological contribution to the $PM_{2.5}$ declining trend is quantitatively assessed with Eq. (10) in the revised manuscript (Lines 338–340) as follows:

$$\mathrm{Con_{met}} = \frac{\mathrm{k_{LT}} - \mathrm{k_{emiss}}}{\mathrm{k_{LT}}} \times 100\%. \quad (10)$$

$\mathrm{Con_{met}}$ (in %) is estimated with the linear trends $\mathrm{k_{LT}}$ of long-term component $\mathrm{PM_{2.5LT}(t)}$ and $\mathrm{k_{emiss}}$ of emission-related long-term component $\mathrm{PM_{2.5}}_{\mathrm{LT}}^{\mathrm{emiss}}(\mathrm{t})$.

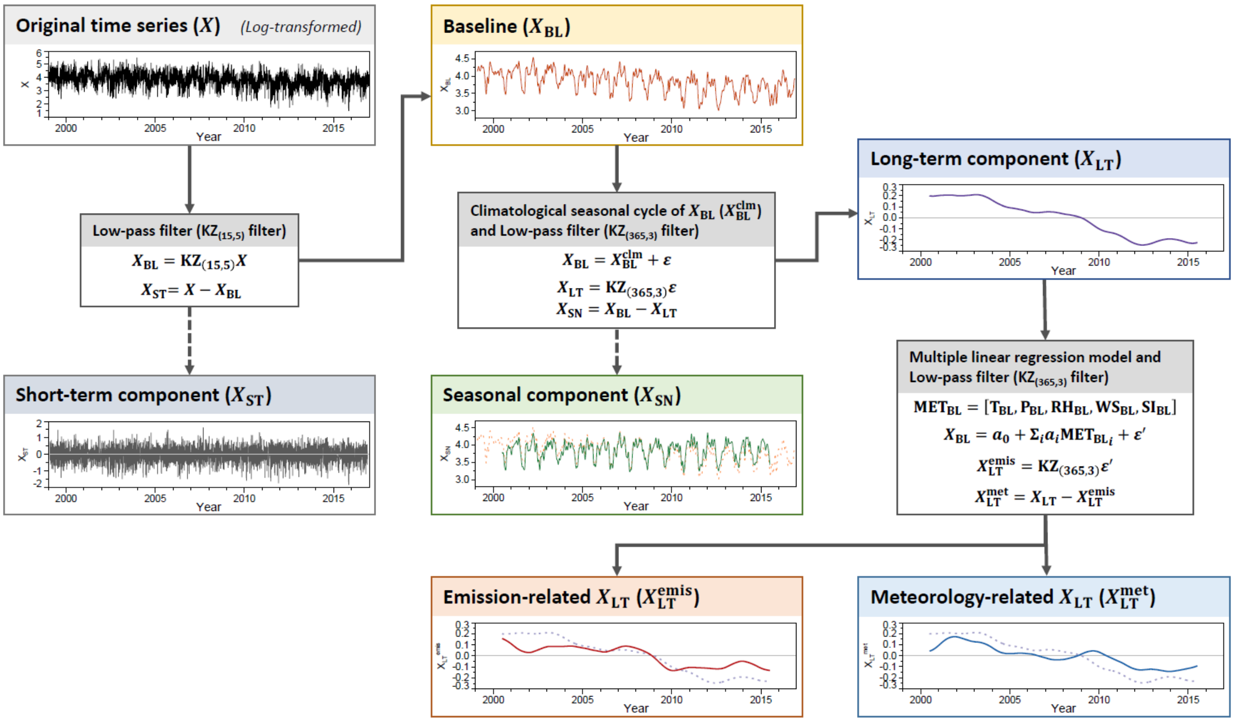

Figure R2 Schematic flowchart of time series decomposition of any environmental variable X into short-term, seasonal, and emission-related and meteorology-related long-term components (Seo et al., 2018).

References:

Seo, J., Park, D. S. R., Kim, J. Y., Youn, D., Lim, Y. B., and Kim, Y.: Effects of meteorology and emissions on urban air quality: a quantitative statistical approach to long-term records (1999–2016) in Seoul, South Korea, Atmospheric Chemistry and Physics, 18, 16121-16137, 2018.

[7. L217-218, this can be testified by checking the trend of SO_2 emissions in this region from the emission estimate. Do the emissions support your explanations here?]

Response 7: Following the reviewer's comment, we have testified by checking the trend of SO_2 emissions in this region from the emission estimate in the revised manuscript (Lines 259–263) as follows:

The interannual variations in emissions for China were calculated from MEIC (Zheng et al., 2018), as well as the annual total emissions of SO_2 and NO_x, PM in THB region reported by National Bureau of Statistic of China (http://www.stats.gov.cn/tjsj/ndsj/, last access: January 17, 2022), presenting the rapid decline of SO_2 emissions in the THB than changes of $PM_{2.5}$ and NO_x emissions (Fig. S2). The declining trend of anthropogenic emissions estimated from emission inventories can support the explanation of the changes in air pollutant concentrations.

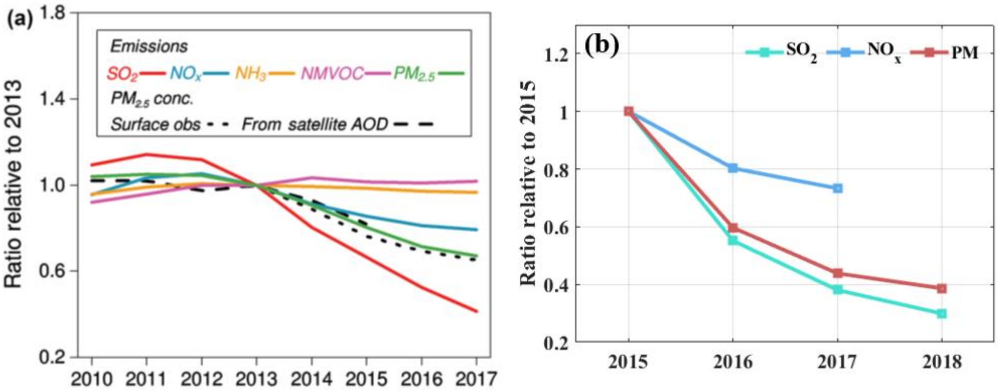

Figure S2 (a) Interannual variations in the ratios of MEIC emissions for 2010–2017 compared with satellite- and ground- based observations relative to those in 2013 (Zheng et al., 2018), (b) interannual variations in the ratios of annual total emission of SO_2, NO_x and PM relative to those in 2015 averaged over the THB reported by National Bureau of Statistic of China.

Reference:

Zheng, B., Tong, D., Li, M., Liu, F., Hong, C. P., Geng, G. N., Li, H. Y., Li, X., Peng, L. Q., and Qi, J.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmospheric Chemistry and Physics, 18, 14095-14111, 2018.

Responses to Referee #2

*[**1. General comments:** This study investigates the relative contribution from meteorological effect and emission changes to $PM_{2.5}$ variation over the Twain-Hu Basin (THB) based on the Kolmogorov–Zurbenko (KZ) filtering of long-term air quality measurement data. It is indicated that the reduction in anthropogenic emissions was the primary cause for the long-term decline in $PM_{2.5}$ concentrations and the meteorological changes moderated the $PM_{2.5}$ variations in the THB. However, in terms of novelty and broad interest, this work still needs to be improved. Besides, there could be great uncertainties associated with the multiple linear regression and KZ filtering method, but the authors have not validated the method and touched on the uncertainties in the conclusion. Here list some of my main concerns.]*

Response 1.1: Thanks for the referee's comments and suggestions. Please find our response as follows and the subsequent **Response 1.2** to the referee's comments and suggestions.

We have clarified the highlights and implications for novelty and broad interest in the revised *Abstract* and *Introduction* as follows:

The THB covering a large region of two provinces, Hubei and Hunan in central China, is surrounded by the high air pollutant emission regions in North China Plain (NCP) to the north, Yangtze River Delta (YRD) to the east, Pearl River Delta (PRD) to the south and Sichuan Basin (SB) to the west (Lin et al., 2018). Driven by East Asian monsoonal winds over Central Eastern China, THB is a major receptor region in regional transport of air pollutants over China (Shen et al., 2020). Governed by the multi-scale atmospheric circulations, air pollutants emitted from the upwind source regions can be transported easily to the downstream receptor region exacerbating the regional air quality, which can result in a complicated relation of source and receptor in regional transport of air pollutants (Hu et al., 2021). However, the previous studies mostly focused on the atmospheric environment change in the source regions with high anthropogenic emissions of air pollutants, and there have been few assessments on multi-scale changes of atmospheric environment over the receptor region in regional transport of air pollutants. In the present study of 5-year observations and modeling, we targeted the THB, a large region of heavy $PM_{2.5}$ pollutions over central China, to assess the meteorological effect on $PM_{2.5}$ changes over a receptor region in regional transport of air pollutants, and we assessed the contributions of air pollutant emissions and meteorological conditions to air quality change over this receptor region with the long-term observations over recent years. Our results highlight the effects of emission mitigation and meteorological changes on source-receptor relationship of region transport of air pollutants with the implication of long-range transport of air pollutants for regional and global environment changes. Therefore, the results in this paper have broader implications for regional - global air pollution issues.

*[**1. General comments:** … … … … Besides, there could be great uncertainties associated with the multiple linear regression and KZ filtering method, but the authors have not validated the method and touched on the uncertainties in the conclusion.]*

Response 1.2: The multiple linear regression is done stepwise, adding and deleting meteorological factors based on their independent statistical significance to obtain the best regression fit for air pollutants. For meteorological variables not in the final multiple linear regression model, the regression coefficients are zero. The selected meteorological variables differ by sites and all regression coefficients pass the confidence of 99%. The multiple linear regressions explained $PM_{2.5BL}$, SO_{2BL} and NO_{2BL} with adjusted determination coefficients (Adj. R^2) of 0.5695–0.8093, 0.0630–0.4592 and 0.6304–0.8669 passing the confidence level of 99 % in all the THB sites, confirming the reasonable construct of multiple linear regressions. The detailed justification and validation of selecting the meteorological parameters and discussions about validating the multiple linear regressions are clarified in *Sect. 3.2* of the revised manuscript.

To verify the results using KZ filter, we have added more discussions by clarifying the reasonable decomposition of multi-time scale components in Lines 164–167 and Lines 179–184 based on the previous studies as follows:

The larger the total variance, the more independent the three components are of each other (Chen et al., 2019). The sum of the long-term, seasonal and short-term components contributed 91.4–94.4 % to the total variance with the regional averages of 92.7 % (Fig. 2), reflecting a satisfactory verification of the KZ filtering results. (lines 164–167)

… … … …

The correlation coefficients of 0.05, 0.01 and 0.04 among the decomposed short-term, seasonal and long-term components were near zero, indicating the orthogonal decomposition of multi-time scale components (Eskridge et al., 1997). According to the decomposed long-term, seasonal and short-term components demonstrated in Fig. 3, the notable peaks of decomposed seasonal and short-term components were highly consistent with the peaks of $PM_{2.5}$ concentrations in the original observed data, which further proved a reasonable decomposition of the multi-scale components of $PM_{2.5}$ change over 2015–2019. (Lines 179–184)

To further validate the accuracy of our results with KZ filter, we have conducted the simulation experiments with Weather Research and Forecasting model with Chemistry (WRF-Chem), which is added in the new *Sect. 3.6* as follows:

3.6 Meteorological contribution to $PM_{2.5}$ changes validated with WRF-Chem modeling

The above observational study investigated the meteorological influence on the changes in $PM_{2.5}$ concentrations in the THB using KZ filter, with concluding the large impact of meteorology on the $PM_{2.5}$

changes over 2015–2019. To validate this conclusion of analyses with KZ filter, we designed three sets of modeling experiments CTRL, SENS-MET and SENS-EMI (Table S6) for December of 2015–2019, respectively driven with the changing meteorology and anthropogenic emissions over 2015–2019, the fixed meteorological conditions and anthropogenic emissions of 2015 with atmospheric chemical model WRF-Chem (Weather Research and Forecasting model with Chemistry). Air pollutant emission inventories, modeling configuration, experiment design and modeling verification were described in the supplement. The modeling verification of experiments CTRL indicated that $PM_{2.5}$ and meteorology were reasonably reproduced by the WRF-Chem simulation (Figs.S4–S5, Table S7), and the designed three sets of modeling experiments CTRL, SENS-MET and SENS-EMI could be used in the further analyses of emission and meteorological impact on $PM_{2.5}$ change over 2015–2019 to confirm the results of KZ filter.

We derived the effect of meteorology by comparing the simulated $PM_{2.5}$ concentrations in the three sets of experiments CTRL, SENS-MET and SENS-EMI (Table S6). The relative contribution of meteorology to the interannual changes of $PM_{2.5}$ concentrations was calculated with a linear additive relationship of contributions of meteorology and emission in the following equations:

$$Con_{MET} = \frac{k_{MET}}{k_{CTRL}} \tag{11}$$

$$Con_{EMI} = \frac{k_{EMI}}{k_{CTRL}} \tag{12}$$

$$RCon_{MET} = \frac{Con_{MET}}{Con_{MET} + Con_{EMI}} \times 100\% \tag{13}$$

k_{CTRL}, k_{MET} and k_{EMI} represent the trends in interannual changes of $PM_{2.5}$ concentrations simulated by the experiments CTRL, SENS-MET and SENS-EMI, respectively. Con_{MET} and Con_{EMI} are the contribution of meteorology and emission, and $RCon_{MET}$ is the contribution rate (%) of meteorology to interannual changes of $PM_{2.5}$ concentrations (Zhang et al., 2020).

Based on WRF-Chem modeling experiments, we assessed the impact of meteorological changes on interannual $PM_{2.5}$ variations from 2015 to 2019 with *Eqs. (11–13)*. The relative contribution of meteorology to interannual $PM_{2.5}$ variations displayed the regional pattern of northern positive and southern negative values over the THB (Fig. 10), confirming the impact of meteorological changes by accelerating and offsetting the effects of emission reductions on $PM_{2.5}$ declining trends in the northern and southern THB, respectively. The general spatial distribution of meteorological contribution rates to $PM_{2.5}$ declining trends from the WRF-Chem simulation was consistent with the results using KZ filter (Figs. 9 and 10), validating the results with KZ filter that meteorological drivers exerted a contrary impact of northern positive and southern negative contribution on long-term changes of $PM_{2.5}$ concentrations in the THB.

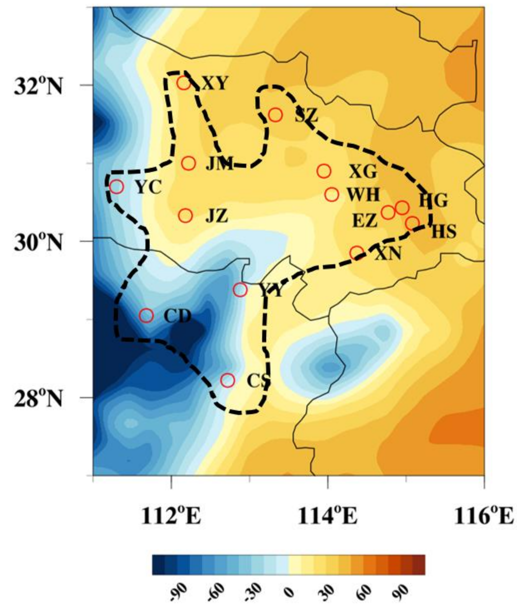

Figure 10 Spatial distribution of contribution rates of meteorological variations to $PM_{2.5}$ reductions based on WRF-Chem modeling experiments (contour, unit: %) in the THB outlined with black dashed line and surrounding regions for December of 2015–2019.

References:

Chen, Z. Y., Chen, D. L., Zhao, C. F., Kwan, M.-P., Cai, J., Zhuang, Y., Zhao, B., Wang, X. Y., Chen, B., and Yang, J.: Influence of meteorological conditions on $PM_{2.5}$ concentrations across China: A review of methodology and mechanism, Environment International, 139, 105558, 2020.

Eskridge, R. E., Ku, J. Y., Rao, S. T., Porter, P. S., and Zurbenko, I. G.: Separating different scales of motion in time series of meteorological variables, Bulletin of the American Meteorological Society, 78, 1473-1484, 1997.

Hu, W. Y., Zhao, T. L., Bai, Y. Q., Kong, S. F., Xiong, J., Sun, X. Y., Yang, Q. J., Gu, Y., and Lu, H. C.: Importance of regional PM2. 5 transport and precipitation washout in heavy air pollution in the Twain-Hu Basin over Central China: Observational analysis and WRF-Chem simulation, Science of the Total Environment, 758, 143710, 2021.

Lin, C. Q., Liu, G. H., Lau, A. K. H., Li, Y., Li, C. C., Fung, J. C. H., and Lao, X. Q.: High-resolution satellite remote sensing of provincial PM2. 5 trends in China from 2001 to 2015, Atmospheric Environment, 180, 110-116, 2018.

Shen, L. J., Wang, H. L., Zhao, T. L., Liu, J., Bai, Y. Q., Kong, S. F., and Shu, Z. Z.: Characterizing regional aerosol pollution in central China based on 19 years of MODIS data: Spatiotemporal variation and aerosol type discrimination, Environmental Pollution, 263, 114556, 10.1016/j.envpol.2020.114556, 2020.

Zhang, W. J., Wang, H., Zhang, X. Y., Peng, Y., Zhong, J. T., Wang, Y. Q., and Zhao, Y. F.: Evaluating the contributions of changed meteorological conditions and emission to substantial reductions of PM2.5 concentration from winter 2016 to 2017 in Central and Eastern China, Science of The Total Environment, 716, 136892, 2020.

[2. There are many parameters used in KZ filtering and multiple linear regression. The justification and validation of the selection of them should be provided. I think the changes in data coverage or the parameter selection would largely influence the final quantitative estimation of contributions, which is suggested to be elaborated.]

Response 2: Following the reviewer's suggestion, we have justified and validated the selection of meteorological parameters in *Sect. 3.2* (Lines 215–221 and Lines 234–241) as follows:

Based on our understanding of chemical and physical processes of diffusive transport, chemical transformation, emissions and depositions of $PM_{2.5}$ in the atmosphere, the dominant meteorological factors for changing $PM_{2.5}$ concentrations over china are wind speed, relative humidity, air temperature, atmospheric pressure and precipitation (Chen et al., 2020). We examined the significant correlations between baseline components of air pollutant concentrations and selected a set of meteorological factors, including air temperature, wind speed, precipitation, relative humidity, and air pressure (Tables S1-S3 in the *Supplement*). The meteorological parameters selected in this study are consistent with the previous studies (Chen et al., 2020). (Lines 215–221)

… … … …

The multiple linear regression is done stepwise, by adding and deleting meteorological factors based on their independent statistical significance to obtain the best regression fit for air pollutants (Draper, 1998). The multiple linear regressions explained $PM_{2.5BL}$, SO_{2BL} and NO_{2BL} with adjusted determination coefficients (Adj. R^2) of 0.5695–0.8093, 0.0630–0.4592 and 0.6304–0.8669 passing the confidence level of 99 % in all the THB sites, confirming the reasonable construct of multiple linear regressions. (Lines 234–241)

Following the reviewer's comments, we have elaborated that the changes in data coverage or the parameter selection would largely influence the final quantitative estimation of contributions of meteorology and emissions for the limitation and outlook of our study in the revised *Conclusions* (Lines 429–435) as follows:

The changes in data coverage and the meteorological parameter selection would largely influence the final quantitative estimation of contributions of meteorology and emissions. Due to the limitation of the data coverage of observational data, further work could be desired with climate analyses of long-term data of fine meteorological and environmental observations and more comprehensively modeling of chemical and physical processes in the atmosphere to generalize the assessment on the effects of emission mitigation and meteorological changes on source-receptor relationship of region transport of air pollutants.

References:

Chen, Z. Y., Chen, D. L., Zhao, C. F., Kwan, M.-P., Cai, J., Zhuang, Y., Zhao, B., Wang, X. Y., Chen, B., and Yang, J.: Influence of meteorological conditions on $PM_{2.5}$ concentrations across China: A review of methodology and mechanism, Environment International, 139, 105558, 2020.

Draper, N. R.: Applied regression analysis, Technometrics, 9, 182-183, 1998.

[3. Another issue is the estimation of the effects of NO_2 and SO_2 emission reductions on $PM_{2.5}$ change trends based on long-term (k_{LT}) and emission-related long-term (k_{emiss}) components of $PM_{2.5}$, SO_2 and NO_2. The long-term changes in $PM_{2.5}$ are also caused by the emission variation of primary components like black and organic carbon, in addition to the chemical transformation of gaseous precursors. The difference in the emission of different primary pollutants may also lead to modifications in Klt/Kemis of $PM_{2.5}$. How was this impact/bias included and quantified in the present work?]

Response 3: We agree with the referee's comment. In the revised manuscript (lines 326–332), we have added the according discussions as follows:

The long-term changes in $PM_{2.5}$ are also caused by the emission variations of primary components like black and organic carbon, in addition to the chemical transformation of gaseous precursors. The difference in the emission of different primary pollutants may also lead to modifications in ${k_{\mathrm{LT}}}/{k_{emiss}}$ of $PM_{2.5}$. However, due to the current lack of long-term observation of $PM_{2.5}$ components in the THB, the influence of emissions variations of primary components on long-term changes in $PM_{2.5}$ concentrations is not assessed in our study. Further work with long-term observational data of $PM_{2.5}$ components like black and organic carbon could be conducted to quantify the influence of emissions of primary components and chemical transformation of gaseous precursors on $PM_{2.5}$ changes.

[4. Figure 9: Why did the contribution rates of meteorological variations show great spatial disparities at a small scale, i.e., EZ, HG and HS. It seems not very likely that the variation in synoptic weather or meteorological conditions has such a large heterogeneity at such a small spatial scale.]

Response 4: Thanks for the reviewer's careful review. In the revised manuscript, we have added the according discussions in *Sect. 3.5* (Lines 352–360) as follows:

It seems not very likely that the variation in synoptic weather or meteorological conditions has such a large heterogeneity at such a small spatial scale over EZ, HG and HS. However, the underlying surface conditions dominate the near-surface meteorological conditions in the atmospheric boundary layer at a small scale (Wang et al., 2017). The topography and land use of HG, HS, EZ and surrounding regions vary distinctly with underlying surface conditions of plain, lakes and hilly area (Fig. R1). The underlying surface of observational sites with different near-surface meteorology effectively influence the local accumulation, chemical transformation, dry and wet depositions of air pollutants (Bai et al., 2022). Therefore, the heterogeneity of meteorological contribution to $PM_{2.5}$ at such a small spatial scale might be attributed to the local meteorological conditions in the atmospheric boundary layer, which is largely affected by the underlying surface changes.

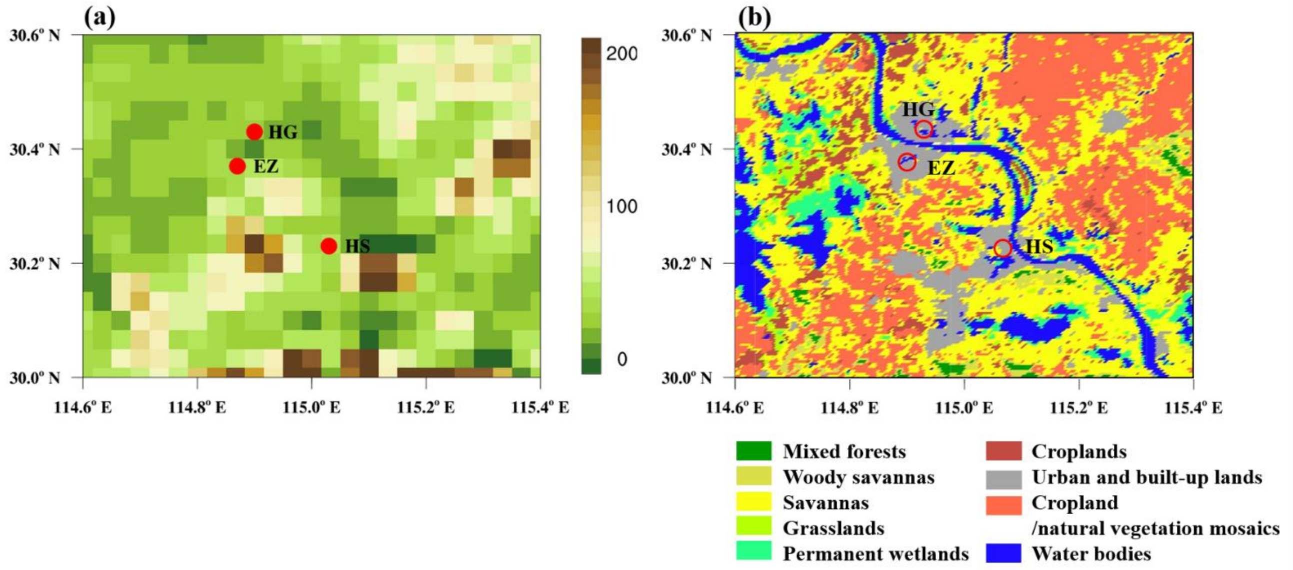

Figure R1 Distribution of (a) topographical height (color contours, m, in a. s. l.) and (b) land use over HG, EZ, HS and the surrounding regions in the THB (https://lpdaac.usgs.gov/products/mcd12q1v006/, last access: January 17, 2022).

References:

Bai, Y., Zhao, T., Hu, W., Zhou, Y., Xiong, J., Wang, Y., Liu, L., Shen, L., Kong, S., Meng, K., and Zheng, H.: Meteorological mechanism of regional PM2.5 transport building a receptor region for heavy air pollution over Central China, Science of The Total Environment, 808, 151951, 10.1016/j.scitotenv.2021.151951, 2022.

Wang, Y., Di Sabatino, S., Martilli, A., Li, Y., Wong, M. S., Gutiérrez, E., and Chan, P. W.: Impact of land surface heterogeneity on urban heat island circulation and sea-land breeze circulation in Hong Kong, Journal of Geophysical Research: Atmospheres, 122, 4332-4352, 10.1002/2017jd026702, 2017.