Meteorological effects on PM$_{2.5}$ change over a receptor region in regional transport of air pollutants: observational study of recent year emission reduction in central China

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Abstract. 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 this study on observations of environment and meteorology over 2015-2019, the Kolmogorov–Zurbenko (KZ) filter was performed to decompose the PM$_{2.5}$ variations into multi-time scale components over the Twain-Hu Basin (THB), a receptor region in regional transport of air pollutants in central China, where the short-term, seasonal and long-term components accounted for respectively 47.5%, 41.4% and 3.7% to 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. The emission- and meteorology-related long-term PM$_{2.5}$ components over the THB were identified. The meteorological contribution to PM$_{2.5}$ declining trend presented the 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 the receptor region in regional PM$_{2.5}$ transport. It is noteworthy that the increasing conversion efficiencies of SO$_2$ and NO$_x$ to sulfate and nitrate for secondary PM$_{2.5}$ could offset the effects 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 with controlling anthropogenic emissions of PM$_{2.5}$ and the gaseous precursors over the receptor region in regional transport of air pollutants.

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 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, the large areas over Central and Eastern China (CEC) have undergone haze pollution with unprecedentedly high PM$_{2.5}$ levels in the regions covering North China Plain (NCP), Yangtze River Delta (YRD), Pearl River Delta (PRD) and Sichuan Basin (SB) (Zhang et al., 2012; Lin et al., 2018; Guo et al., 2017). In order to improve air quality with reducing air pollutant emissions, Chinese government has implemented an Action Plan of controlling anthropogenic emissions since September 2013 (http://www.gov.cn/xinwen/2018-02/01/content_5262720.htm, last access: August 21, 2021). Surface PM$_{2.5}$ concentrations 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.

In addition to anthropogenic emissions of air pollutants, the meteorological conditions can alter the local accumulation, regional transport, chemical conversion, wet and dry depositions of air pollutants (Lu et al., 2017; Li et al., 2018). Severe haze pollution always occurs in the wintertime under the stagnant meteorological conditions with weak near-surface wind, strong
temperature inversion, and high relative humidity in the atmospheric boundary layer, which are favorable for the accumulation of air pollutants to form air pollution (Li et al., 2018; Miao et al., 2015; Tang et al., 2016). Meteorological conditions are closely governed by synoptic circulations, modulating the atmospheric physical and chemical processes including regional transport of air pollutants (Miao et al., 2017; Ning et al., 2019). The climate changes of East Asian monsoons largely influence the seasonal and interannual variations of aerosol concentrations for air pollution over China (Zhu et al., 2012; Jeong and Park, 2017).

Assessments on contributions of anthropogenic emissions and meteorological changes to air quality improvement are an important issue in environmental changes (Pearce et al., 2011; Zhang et al., 2018; Chen et al., 2019). The chemical transport models have been widely used to quantify the meteorological effects on PM$_{2.5}$ variations by a linear additive relationship between sensitivity and base simulations (Mueller and Mallard, 2011; Li et al., 2015b; Zhang et al., 2020). The contribution of meteorological changes to PM$_{2.5}$ decreases was estimated at the averages of 10–20% with the interannual fluctuations of about 5% in CEC from 2015 to 2019 through a model-based environmental meteorology index (Gong et al., 2021). The accuracy of modeling assessments can be influenced by the uncertainties in emission inventories and the incomplete chemical and physical mechanisms in air pollution simulation (Li et al., 2011). Based on statistical analysis on long-term observational data, it was quantified that the emission control 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, the modeling and observational studies have mostly assessed the contribution of emissions and meteorology to regional PM$_{2.5}$ variations 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.

The Twain-Hu Basin (THB), featuring the lower lands (mainly less than 200 m in a. s. l.) of two provinces Hubei and Hunan in central China (Fig. 1), is surrounded by the high air pollution regions NCP, YRD, PRD and SB. As such, it is the receptor region in regional transport of air pollutants from the upstream region driven by East Asian monsoonal winds over CEC (Shen et al., 2020). The heavy air pollution in the THB with a unique “non-stagnation” atmospheric boundary layer is
aggravated by regional PM$_{2.5}$ transport over CEC (Zhong et al., 2019; Yu et al., 2020). By cohesion with the heavy pollution region of NCP through distinct transport channels, the regional transport from northern China to the THB 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 air pollution region in central China need to be specifically assessed with the long-term observations over recent years.

In this observational study, we investigated the multi-scale changes of PM$_{2.5}$ concentrations over the THB from 2015 to 2019 by establishing the statistic model with Kolmogorov–Zurbenko (KZ) filter, and then evaluated the contributions of anthropogenic emissions and meteorological changes to the declining trends in PM$_{2.5}$ concentrations in this receptor region in regional PM$_{2.5}$ transport over CEC during the past 5-year emission control. 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: August 21, 2021). 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: August 21, 2021), including air temperature, relative humidity (RH), sea level pressure (SLP), wind speed (WS) and precipitation with temporal resolutions of 3 h.

2.2 KZ filter
To better understand the multi-time scale variations of PM\(_{2.5}\) and the relation to air pollutant emissions and meteorological drivers, KZ filter (Rao and Zurbenko, 1994; Seo et al., 2018) is used to separate the daily data into multi-scale components, based on an iterative moving average that removes high frequency variations in the data with the applications in 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 the length of moving average window \(m\) and the number of iterations \(p\), can remove the high-frequency component of period 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 a high accuracy level to compare with the wavelet transform method (Eskridge et al., 1997). In this study, we applied KZ\(_{15,5}\) and KZ\(_{365,3}\) filters to remove the variations with the periods shorter than 33 days and 1.7 years.

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:

\[
X(t) = X_{ST}(t) + X_{BL}(t). \tag{1}
\]

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 days from the observed data, expressing with:

\[
X_{BL}(t) = KZ_{15,5}[X(t)]. \tag{2}
\]

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

\[ X_{SN}(t) = X_{BL}(t) - X_{LT}(t). \] (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 regional transport of air pollutants (Seo et al., 2017), partly associated with short-term fluctuations in air pollutant emissions (Russell et al., 2010). The seasonal and long-term components are attributable to the variations in air pollutant emissions related to human activities as well as the seasonal and interannual changes in meteorological conditions (Kim et al., 2018).

### 2.3 Multiple linear regression of air pollutant changes with meteorological variables

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

\[ PM_{2.5BLMLR}(t) = a_0 + \sum_i a_i MET_{BLi}(t), \] (7)

where MET\(_{BLi}(t) \ (i \in \{1,5\})\) 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\(_{BL}(t)\), Pre\(_{BL}(t)\). We fit the regression coefficient \(a_i\) for each meteorological variable and the intercept \(a_0\). The residual \( \epsilon_{PM_{2.5}} \) between \( PM_{2.5BL}(t) \) and \( PM_{2.5BLMLR}(t) \) regressed with the multiple linear equation (7) is given as:

\[ \epsilon_{PM_{2.5}}(t) = PM_{2.5BL}(t) - PM_{2.5BLMLR}(t). \] (8)

\( \epsilon_{PM_{2.5}} \) 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 emission-related long component $PM_{2.5,LT}^{emiss}(t)$ can be isolated as follows:

$$PM_{2.5,LT}^{emiss}(t) = KZ_{365,3}[\varepsilon_{PM_{2.5}}(t)].$$

(9)

Here the long-term component of surface PM$_{2.5}$ concentration can be further separated into the emission- and meteorology-related long-term components with Eqs. (9) and (4) (Seo et al., 2018). Similarly, the multi-time scale variations in SO$_2$ and NO$_2$ with long-term variations related to changes in air pollutant emissions and meteorological drivers are decomposed by KZ filter with multiple linear regression. Seo et al. (Seo et al., 2018) described the details of this method.

3. Results and discussion

3.1 Verification of PM$_{2.5}$ decompositions in multi-scale variations

The daily PM$_{2.5}$ concentrations observed in 14 sites over the THB (Fig. 1) were decomposed into short-term, seasonal and long-term components with Eqs. (4), (5) and (6) of the KZ filter. To verify the decomposition results, the spatial distribution of total contributions of short-term, seasonal and long-term PM$_{2.5}$ components to the total variances of observed daily changes in PM$_{2.5}$ concentrations over 2015–2019 were shown in Figure 2a. The total contribution of short-term, seasonal and long-term components demonstrated the regional distribution with the high values exceeding 90% and the regional average of 92.7% in the THB (Fig. 2a), which presented a good decomposition of the multi-time scale components from the observed daily PM$_{2.5}$ with the KZ filter.

Based on the PM$_{2.5}$ decomposition results of KZ filter, the short-term, seasonal and long-term components respectively accounted for 34.8%–53.8%, 29.2%–56.3% and 0.2%–9.8% of the total variances of daily PM$_{2.5}$ changes in the THB over recent years (Figs. 2b, 2c and 2d), reflecting the different patterns of multi-time scale variations of PM$_{2.5}$ over this region in central China with diverse effects of emissions and meteorology. The regional contributions of short-term, seasonal and long-term components were averaged respectively with 47.5%, 41.4% and 3.7% to daily PM$_{2.5}$ changes over the THB (Fig. 2), which could be reasonably verified that the daily variation in atmospheric pollutant was generally dominated by short-term
and seasonal components with 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 in 14 sites of the THB to characterize the temporal variations of three components in the THB for 2015–2019 (Fig. 3). 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 an important role of the short-term component with the temporal period $< 33$ days in the day-to-day variations of PM$_{2.5}$ concentrations in the THB (Fig. 3a).

The notable peaks of PM$_{2.5}$ seasonal components emerged in winters were highly in keeping 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 components and daily PM$_{2.5}$ concentrations, which could reflect a significant modulation of the PM$_{2.5}$ seasonal oscillations to 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 long-term component of PM$_{2.5}$ exhibited a steadily declining trend over 2015-2019 (Fig. 3c), which might be caused by the sustained impact of emission control. The correlation coefficient ($r = 0.24$, $p<0.05$) of long-term PM$_{2.5}$ component with the observed daily PM$_{2.5}$ change was much smaller than those of 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.

### 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 regional climate of East Asian monsoons rather than synoptic-scale meteorological processes. Generally, the baseline components of
air pollutants were negatively correlated with baseline components of wind speed (WS\textsubscript{BL}) and positively correlated with baseline components of sea level pressure (SLP\textsubscript{BL}) (Table S1–S3), which could be attributed to the ventilation effect of wind and stagnant condition 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\textsubscript{2.5} concentrations over the emission source region, increasing wind speed might cause the accumulation of PM\textsubscript{2.5} concentrations over the downwind region of emission sources (Chen et al., 2020), which led to the inconsistent influence of WS\textsubscript{BL} in the region of central China (Table S1–S3). Under surface high air temperature conditions, there are strong thermal activities such as turbulence, making an accelerated dispersion of air pollutants (Yang et al., 2016b). The negative influence of RH\textsubscript{BL} and T\textsubscript{BL} on PM\textsubscript{2.5BL}, SO\textsubscript{2BL} and NO\textsubscript{2BL} mainly reflected the effect of seasonal cycle in East Asian winter and summer monsoons, whereas the influence of precipitation on air pollutants was more straightforward than other meteorological parameters, negatively influencing surface pollutant concentrations through the precipitation washout of air pollutants (Table S1–S3).

To isolate emission-related long-term components from long-term components of PM\textsubscript{2.5}, NO\textsubscript{2} and SO\textsubscript{2}, the stepwise multiple linear regressions of PM\textsubscript{2.5BL}, SO\textsubscript{2BL} and NO\textsubscript{2BL} respectively with baseline components of meteorological parameters (T\textsubscript{BL}, WS\textsubscript{BL}, RH\textsubscript{BL}, SLP\textsubscript{BL} and Pre\textsubscript{BL}) were conducted with Eq. (7) in 14 sites, by adding and deleting meteorological variables based on the independent statistical significance to obtain the best model fit (Draper, 1998). We evaluated the PM\textsubscript{2.5BL}, SO\textsubscript{2BL} and NO\textsubscript{2BL} fitted by the multiple linear regression models with the KZ decomposition (Table 1). The multiple linear regressions explained PM\textsubscript{2.5BL}, SO\textsubscript{2BL} and NO\textsubscript{2BL} with adjusted determination coefficients (Adj. R\textsuperscript{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. The Adj. R\textsuperscript{2} of multiple linear regression for SO\textsubscript{2BL} were lower than those of PM\textsubscript{2.5BL} and NO\textsubscript{2BL}, which might be attributed to the larger impact of SO\textsubscript{2} emission control on the seasonal and long-term SO\textsubscript{2} variations. In general, the variations of meteorological drivers can well reproduce the meteorology-related seasonal and long-term variations of PM\textsubscript{2.5}, SO\textsubscript{2} and NO\textsubscript{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 (Li et al., 2015a). 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 with chemical reactions of the precursor gases (e.g., SO$_2$ and NOx), which are mainly produced by human activities (Li et al., 2015a; Yang et al., 2016a). 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 Figure 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).

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 Figure 5. The 5-year averaged PM$_{2.5}$ concentrations over the THB exceeded the Chinese National secondary air quality standard of 35 $\mu$g m$^{-3}$ for annual mean PM$_{2.5}$ concentration (Fig. 5a), while SO$_2$ and NO$_2$ concentrations reached the secondary standards of 60 $\mu$g m$^{-3}$ and 40 $\mu$g m$^{-3}$ in annual mean SO$_2$ and NO$_2$ concentrations at most sites over the THB (Figs. 5d and 5g). Specifically, the 5-year averaged NO$_2$ concentrations exceeded 40 $\mu$g m$^{-3}$ in WH (Wuhan), the mega-city in central China, that 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 (Figs. 5b and 5e), whereas NO$_2$ trends were changed from mostly negative to positive in
some sites (Fig. 5h), possibly due to the spatial disparity of NOx 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 -20% – -40% yr$^{-1}$ over the five years (Figs. 5c, 5f and 5i), 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 (Figs. 5c, 5f and 5i). These interannual changes of air pollutants in the THB over recent years were investigated with the emission- and meteorology-related long-term components of 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 the secondary aerosols (Zhang et al., 2018). The reduction rates of anthropogenic emissions have markedly accelerated after 2013, decreasing by 59% for SO$_2$, 21% for NO$_x$ and 33% for PM$_{2.5}$ during 2013–2017 over the THB region (Zheng et al., 2018). In order to assess the effect of changing precursor pollutant emissions on PM$_{2.5}$ declines, we compared the linear trends of emission-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 trends of emission-related long-term PM$_{2.5}$ and SO$_2$ components as well as the variable trends of emission-related long-term NO$_2$ components were distributed basically consistent with the positive and negative trends in the interannual variations of air pollutant concentrations in the THB (Fig. 5 (middle column); 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 PM$_{2.5}$’s gaseous precursors. As major gaseous precursors, SO$_2$ and NO$_2$ can be oxidized to convert nitrate and sulfate for secondary PM$_{2.5}$ (Li et al., 2015a). 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 emission-related long-term ($k_{emiss}$) components of PM$_{2.5}$, SO$_2$ and NO$_2$, in the THB over 2015–2019 were demonstrated in Figure 7, where the long-term and emission-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 emission-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 air quality change, whereas the long-term trend of air pollutant concentrations was more significant than the emission-related long-term trend with $k_{LT}/k_{emiss} > 1$, 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 PM$_{2.5}$’s gaseous precursors 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 Figure 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, indicating 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 for 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), 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 obviously facilitating effect of meteorology on 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 7 and 9 sites in the THB with the decreasing trends of emission-related long-term SO$_2$ and NO$_2
components below -0.5 and 0.0 μg m⁻³ 100d⁻¹ respectively (Table S4) to compare the trend ratios \( \frac{k_{LT}}{k_{emiss}} \) of PM₂.₅, NO₂ and SO₂ for 2015–2019 (Fig. 8). The significantly negative linear correlations between changes in \( \frac{k_{LT}}{k_{emiss}} \) of gaseous precursors (SO₂ and NO₂) and PM₂.₅ could present the connection of \( \frac{k_{LT}}{k_{emiss}} > 1 \) for NO₂ and SO₂ with \( \frac{k_{LT}}{k_{emiss}} < 1 \) for PM₂.₅, which confirmed the fact that the high conversion efficiency of SO₂ and NO₂ to sulfate and nitrate could offset the role of PM₂.₅ emission reduction in controlling PM₂.₅ pollution. This study identified the enhancing contribution of gaseous precursor emissions to PM₂.₅ concentrations with reducing anthropogenic emissions of air pollutants over the receptor region in regional PM₂.₅ transport.

### 3.5 Meteorological contribution to PM₂.₅ 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 \( Con_{met} \) to long-term PM₂.₅ change trend is calculated with the following equation:

\[
Con_{met} = \frac{k_{LT} - k_{emiss}}{k_{LT}} \times 100\% \tag{10}
\]

Here, \( Con_{met} \) (in %) is estimated with the linear trends \( k_{LT} \) of long-term component \( PM_{2.5LT}(t) \) and \( k_{emiss} \) of emission-related long-term component \( PM_{2.5emiss}(t) \). \( PM_{2.5LT}(t) \) and \( PM_{2.5emiss}(t) \) are respectively calculated with Eqs. (4) and (9).

To quantitatively assess the meteorological contributions to the PM₂.₅ declining trends, the linear trends \( k_{LT} \) and \( k_{emiss} \) with the meteorological contribution rate \( Con_{met} \) in Eq. (10) were presented in Table S5 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 PM₂.₅ declining trends for improving regional air quality in central China. By comparing the PM₂.₅ declining trends \( k_{emiss} \) and \( k_{LT} \) from site to site (Table S5), the positive and negative contributions of meteorological variations to PM₂.₅ 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 THB’s regional environment.
The spatial distribution of meteorological contribution rates $Con_{met}$ to long-term PM$_{2.5}$ declining trend presented the unique pattern of northern positive and southern negative values over the THB (Fig. 9), with the high positive contributions in northern sites XY (61.92%) and EZ (37.31%) as well as low negative contributions in southern sites CD (-24.93%) and CS (-23.03%). Comparing with the statistical studies of meteorological influence on regional PM$_{2.5}$ changes in other regions over China with the meteorological contribution of about 20% over recent years (Chen et al., 2019; Zhai et al., 2019), 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). The meteorological change could accelerate and offset the effects of emission reductions on PM$_{2.5}$ declining trends in the northern and southern THB, which might be attributed to 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).

4. Conclusions

The observational data of environment and meteorology from 2015 to 2019 were achieved to investigate the characteristics and causes of PM$_{2.5}$ reductions in the THB, a receptor region in regional transport of air pollutants in central China. This study decomposed the observed PM$_{2.5}$ concentrations into multi-time scale 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% to 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, to figure out 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 for 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 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 emission reductions on PM$_{2.5}$ declining trends in the northern and southern THB, which could be attributed to the upwind diffusing and downward accumulating roles of regional transport pathway on 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 offset the role of PM$_{2.5}$ emission reduction in controlling air pollution, and the contribution of gaseous precursor emissions to secondary PM$_{2.5}$ enhanced with the reducing anthropogenic emissions of air pollutants over this receptor region.

This study exposed the impact of anthropogenic emissions and meteorological conditions on the PM$_{2.5}$ decline over a receptor region in regional transport of air pollutants in central China. The effect of regional transport on PM$_{2.5}$ pollution over the receptor region was found differing from that over the source regions with high anthropogenic emissions. To generalize our finding, further work could be desired with climate analyses of long-term observations of air pollutants and more comprehensively modeling of air quality and meteorology.

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).

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

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

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Table 1 Adjusted determination coefficients (Adj. R^2) between the baseline components decomposed by KZ filter and fitted with multiple linear regressions respectively for PM_{2.5BL}, SO_{2BL} and NO_{2BL} in 14 sites over the THB. All Adj. R^2 passing the confidence level of 99%.
<table>
<thead>
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<th>Sites</th>
<th>Adj. R² of multiple linear regressions</th>
</tr>
</thead>
<tbody>
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<td></td>
<td>PM₂.₅BL</td>
</tr>
<tr>
<td>JZ</td>
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</tr>
<tr>
<td>XN</td>
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</tr>
<tr>
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<td>0.6755</td>
</tr>
<tr>
<td>CD</td>
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</tr>
</tbody>
</table>
Figure 1 Topographical height (color contours, m, in a. s. l.) over the THB (outlined with black dashed line) with the locations of 14 sites (red dots) and the surrounding regions in central China.

Figure 2 Spatial distributions of the (a) total and relative contributions of (b) short-term, (c) seasonal and (d) long-term
components to the total variances of daily PM$_{2.5}$ changes observed at 14 sites in the THB with the regional averages of 92.7%, 47.5%, 41.4% and 3.7%.

Figure 3 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.
Figure 4 Interannual variations in the ratios of observed annual mean concentrations of SO$_2$, NO$_2$ and PM$_{2.5}$ relative to those in 2015 averaged over the THB.

Figure 5 Spatial distributions of (left column) 5-year averages of (a) PM$_{2.5}$, (d) SO$_2$ and (g) NO$_2$ concentrations (μg m$^{-3}$, unit: μg m$^{-3}$).
3), (middle column) the linear trends in interannual variations of (b) PM$_{2.5}$, (e) SO$_2$ and (h) NO$_2$ ($k$, unit: μg m$^{-3}$ yr$^{-1}$), as well as (right column) the change rates ($R_t=k/A$, unit: % yr$^{-1}$) of (c) PM$_{2.5}$, (f) SO$_2$ and (i) NO$_2$ in the THB over 2015–2019.

**Figure 6** Spatial distributions of the linear trends in emission-related long-term components of (a) PM$_{2.5}$, (b) SO$_2$ and (c) NO$_2$ (unit: μg m$^{-3}$ d$^{-1}$) over 2015–2019 in the THB.

**Figure 7** Spatial distributions of the ratios of linear trends in long-term components ($k_{LT}$) and emission-related long-term components ($k_{emiss}$) of (a) PM$_{2.5}$, (b) SO$_2$ and (c) NO$_2$ at 14 sites in the THB over 2015–2019.
Figure 8 Scatter plots of the ratios between $k_{LT}$ and $k_{emiss}$ of (a) SO$_2$, (b) NO$_2$ and PM$_{2.5}$ in the THB from 2015 to 2019 with red lines for the linear fitting equations.

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