Meteorology impact on PM$_{2.5}$ change over a receptor region in the regional transport of air pollutants: observational study of recent emission reductions in central China

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Received: 22 August 2021 – Discussion started: 30 September 2021
Revised: 11 February 2022 – Accepted: 14 February 2022 – Published: 17 March 2022

Abstract. As an important issue in atmospheric environment, the contributions of anthropogenic emissions and meteorological conditions to air pollution have been little assessed over receptor regions 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$_{2.5}$ pollution in central China, to assess the effects of meteorology on PM$_{2.5}$ change over a receptor region in the regional transport of air pollutants. Based on observations of environment and meteorology over 2015–2019, the Kolmogorov–Zurbenko (KZ) filter was performed to decompose the PM$_{2.5}$ variations into multiple timescale components over the THB, where the short-term, seasonal and long-term components accounted for, respectively, 47.5 %, 41.4 % and 3.7 % of daily PM$_{2.5}$ changes. The short-term and seasonal components dominated the day-to-day PM$_{2.5}$ variations with long-term component determining the change trend of PM$_{2.5}$ concentrations over recent years. As the emission- and meteorology-related long-term PM$_{2.5}$ components over the THB were identified, the meteorological contribution to the declining PM$_{2.5}$ trend presented a distinct spatial pattern over the THB with northern positive rates up to 61.92 % and southern negative rates down to $-24.93$ %. The opposite effects of meteorology on PM$_{2.5}$ pollution could accelerate and offset the effects of emission reductions in the northern and southern THB, which is attributed to the upwind diffusion and downward accumulation of air pollutants over receptor regions in regional PM$_{2.5}$ transport. It is noteworthy that the increasing conversion efficiencies of SO$_2$ and NO$_2$ to sulfate and nitrate for secondary PM$_{2.5}$ could have offset the effect of PM$_{2.5}$ emission reduction on air pollution in the THB during recent years, revealing the enhancing contribution of gaseous precursor emissions to PM$_{2.5}$ concentrations under control of anthropogenic emissions of PM$_{2.5}$ and the gaseous precursors over receptor regions in the regional transport of air pollutants. Our results highlight the effects of emission mitigation and meteorological changes on the source–receptor relationship of the regional transport of air pollutants with the implication of long-range transport of air pollutants for regional and global environment changes.
1 Introduction

Haze pollution with high levels of PM$_{2.5}$ (fine particulate matters with aerodynamic diameters equal to or less than 2.5 $\mu$m) has been a serious problem in the atmospheric environment (Peng et al., 2016; Wang et al., 2016) with adverse influences on air quality and human health (Cao et al., 2012; Crouse et al., 2012). In recent years, large areas in central and eastern China (CEC) have experienced haze pollution with unprecedentedly high PM$_{2.5}$ levels in the regions covering the North China Plain (NCP), Yangtze River Delta (YRD), Pearl River Delta (PRD) and Sichuan Basin (SB) (Zhang et al., 2012; C. Q. Lin et al., 2012; Guo et al., 2017). In order to improve air quality by reducing air pollutant emissions, the Chinese government has implemented an action plan for controlling anthropogenic emissions since September 2013 (http://www.gov.cn/xinwen/2018-02/01/content_5262720.htm, last access: 17 January 2022). Surface PM$_{2.5}$ concentrations have exhibited 30%–40% decreases in CEC over recent years (Xue et al., 2019; Zhang et al., 2019). However, the changes of air pollution are generally co-determined by air pollutant emissions and meteorological conditions. The contributions of changes in meteorology and anthropogenic emissions to the improvement of air quality need to be comprehensively investigated.

PM$_{2.5}$ includes primary particles emitted directly from various sources and secondary particles generated by homogeneous and heterogeneous chemical reactions of gaseous precursor in the atmosphere, depending on the emissions of the primary PM$_{2.5}$ and its gaseous precursors (Li et al., 2011). Based on the statistical analysis of the long-term observational data, it was quantified that the emission controls could explain more of the variances in PM$_{2.5}$ than meteorology (Gui et al., 2019), and 12% of the observed PM$_{2.5}$ decrease was attributed to meteorological drivers in China since 2013 (Zhai et al., 2019). However, modeling and observational studies have mostly assessed the contribution of emissions and meteorology to regional PM$_{2.5}$ variations in the emission source regions with high anthropogenic emissions of air pollutants, and there have been few assessments of multi-scale changes of atmospheric environment over receptor regions in the regional transport of air pollutants.

The Twain-Hu Basin (THB), featuring the lowlands (mainly less than 200 m a.s.l.) of two central Chinese provinces, Hubei and Hunan (Fig. 1), is surrounded by the high air pollution regions of NCP, YRD, PRD and SB. As such, it is a key receptor region in the regional transport of air pollutants from the upstream region driven by East Asian monsoonal winds over CEC (Shen et al., 2020). Heavy air pollution in the THB with a unique “non-stagnation” atmospheric boundary layer is aggravated by the regional PM$_{2.5}$ transport over CEC (Zhong et al., 2019; Yu et al., 2020). In combination with the heavy pollution region of NCP through distinct transport channels, the regional transport from northern China to central China contributed 70.5% PM$_{2.5}$ concentrations to a wintertime heavy pollution episode in the THB (Hu et al., 2021). Thus, the contributions of air pollutant emissions and meteorological conditions to air quality change over this region in central China need to be specifically assessed with the long-term observations over recent years.

In this study, we investigated the multi-scale changes of PM$_{2.5}$ concentrations over the THB, a key receptor region of regional PM$_{2.5}$ transport in China, from 2015 to 2019 by establishing a statistic model with a Kolmogorov–Zurbenko (KZ) filter. We then evaluated the contributions of anthropogenic emissions and meteorological changes to the declining trends in PM$_{2.5}$ concentrations over this receptor region in regional PM$_{2.5}$ transport over CEC during the past 5 years of emission controls. The analysis of THB’s multi-scale air quality changes can improve the understanding of the effects of emission mitigation and meteorological changes on environmental change with regional transport of air pollutants.
2 Data and methods

2.1 Data

In order to analyze air quality changes in the THB, the observational data of hourly NO$_2$, SO$_2$ and PM$_{2.5}$ concentrations from 2015 to 2019 were collected from the national air quality monitoring network (http://www.mee.gov.cn/, last access: 17 January 2022). The air quality observation data are under quality control, based on China’s national standard of air quality observation.

The data of meteorological observations in the THB were sourced from the weather monitoring network of China Meteorological Administration (http://data.cma.cn/, last access: 17 January 2022), including air temperature ($T$), relative humidity ($RH$), sea level pressure ($SLP$), wind speed ($WS$) and precipitation ($Pre$), with temporal resolutions of 3 h.

2.2 KZ filter

To better understand the multiple timescale variations of PM$_{2.5}$ and the relations to air pollutant emissions and meteorological drivers, a KZ filter (Rao and Zurbenko, 1994; Seo et al., 2018) is used to separate the daily data into multiscale components, based on an iterative moving average that removes high-frequency variations in the data with applications in the study of air pollutants, especially O$_3$ and PM$_{2.5}$ variations (Chen et al., 2019; Ma et al., 2016; Seo et al., 2014; Zheng et al., 2020).

The KZ filter $KZ_{m,p}$ with a length of moving average window $m$ and number of iterations $p$ can remove the high-frequency component of periods smaller than the effective filter width $N \geq m \times p^{1/2}$. The KZ filter is applicable to the time series with missing data owing to the iterative moving average process, which provides high accuracy when compared with the wavelet transform method (Eskridge et al., 1997). By comparing different sets of moving average $m$ and number of iterations $p$, it was found that the decomposed time series using the KZ$_{15,5}$ filter exhibited no white noise (short-term component), and the trend of the long-term component derived with the 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 spectra of daily observational data in periods of less than 33 d and longer than 632 d (1.7 years) have been well reproduced by short-term and long-term components, and the seasonal component well represents the seasonal variations, i.e., periods between 33 d and 1.7 years (Seo et al., 2018). Thus we applied KZ$_{15,5}$ and KZ$_{365,3}$ filters to remove the variations with periods shorter than 33 d and 1.7 years in this study.

A meteorological or environmental variable $X(t)$ observed in time series $t$ can be decomposed into the short-term component $X_{ST}(t)$ and the baseline component $X_{BL}(t)$ presenting as

$$X(t) = X_{ST}(t) + X_{BL}(t).$$

The baseline component $X_{BL}(t)$ is obtained by applying the KZ$_{15,5}$ filter to $X(t)$, removing the short-term component $X_{ST}(t)$ with the temporal period shorter than 33 d from the observed data, expressed as

$$X_{BL}(t) = KZ_{15,5}[X(t)].$$

The baseline component $X_{BL}(t)$ also can be separated into the daily climatic averages $X_{BL}^{clm}$ over the study period, occupying most of the seasonality in $X_{BL}(t)$ and the residual $\varepsilon(t)$:

$$\varepsilon(t) = X_{BL}(t) - X_{BL}^{clm}.\tag{3}$$

To obtain the long-term component $X_{LT}(t)$ by removing the variations with the temporal period shorter than 1.7 years, the KZ$_{365,3}$ filter is applied to $\varepsilon(t)$ expressed as follows:

$$X_{LT}(t) = KZ_{365,3}[\varepsilon(t)],\tag{4}$$

with the short-term component

$$X_{ST}(t) = X(t) - X_{BL}(t)\tag{5}$$

and the seasonal component

$$X_{SN}(t) = X_{BL}(t) - X_{LT}(t).\tag{6}$$

The KZ filter was used to separate the daily surface PM$_{2.5}$, NO$_2$ and SO$_2$ concentrations into short-term, seasonal and long-term components in this study. The short-term component presents a synoptic-scale variation of meteorological influences, which could control local accumulation and
2.3 Multiple linear regression of air pollutant changes with meteorological variables

By altering the local accumulation, regional transport, chemical conversion and wet and dry depositions of air pollutants, the meteorological factors such as wind, RH, T, air pressure and Pre could exert significant impacts on PM$_{2.5}$ changes (Sun et al., 2013; Li et al., 2018; Z. Y. Chen et al., 2020). Therefore, with the multiple factors of the baseline components of 10 m WS, 2 m RH, 2 m $T$, SLP and Pre calculated by Eq. (2), a multiple linear regression equation was stepwise established for the baseline component of PM$_{2.5}$ as follows:

$$PM_{2.5BL\text{MLR}}(t) = a_0 + \sum_i a_i MET_{BL_i}(t),$$

where $MET_{BL_i}(t)$ (i.e. $[1, 5]$) is the baseline component of the meteorological variable $i$ with $i = 1, 2, 3, 4, 5$, respectively, for WS$_{BL}(t)$, RH$_{BL}(t)$, $T_{BL}(t)$, SLP$_{BL}(t)$, and Pre$_{BL}(t)$. We fit the regression coefficient $a_i$ for each meteorological variable and the intercept $a_0$. The residual $\varepsilon_{PM_{2.5}}$ between PM$_{2.5BL}$ and PM$_{2.5BL\text{MLR}}$ regressed with the multiple linear Eq. (7) is given as

$$\varepsilon_{PM_{2.5}}(t) = PM_{2.5BL}(t) - PM_{2.5BL\text{MLR}}(t).$$

$\varepsilon_{PM_{2.5}}$ contains not only the variability of PM$_{2.5}$ related to long-term changes in air pollutant emissions but also the minor seasonal change of PM$_{2.5}$ attributable to unconsidered meteorological influences in the multiple linear regression. By removing the minor seasonal change from $\varepsilon_{PM_{2.5}}$ with the KZ$_{365.3}$ filter, the emissions-related long-term component PM$_{2.5\text{emiss}}(t)$ can be isolated as follows:

$$PM_{2.5\text{emiss}}(t) = KZ_{365.3}[\varepsilon_{PM_{2.5}}(t)].$$

Here the long-term component of surface PM$_{2.5}$ concentrations can be further separated into the emission- and meteorology-related long-term components with Eqs. (9) and (4) (Seo et al., 2018). Similarly, the multiple timescale variations in SO$_2$ and NO$_2$ with long-term variations related to changes in air pollutant emissions and meteorological drivers are decomposed by the KZ filter with multiple linear regression. Seo et al. (2018) described the details of this method.
nents were, respectively, with 47.5%, 41.4% and 3.7% of the daily PM$_{2.5}$ changes over the THB (Fig. 2), and thus it could be reasonably verified that the daily variation in atmospheric pollutant was generally dominated by the short-term and seasonal components, with the long-term component determining the change trend (Ma et al., 2016; Yin et al., 2019a).

The short-term, seasonal and long-term PM$_{2.5}$ components were averaged for 14 sites of the THB to characterize the temporal variations of three components in the THB for 2015–2019 (Fig. 3). 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 multiple timescale components (Esford 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 the reasonable decomposition of the multi-scale components of PM$_{2.5}$ change over 2015–2019.

The observed daily PM$_{2.5}$ exhibited a distinct daily variation, with an overlapping of high-frequency variations, which could be caused by mesoscale and synoptic-scale meteorological processes (Ma et al., 2016). The short-term component of PM$_{2.5}$ fluctuated frequently with a significantly positive correlation to the daily change of PM$_{2.5}$ ($r = 0.68$, $p<0.05$), indicating the important role of the short-term component with the temporal period $<33$ d in the day-to-day variations of PM$_{2.5}$ concentrations in the THB (Fig. 3a).

The notable peaks of PM$_{2.5}$ seasonal component that emerged in winters were highly consistent with the peaks of observed daily PM$_{2.5}$ concentrations (Fig. 3b). A close linkage with the significant correlation coefficient of 0.75 ($p<0.05$) was found between the changes of PM$_{2.5}$ seasonal component and daily PM$_{2.5}$ concentrations, which could reflect a significant modulation of the PM$_{2.5}$ seasonal oscillations in the day-to-day variations of PM$_{2.5}$, driven by the seasonal shift of East Asian summer and winter monsoons, as well as the seasonal change of anthropogenic emissions (Zhu et al., 2012; Jeong and Park, 2017). The change of the 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 PM$_{2.5}$ concentrations under the sustained impact of emission controls (Zhang et al., 2019; Xu et al., 2020). The correlation coefficient ($r = 0.24$, $p<0.05$) of the long-term PM$_{2.5}$ component with the observed daily PM$_{2.5}$ change was much smaller than those of the short-term and seasonal PM$_{2.5}$ components, implying less influence of emission reduction on the daily PM$_{2.5}$ change and air pollution frequency, although the declining trend in PM$_{2.5}$ was determined by anthropogenic emission reduction.

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 emissions-related PM$_{2.5}$ concentrations occurred in central China (Wang et al., 2019; L. 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 and the long-term component over the THB are displayed in Fig. S1a in the Supplement, and suggest the steadily declining trend of PM$_{2.5}$ and the dominant impact of emission reduction on long-term PM$_{2.5}$ changes, which is consistent with previous studies using multiple linear regression models for central China (Fig. S1b). The meteorology-related long-term component has a positive value in certain periods, implying the significant modulating effect of meteorology on PM$_{2.5}$ decline in the THB.

### 3.2 Multiple linear regressions of PM$_{2.5}$, SO$_2$ and NO$_2$ with meteorological drivers

Since the short-term variations in meteorological variables were excluded, the correlations between baseline components of PM$_{2.5}$ and meteorological variables were only related to their seasonal and long-term components, affected by the regional climate of East Asian monsoons rather than synoptic-scale meteorological processes. 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 (Z. Y. 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 previous studies (Z. Y. Chen et al., 2020).

Generally, the baseline components of air pollutants were negatively correlated with the baseline components of wind speed (WS$_{BL}$) and positively correlated with the baseline components of sea level pressure (SLP$_{BL}$; Tables S1–S3), which could be attributed to the ventilation effect of wind and the stagnant conditions of meteorology in high-pressure systems, restraining the horizontal and vertical dispersions of air pollutants (Hsu and Cheng, 2016; Wang et al., 2016; Miao et al., 2017). Although wind speed exerts a negative influence of on PM$_{2.5}$ concentrations over the emission source region, increasing wind speed might cause the accumulation of PM$_{2.5}$ concentrations over the downwind region of emission sources (Z. Y. Chen et al., 2020), which led to the inconsistent influence of WS$_{BL}$ in the region of central China (Tables S1–S3). Under high surface air temperature conditions, there are strong thermal activities such as turbulence, mak-

https://doi.org/10.5194/acp-22-3579-2022  Atmos. Chem. Phys., 22, 3579–3593, 2022
The relations of regional averages of (a) short-term (PM$_{2.5}$-ST), (b) seasonal (PM$_{2.5}$-SN) and (c) long-term (PM$_{2.5}$-LT) components with the observed daily PM$_{2.5}$ concentrations (PM$_{2.5}$) over the THB from 2015 to 2019.

To isolate emissions-related long-term components from long-term components of PM$_{2.5}$, NO$_2$ and SO$_2$, the stepwise multiple linear regressions of PM$_{2.5BL}$, SO$_2BL$ and NO$_2BL$, respectively, with the baseline components of the meteorological parameters ($T_{BL}$, $WS_{BL}$, RH$_{BL}$, SLP$_{BL}$ and Pre$_{BL}$) were conducted with Eq. (7) for 14 sites, by adding and deleting meteorological variables based on their independent statistical significance to obtain the best model fit (Draper, 1998). We evaluated PM$_{2.5BL}$, SO$_2BL$ and NO$_2BL$ fitted by multiple linear regression models with KZ decomposition (Table 1). The multiple linear regressions explained PM$_{2.5BL}$, SO$_2BL$ and NO$_2BL$ with the 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 construction of the multiple linear regressions. The Adj. $R^2$ of multiple linear regression for SO$_2BL$ were lower than those of PM$_{2.5BL}$ and NO$_2BL$, which might be attributed to the larger impact of SO$_2$ emission controls on the seasonal and long-term SO$_2$ variations. In general, the variations of meteorological drivers can well reproduce the meteorology-related seasonal and long-term variations of PM$_{2.5}$, SO$_2$ and NO$_2$ in the THB (Table 1).

### 3.3 Interannual variations in air pollutants observed over the THB

PM$_{2.5}$ consists of chemical components generated in the complex physical and chemical processes (S. Li et al., 2015). Primary particles are emitted directly from anthropogenic (e.g., industry, power plants and vehicles) and natural (e.g., outdoor biomass burning and dust storms) sources. Secondary particles (e.g., sulfate and nitrate) are converted in the chemical reactions of the precursor gases (e.g., SO$_2$ and NO$_x$), which are mainly produced by human activities (S. Li et al., 2015; H. Yang et al., 2016). Therefore, in addition to the reductions in primary particulate emissions, control of the secondary aerosol precursor emissions is of great importance in mitigating air pollution.

The interannual variations of the ratios in annual mean PM$_{2.5}$, SO$_2$ and NO$_2$ concentrations relative to the annual averages in 2015 over the THB are displayed in Fig. 4.
The declines of PM$_{2.5}$ and SO$_2$ in 2019 averaged over the THB were $-26\%$ and $-68\%$ relative to 2015, while the decrease ratio in NO$_2$ was only $-8\%$ over this region. The observed SO$_2$ concentrations had a steeper decrease than PM$_{2.5}$ and NO$_2$, possibly because the dominant source sectors (i.e., power and industry) of SO$_2$ significantly reduced their emissions (Zheng et al., 2018). The power sector was the major contributor to emission reduction but only accounted for one-third of NO$_x$ emissions and the contribution of transportation to NO$_x$ emissions was estimated to have increased over recent years (Zheng et al., 2018). The interannual variations in emissions for China were calculated from MEIC (Zheng et al., 2018), the annual total emissions of SO$_2$ and NO$_x$ and the PM in the THB region reported by the National Bureau of Statistic of China (http://www.stats.gov.cn/tjsj/hndsjs/, last access: 17 January 2022), presenting a more rapid decline of SO$_2$ emissions in the THB than the changes of PM$_{2.5}$ and NO$_x$ emissions (Fig. S2 in the Supplement). The declining trend of anthropogenic emissions estimated from emission inventories can support the explanation of the changes in air pollutant concentrations.

Figure 5 shows the spatial distributions of 5-year averaged concentrations, the linear trends and the change rates in interannual variations of PM$_{2.5}$, SO$_2$ and NO$_2$ observed in the THB over 2015–2019. The change rates (% yr$^{-1}$) were calculated with the linear trends by dividing with temporal-mean concentrations of air pollutants at the observation sites for the analysis period in Fig. 5. The 5-year averaged PM$_{2.5}$ concentrations over the THB exceeded the Chinese National secondary air quality standard of 35 µg m$^{-3}$ for annual mean PM$_{2.5}$ concentrations (Fig. 5a), while SO$_2$ and NO$_2$ concentrations reached the secondary standards of 60 and 40 µg m$^{-3}$ in annual mean SO$_2$ and NO$_2$ concentrations at most sites over the THB (Fig. 5d and g). Specifically, the 5-year averaged NO$_2$ concentrations exceeded 40 µg m$^{-3}$ in WH (Wuhan), a megacity in central China, which might be attributable to the large amounts of traffic transportation. From 2015 to 2019, both PM$_{2.5}$ and SO$_2$ decreased at all sites over the THB (Fig. 5b and e), whereas NO$_2$ trends changed from mostly negative to positive in some sites (Fig. 5h), possibly due to the spatial disparity of NO$_x$ emissions in traffic sectors (Zheng et al., 2018). The comparison among the change rates of PM$_{2.5}$, SO$_2$ and NO$_2$ in the THB presented the largest decreases of SO$_2$ with $-40\%$–$-20\%$ yr$^{-1}$ over the 5 years (Fig. 5c, f and i), reflecting the effective control of SO$_2$ emissions in terms of primary gaseous pollutants. There were obvious decreases in regional mean PM$_{2.5}$, SO$_2$ and NO$_2$ concentrations over the THB (Fig. 4), while the declining degree of PM$_{2.5}$ and SO$_2$ varied from site to site over the THB and the change trends in NO$_2$ were weak negative and even positive in certain sites (Fig. 5c, f and i). These interannual changes of air pollutants in the THB over recent years were investigated using the emission- and meteorology-related long-term components of the air pollutants in the next sections.

### 3.4 Effects of NO$_2$ and SO$_2$ emission reductions on PM$_{2.5}$ change trends

The declining trend of PM$_{2.5}$ in China could be partly attributed to the reduced NO$_x$ and SO$_2$ concentrations for producing secondary aerosols (Zhang et al., 2018). The reduction rates of anthropogenic emissions markedly accelerated after 2013, decreasing by 59% for SO$_2$, 21% for NO$_x$, and 33% for PM$_{2.5}$ during 2013–2017 over China (Zheng et al., 2018). In order to assess the effect of changing precursor pollutant emissions on PM$_{2.5}$ reductions, we compared the linear trends of emissions-related long-term components of PM$_{2.5}$, NO$_2$ and SO$_2$ decomposed based on Eq. (9) over the THB for 2015–2019 (Fig. 6). The distinct declining

| Sites | Adj. $R^2$ of multiple linear regressions |
|-------|------------------------------------------|
|       | PM$_{2.5BL}$ | SO$_{2BL}$ | NO$_{2BL}$ |
| JZ    | 0.6776       | 0.4166     | 0.8358     |
| XN    | 0.6899       | 0.0630     | 0.7408     |
| XY    | 0.7971       | 0.6741     | 0.8181     |
| JM    | 0.7872       | 0.3612     | 0.6480     |
| YC    | 0.7168       | 0.2980     | 0.6304     |
| SZ    | 0.7175       | 0.3612     | 0.8669     |
| WH    | 0.7289       | 0.2718     | 0.6653     |
| EZ    | 0.7162       | 0.4592     | 0.7523     |
| HG    | 0.6937       | 0.1901     | 0.7220     |
| HS    | 0.5695       | 0.2787     | 0.6952     |
| CS    | 0.7307       | 0.1255     | 0.7012     |
| YY    | 0.7501       | 0.1047     | 0.7592     |
| XG    | 0.6755       | 0.4389     | 0.7692     |
| CD    | 0.7017       | 0.1730     | 0.6937     |

https://doi.org/10.5194/acp-22-3579-2022 Atmos. Chem. Phys., 22, 3579–3593, 2022
trends of emissions-related long-term PM$_{2.5}$ and SO$_2$ components, as well as the variable trends of emissions-related long-term NO$_2$ components were distributed basically consistently with the positive and negative trends in the interannual variations of air pollutant concentrations in the THB (middle column of Fig. 5; Fig. 6), demonstrating that the local emissions of air pollutants could spatially dominate the long-term variations of air pollutants in central China, especially the increasing trends in NO$_2$ at some THB sites.

PM$_{2.5}$ concentrations are changed by emissions of both primary PM$_{2.5}$ and its gaseous precursors. As major gaseous precursors, SO$_2$ and NO$_2$ can be oxidized to convert nitrate and sulfate for secondary PM$_{2.5}$ (S. Li et al., 2015). To investigate the effects of emission reductions on the interannual variations of PM$_{2.5}$, NO$_2$ and SO$_2$ over recent years, the ratios of change trends in long-term ($k_{LT}$) and emissions-related long-term ($k_{emiss}$) components of PM$_{2.5}$, SO$_2$ and NO$_2$, in the THB over 2015–2019 were demonstrated in Fig. 7, where the long-term and emissions-related long-term components of PM$_{2.5}$, SO$_2$ and NO$_2$ were calculated with Eqs. (4) and (9). The trend ratios $k_{LT}/k_{emiss}$<1 indicated the more obvious downward trend of emissions-related long-term variations than the long-term trend of air pollutant concentrations, which might be attributed to the offsetting effect of meteorological conditions on emission reduction in the air quality change, whereas the long-term trend of air pollutant concentrations was more significant than the emissions-related long-term trend as $k_{LT}/k_{emiss}$>1, reflecting the synchronous impacts of anthropogenic emissions and meteorology on the long-term trend in air pollutant change. In addition, the trend ratios $k_{LT}/k_{emiss}>1$ and $k_{LT}/k_{emiss}<1$ of the gaseous precursors of PM$_{2.5}$, SO$_2$ and NO$_2$, could reflect the high and weak efficiencies of SO$_2$ and NO$_2$, converting to sulfate and nitrate in the production of secondary PM$_{2.5}$ during air pollutant emission reduction. The notable differences in Fig. 7 were spatially distributed with the trend ratios $k_{LT}/k_{emiss}>1$ and $k_{LT}/k_{emiss}<1$ in PM$_{2.5}$, SO$_2$ and NO$_2$ concentrations under the same meteorological conditions, indi-
cating the different influences of emissions on the long-term variations of PM$_{2.5}$, SO$_2$ and NO$_2$ in the THB during recent years. The reduction in PM$_{2.5}$ emissions was a primary cause of the long-term declines in PM$_{2.5}$ concentrations in the THB, even though the meteorological changes might offset the effects of emission reduction on air quality improvement over the southern THB (Figs. 6 and 7). It is noteworthy that the trend ratios $k_{LT}/k_{emiss} < 1$ of PM$_{2.5}$ were accompanied with $k_{LT}/k_{emiss} > 1$ of SO$_2$ and NO$_2$ at the downwind southern THB sites with both negative $k_{LT}$ and $k_{emiss}$ (Fig. 7, Table S4 in the Supplement), which could imply the increasing conversion efficiency of SO$_2$ and NO$_2$ to sulfate and nitrate for secondary PM$_{2.5}$ during the reductions of air pollutant emissions over recent years. In the upwind northern THB sites, the $k_{LT}/k_{emiss} > 1$ of PM$_{2.5}$ were accompanied with $k_{LT}/k_{emiss} > 1$ of SO$_2$ and NO$_2$ with an obvious facilitating effect of meteorology on the PM$_{2.5}$ decline (Fig. 7, Table S4), revealing the underlying effect of regional transport of air pollutants on the spatial distribution of conversion efficiency of gaseous precursor to secondary PM$_{2.5}$.

In order to further assess the effect of gaseous precursor emissions on PM$_{2.5}$ declines during recent 5-year air pollution mitigation, we selected seven and nine sites in the THB with decreasing trends of emissions-related long-term SO$_2$ and NO$_2$ components below -0.5 and 0.0 $\mu$g m$^{-3}$ 100 d$^{-1}$, respectively, (Table S4) to compare the trend ratios $k_{LT}/k_{emiss}$ of PM$_{2.5}$, NO$_2$ and SO$_2$ for 2015–2019 (Fig. 8). The significantly negative linear correlations between the changes in $k_{LT}/k_{emiss}$ of gaseous precursors (SO$_2$, NO$_2$) and PM$_{2.5}$ could present the connection of $k_{LT}/k_{emiss} > 1$ of NO$_2$ and SO$_2$ with $k_{LT}/k_{emiss} < 1$ of PM$_{2.5}$, which confirms the fact that the high conversion efficiency of SO$_2$ and NO$_2$ to sulfate and nitrate could have have offset the role of PM$_{2.5}$ emission reduction in controlling PM$_{2.5}$ pollution. The decreasing emissions of gaseous precursors drove faster oxidation of NO$_2$ and SO$_2$ to nitrate and sulfate components of PM$_{2.5}$ in the source regions of air pollution in China (Zhai et al., 2021; Huang et al., 2021). This study identified the enhancing contribution of gaseous precursors to PM$_{2.5}$ concentrations with reduced anthropogenic emissions of air pollutants over receptor regions in regional PM$_{2.5}$ transport.

There are a few sources of uncertainty in the discussion of chemical transformation, for example in the separation of emission- and meteorology-related long-term components and in the selection of observational sites. Our results point to a better understanding of the offsetting effect of SO$_2$ and NO$_2$ oxidized to secondary particles in PM$_{2.5}$ mitigation during the emission reduction in the THB. 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 $k_{LT}/k_{emiss}$ of PM$_{2.5}$. However, due to the current lack of long-term observations of PM$_{2.5}$ components in the THB, the influence of emission 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.

### 3.5 Meteorological contribution to PM$_{2.5}$ change trends

As the air pollutant change trend is assumed to generally consist of emission- and meteorology-related changes (Seo et al., 2018; Yin et al., 2019b), the meteorological contribution rate $\text{Con}_{\text{met}}$ to the long-term PM$_{2.5}$ change trend is calculated with the following equation:

$$\text{Con}_{\text{met}} = \frac{k_{LT} - k_{emiss}}{k_{LT}} \times 100\%.$$  \hspace{1cm} (10)

Here, $\text{Con}_{\text{met}}$ (in $\%$) is estimated with the linear trends $k_{LT}$ of the long-term component PM$_{2.5LT}(t)$ and $k_{emiss}$ of the emissions-related long-term component PM$_{2.5LT}(t)$, PM$_{2.5LT}(t)$ and PM$_{2.5emiss}(t)$ are, respectively, calculated with Eqs. (4) and (9).

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**Figure 6.** Spatial distributions of the linear trends in emissions-related long-term components of (a) PM$_{2.5}$, (b) SO$_2$ and (c) NO$_2$ (unit: $\mu$g m$^{-3}$ d$^{-1}$) over 2015–2019 in the THB.
To quantitatively assess the meteorological contribution to the declining PM$_{2.5}$ trends, the linear trends $k_{LT}$ and $k_{emiss}$ with the meteorological contribution rate $C_{met}$ in Eq. (10) were presented in Table S5 in the Supplement for 14 sites over the THB during 2015–2019. All the trends $k_{LT}$ and $k_{emiss}$, respectively, in PM$_{2.5LT}(t)$ and PM$_{2.5emiss}(t)$ were negative over the THB (Table S5), indicating the significant effect of emission reductions on declining PM$_{2.5}$ trends for improving regional air quality in central China. By comparing the declining PM$_{2.5}$ trends $k_{emiss}$ and $k_{LT}$ from site to site (Table S5), the positive and negative contributions of meteorological variations to PM$_{2.5}$ change trends over recent years were determined with the positive and negative differences between $k_{emiss}$ and $k_{LT}$ with the distinct meteorological influences on the change of the THB regional environment.

The spatial distribution of meteorological contribution rates $C_{met}$ regarding the long-term declining PM$_{2.5}$ trend presented a unique pattern of northern positive and southern negative values over the THB (Fig. 9), with high positive contributions in the northern sites XY (61.92 %) and EZ (37.31 %), and low negative contributions in the southern sites CD (−24.93 %) and CS (−23.03 %). It is worth mentioning that the contribution rates of meteorological variations show great spatial disparities at a small scale, i.e., EZ, HG and HS, which seems not be induced by the variation in synoptic weather or meteorological conditions. 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 the surrounding regions vary distinctly with the underlying surface conditions of plains, lakes and hilly areas. The underlying surface of observational sites with different near-surface meteorology effectively influences the local accumulation, chemical transformation, and dry and wet depositions of air pollutants (Bai et al., 2022). Therefore, the heterogeneity of meteorological contributions to PM$_{2.5}$ at such a small spatial scale might be attributed to the local meteorological conditions in the atmospheric boundary layer, which are largely affected by the underlying surface changes.
Compared with the statistical studies using synthetic data of meteorological influence on regional PM$_{2.5}$ changes in central China and the meteorological contribution from $-45.5\%$ to $29.0\%$ over recent years (Gong et al., 2021; L. Chen et al., 2020), the PM$_{2.5}$ pollution over the THB was affected contrarily by meteorological drivers with the northern positive and southern negative contributions from 2015 to 2019 (Fig. 9). The meteorological change could accelerate and offset the effects of emission reductions on declining PM$_{2.5}$ trends in the northern and southern THB, which might be attributed to the regional transport of air pollutants conducive to the upwind diffusion and downward accumulation of air pollutants, respectively, over the northern and southern THB under the declining wind of East Asian monsoons over recent years (Hu et al., 2020; Zhong et al., 2019).

![Figure 9](https://example.com)  

**Figure 9.** Spatial distribution of contribution rates (colored dots, unit: %) of meteorological variations regarding PM$_{2.5}$ reductions with topographical height (color contours, m a.s.l.) in the THB (outlined with orange dashed line) and surrounding regions from 2015 to 2019.

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 the KZ filter, concluding on the large impact of meteorology on the PM$_{2.5}$ changes over 2015–2019. To validate this conclusion of analyses with the KZ filter, we designed three sets of modeling experiments CTRL, SENS-MET and SENS-EMI (Table S6 in the Supplement) for December of 2015–2019, respectively, driven with the changing meteorology and anthropogenic emissions over 2015–2019, 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 are 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 in the Supplement), and the three designed sets of modeling experiments CTRL, SENS-MET and SENS-EMI can be used in the further analyses of emission and meteorological impact on PM$_{2.5}$ change over 2015–2019 to confirm the results of the KZ filter.

We derived the impact of meteorology by comparing the simulated PM$_{2.5}$ concentrations in 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:

\[
\text{Con}_{\text{MET}} = \frac{k_{\text{MET}}}{k_{\text{CTRL}}}, \quad (11)
\]

\[
\text{Con}_{\text{EMI}} = \frac{k_{\text{EMI}}}{k_{\text{CTRL}}}, \quad (12)
\]

\[
R\text{Con}_{\text{MET}} = \frac{\text{Con}_{\text{MET}}}{\text{Con}_{\text{MET}} + \text{Con}_{\text{EMI}}} \times 100\%. \quad (13)
\]

$k_{\text{CTRL}}, k_{\text{MET}}$ and $k_{\text{EMI}}$ represent the trends in interannual changes of PM$_{2.5}$ concentrations simulated by the experiments CTRL, SENS-MET and SENS-EMI, respectively. $\text{Con}_{\text{MET}}$ and $\text{Con}_{\text{EMI}}$ are the contributions of meteorology and emission, and $R\text{Con}_{\text{MET}}$ 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 a 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 declining PM$_{2.5}$ trends in the northern and southern THB, respectively. The general spatial distribution of meteorological contribution rates regarding declining PM$_{2.5}$ trends from the WRF-Chem simulation was consistent with the results using the KZ filter (Figs. 9 and 10), validating these results and confirming that meteorological drivers exerted a contrary impact between the northern positive and southern negative contributions to long-term changes of PM$_{2.5}$ concentrations in the THB.

4 Conclusions

The meteorological effect on multi-scale changes of the atmospheric environment has been rarely assessed for the re-
receptor region in the regional transport of air pollutants. In this study of observations and modeling, we targeted the THB, a large region of heavy PM$_{2.5}$ pollution in central China, to assess the effects of meteorology on PM$_{2.5}$ changes over a receptor region in the regional transport of air pollutants during recent years. The study results provide insights into the effects of emission mitigation and meteorological changes on the source–receptor relationship of the long-range transport of air pollutants for regional and global environment changes.

This study decomposed the observed PM$_{2.5}$ concentrations into multiple timescale components with a modified KZ filter to better understand the PM$_{2.5}$ variations, with the short-term, seasonal and long-term components accounting for, respectively, 47.5%, 41.4% and 3.7% of observed PM$_{2.5}$ changes. The short-term and seasonal PM$_{2.5}$ components dominated the daily PM$_{2.5}$ changes and long-term component determined the trend of PM$_{2.5}$ change over recent years. The long-term components of PM$_{2.5}$, SO$_2$ and NO$_2$ were further isolated into emission- and meteorology-related long-term components with multiple linear regressions in order to determine the contributions of emission and meteorology to PM$_{2.5}$ decline in the THB over 2015–2019. The reduction in anthropogenic emissions was the primary cause of the long-term decline in PM$_{2.5}$ concentrations, and the meteorological changes moderated the PM$_{2.5}$ variations in the THB. As the receptor region of regional PM$_{2.5}$ transport, the impact of diverse meteorological conditions on long-term trend of PM$_{2.5}$ changes displayed a unique regional pattern of northern positive rates up to 61.92% and southern negative rates down to -24.93%. The change of meteorological conditions could accelerate and offset the effects of the emission reductions on declining PM$_{2.5}$ trends in the northern and southern THB, which can be attributed to the upwind diffusing and downward accumulating roles of regional transport pathways in air pollutants in the THB. In terms of gaseous precursor emissions, the increasing conversion efficiency of SO$_2$ and NO$_2$ to sulfate and nitrate for secondary PM$_{2.5}$ could have offset the role of emission reduction in controlling air pollution, and the contribution of gaseous precursors to secondary PM$_{2.5}$ was enhanced with the reducing anthropogenic emissions of air pollutants over this receptor region.

This study revealed the impact of anthropogenic emissions and meteorological conditions on PM$_{2.5}$ decline over a receptor region in the regional transport of air pollutants in central China. The effect of regional transport on PM$_{2.5}$ pollution over receptor regions is found to differ from that over source regions with high anthropogenic emissions. We considered the uncertainties induced by statistical methods as systematic biases and explained the offsetting effect of enhancing oxidation of gaseous precursors to secondary particles on PM$_{2.5}$ decline during stringent emission controls. The changes in data coverage and meteorological parameter selection largely influence the quantitative estimation of the contributions of meteorology and emissions. Further work should be undertaken regarding climate analysis of the long-term data of fine meteorological and environmental observations and more comprehensive modeling of chemical and physical processes in the atmosphere to generalize the assessment of the effects of emission mitigation and meteorological changes on the source–receptor relationship of the regional transport of air pollutants.

Data availability. Data used in this paper can be provided upon request from Xiaoyun Sun (sunxy6362@126.com) or Tianliang Zhao (tlzhao@nuist.edu.cn).

Supplement. The supplement related to this article is available online at: https://doi.org/10.5194/acp-22-3579-2022-supplement.

Author contributions. TZ and XS conceived the study. YB provided the observation data and analysis. XS designed the graphics and wrote the manuscript with help from TZ, YB and SK. HZ, WH, XM and JX were involved in the scientific discussion. All authors commented on the paper.

Competing interests. The contact author has declared that neither they nor their co-authors have any competing interests.
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Acknowledgements. This research was financially funded by grants from National Natural Science Foundation of China (grant nos. 41830965, 42075186 and 91744209) and the National Key R & D Program Pilot Projects of China (grant no. 2016YFC0203304).

Financial support. This research has been supported by the National Natural Science Foundation of China (grant nos. 41830965, 42075186 and 91744209) and the National Key Research and Development Program of China (grant no. 2016YFC0203304).

Review statement. This paper was edited by Leiming Zhang and reviewed by three anonymous referees.

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