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 #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:

$$Con_{MET} = \frac{k_{MET}}{k_{CTRL}} \quad (11)$$

$$Con_{EMI} = \frac{k_{EMI}}{k_{CTRL}} \quad (12)$$

$$RCon_{MET} = \frac{Con_{MET}}{Con_{MET} + Con_{EMI}} \times 100\% \quad (13)$$

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.

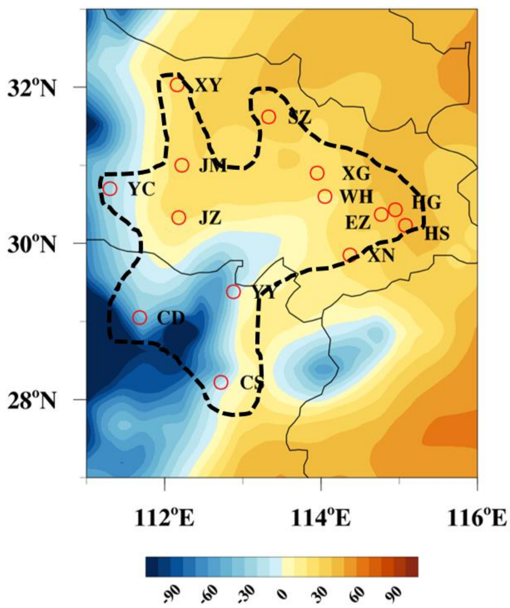

Figure 10 Spatial distribution of contribution rates of meteorological variations to $PM_{2.5}$ reductions based

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.

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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.

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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.

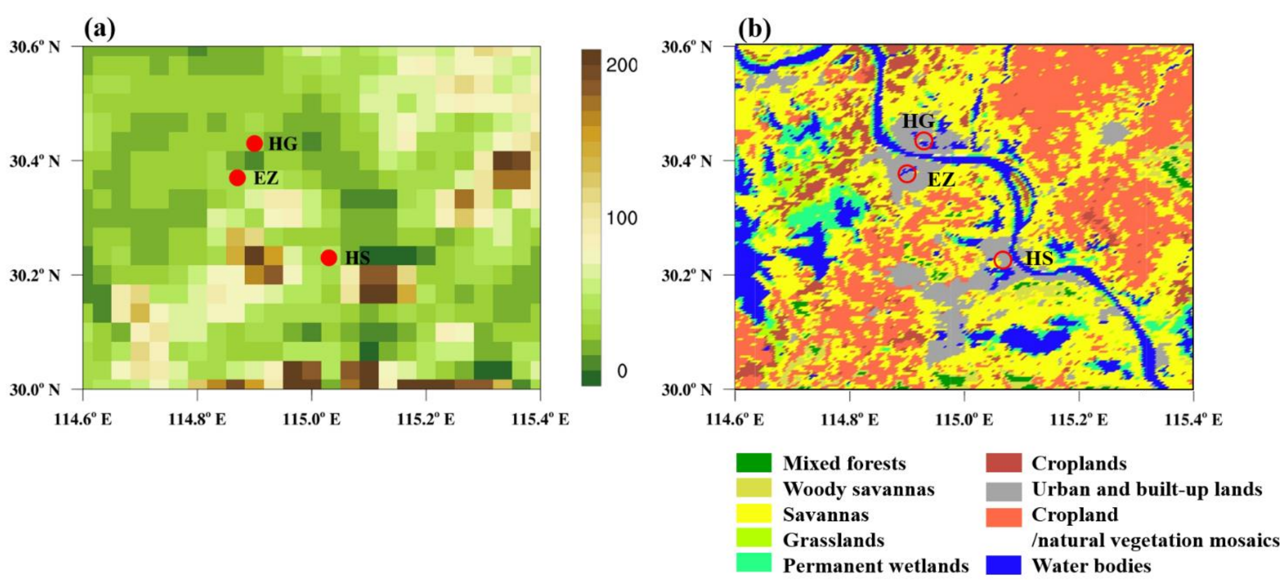

Figure R1 Distribution of (a) topographical height (color contours, m, in a. s. l.) and (b) land use over HG, EZ, HS and the surrounding regions in the THB (https://lpdaac.usgs.gov/products/mcd12q1v006/, last access: January 17, 2022).

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.