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Dynamics-based estimates of decline trend with fine temporal variations in China's $PM_{2.5}$ emissions

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Abstract

Timely, continuous, and dynamics-based estimates of $PM_{2.5}$ emissions with a high temporal resolution can be objectively and optimally obtained by assimilating observed surface $PM_{2.5}$ concentrations using flow-dependent error statistics. Annual $PM_{2.5}$ emissions in China have consistently decreased of approximately 3% to 5% from 2017 to 2020. Significant $PM_{2.5}$ emission reductions occurred frequently in regions with large $PM_{2.5}$ emissions. COVID-19 could cause a significant reduction of $PM_{2.5}$ emissions in the north China plain and northeast of China in 2020. The magnitudes of $PM_{2.5}$ emissions were greater in the winter than in the summer. $PM_{2.5}$ emissions show an obvious diurnal variation that varies significantly with the season and urban population. Improved representations of $PM_{2.5}$ emissions across time scales can benefit emission inventory, regulation policy and emission trading schemes, particularly for especially for high temporal

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resolution air quality forecasting and policy response to severe haze pollutions or rare human events with significant socioeconomic impacts.

1. Introduction

Anthropogenic emissions have imposed essential influences on the earth system, from hourly air quality and human health to long-time climate and environment. To reduce anthropogenic emissions, the Chinese government has enforced the Clean Air Action (2013) since 2013. Studies to date that evaluated the emission controls and understood the climate responses from emission reductions often have used either a fixed meteorology with emission changes or *vice versa* (Li et al., 2019a; Li et al., 2021, Zhai et al., 2021). Estimated emissions from empirical extrapolation were commonly applied to analyze the meteorological-chemical mechanisms and associated social-economic impacts from occasional events like the 2015 China Victory Day Parade and Coronavirus Disease 2019 (COVID-19) pandemic (Wang et al., 2017; Liu et al., 2020; Huang et al., 2020; Zhu et al., 2021). But to better understand both long-term and short-term influences from emission changes, the continuous, up-to-date, and high temporal-/spatial-resolution emission estimates with coherent interactions of meteorology and emission changes are needed.

The complex contributions from energy production, industrial processes, transportation, and residential consumptions have imposed great challenges to accurately estimate the emissions. The emission inventories created by the traditional bottom-up techniques were typically outdated from the present day due to the lack of accurate and timely statistics, and often with coarse temporal resolutions from monthly to annual (Zhang et al., 2009; Li et al., 2014; Janssens-Maenhout et al., 2015; Zheng et al., 2018). Alternatively, update-to-date emission estimates with high temporal-spatial resolutions could be provided by top-down techniques (Miyazaki et al., 2017), but most emissions estimated by top-down techniques were intermittent and analyzed at monthly scale or longer longer (Zhang et al., 2016; Jiang et al., 2017; Qu et al., 2017; Cao et al., 2018; Müller et al., 2018; Chen et al., 2019; Li et al., 2019b; Miyazaki et al., 2020). Moreover, emissions updated by the top-down techniques based on satellite observations could be insufficient to capture realistic near-surface characteristics (Li et al., 2019b; Liu et al., 2011; Choi et al., 2020).

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Given the development of observation networks and advanced data assimilation strategies, timely and dynamics-based emission estimates with high temporal resolution can be achieved by harmonically constraining the atmospheric-chemical model with dense observations of trace gas compounds through an optimal assimilation methodology. The ensemble Kaman smoother (EnKS) (Whitaker et al., 2002; Peters et al., 2007; Peng et al., 2015), as a four-dimensional (4D) assimilation algorithm, makes use of chemical observations from past to future to provide an optimal estimate of source emissions, and it can capture the "error of the day" and construct fine emission characteristics with high temporal-spatial resolutions by using short-term ensemble forecasts (Kalnay, 2002). Since 2013, the fine particulate matter pollution ($PM_{2.5}$, particles smaller than 2.5 μm in diameter) as the most urgent threat to public health has been persistently decreased, and ground-based observations of $PM_{2.5}$ have been progressively increased (Huang et al., 2018). Thus by harmonically assimilating dense surface $PM_{2.5}$ observations into an atmospheric-chemical model through an EnKS, hourly estimates of $PM_{2.5}$ emission that were continuously cycled for years 2016-2020 are presented in this study.

The timely estimated emissions can provide guidance for emission inventories that usually have time lags and emission trading schemes that often require up-to-date source emissions. Based on the dynamics-based estimated emissions with harmonic combination of the model and observations, better evaluation of the emission controls and more comprehensive understanding of the consequent climate responses can be obtained. The high temporal-resolution estimated emissions can reveal features of emissions that are absent from the traditional ones with coarse temporal resolutions. Moreover, the timely and dynamics-based emission estimates with high temporal resolution are essential for regional air quality modeling, especially for the occurrence of severe haze pollutions associated with timely evaluation for the impact on public health (Attri et al., 2001; Wang et al., 2014; Ji et al., 2018; Wang et al., 2020; Liu et al., 2021) and events that lead to large changes of emissions and significant socioeconomic impacts such as the COVID-19 pandemic (Huang et al., 2020; Le et al., 2020).

2. Data assimilation and experimental design

The estimate of $PM_{2.5}$ emission can be successfully constrained by the $PM_{2.5}$ concentration observations through an ensemble Kalman filter (EnKF; Peng et al., 2017, 2018, 2020). For a retrospective 'reanalysis' mode here, all available $PM_{2.5}$ concentration observations, including

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those data collected after the analysis time, can be used. Thus a EnKS, a direct generalization of the EnKF, is applied to incorporate $PM_{2.5}$ concentration observations both before and after the analysis time, aiming to provide an optimal estimate of the $PM_{2.5}$ emission. Detailed procedures of the EnKS are described in section 2.1.

2.1 An ensemble Kalman smoother to update the source emission

The ensemble priors of source emissions $\mathbf{e}^f$ is created by multiplying a scaling factor $\boldsymbol{\lambda}^f$ to the prescribed emission $\mathbf{e}^{\mathrm{P}}$ (Peng et al., 2017, 2018, 2020), where the superscript f denotes priors. Given a constant $\mathbf{e}^{\mathrm{P}}$, the update of $\mathbf{e}^f$ is equivalent to the update of $\boldsymbol{\lambda}^f$. Due to a time lag, the prior scaling factor at time t-1 ($\boldsymbol{\lambda}^f_{t-1}$) is updated by chemical observations at time t ($\mathbf{y}^c_t$). At time t-1, the prior scaling factor for the i^{th} member is written as

$$\boldsymbol{\lambda}^f_{i,t-1} = \frac{1}{M}\left[\left(\beta\frac{\mathbf{c}^f_{i,t-1}}{\overline{\mathbf{c}}^f_{t-1}} + 1 - \beta\right) + \sum_{j=t-M}^{t-2}\boldsymbol{\lambda}^a_{i,j|j+1:t-1}\right]. \quad (1)$$

The first term is the concentration ratio given by the prior of the chemical fields ($\mathbf{c}^f_{i,t-1}$) normalized by the ensemble mean ($\overline{\mathbf{c}}^f_{t-1}$), where β is an inflation factor used to compensate the insufficient ensemble spread (Peng et al., 2017). Through using the concentration ratio, each ensemble member of the source emissions naturally has the spatial correlations given by the chemical fields. The second term is the mean of the posterior scaling factors at previous assimilation cycles, where the superscript a denotes posteriors, M is the length of smoothing, and the subscript j+1:t-1 indicates that the scaling factor at time j is updated by future observations from j+1 to t-1. The assimilation of future observations will be described below.

The ensemble square-root filter (EnSRF) (Peng et al., 2017) is used to update $\boldsymbol{\lambda}^f_{t-1}$ by assimilating $\mathbf{y}^c_t$. For the scaling factor at time t-1, posterior ensemble mean is given by

$$\overline{\boldsymbol{\lambda}}^a_{t-1} = \overline{\boldsymbol{\lambda}}^f_{t-1} + \rho \circ \mathbf{P}^{ec}_{t-1,t}\mathbf{H}_t^{c\mathbf{T}}\left(\mathbf{H}^c_t\mathbf{P}^c_t\mathbf{H}_t^{c\mathbf{T}} + \mathbf{R}^c_t\right)^{-1}\left(\mathbf{y}^c_t - H^c_t\overline{\mathbf{c}}^f_t\right), \quad (2)$$

and posterior ensemble perturbations are given by

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$$\boldsymbol{\lambda}_{i,t-1}^{'a} = \boldsymbol{\lambda}_{i,t-1}^{'f} - \rho \circ \mathbf{P}_{t-1,t}^{ec} \mathbf{H}_t^{c\mathbf{T}} \left[\left(\sqrt{\mathbf{H}_t^c \mathbf{P}_t^c \mathbf{H}_t^{c\mathbf{T}} + \mathbf{R}_t^c} \right)^{-1} \right]^{\mathbf{T}} \left[\sqrt{\left(\mathbf{H}_t^c \mathbf{P}_t^c \mathbf{H}_t^{c\mathbf{T}} + \mathbf{R}_t^c \right)} + \sqrt{\mathbf{R}_t^c} \right]^{-1} \mathbf{H}_t^c \boldsymbol{\lambda}_{i,t-1}^{'f} \quad , \tag{3}$$

where $\mathbf{P}_{t-1,t}^{ec}$ denotes the background error covariance matrix of $\boldsymbol{\lambda}_{t-1}^{f}$ and $\mathbf{c}_t^f$, $\mathbf{P}_t^c$ indicates the background error covariance matrix of $\mathbf{c}_t^f$, H_t^c, $\mathbf{H}_t^c$ and $\mathbf{R}_t^c$ are the observation forward operator, Jacobian matrix and observation error covariance matrix of the chemical fields at time t, ρ is the localization matrix and $\circ$ denotes the Schur (elementwise) product.

By applying the ensemble Kalman smoother (EnKS) (Whitaker et al., 2002; Peters et al., 2007), the chemical observation $\mathbf{y}_t^c$ is also assimilated to update the posterior scaling factor at previous assimilation cycles $j\left(j = t-K,\ldots,t-2\right)$. After assimilating the future chemical observation at time t, posterior ensemble mean of the scaling factor at j is given by

$$\overline{\boldsymbol{\lambda}}_{j|j+1:t}^{a} = \overline{\boldsymbol{\lambda}}_{j|j+1:t-1}^{a} + \rho \circ \mathbf{P}_{j|j+1:t-1,t}^{ec} \mathbf{H}_t^{c\mathbf{T}} \left(\mathbf{H}_t^c \mathbf{P}_t^c \mathbf{H}_t^{c\mathbf{T}} + \mathbf{R}_t^c \right)^{-1} \left(\mathbf{y}_t^c - H_t^c \overline{\mathbf{c}}_t^f \right), \tag{4}$$

and posterior ensemble perturbations are given by

$$\boldsymbol{\lambda}_{i,j|j+1:t}^{'a} = \boldsymbol{\lambda}_{i,j|j+1:t-1}^{'a} - \rho \circ \mathbf{P}_{j|j+1:t-1,t}^{ec} \mathbf{H}_t^{c\mathbf{T}} \left[\left(\sqrt{\mathbf{H}_t^c \mathbf{P}_t^c \mathbf{H}_t^{c\mathbf{T}} + \mathbf{R}_t^c} \right)^{-1} \right]^{\mathbf{T}} \left[\sqrt{\left(\mathbf{H}_t^c \mathbf{P}_t^c \mathbf{H}_t^{c\mathbf{T}} + \mathbf{R}_t^c \right)} + \sqrt{\mathbf{R}_t^c} \right]^{-1} \mathbf{H}_t^c \boldsymbol{\lambda}_{i,t-1}^{'f}, \tag{5}$$

where $\mathbf{P}_{j|j+1:t-1,t}^{ec}$ denotes the background error covariance matrix of $\boldsymbol{\lambda}_{j|j+1:t-1}^{a}$ and $\mathbf{c}_t^f$. After (2)-(5), the updated $\boldsymbol{\lambda}_{j|j+1:t}^{a}, j\left(j = t-M+1,\ldots,t-1\right)$ will be used to construct the prior scaling factor at next time t+1 (1).

As a Monte Carlo approach, the EnKS uses the forecast-analysis error covariances based on ensemble forecasts / analyses to compute the Kalman gain matrix with time lags, to incorporate observations from the past to the future. The first iteration of EnKS is equivalent to EnKF that assimilates observations up to the analysis time. The following iterations of EnKS assimilate observations in the future to update the state at the analysis time. The hourly forecasts of $PM_{2.5}$ concentration from the cycling assimilation experiment matched the independent observed

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quantities (Figure 1). Therefore, the ability of EnKS to retrieve the source emissions has been demonstrated.

2.2 WRF-Chem model, observations and emissions

To simulate the transport of aerosol and chemical species, the WRF-Chem model version 3.6.1 (Grell et al., 2005) that has the meteorological and chemical components fully coupled is used. The model parameterization schemes follow Peng et al. (2017). Figure 2 shows the model domain that covers most east Asia regions. Horizontal grid spacing is 45 km with 57 vertical levels and model top at 10 hPa.

Experiments are conducted for each year from 2016 to 2020 separately. The 6-h meteorological observations, including all in-situ observations and cloud motion vectors from the National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS; http://www.emc.ncep.noaa.gov/mmb/data_processing/prepbufr.doc/table_2.htm), are assimilated every 6 h. The hourly observed chemical quantities, which contain PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3, and CO from the Ministry of Ecology and Environment of China (https://aqicn.org/map/china/cn/), are assimilated every hour. Figure 2 shows the assimilated chemical observation network, which has 560 randomly chosen stations from 1576 stations in total. The thinning of observations is applied to avoid correlated errors of observations (Peng et al., 2017). The observation priors are computed by the "observer" portion of the Grid-point Statistical Interpolation system (GSI) (Kleist et al., 2009).

The hourly and constantly prescribed anthropogenic emissions are obtained from the EDGAR-HTAP (Emission Database for Global Atmospheric Research for Hemispheric Transport of Air Pollution v2.2) v2.2 inventory (Janssens-Maenhout et al., 2015), in which the Chinese emissions are derived from MEIC in 2010 (Lei et al., 2011; Li et al., 2014). Natural emissions, including the biogenic (Guenther et al., 1995), dust (Ginoux et al., 2001), dimethyl sulfide and sea salt emissions (Chin et al., 2000), are computed online.

2.3 Assimilation and ensemble configurations

The $PM_{2.5}$ emission directly gives the primary $PM_{2.5}$, and then the primary $PM_{2.5}$ along with other precursor emissions could contribute to the secondary $PM_{2.5}$. The observations of $PM_{2.5}$ concentrations that contain both primary and secondary $PM_{2.5}$, are used to constrain the $PM_{2.5}$

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emission through data assimilation. Thus the correlations between the concentration observations and source emissions might be contaminated by the secondary $PM_{2.5}$. In the presnt study, the impact of the secondary $PM_{2.5}$ is ignored. One possible way to untangle the impact of secondary $PM_{2.5}$ on the estimates of $PM_{2.5}$ emission is to jointly estimate the source emission, primary and secondary $PM_{2.5}$ given the concentration observations.

The National Oceanic and Atmospheric Administration (NOAA) operational EnKF system (https://dtcenter.ucar.edu/com-GSI/users/docs/users_guide/GSIUserGuide_v3.7.pdf), which is an EnSRF and modified with the EnKS feature, is used to assimilate the observations. Ensemble size is set to 50. To combat the sampling error resulted from a limited ensemble size, covariance localization and inflation are applied. The Gaspari and Cohn (GC) (1999) function with a length scale of 675 km is used to localize the impact of observations and mitigate the spurious error correlations between observations and state variables. The constant multiplicative posterior inflation (Whitaker and Hamill 2012) with coefficients 1.12 for all meteorological and chemical variables is applied to enlarge the ensemble spread. The inflation β for advancing the scale factor is 1.2. The smoothing length M for source emissions is 4, and the EnKS lagged length K is 6.

At 0000 UTC 26 December of previous year, ensemble initial conditions (ICs) of the meteorological fields are generated by adding random perturbations that sample the static background error covariances (Barker et al., 2012) on the NCEP FNL (Final) analyses (Torn et al., 2006). Ensemble ICs of the chemical fields are 0, and source emissions of each ensemble member are adopted from the EDGAR-HTAP v2.2 inventory with random perturbations of mean 0 and variances of 10% of the emission values. Hourly ensemble lateral boundary conditions (LBCs) are generated using the same fixed-covariance perturbation technique as the ensemble ICs. After 6-d spin up, ensemble data assimilation experiments start cycling for each year.

3. $PM_{2.5}$ emission for years 2016-2020

Starting from the constant source emission PR2010 (Janssens-Maenhout et al., 2015), the annual dynamics-based estimates of $PM_{2.5}$ emission (DEPE) averaged over mainland China for years 2016-2020 are 8.17, 7.91, 7.53, 7.13 and 6.89 Tg, respectively. For years 2016 and 2017, the annual DEPE are very closed to 8.1 and 7.6 Tg from the Multi-resolution Emission Inventory (MEIC) (Zheng et al., 2018). From year 2017 to 2020, the estimated annual $PM_{2.5}$ emissions are reduced 3.2%, 7.8%, 12.7% and 15.7% respectively compared to that of year 2016. There has been

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3%-5% persistent reduction of annual $PM_{2.5}$ emission from year 2017 to 2020, which demonstrates the effectiveness of China's Clean Air Action (2013) implemented since 2013 and China Blue Sky Defense War Plan (2018) enforced since 2018 with strengthened industrial emission standards, phased out outdated industrial capacities, promoted clean fuels in residential sector and so on (Zhang et al., 2019).

The monthly DEPE show reduction of $PM_{2.5}$ emission nearly in each month from years 2016 to 2020 (Figure 3a), which further demonstates the effectiveness of China's national plan, rather than the role of weather effects alone. Compared to year 2016, both the reduction amount and reduction ratio of $PM_{2.5}$ emission are more prominent for February, March, June-September, and November than the other months (Figure3b). Given larger magnitudes of $PM_{2.5}$ emission in winter than in summer, emission controls with a focus from October to May should be considered in the design of future clean air actions in China, since total $PM_{2.5}$ emission during this period accounts for approximate 75% annual amount. Spatial distributions of the changes of $PM_{2.5}$ emission from year 2017 to 2020 compared to year 2016 show significant decreases occurred at Beijing-Tianjin-Hebei region (BTH), Yangtze River Delta region (YRD), Pearl River Delta region (PRD) and Sichuan-Chongqing Region (SCR), especially for years 2019-2020 (Figure 4). From year 2016 to 2020, BTH, YRD and SRC have larger reductions of $PM_{2.5}$ emission than PRD, but SCR has larger reduction ratio compared to year 2016 than BTH and YRD (Figure 5). Therefore, BTH and YRD have more potentials for $PM_{2.5}$ emission controls than PRD and SCR, which can give a guidance for future clean air actions. More specifically, most provinces have $PM_{2.5}$ emission reduction from year 2016 to 2020, and the reduction ratios generally increase from year 2017 to 2020 (Table 1), which confirms continuous and effective emission controls from Clean Air Action to Blue Sky Defense War Plan in China.

Despites the trend in $PM_{2.5}$ emissions from year 2016 to 2020, the DEPE of year 2016 has similar monthly distributions to MEIC2016 in general (Figure 3a). MEIC2016 has a "Pan-shape" monthly distribution with nearly constant $PM_{2.5}$ emissions from April to October. This seasonal dependence of emissions is mainly contributed by the variations of residential energy use, which are empirically dependent on coarse monthly mean temperature intervals and thus cannot reflect the realistic monthly variations (Streets et al., 2003; Li et al., 2017). However, the DEPE yet shows a "V-shape" monthly distribution, with the minimum occurring in August. The estimated $PM_{2.5}$ emission is 11.8% higher than MEIC2016 in April but 12.1% lower than MEIC2016 in August,

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and these different monthly distributions can influence the consequent climate responses including the radiative forcing and energy budget (Yang et al., 2020) and also impact the health issues (Liu et al., 2018). Moreover, monthly fractions of the DEPE are consistent cross years (Figure 3c). The absence of interannual variations of monthly $PM_{2.5}$ emission fraction provides basis for previous studies that follow the same monthly changes of source emissions from different years (Zhang et al., 2009; Zheng et al., 2020, 2021). Monthly allocations of $PM_{2.5}$ emission can be directly and objectively obtained given an estimated total annual amount based on the estimated monthly fractions of DEPE, which is valuable for emission inventory, air quality simulation, and potentially applications for future scenarios due to more accurate month fractions of DEPE. Since the hourly priors of $PM_{2.5}$ concentrations from the cycling assimilation for optimally estimating $PM_{2.5}$ emission fit to the observed $PM_{2.5}$ quantities (Figure 1), the monthly DEPE provides more realistic monthly fluctuations than the empirical estimate.

4. Diurnal variations of $PM_{2.5}$ emission

The DEPE with high temporal-resolution given the constant prior PR2010 can reveal features that are unable to represent in the commonly used emission estimates. Although the prior PR2010 has no diurnal variations, hourly posteriors of $PM_{2.5}$ emission provide the first objectively estimated diurnal variations for different seasons for years 2016-2020. To statistically present the diurnal variations, the fractions of hourly $PM_{2.5}$ emissions divided by the daily amount are averaged over different years and regions (Figures 6 and 7, and Table 2). The diurnal variations of $PM_{2.5}$ emission are critical for understanding the mechanisms of $PM_{2.5}$ formation and evolution and are also essential for $PM_{2.5}$ simulation and forecast.

Five-year mean diurnal variations of the estimated $PM_{2.5}$ emission fraction for mainland China show that despite the monthly variations of $PM_{2.5}$ emission, the diurnal-variation fractions for November, December, January and February are similar, while those for June, July and August are similar (Figure 6a). There are stronger diurnal variations of $PM_{2.5}$ emission in summer than in winter, which are represented by larger $PM_{2.5}$ emission fractions during morning and less $PM_{2.5}$ emission fractions during evening. The diurnal variations of $PM_{2.5}$ emission from March to May gradually transform from the patterns of winter to those of summer, and *vice versa* for the diurnal variations of $PM_{2.5}$ emission from September to November. The monthly changes of diurnal variations of $PM_{2.5}$ emission are consistent with the seasonal dependence, since monthly variations

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of $PM_{2.5}$ emission are mainly related to the variations of residential consumptions (Li et al., 2017) in which the space-heating has nearly no diurnal variations and then larger $PM_{2.5}$ emissions during winter lead to reduced diurnal variations than summer. Similar to the monthly fractions of estimated $PM_{2.5}$ emission for mainland China, diurnal variations of $PM_{2.5}$ emission fraction are consistent cross years for a given month (Figure 7). Table 2 gives five-year mean diurnal variations of the estimated $PM_{2.5}$ emission fraction for each month. Based on these high-resolution diurnal-variation fractions, hourly estimates of $PM_{2.5}$ emission can be objectively obtained for a given monthly estimated $PM_{2.5}$ emission.

Despite the high temporal resolution, the DEPE also has the ability to analyze diurnal variations for specific cities. The monthly changes of diurnal variations of $PM_{2.5}$ emission estimated for megacities with urban populations larger than 5 million and non-megacities with urban populations smaller than 5 million (Notice of the State Council on Adjusting the Standards for Categorizing City Sizes, 2014) are consistent with those estimated from mainland China (Figure 6). Compared to the diurnal variations of $PM_{2.5}$ emission estimated for mainland China, the megacities have stronger diurnal variations, while the non-megacities have weaker diurnal variations. These detailed descriptions of $PM_{2.5}$ emission that are usually absent in common emission estimates can be essential for $PM_{2.5}$ simulation, especially for providing timely and realistic guidance for severe haze events.

There has been lack of local measurements for diurnal variations and widely adopted diurnal variation profiles of $PM_{2.5}$ emission in China. Compared to the diurnal variations of $PM_{2.5}$ emission fractions estimated based on diurnal variation profiles from US and EU (Wang et al., 2010), the noon and evening peaks estimated from DEPE have smaller $PM_{2.5}$ emission fractions, with mean underestimations of $PM_{2.5}$ emission fraction of 0.31% and 1.05% for noon peak and evening peak respectively (Figures 6a and 8). The morning peak of Wang et al. (2010) is similar to that of DEPE for spring and fall, but the former overestimates $PM_{2.5}$ emission fraction of 0.81% for winter while underestimates $PM_{2.5}$ emission fraction of 0.79% for summer. Due to the overestimated peaks, diurnal variations of Wang et al.(2010) have sharper appearance rate for morning peak and disappearance rate for evening peak. Compared to the diurnal variations based on diurnal variation profiles from ES and EU (Wang et al., 2010), the diurnal variations of the DEPE are constrained by the atmospheric-chemical model and observed $PM_{2.5}$ concentrations,

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which can objectively determine the diurnal variations of $PM_{2.5}$ emission for specific regions and seasons.

5. Impact of COVID-19 on $PM_{2.5}$ emissions

The abrupt outbreak of the COVID-19 pandemic has produced dramatically socioeconomic impacts in China. To prevent the virus spread, a lockdown was first implemented on 23 January 2020 in Wuhan, Hubei province, and subsequently the national lockdown has been enforced in China (Liu et al., 2020; Huang et al., 2020; Zhu et al., 2021). Consequently, the total $PM_{2.5}$ emission of February 2020 for China shows an obvious decrease compared to those of previous years (Figure 3). The high temporal-resolution DEPE reveals the detailed changes of $PM_{2.5}$ emission with time (Figure 9). The $PM_{2.5}$ emission started to decrease right around the COVID outbreak, and had been smaller than those of year 2019 till early March. During February 2020, the DEPE shows significant reductions at the north China plain and northeast of China where prominent $PM_{2.5}$ emission occurred, while spotted $PM_{2.5}$ emission differences with small magnitudes showed at the other regions (Figures 10a-b). Along with recovery from the COVID-19, the estimated $PM_{2.5}$ emission rebounded in March (Figures 3a, 9, 10c-d), which is contributed to the national work resumption. Thus, the DEPE is able to timely reflect the dynamic response of $PM_{2.5}$ emission to the COVID-19.

To avoid fluctuations due to diurnal variations and monthly changes of $PM_{2.5}$ emission, 7-day averaged $PM_{2.5}$ emission differences between year 2020 and 2019 are used to analyze the dynamic impact of COVID-19 on $PM_{2.5}$ emission (Figure 11). Before the lockdown, there were slight $PM_{2.5}$ emission differences over several provinces (Figures 11a-b). During the first week of lockdown, $PM_{2.5}$ emission reduction larger than $5x10^{-2}$ ($\mu g \cdot m^{-2} \cdot s^{-1}$) that is about 60%-70% emission reduction, occurred at Hubei, Hunan, Guangdong, Anhui and Zhejiang provinces (Figure 11c). The $PM_{2.5}$ emission reduction extended to BTH and Shandong province during the second week of lockdown (Figure 11d), and continuously spread to the three northeast provinces of China during the third week of lockdown (Figure 11e). During the third week of lockdown, the increased $PM_{2.5}$ emissions for BTH and SCR are possibly caused by the massive emissions from high-profile firework burning on the Chinese New Year Eve of year 2019 (Ji et al., 2018). The $PM_{2.5}$ emission reduction had been maintained over the central and northern China till early March when the

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lockdown was lift (Figures 11f-i). Thus, the timely DEPE can provide up-to-date guidance for quantifying

6. Discussion

High temporal-resolution and dynamics-based estimations of $PM_{2.5}$ emission can be objectively and optimally obtained by assimilating past and future observed surface $PM_{2.5}$ concentrations through flow-dependent error statistics. This advanced assimilation strategy can be applied for emission estimates of other chemical species when corresponding observations are available, and extend to observation types besides the surface concentrations, like the aerosol optical depth (Liu et al., 2011; Choi et al., 2020). Moreover, current estimates of $PM_{2.5}$ emission are lack of explicitly representations of primary and secondary $PM_{2.5}$, which could be resolved by joint estimation of the source emission, primary and secondary $PM_{2.5}$ given the concentration observations. Another deficiency of this top-down technique is that it cannot directly determine dynamics-based $PM_{2.5}$ emissions for different sectors as the bottom-up techniques. But this top-down technique can be integrated into the bottom-up technique to retain advantages of both methods. The annual emission estimate from the bottom-up technique can be further downscaled to hourly estimates by first distributing the annual amount to each month through the monthly allocations estimated from the top-down technique, and then assuming evenly daily distribution, finally applying the fractions of diurnal variation estimated from the top-down technique. The information collected by the bottom-up technique is retained, while the common drawback of coarse temporal resolution for the bottom-up technique is remedied. The integrated bottom-up and top-down technique can improve spatiotemporal representations of source emissions cross time scales and sectors, which is beneficial for emission inventory, air quality forecast, regulation policy and emission trading scheme.

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Data availability

The meteorological data used for meteorological initial conditions and boundary conditions is available from the University Corporation for Atmospheric Research (UCAR) Research Data Archive (https://rda.ucar.edu/datasets/ds083.3/). The assimilated meteorological observations are available from the UCAR Research Data Archive (https://rda.ucar.edu/datasets/ds337.0/), and the assimilated chemical observations are available from https://aqicn.org/map/china/cn/. The prescribed constant anthropogenic emissions are available from the Emission Database for Global Atmospheric Research for Hemispheric Transport of Air Pollution (EDGAR-HTAP) inventory (https://data.jrc.ec.europa.eu/dataset/jrc-edgar-htap_v2-2) and the Multi-resolution Emission Inventory (MEIC; http://meicmodel.org/?page_id=560).

The WRF-Chem model version 3.6.1 is available from https://www2.mmm.ucar.edu/wrf/users/download/get_sources.html#WRF-Chem. The NOAA operational EnKF system is available from https://dtcenter.org/community-code/gridpoint-statistical-interpolation-gsi.

Competing interests

The contact author has declared that none of the authors has any competing interests.

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Figures and Tables

Captions:

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Figure 1. Times series of hourly $PM_{2.5}$ concentration biases ($\mu g \cdot m^{-3}$). The ensemble mean priors compared to the observed quantities for December of years 2016-2020 (gray and black), and the mean biases of years 2016-2020 (blue).

Figure 2. Model domain and observation sites for cycling assimilation. Red and blue dots denote the assimilated and unassimilated observational sites, respectively.

Figure 3. (a) Dynamics-based monthly $PM_{2.5}$ emission estimates ($Tg \cdot day^{-1}$) summed over mainland China of each year from 2016 to 2020 (colored) and the estimated $PM_{2.5}$ emission from MEIC (gray); (b) Ratio of $PM_{2.5}$ emission changes between two adjacent years from year 2016 to 2020 normalized by the $PM_{2.5}$ emission of year 2016; (c) Monthly fractions of dynamics-based $PM_{2.5}$ emission estimates for years 2016-2020 (light blue), the five-year mean fractions of dynamics-based monthly $PM_{2.5}$ emission estimates with bars denoting one standard deviation of the five-year variations (dark blue), and the monthly fractions of estimated $PM_{2.5}$ emission from MEIC (gray).

Figure 4. (a) Spatial distribution of dynamics-based $PM_{2.5}$ emission estimates ($\mu g \cdot m^{-2} \cdot s^{-1}$) for year 2016, and compared to that of year 2016, spatial distributions of dynamics-based $PM_{2.5}$ emission changes of year (b) 2017, (c) 2018, (d) 2019 and (e) 2020.

Figure 5. (a) The differences of dynamics-based $PM_{2.5}$ emission estimates between years 2017-2020 and 2016, and (b) the differences normalized by that of year 2016.

Figure 6. Five-year mean diurnal variations of dynamics-based $PM_{2.5}$ emission fraction averaged over (a) mainland China, (b) megacities with urban population $\geq$ 5 million, and (c) non-megacities with urban population < 5 million.

Figure 7. Diurnal variations of dynamics-based $PM_{2.5}$ emission fractions for years 2016-2020 (light blue) and five-year mean fractions with bars denoting one standard deviation of the five-year variations (dark blue) are averaged over mainland China for (a) January, (b) April, (c) July, and (d) October.

Figure 8. Diurnal variations of $PM_{2.5}$ emission fraction for each month based on diurnal variation profiles from ES and EU (Wang et al. 2010).

Figure 9. Hourly (light red and blue) and daily (dark red and blue) dynamics-based $PM_{2.5}$ emission estimates ($kg \cdot h^{-1}$) summed over mainland China from January to March of years 2019 and 2020.

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Figure 10. Spatial distributions of dynamics-based $PM_{2.5}$ emission estimates ($\mu g \cdot m^{-2} \cdot s^{-1}$) on (b) February and (d) March of year 2019, and spatial distributions of dynamics-based $PM_{2.5}$ emission reduction of year 2020 compared to year 2019 for (c) February and (e) March.

Figure 11. Mean spatial distributions of $PM_{2.5}$ emission differences ($\mu g \cdot m^{-2} \cdot s^{-1}$) between year 2020 and 2019 for 9 weeks starting at 9 January 2020. Negative (positive) values indicate that $PM_{2.5}$ emission of year 2020 is smaller (larger) than that of year 2019. The numbers in (a) denote provinces as: 1 Heilongjiang, 2 Neimenggu, 3 Xinjiang, 4 Jilin, 5 Liaoning, 6 Gansu, 7 Hebei, 8 Beijing, 9 Shanxi, 10 Tianjin, 11 Shanxi, 12 Ningxia, 13 Qinghai, 14 Shandong, 15 Xizang, 16 Henan, 17 Jiangsu, 18 Anhui, 19 Sichuan, 20 Hubei, 21 Chongqing, 22 Shanghai, 23 Zhejiang, 24 Hunan, 25 Jiangxi, 26 Yunnan, 27 Guizhou, 28 Fujian, 29 Guangxi, 30 Guangdong, 31 Taiwan, 32 Hongkong, 33 Macao, 34 Hainan.

Table 1. Dynamics-based $PM_{2.5}$ emission estimates of year 2016 for each province whose value is larger than 0.01 $\mu g \cdot m^{-2} \cdot s^{-1}$ are shown in the second column. Ratios of $PM_{2.5}$ emission changes of years 2017-2020 compared to year 2016 are shown from the third to the sixth column, with negative (positive) values indicating decrease (increase) of $PM_{2.5}$ emission.

Table 2. Five-year mean diurnal fractions (%) of the dynamics-based $PM_{2.5}$ emission estimates over mainland China on local solar time (LST) for each month.

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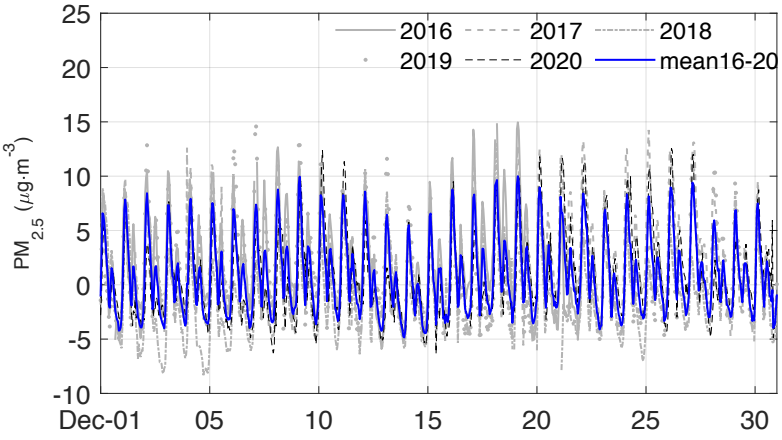

Figure 1. Times series of hourly $PM_{2.5}$ concentration biases ($\mu g \cdot m^{-3}$). The ensemble mean priors compared to the observed quantities for December of years 2016-2020 (gray and black), and the mean biases of years 2016-2020 (blue).

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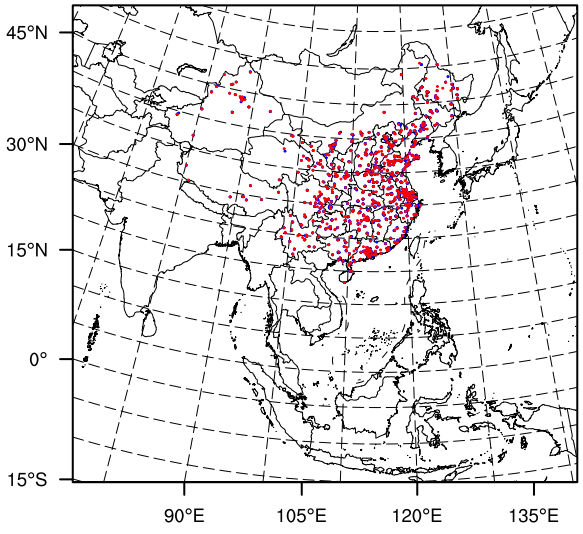

Figure 2. Model domain and observation sites for cycling assimilation. Red and blue dots denote the assimilated and unassimilated observational sites, respectively.

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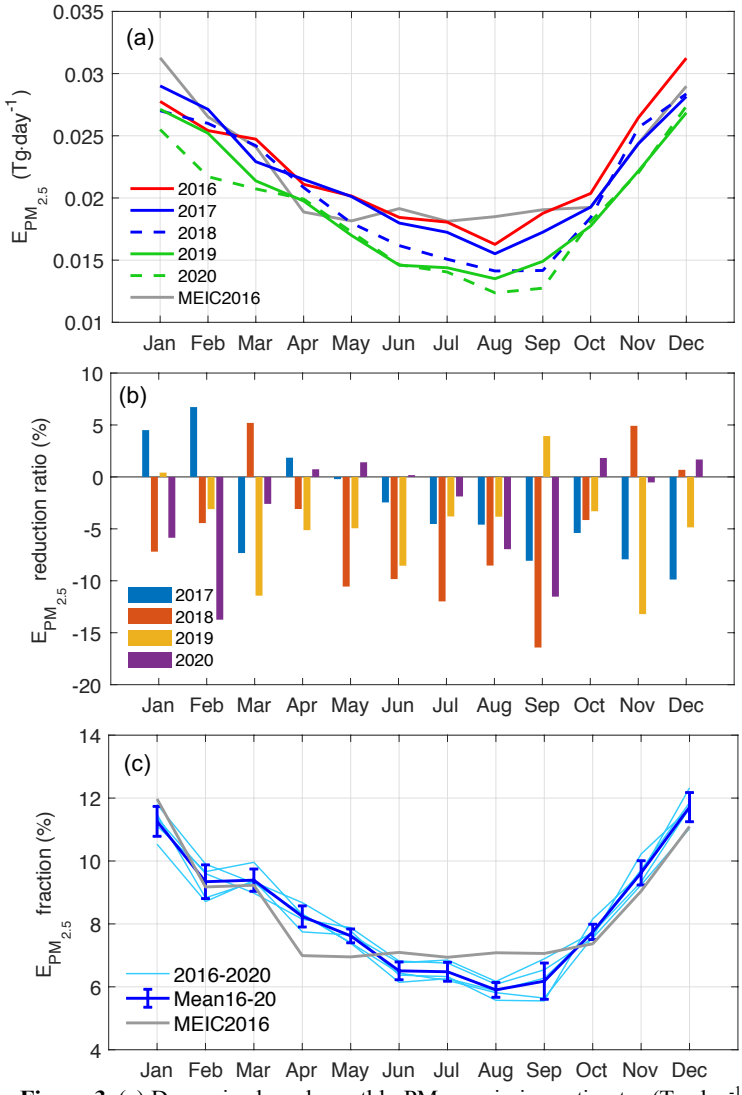

Figure 3. (a) Dynamics-based monthly $PM_{2.5}$ emission estimates (Tg·day^{-1}) summed over mainland China of each year from 2016 to 2020 (colored) and the estimated $PM_{2.5}$ emission from MEIC (gray); (b) Ratio of $PM_{2.5}$ emission changes between two adjacent years from year 2016 to 2020 normalized by the $PM_{2.5}$ emission of year 2016; (c) Monthly fractions of dynamics-based $PM_{2.5}$ emission estimates for years 2016-2020 (light blue), the five-year mean fractions of dynamics-based monthly $PM_{2.5}$ emission estimates with bars denoting one standard deviation of the five-year variations (dark blue), and the monthly fractions of estimated $PM_{2.5}$ emission from MEIC (gray).

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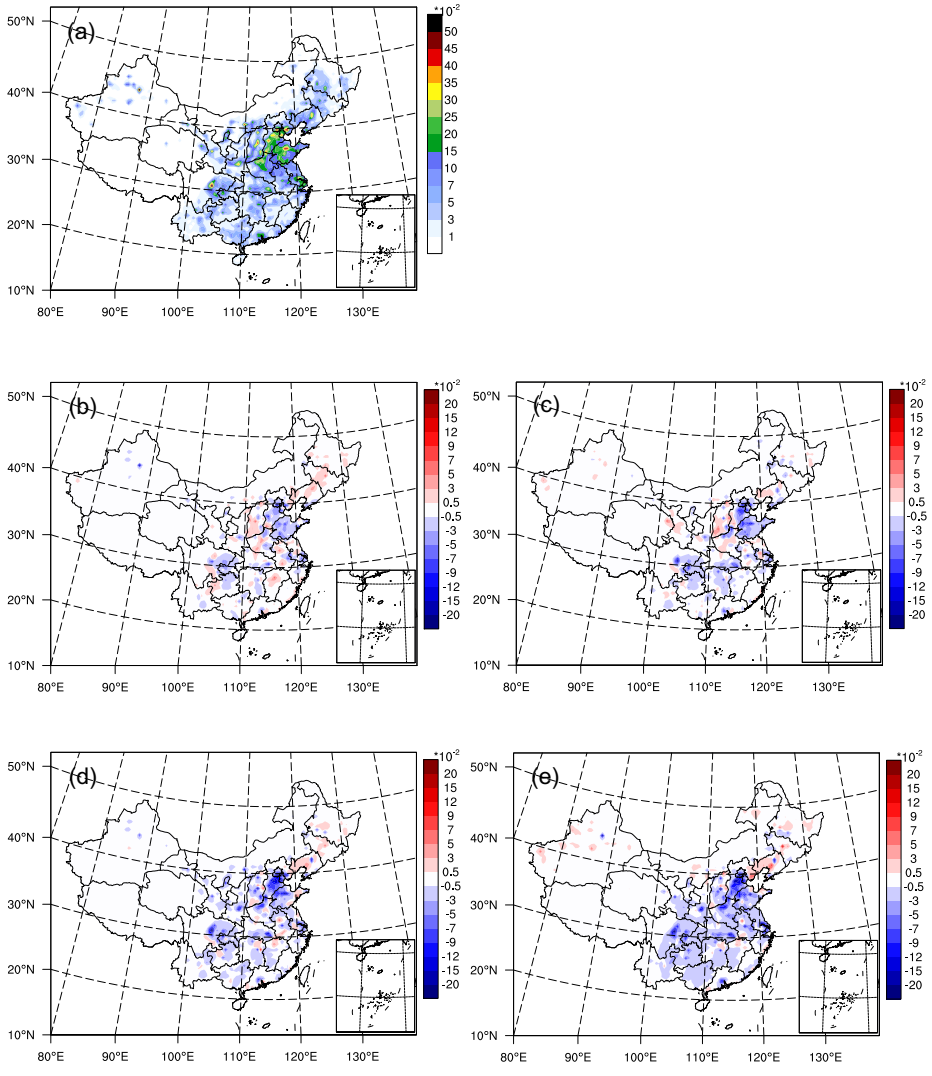

Figure 4. (a) Spatial distribution of dynamics-based $PM_{2.5}$ emission estimates ($\mu g \cdot m^{-2} \cdot s^{-1}$) for year 2016, and compared to that of year 2016, spatial distributions of dynamics-based $PM_{2.5}$ emission changes of year (b) 2017, (c) 2018, (d) 2019 and (e) 2020.

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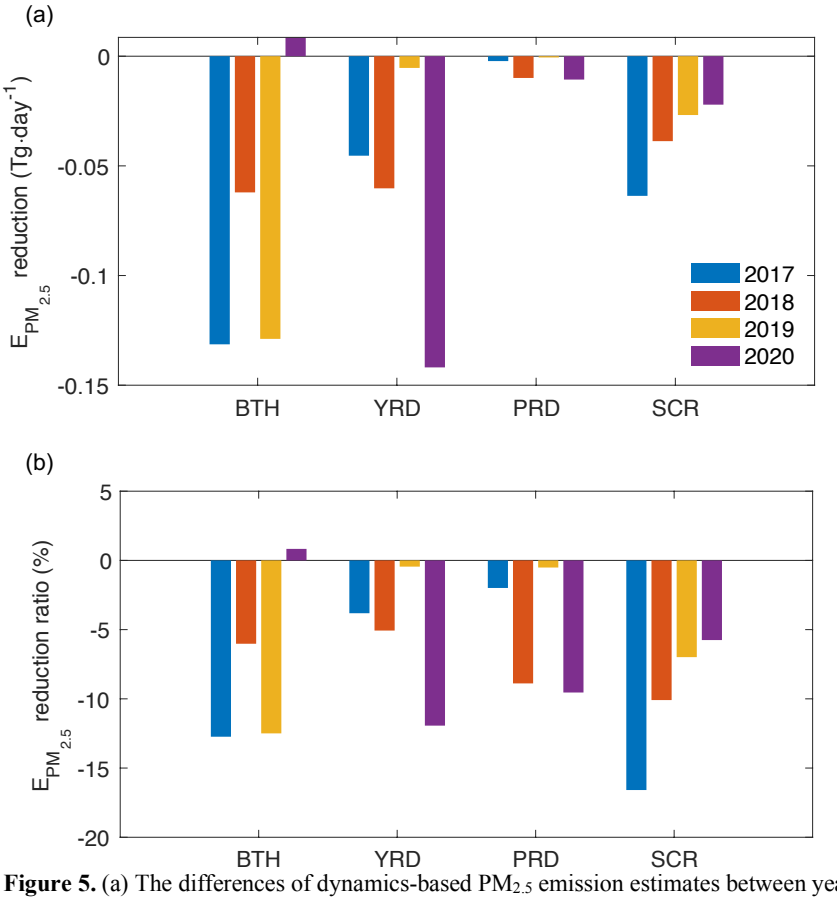

624
625 **Figure 5.** (a) The differences of dynamics-based $PM_{2.5}$ emission estimates between years 2017-2020 and 2016,
626 and (b) the differences normalized by that of year 2016.
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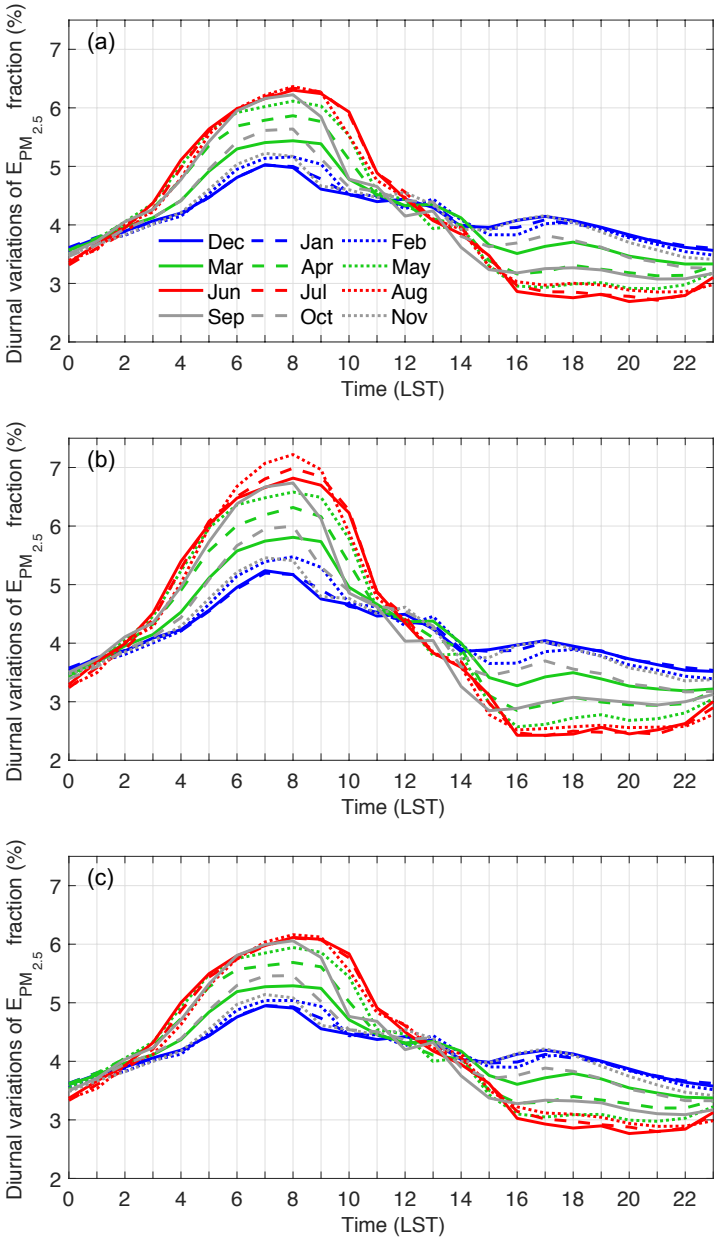

Figure 6. Five-year mean diurnal variations of dynamics-based $PM_{2.5}$ emission fraction averaged over (a) mainland China, (b) megacities with urban population ≥ 5 million, and (c) non-megacities with urban population < 5 million.

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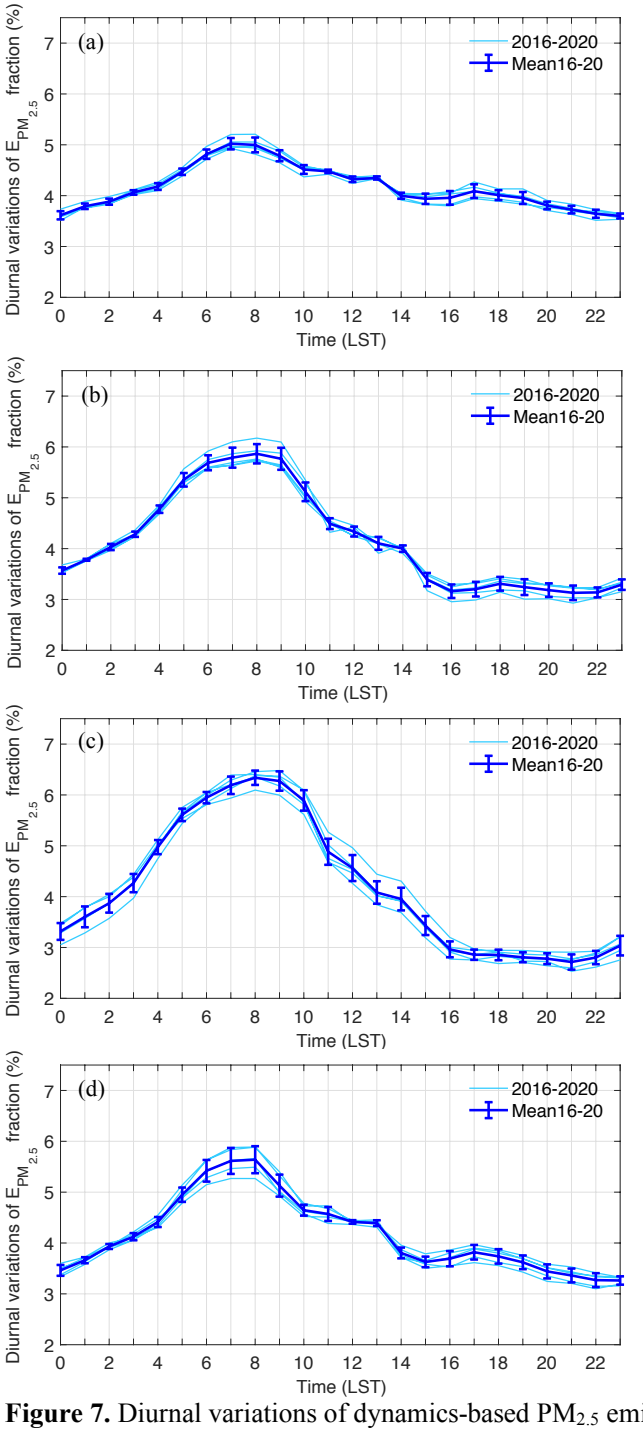

Figure 7. Diurnal variations of dynamics-based $PM_{2.5}$ emission fractions for years 2016-2020 (light blue) and five-year mean fractions with bars denoting one standard deviation of the five-

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year variations (dark blue) are averaged over mainland China for (a) January, (b) April, (c) July, and (d) October.

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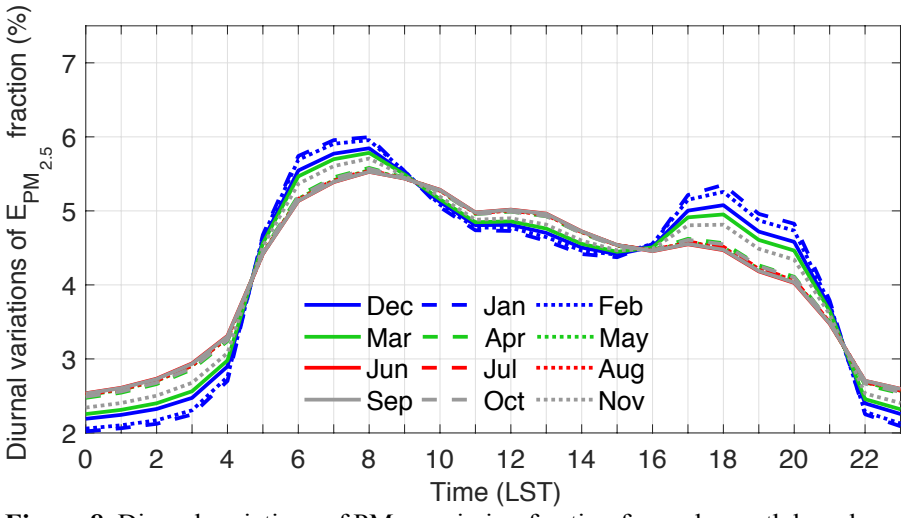

Figure 8. Diurnal variations of $PM_{2.5}$ emission fraction for each month based on diurnal variation profiles from ES and EU (Wang et al. 2010).

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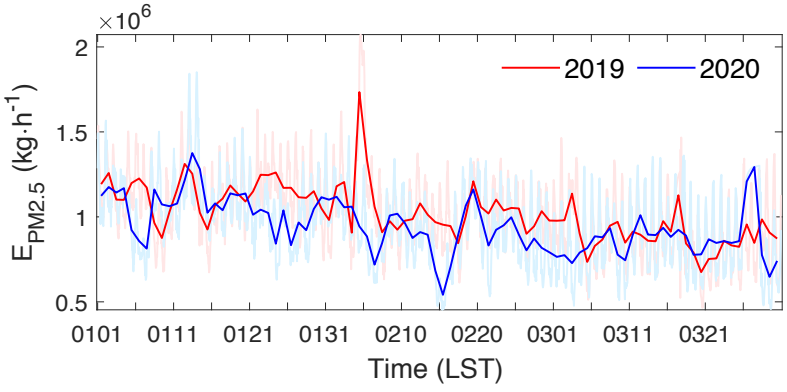

Figure 9. Hourly (light red and blue) and daily (dark red and blue) dynamics-based $PM_{2.5}$ emission estimates ($kg \cdot h^{-1}$) summed over mainland China from January to March of years 2019 and 2020.

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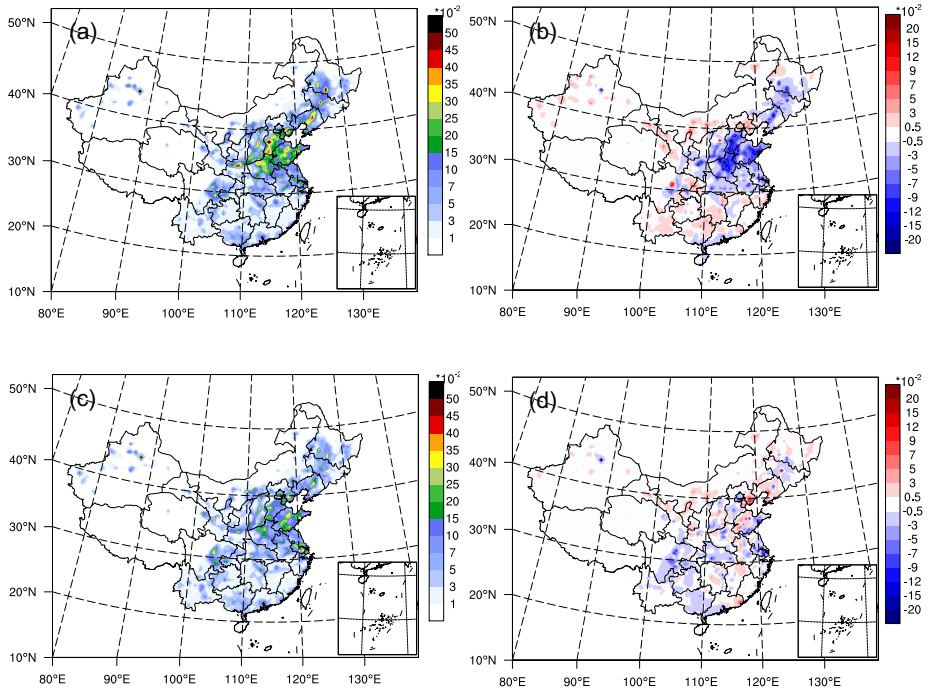

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653 **Figure 10.** Spatial distributions of dynamics-based $PM_{2.5}$ emission estimates ($\mu g \cdot m^{-2} \cdot s^{-1}$) on (b) February and (d)
654 March of year 2019, and spatial distributions of dynamics-based $PM_{2.5}$ emission reduction of year 2020 compared to
655 year 2019 for (c) February and (e) March.

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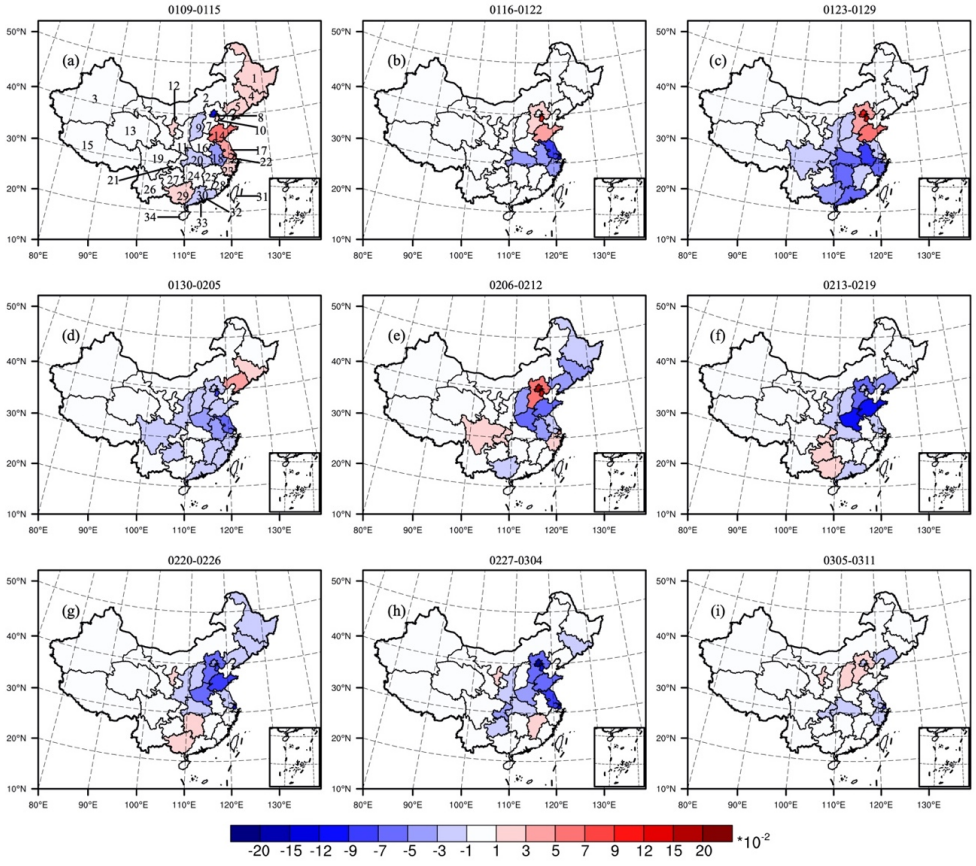

658 **Figure 11.** Mean spatial distributions of $PM_{2.5}$ emission differences ($\mu g \cdot m^{-2} \cdot s^{-1}$) between year
659 2020 and 2019 for 9 weeks starting at 9 January 2020. Negative (positive) values indicate that
660 $PM_{2.5}$ emission of year 2020 is smaller (larger) than that of year 2019. The numbers in (a) denote
661 provinces as: 1 Heilongjiang, 2 Neimenggu, 3 Xinjiang, 4 Jilin, 5 Liaoning, 6 Gansu, 7 Hebei, 8
662 Beijing, 9 Shanxi, 10 Tianjin, 11 Shanxi, 12 Ningxia, 13 Qinghai, 14 Shandong, 15 Xizang, 16
663 Henan, 17 Jiangsu, 18 Anhui, 19 Sichuan, 20 Hubei, 21 Chongqing, 22 Shanghai, 23 Zhejiang,
664 24 Hunan, 25 Jiangxi, 26 Yunnan, 27 Guizhou, 28 Fujian, 29 Guangxi, 30 Guangdong, 31
665 Taiwan, 32 Hongkong, 33 Macao, 34 Hainan.
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Table 1. Dynamics-based $PM_{2.5}$ emission estimates of year 2016 for each province whose value is larger than 0.01 $\mu g \cdot m^{-2} \cdot s^{-1}$ are shown in the second column. Ratios of $PM_{2.5}$ emission changes of years 2017-2020 compared to year 2016 are shown from the third to the sixth column, with negative (positive) values indicating decrease (increase) of $PM_{2.5}$ emission.

Province	$PM_{2.5}$ emission of year 2016 ($\mu g \cdot m^{-2} \cdot s^{-1}$)	Percentage of $PM_{2.5}$ emission change for year 2017 (%)	Percentage of $PM_{2.5}$ emission change for year 2018 (%)	Percentage of $PM_{2.5}$ emission change for year 2019 (%)	Percentage of $PM_{2.5}$ emission change for year 2020 (%)
Tianjin	0.2083	-14.07	-22.99	-38.70	-26.98
Shanghai	0.2067	-24.39	-30.21	-21.46	-30.05
Shandong	0.1631	-15.26	-21.02	-15.57	-19.41
Beijing	0.1598	-26.64	-25.75	-41.92	-45.27
Hebei	0.1178	-7.47	-11.98	-26.39	-22.87
Jiangsu	0.1088	-6.52	-3.98	-12.69	-28.20
Henan	0.1064	-1.41	-3.68	-12.15	-24.91
Shanxi	0.0885	6.17	7.90	-13.18	-13.85
Liaoning	0.0742	6.32	-2.58	3.22	11.42
Anhui	0.0687	1.92	-5.63	-6.23	-21.57
Hubei	0.0574	-5.87	-17.69	-19.76	-36.48
Zhejiang	0.0557	-3.62	-9.32	-9.99	-18.05
Chongqing	0.0525	-22.24	-29.81	-24.63	-38.41
Shanxi	0.0498	0.62	-1.97	-18.05	-17.85
Guangdong	0.0481	1.21	-6.01	-6.69	-14.37
Ningxia	0.0481	-8.17	-5.93	-24.46	-12.95
Hunan	0.0417	-6.40	-19.35	-9.91	-20.62
Guangxi	0.0390	-2.42	-3.52	-12.47	-22.31
Guizhou	0.0365	-4.01	-15.82	-21.74	-46.41
Jilin	0.0360	12.30	-3.22	7.37	4.76
Jiangxi	0.0353	13.22	-9.67	-7.19	-11.91
Sichuan	0.0337	-7.66	-15.66	-27.68	-37.93
Fujian	0.0244	3.13	-2.73	-8.13	-13.41
Heilongjiang	0.0231	7.30	-0.21	3.14	3.91
Yunnan	0.0221	-1.26	-7.16	-9.93	-15.35
Gansu	0.0177	-4.26	5.28	-17.89	-16.49
Hainan	0.0173	3.93	-0.41	-5.04	-4.78
Neimenggu	0.0141	-0.00	-3.63	-8.16	3.55

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675 **Table 2.** Five-year mean diurnal fractions (%) of the dynamics-based $PM_{2.5}$ emission estimates
676 over mainland China on local solar time (LST) for each month.

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
0	3.61	3.53	3.57	3.57	3.50	3.36	3.33	3.39	3.51	3.46	3.49	3.59
1	3.79	3.71	3.74	3.78	3.76	3.67	3.60	3.58	3.72	3.66	3.66	3.77
2	3.88	3.82	3.97	4.03	4.05	3.95	3.87	4.01	4.05	3.93	3.83	3.89
3	4.06	4.02	4.12	4.28	4.37	4.37	4.27	4.22	4.26	4.12	4.01	4.07
4	4.18	4.15	4.42	4.78	5.01	5.11	4.97	4.80	4.77	4.41	4.21	4.20
5	4.47	4.53	4.92	5.35	5.61	5.64	5.61	5.55	5.42	4.95	4.60	4.47
6	4.82	4.95	5.30	5.69	5.92	5.98	5.95	5.98	5.95	5.42	5.04	4.81
7	5.02	5.14	5.41	5.79	6.02	6.16	6.19	6.22	6.16	5.61	5.22	5.03
8	5.00	5.16	5.44	5.87	6.12	6.30	6.34	6.36	6.22	5.64	5.17	4.98
9	4.78	5.04	5.39	5.77	6.03	6.24	6.27	6.27	5.85	5.13	4.66	4.61
10	4.51	4.51	4.78	5.12	5.53	5.93	5.89	5.52	4.78	4.66	4.60	4.52
11	4.48	4.50	4.52	4.49	4.55	4.89	4.88	4.76	4.66	4.57	4.46	4.40
12	4.32	4.28	4.33	4.34	4.43	4.45	4.56	4.47	4.15	4.42	4.56	4.44
13	4.35	4.44	4.35	4.10	3.94	4.07	4.08	4.10	4.24	4.39	4.34	4.30
14	4.00	4.09	4.12	4.00	3.99	3.84	3.95	3.93	3.62	3.80	3.93	3.98
15	3.94	3.83	3.66	3.39	3.30	3.48	3.43	3.29	3.24	3.63	3.91	3.96
16	3.96	3.83	3.51	3.16	2.95	2.86	2.96	3.03	3.18	3.69	4.09	4.08
17	4.09	4.02	3.63	3.20	2.93	2.80	2.86	2.97	3.25	3.82	4.16	4.15
18	4.01	4.05	3.70	3.31	2.99	2.76	2.85	3.00	3.27	3.74	4.05	4.07
19	3.95	3.93	3.61	3.24	3.01	2.82	2.81	2.97	3.23	3.62	3.88	3.96
20	3.81	3.78	3.46	3.18	2.92	2.69	2.78	2.89	3.14	3.44	3.70	3.83
21	3.73	3.67	3.39	3.13	2.91	2.74	2.71	2.85	3.08	3.36	3.57	3.71
22	3.64	3.54	3.33	3.14	2.98	2.79	2.80	2.86	3.08	3.27	3.45	3.62
23	3.60	3.48	3.33	3.29	3.18	3.10	3.04	2.98	3.17	3.26	3.41	3.56

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