Replies to Questions by Referee 1

General comments
Based the MM5/CUACE model and observational data, an environmental meteorological index EMI2.5 and an assessment framework were developed in the present work. The roles of meteorology and control measures in China fine particular matter trend from 2013 to 2019 were separately assessed. It was found that the nationally aver- aged PM2.5 concentration had declined about 50

Specific comments
1. The current framework considers only the effects of emissions and meteorological conditions on PM 2.5 change. Actually, atmospheric chemistry plays a crucial role in shaping PM2.5 concentration. Can the authors include this factor in the framework? It would be more nice and convincing. Otherwise, the conclusions could not be so solid.

Reply: The same question was raised by another referee. The answer to the question is as follows:

The EMI index was focused explicitly on three major physical processes of iTran, iAccu, and iEmid that are closely related to the meteorological influences. However, the secondary formation of aerosols is implicitly considered in the EMI as the three major physical processes are calculated from the concentrations of aerosols (C) as indicated in Equation (3), which are resulted from the full processes of chemical mechanisms or "a highly parameterized method" that accounts for the secondary aerosol formations. Furthermore, we have done a comparison of simulated PM2.5 with full processes and the EMI with the parameterized method, and the correlation coefficients between them range 0.72 to 0.93 for the regions in this study. The limitation of non-inclusion of explicit chemical terms in the EMI is pointed out in the manuscript.
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2. In the model simulations, both primary and pre-cursor emissions of PM are based on the 2016 MEIC inventory. However, the present work focused on the tendency of PM2.5 from 2013 to 2019. Did the authors use the same inventory for every year or change the inventory year by year?

Reply: In order to isolate the meteorological impacts, we have used the 2016 MEIC emissions throughout the simulations, i.e. from 2013 to 2019, resulting the differences caused by meteorological changes only.

3. Table 2 shows the observed PM2.5 difference between 2019 and 2015, why not 2019 and 2013 to be consistent with the title and other parts?

Reply: The same question was raised by another referee. The answer to the question is as follows:

There are two issues here that prompted us to use two different time intervals for the comparisons in the paper. The first issue is the completeness of the network observational data series of $PM_{2.5}$ in China. The systematical and network observations of $PM_{2.5}$ started in China from 2013. However, it took about two years (until 2015) to develop to the current status. Number of monitoring stations national-wide in 2013 was less than 900, reached to about 1400 in early 2015 and maintained the same up to now. To show the completeness of the observed PM2.5 time series and for most part of the paper, we made the comparison starting from 2013 as graph illustrations. The second issue is the data consistence and policy relevance of the assessment. Statistically, because the national observation site numbers are relative constant from 2015 to 2019, it makes more sense to use the 2015-2019 data for numerical assessment such as those shown in Table 2. The use of 2015-2019 data for Table 2 was also motivated by the introduction of the Environmental Protection Law of People's Republic of China in January 2015. For the regulation assessment point of view, the comparison Table 2 was in line with the date of the law introduction and the impact assessment by emission changes was more relevant to the interests of management to show how effective the law was.

4. In Eq.(3) why the integration is just over dz, not dxdydz?

Reply: The Equation (3) was introduced to account for the column loading of aerosols in the PBL, that contains most of the aerosol masses, for a grid. The dxdy, i.e. advection terms, is done in the 3-D transport part of the model.

Technical corrections
There are numerous typos need to be corrected. I suggest the authors carefully proof read the manuscript to make sure all language problems are fixed.

1. Page 8 Line 2: "Results and Discussion secession" should be "Results and Discussion section". This should be corrected all over the entire manuscript.

Reply: Thanks. Corrected!

2. Page 8 in Figure 2: "Natiaonal" should be "National"; "t0" should be "to"!

Reply: Thanks. Corrected!

3. Page 11 Line 17: "sing" should be "sign"; Line 18 "vise visa" should be "vice versa"
Reply: Thanks. Corrected!

4. Page 12 Line 11: I think Kx, Ky and Kz should be turbulent diffusion coefficients; Line 16-17: "on and to" should exchange position; Line 19 "compared" might be "computed"

Reply: Done for the "on and to" exchange. Line 19 "compared" was not changed as we indeed meant to compare.

5. Page 13: Line 5 and Lines 10-11. The explanation on /EMI(p)2.5 is not consistent; Line 13 “Figure 2” should be “Figure 3”.

Reply: Thanks. Corrected!

6. Page 14: Lines 7 to 9, why January 2103 (should be 2013!) to January 2016?; Line 15 “combined” should be “combine”.

Reply: Thanks. Corrected!

7. Page 17: Lines 3-10, I could not understand what the authors meant. Line 8 (Wang et al.) is not a proper citation; Line 13 the introduction of Figure 4 is not consistent with the actual caption in Page 18 Lines 2-4.

Reply: As in the Question 2 above, we have used the MEIC emissions for 2016 for all simulations, but applied a monthly variation based on Wang et al. 2011: (Verification of anthropogenic emissions of China by satellite and ground observations). We have checked with the authors of this paper and been assured that the monthly variations were discussed.

We have corrected the Line 13 to “spatial distribution of correlation coefficients between…”

8. Page 19: In Figure 5 the subcaptions of (c) and (d), “contributions” should be “con-tributions”.

Reply: Thanks. Corrected! The caption for Figure 5 has been re-written.

9. Page 22: Line 4 “2103” should be “2013”; Line 6 (Wang et al.) is not a proper citation; “to” should be removed from “pointing out to”.

Reply: Thanks. Corrected!

10. Page 25: Line 20, “favorite and un-favorite” might be “favorable and unfavorable”. This is also true for other statements hereafter.

Reply: Thanks. Corrected: 4 in total.

11. Page 29: Line 14 “2105” should be “2015”!

Reply: Thanks. Corrected!

12. Page 30: Line 10, to judge whether the meteorological conditions are favorite or not to. . .

Reply: Thanks. Corrected!

Replies to Questions by Referee 2

It has been a long-standing problem to quantify the roles of meteorology and emission change in regional air pollution variations. Different modeling tools and techniques have been developed and utilized to address this problem. In this study, the authors developed a process analysis-based framework in a chemical transport model named CUACE to identify the driving factors of PM2.5 changes in China during 2013-2019. They defined an Environmental Meteorological Index (EMI) by tracking the contributions of different physical processes including transport, diffusion, emission, and deposition to simulated PM2.5 concentrations in the model. The topic is within the scope of the journal and the research question is of broad interest in the community. In general, the manuscript is well-structured, but the English writing in some parts (especially the methodology section) can be improved for clear description. Based on the current version, I have some major concerns about the theoretical basis of this EMI framework. Please see below the detailed comments to be addressed.

Reply: The methodology section has been revised to clarify some descriptions, which may have caused certain confusions for the referee to raise the questions (1) and (2) below.

(1) The EMI-based analytical framework is based on the continuity equation and is similar to some mature and widely-used probing tools in other CTMs such as the integrated process rate (IPR) analysis in CMAQ and the process analysis (PA) tool in CAMx. However, the major difference between this EMI method and other probing tools is that secondary aerosol formation is missing in the EMI framework. Let's revisit a simplified continuity equation (Seinfeld and Pandis, 2016) and compare it with EMI defined in this study:

$$\frac{\partial \langle c \rangle}{\partial t} + \bar{u}_j \frac{\partial \langle c \rangle}{\partial x_j} = \frac{\partial}{\partial x_j}\left(K_{jj} \frac{\partial \langle c \rangle}{\partial x_j} \right) + R + S$$

Above is the atmospheric diffusion equation that is based on the mixing-length theory and two assumptions (negligible molecular diffusion and incompressible atmosphere). CTMs, including CUACE used in this study, use this equation to describe the spatiotemporal evolution of air pollutant concentrations. According to the EMI definition in Section 2.3 of this study, EMI is a time integral of atmospheric pollution changing tendency that consists of three parts: iTran, iAccu, and iEmid (Eqs. (1)-(3) on page 11/12). The first part iTran corresponds to the advection term on the LHS of the diffusion equation but with an opposite sign (after moving the advection term from the LHS to the RHS), the second part iAccu corresponds to the turbulent diffusion term (much greater than molecular diffusion) on the RHS of the diffusion equation, and the third part iEmid corresponds to the last source and sink term S on the RHS of the diffusion equation. Since ΔEMI only includes these three parts without the chemical generation term R

(which also depends on meteorological factors such as temperature and relative humidity), it only approximates the direct physical processes modulating aerosol concentrations and ignores other meteorological impacts on chemical reactions and secondary formation of PM2.5. Given the large contribution of secondary formation to PM2.5 concentrations in China (Huang et al., 2014), it's inevitable to include the comprehensive aerosol processes regarding chemical formation, nucleation, condensation, coagulation, and gas–particle partitioning etc. in this kind of probing tools to conserve the mass balance in CTMs. For instance, the IPR analysis in CMAQ considers the effects of individual physical processes and the net effect of chemistry (aerosol processes) on gas-phase air pollutants (PM2.5). It also provides more details of the chemical transformations associated with the model's chemical mechanism in the integrated reaction rate (IRR) analysis. It's noted that the CUACE model considers secondary aerosol formation in its aerosol module (line 7-8 on page 16), but it uses "a highly parameterized method" to directly estimate secondary aerosol formation from precursors including SO2, NO2, and VOC for the EMI application (line 8-11 on page 16). Therefore, it seems the EMI framework can only take account of the three listed physical processes (iTran, iAccu, and iEmid) without consideration of meteorological effects on secondary inorganic and organic aerosol formation and transformation, which is a limitation of the current framework that should be pointed out in the manuscript.

Reply: Thank you for pointing this issue out. The same question was raised by another referee. The answer to the question is as follows:

The EMI index was focused explicitly on three major physical processes of iTran, iAccu, and iEmid that are closely related to the meteorological influences. However, the secondary formation of aerosols is implicitly considered in the EMI as the three major physical processes are calculated from the concentrations of aerosols (C) as indicated in Equation (3), which are resulted from the full processes of chemical mechanisms or "a highly parameterized method" that accounts for the secondary aerosol formations. Furthermore, we have done a comparison of simulated PM2.5 with full processes and the EMI with the parameterized method, and the correlation coefficients between them range 0.72 to 0.93 for the regions in this study, indicating that the parameterized method used in this study for EMI largely approximates the variation of PM2.5 with full processes. The limitation of non-inclusion of explicit chemical terms in the EMI is pointed out in the manuscript.

(2) Even focusing on EMI itself without considering complex aerosol processes, the EMI framework is still problematic to be applied for assessing meteorological contributions to PM2.5 concentration changes. A simple way to demonstrate this is to consider two idealized extreme conditions: the first is an extreme stagnation case with zero wind and the second is an extreme dispersion case with single-direction high winds (time invariant). In both cases we

assume no precipitation, no wet deposition (L_d = 0 in iEmid), and negligible dry deposition compared with emissions ($Emis \gg V_d$ in iEmid).

In the first stagnation case, the first two parts of ΔEMI (iTran and iAccu) would diminish to zero since there is no wind (no advection) and no turbulence (no turbulent diffusion). The third part iEmid would be dominated by the constant emission term (assumed in line 12 on page 12) given $Emis \gg V_d$ and L_d = 0. In this case, ΔEMI approximates to an emission-based constant that is irrelevant to meteorology ($\Delta EMI = iTran + iAccu + iEmid = 1/C_0 \int (Emis) dz = 1/C_0\ Emis \cdot \hbar$, where C_0, $Emis$, and $\hbar$ are constants). After applying this approximation to Eqs. (5)-(6) in the manuscript, the ratio of

$$\overline{EMI(p0)_{2.5}}/\overline{EMI(p1)_{2.5}} = EMI(p0)_{2.5}/\left(EMI(p0)_{2.5} + \frac{1}{c_0} Emis \cdot h \cdot (p1 - p0)\right)$$

becomes a variable that only depends on the initial value of EMI ($EMI(p0)_{2.5}$), scaling constant C_0, constant $Emis$ intensity, vertical height $\hbar$, and the time interval between $p0$ and $p1$, which does not reflect the meteorological impact on $PM_{2.5}$ concentration changes from $p0$ and $p1$ (though in this case the meteorological impact should be zero as all considered meteorological processes have been turned off or neglected and PM2.5 concentrations solely depends on emission intensity and time intervals) as alleged in line 19-20 on page 14.

In the second case with extreme dispersion conditions, ΔEMI would be dominated by the first advection term iTran due to constant high winds (the concentration gradient still exists because of the constant emission source), and $\overline{EMI(p1)_{2.5}}$ would keep increasing to a huge number after a long time integral of ΔEMI. Given a predetermined initial value of $\overline{EMI(p0)_{2.5}}$ at $p0$, the ratio of $\overline{EMI(p0)_{2.5}}/\overline{EMI(p1)_{2.5}}$ in Eq. (6) approaches to zero after a long time because of the much greater denominator, which again fails to represent the meteorological impact on concentration changes from $p0$ and $p1$ (in this case the right answer for the meteorological impact should be ~100% because of the dominant role of strong advection, while the emission impact reduces to nearly zero).

The failure of the EMI framework to describe meteorological impacts on $PM_{2.5}$ concentrations results from the incorrect inclusion of emissions ($Emis$) in EMI, which contradicts its objective to separate meteorological effects from emissions. Given such defect in its theoretical basis, there is no need to further discuss the EMI-based modeling results.

Reply: To answer the questions, we first have to clarify three levels of EMI definitions:

Level 1: ΔEMI, the tendency that causes the changes of pollution level at each time step Δt.

Level 2: EMI(t), the index as a function of time t.

Level 3: $\overline{EMI(p)_{2.5}}$, averaged EMI for a period of time (p), i.e. a week or a month.

This is illustrated in Figure 3 of the manuscript. Therefore, P0 and p1 are two time periods defined for the comparisons of averaged meteorological impacts by EMI, i.e. p0 represents the month of January in 2015, and p1 represents the month of January in 2019. The p0 and p1 do not represent one period of time from p0 to p1. The focus of EMI applications is on the comparison of averaged meteorological difference between these two time intervals (p0 and p1).

For the first case of absolute stagnation, if $\Delta EMI = iTran + iAccu + iEmid = 1/c_0 \cdot Emis \cdot \hbar$, as a constant, the equation (5) becomes for the period of p0 (n steps):

$$\overline{EMI(p0)_{2.5}} = EMI(p0) + (n-1) \times \Delta EMI(p0) \times \Delta t$$

which means that averaged EMI will increase and be accumulated as the time goes on for n steps. This is exactly what this kind of meteorological conditions will bring about to the pollution levels.

In order to compare the difference of meteorological impacts, we have to define a new period, i.e. p1. If p 1 has the same initial conditions [EMI(p0)=EMI(p1)] and absolute stagnation, the averaged $\overline{EMI(p)_{2.5}}$ would be determined by the duration of the stagnation (m steps).

$$\overline{EMI(p1)_{2.5}} = EMI(p1) + (m-1) \times \Delta EMI(p1) \times \Delta t$$

The longer of the stagnation, the larger of averaged $\overline{EMI(p)_{2.5}}$. This is exactly what $\overline{EMI(p)_{2.5}}$ is intended to be: a quantitative description of the meteorological impact on pollution levels. If n=m, the ratio of $\overline{EMI(p0)_{2.5}}/\overline{EMI(p1)_{2.5}} = 1$, indicating the same meteorological impact as expected.

If the period p1 is defined as the referee suggested: extreme dispersion conditions (assumed constant) with the same emission, we would expect a huge NEGATIVE $iTran$ for the ΔEMI(p1), resulting $\Delta EMI(p1) \ll \Delta EMI(p0)$ and $\overline{EMI(p1)_{2.5}} \ll \overline{EMI(p0)_{2.5}}$ and reflecting favorite meteorological conditions for P1, i.e. the ratio of $\overline{EMI(p0)_{2.5}}/\overline{EMI(p1)_{2.5}} \gg 1$. Eventually, if

$\Delta EMI(\mathrm{p1}) = iTran + iEmid < 0$, the dispersion would clean the pollutants for a certain period of time, and bring the EMI(p1) to reach zero as the concentration has reached zero by extreme dispersion; if $\Delta EMI = iTran + iEmid > 0$, we can then expect an increase of EMI(p1). The following figure illustrates the concept of the EMI and the areas below each curve (red and black line) is the averaged $\overline{EMI(p)_{2.5}}$ for each period, respectively, for the cases suggested by the referee (Figure 1).

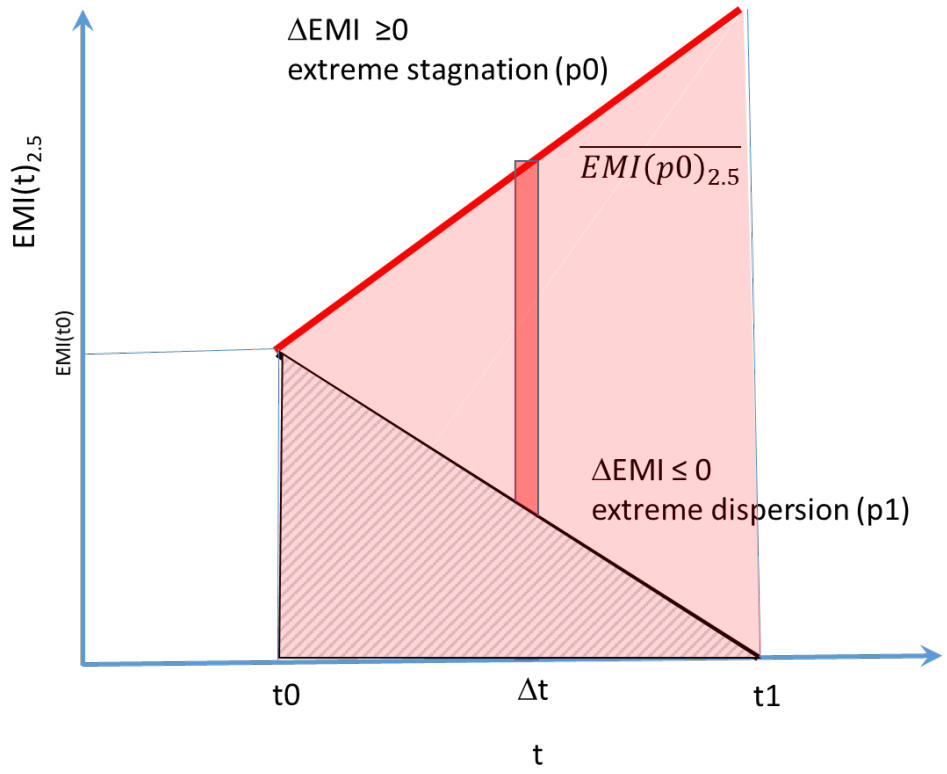

Figure 1: Illustration of relationship between 3 levels of EMI definition for the cases suggested by Referee #2.

Please NOTE that since we used a constant emission at each location to compute the EMI, any changes in $\overline{EMI(p)_{2.5}}$ for two periods (p0 and p1) are solely attributed to the changes in meteorological conditions. If emission changes (+ or -) due to anthropogenic activities from p0 to p1 in conjunction with the meteorological variations, the $\overline{EMI(p)_{2.5}}$ will not be able to fully account for all the contributions, as the observational values are caused by both changes.

In order to separate the impacts of meteorology and emission contributions to the changes in pollutant concentrations from p0 to p1, the equations (6) and (7) are introduced and used to

quantitively assess the emission CHANGES only on the observed levels of pollutants ($PM_{2.5}$ for current study) from p0 to p1. An assumption is made here that under the same emissions at p1 (e1), the ratio of the averaged $PM_{2.5}$ concentrations under meteorology for p0 (m0) to the averaged $PM_{2.5}$ concentrations under meteorology for p1 (m1) is equal to the ratio of averaged EMI for each periods, i.e. $\overline{EMI(p0)_{2.5}}$ / $\overline{EMI(p1)_{2.5}}$, which is exactly what EMI is intended to be. Therefor:

$$PM(m0,e1)\Big/_{PM(m1,e1)} = \overline{EMI(p0)_{2.5}}\Big/_{\overline{EMI(p1)_{2.5}}} \quad (6)$$

The impact of only emission changes from *e0* to *e1* on the concentration changes can be expressed as:

$$\Delta EMIS = \frac{PM(m0,e1)-PM(m0,e0)}{PM(m0,e0)} \times 100\% \quad (7)$$

where PM(*m0*, *e0*) and PM(*m1*, *e1*) are the observed concentrations at p0 and p1, respectively. PM(*m0*, *e1*) is estimated from Equation (6).

In summary, we think the assessment framework is solid based. The questions raised the referee was due to the confusion by the description part of methodology section, which may have misled the referee to derive and come out with the questions. Because of this, we have revised this section extensively to give a clearer description. Thanks for the referee.

Below are some technical corrections and comments:

(1) What are the units of EMI and ΔEMI? Is EMI unitless as shown in Fig. 5/9? You will get different answers after doing dimensional analysis for Eqs. (1)-(3).

Reply: The EMI is unitless. Thanks to the referee who found the problems in Eqs (1)-(3): There is a term (1/h) missing in the equation and we have fixed them. The new equations are as follows:

$$iTran = \frac{1}{hC_0}\int_0^h \left(u\frac{\partial C}{\partial x} + v\frac{\partial C}{\partial y} + w\frac{\partial C}{\partial z}\right) dz$$

$$iAccu = \frac{1}{hC_0}\int_0^h \left[\frac{\partial C}{\partial x}\left(Kx\frac{\partial C}{\partial x}\right) + \frac{\partial C}{\partial y}\left(Ky\frac{\partial C}{\partial y}\right) + \frac{\partial C}{\partial z}\left(Kz\frac{\partial C}{\partial z}\right)\right] \mathrm{dz}$$

$$iEmid = \frac{1}{hC_0}\int_0^h [Emis - (V_d + L_d)]\, dz$$

The calculations in the model was done with the right equations and therefore the results presented in the paper were not impacted by this problem.

(2) How to determine the initial value for EMI(t0)? Here I assume t0 denotes the first day of 2013, which is the start point of the model simulation. But the initial value for EMI is not mentioned in the manuscript.

Reply： In order to compare each year (or month) under the same conditions, the initial value of EMI(t0) was set the same for the first day of each year (or month). We also checked the sensitivity of EMI on the initial values of EMI(t0) and concluded that monthly averaged EMI was hardly impacted by the initial values. Nevertheless, the initial values for each month was set up by the averaged $PM_{2.5}$ concentrations for the first day from 2013 to 2019 divided by a constant C (35 um/m3). This has been added in the manuscript.

(3) What is h in Eq. (3)? Is it boundary layer height or not?

Reply: It is an arbitrary value of 1500 meters but it was a height defined to contain most of aerosol mass in the boundary layer.

(4) What kind of data were used for the correlation in Fig. 4? Monthly? Or Daily?

Reply: They are daily values used for the correlation.

(5) The time intervals for model evaluation are inconsistent throughout the manuscript. For example, Figs. 7-10 show the comparison from 2013 to 2019, but Table 2 shows the attribution results between 2015 and 2019.

Reply: There are two issues here that prompted us to use two different time intervals for the comparisons in the paper. The first issue is the completeness of the network observational data series of $PM_{2.5}$ in China. The systematical and network observations of $PM_{2.5}$ started in China from 2013. However, it took about two years (until 2015) to develop to the current status. Number of monitoring stations national-wide in 2013 was less than 900, reached to about 1400 in early 2015 and maintained the same up to now. To show the completeness of the observed PM2.5 time series and for most part of the paper, we made the comparison starting from 2013 as graph illustrations. The second issue is the data consistence and policy relevance of the assessment. Statistically, because the national observation site numbers are relative constant from 2015 to 2019, it makes more sense to use the 2015-2019 data for numerical assessment such as those shown in Table 2. The use of 2015-2019 data for Table 2 was also motivated by the introduction of the Environmental Protection Law of People's Republic of China in January 2015. For the regulation assessment point of view, the comparison Table 2 was in line with the date of the law introduction and the impact assessment by emission changes was more relevant to the interests of management to show how effective the law was.

Assessment of meteorology vs control measures in China fine particular matter trend from 2013-2019 by an environmental meteorology index

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Abstract

A framework was developed to quantitatively assess the contribution of meteorology variations ~~in~~ to the trend of fine particular matter ($PM_{2.5}$) concentrations and to separate the impacts of meteorology from the control measures in the trend, based upon an Environmental Meteorology Index (EMI). The model-based index EMI realistically reflects the role of meteorology in the trend of $PM_{2.5}$ ~~PM~~ and is explicitly attributed into three major factors: deposition, vertical accumulation and horizontal transports. Based on the 2013-2019 $PM_{2.5}$ observation data and re-analysis meteorological data in China, the contributions of meteorology and control measures in nine regions of China were assessed separately by the EMI-based framework. Monitoring network observations show that the $PM_{2.5}$ concentrations have been declined about 50% on national average and about 35% to 53% for various regions. It is found that the nation-wide emission control measures were the dominant factor in the declining trend of China $PM_{2.5}$ concentrations, contributing to about 47% of the $PM_{2.5}$ decrease from 2013 to 2019 on

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the national average and 32% to the 52% for various regions. The meteorology has a variable and sometimes critical contribution to the year by year variations of $PM_{2.5}$ concentrations, 5% on annual average and 10-20% for the fall-winter heavy pollution seasons.

1. Introduction

Recent observation data from the Ministry of Ecology and Environment of China (MEE) has shown a steady improvement of air quality across the country, especially in particular matter (PM) concentrations (Hou et al., 2019). According to 2013-2019 China Air Quality Improvement Report issued by MEE, compared to 2013, the average concentrations of particulate matter with an aerodynamic diameter of less than 2.5 μm ($PM_{2.5}$) ~~concentrations~~ in 74 major cities of China have decreased by more than 50% in 2019. From scientific and management point of views, a quantitative apportionment of the reasons behind the trend is critical to assess the reduction strategies implemented by the government and to guide future air quality control policy. However, the assessment of the improvements of air quality is a complicated process that involves the quantification of changes in the emission sources, meteorological factors, and other characteristics of the $PM_{2.5}$ pollution, which are also interacting with each other. In order to separate the relative degree of these factors, a comprehensive analysis, including observational data and model simulation, is needed.

Researches have been done extensively on the impacts of weather systems on air quality. Synoptic and local meteorological conditions have been recognized to influence the PM concentrations at various scales (Beaver and Palazoglu, 2006;He et al., 2017a;He et al., 2017b;Pearce et al., 2011a;Pearce et al., 2011b). For the atmospheric aerosol pollution ~~in eastern China~~, the dynamic effect of the downdraft in the "leeward slope" and "weak wind area" of the Qinghai Tibet Plateau in winter is not conducive to the diffusion of air pollution emissions in the urban agglomerations of eastern China (Xu et al., 2015;Xu et al., 2002). The evolution of circulation situation is an important factor driving the change of haze pollution (He et al., 2018). The local circulations, such as mountain and valley wind and urban island circulation, have significant impact on local pollutant concentration (Chen et al., 2009;Yu et al., 2016). Previous studies also revealed that $PM_{2.5}$ concentration is significantly correlated with local meteorological elements, such as temperature, humidity, wind speed, and boundary layer height (He et al., 2017b;Bei et al., 2020;Ma et al., 2019;He et al., 2016).

In the Beijing-Tianjin-Hebei (BTH) Region, a correlation analysis and principal component regression method (Zhou et al., 2014) was used to identify the major meteorological factors that influenced the API (Air Pollution Index) time series in China from 2001-2010, indicating that air pressure, air temperature, precipitation and relative humidity were closely related to air quality ~~and resulting in~~with a series of regression formulas. Yet, the analysis was assumed a relatively unchanged emission whose impacts were not taken into account. On a local scale, an attempt (Zhang et al., 2017) has been made to correlate the air pollutant levels with a combination of meteorological factors

with the development of the Stable Weather Index (SWI) at CMA. The SWI is a composite index which includes the advection, vertical diffusion and humidity and other meteorological factors that are related to the formation of air pollutions in a specific region or city. A higher value of SWI means a weaker diffusion of air pollutants. This index had some success in assessing the meteorological impacts on air pollution, especially calibrated for a specific region, i.e. Beijing. However, when applied to different areas where the emission patterns and meteorological features are different, this index failed to give a universal or comparable indication of meteorological assessment of pollution levels across the nation.

Using the Kolmogorov-Zurbenko (KZ) wave filter method, Bai et al (2015) ~~made an effort to break~~separated the API time series in three Chinese cities into short-term, seasonal and long-term components, and then used the stepwise regression to set up API baseline and short-term components separately and established linear regression models for meteorological variables of corresponding scales. Consequently, with the long-term representing the change of emissions removed from the time series, the meteorological contributions alone were assumed and analyzed, pointing out that unfavorable conditions often lead to an increase by 1-13 whereas the favorable conditions to a decrease by 2-6 in the long-term API series, respectively. Though the contributions of emissions and meteorological variations were separated by the research, it wa~~i~~s only done by mathematical transformations and far from the reality. The mechanisms behind the variation of the time series were not investigated.

A chemical transport model (CTM) is an ideal tool to carry the task of assessment by taking the meteorology, emissions and processes into considerations altogether. Andersson et al. (2007) used a CTM to study the meteorologically induced inter-annual variability and trends in deposition of sulphur and nitrogen as well as concentrations of surface ozone (O_3), nitrogen dioxide (NO_2) and PM and its constituents over Europe during 1958-2001. It is found that the average European interannual variation, due to meteorological variability, ranges from 3% for O_3, 5% for NO_2, 9% for PM, 6-9% for dry deposition, to about 20% for wet deposition of sulphur and nitrogen. A multi-model assessment of air quality trends with constant anthropogenic emissions was also carried out in Europe (Colette et al., 2011) and found that the magnitude of the emission-driven trend exceeds the natural variability for primary compounds, concluding that that emission management strategies have had a significant impact over the past 10 years, hence supporting further emission reductions strategies. Model assessments of air quality trends at various regions and time periods (Wei et al., 2017;Li et al., 2015) in China were also done and yielded some useful results. For the BTH Region, Li et al. (2015) used the Comprehensive Air Quality Model with extensions (CAMx) plus the Particulate Source Apportionment Technology (PSAT) ~~and~~ to simulate~~d~~ the contributions of emission changes in various sectors and changes in meteorology conditions for the $PM_{2.5}$ trend from 2006 to 2013. It was found that the change of source contribution of $PM_{2.5}$ in Beijing and northern Hebei was dominated by the change of local emissions. However, for Tianjin, and central and southern Hebei province, the change of meteorology condition was as important as the change of emissions, illustrating the regional difference of

impacts by meteorology and emissions. However, the emission changes in the simulations were assumed and did not reflect the real spatia-temporal variations.

There is no surprise that previous studies could not systematically catch the meteorological impacts across the whole nation as the controlling meteorological factors involving the characteristics of plenary boundary layers (PBL), wind speed and turbulence, temperature and stability, radiation and clouds, underlying surface as well as pollutant emissions, vary greatly from region to region. A single index or correlation cannot be applied to the entire nation. Obviously, in order to systematically assess the impacts of meteorology on air pollution, these factors have to be taken into consideration in a framework and be assessed simultaneously. This paper presents a methodology to assess the individual impacts of meteorology and emission changes, based on a model-derived index EMI, i.e., Environmental Meteorology Index, and observational data, providing a comprehensive analysis of the air quality trends in various regions of China, with mechanistic and quantitative attributions of various factors.

2. Methodology

The assessment is carried out through the combination of observational data and EMI index from model analysis. Since the emission and air quality characteristics vary greatly from region to region in China, the analysis is divided into 9 focused regions (Figure 1). Regional air quality dada ($PM_{2.5}$) provides the basis for the trend analysis. Separating the

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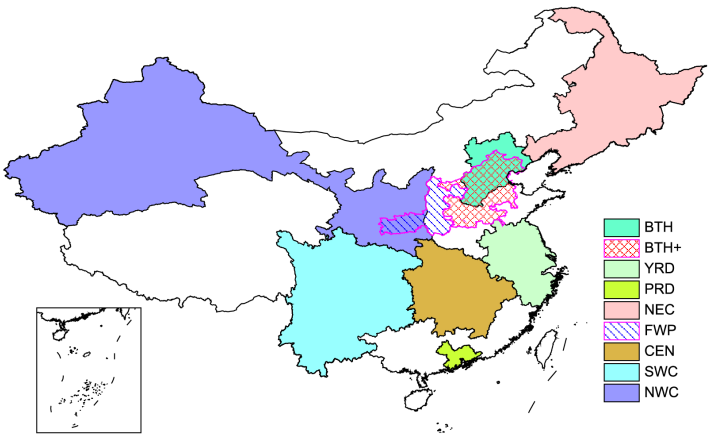
trend contribution from regional emission reduction and meteorological variation ~~needs~~ entails a framework, which is discussed below.



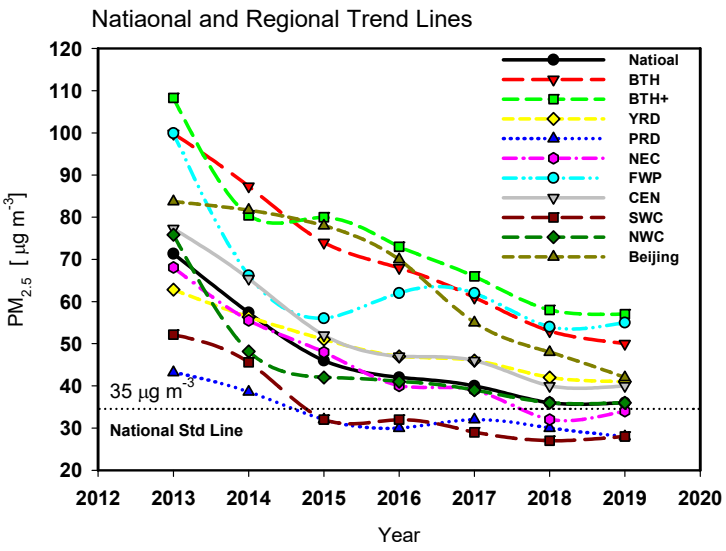
Note： **BTH**: Beijing, Tianjin and Hebei; **BTH+**: BJ, TJ + 26 cities; **YRD**: Shanghai, Jiangsu, Zhejiang and Anhui; **PRD**: 9 cities in Guangdong; **NEC**: Heilongjiang, Jilin and Liaoning; **FWP**: 11 cities in Shanxi, Shannxi and Henan; **CEN**: Hubei, Hunan and Jiangxi; **SWC**: Yunnan, Guizhou, Sichuan, Chongqing; **NWC**: Shannxi, Gansu, Ningxia and Xinjiang

Figure 1: Analysis region separation and definition.

2.1. Particular Matter (PM) Observation Data

The observational pollution data of $PM_{2.5}$ concentrations used in this study were from the monitoring network of the Ministry of Ecology and Environment (MEE) of China (http:// english.mee.gov.cn/). From 2013 to 2019, the concentrations have shown a large change in the country where most regions see a declined trend in the annual concentrations. Data show that from 2013 to 2019, the national annual averaged $PM_{2.5}$

concentrations have dropped about 50% (Fig. 2), where the haze days have been shortened by 21.2 days from the China Meteorological Administration (CMA) monitoring data (Table 1), with some regional differences. Regionally, by 2019, the $PM_{2.5}$ reduction rate from 2013 ranges from 35 to 53%. Detailed analysis will be given in the Results and Discussions secetssion.



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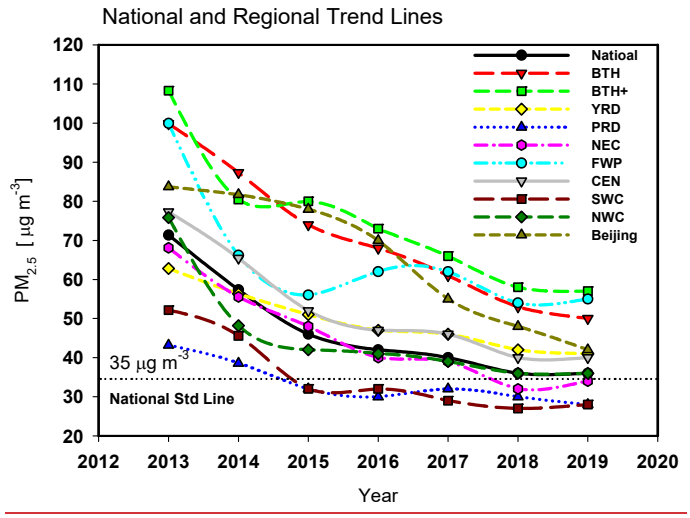

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3 Figure 2: National and regional trend lines of $PM_{2.5}$ in China from 2013 ~~t0~~ to 2019.

It is noted that the $PM_{2.5}$ mass concentrations by MEE are now reported under observation site's actual conditions of temperature and pressure from September 1, 2018 before which the values were reported under the standard state (STP), i.e. 273 K and 101.325 kPa. In order to maintain the consistence of the data series, the $PM_{2.5}$ concentrations used in this study have all been converted according to the new standard (MEE, 2012)(GB3095-2012) under actual conditions. Research has shown that after the change of reporting standard, the $PM_{2.5}$ concentration in most cities decreased, and the number of good days to meet the standard increased (Zhang and Rao, 2019).

2.2. Meteorological Data

Conventional meteorological data can provide qualitative assessment of the contributions of meteorological factors to the changes of air quality. The data used in this study are from 843 national base weather stations of the CMA from 2013 to 2019. The wind speed (WS), day with small wind (DSW), relative humidity (RH) and haze days are used to analyze the pollution meteorological conditions. When the daily average wind speed is less than 2 m s^{-1}, a DSW day is defined. Since the haze formation is always related to stable meteorological conditions and high aerosol mass loading, haze observation from CMA is also used to analyze the haze trends and the impact of air quality on visibility. A haze day is defined with daily averaged visibility less than 10 km and relative humidity less than 85% (Wu et al., 2014), excluding days of low visibility due to precipitation, blowing snow, blowing sand, floating dust, sandstorms and smoke.

~~Data show that from 2013 to 2019, T~~the 2019 national annual averaged WS has increased by ~~12.9~~4.5%, DSW dropped by 15.1%, and RH ~~almost unchanged~~decreased by 3.9% ~~(Table 1)~~C~~,~~ompared with ~~with~~2013, with regional differences (Table 1). Slightly changes occurred when compared with 2015 that WS has decreased by 0.7%, DSW dropped by 11.3%, and RH decreased by 2.2%. Overall, ~~,~~ i~~t~~t can be seen that the annual haze days have a certain degree of correlations negatively ~~positively~~ with WS and positively ~~negatively~~ with DSW. Detailed analysis linking $PM_{2.5}$ and meteorology will be given in the Results and Discussion~~s secession~~section.

Table 1: National and regional environmental meteorology in 2019 and comparisons with ~~2013~~ 2015 and ~~2016~~ 2013

Region	Wind Speed			Days with Small Wind			Relative Humidity			Haze (days)		
	~~AVG~~ AVG (m s^{-1})	vs ~~2016~~ 2015 (%)	vs 2013 (%)	Days	vs ~~2016~~ 2015 (%)	vs 2013 (%)	%	vs ~~2016~~ 2015 (%)	vs 2013 (%)	Days	vs ~~2016~~ 2015	vs 2013
National	~~12~~.29	~~-~~ ~~+03~~.76	+~~412~~.95	129.8	~~-11.3~~ ~~0.8~~	-15.1	60.1	~~-2.2~~ ~~6.3~~	-3.9	25.7	~~-~~ ~~19.0~~ ~~12.1~~	-21.2
BTH	~~21~~.09	~~-8.6~~ ~~2.7~~	-2.2	131.0	~~+14.7~~ ~~+5.8~~	+9.0	56.7	~~-2.6~~ ~~3.3~~	-4.2	45.2	~~-~~ ~~20.4~~ ~~15.2~~	-26.1
BTH+	2.0~~1~~	~~-9.9~~ ~~2.2~~	~~-~~ ~~+01.0~~ ~~8~~	114.4	~~+11.4~~ ~~+4.3~~	-5.6	58.3	~~-~~ ~~3.9+0~~ ~~.2~~	+0.6	54.5	~~-~~ ~~34.8~~ ~~21.2~~	-30.3
YRD	2.1	~~+2.1~~ ~~-2.2~~	-4.7	114.1	~~-~~ ~~11.2+~~ ~~1.9~~	+5.2	76.3	~~-0.9~~ ~~1.5~~	+5.5	34.0	~~-~~ ~~43.8~~ ~~36.9~~	-54.9
FWP	~~1.9~~ ~~2.0~~	~~+0.3~~ ~~-1.6~~	~~+~~+10.~~99~~	122.8	~~-~~ ~~12.1+~~ ~~7.2~~	-25.2	59.9	~~-2.9~~ ~~1.5~~	+3.3	51.6	~~-~~ ~~44.2~~ ~~32.7~~	-43.8
PRD	2.0	~~+1.9~~ ~~+0.9~~	-10.4	118.5	~~+16.2~~ ~~+12.7~~	+14.4	79.7	~~-8.0~~ ~~3.0~~	+10.3	3.1	~~-~~ ~~10.3~~ ~~6.4~~	-34.3
NEC	2.7	~~+3.6~~ ~~+3.6~~	+12.9	55.8	~~-33.7~~ ~~10.1~~	-38.4	61.6	~~-2.8~~ ~~3.4~~	-5.8	13.6	~~-~~ ~~30.8~~ ~~19.4~~	-12.4
CEN	1.8	~~+3.2~~ ~~-2.1~~	+0.4	172.1	~~-~~ ~~9.4+2~~ ~~.2~~	-2.8	77.9	~~-~~ ~~1.9+0~~ ~~.4~~	+6.9	30.3	~~-~~ ~~27.9~~ ~~19.7~~	-23.2
SWC	1.7	~~+3.7~~ ~~+4.4~~	+12.2	180.7	~~-13.3~~ ~~6.9~~	-16.3	74.7	~~-~~ ~~0.9+1~~ ~~.5~~	+5.7	11.1	~~-~~ ~~12.1~~ ~~13.7~~	-12.4

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NWC	1.9	~~-~~8.4~~-~~3.3	+4.3	146.8	~~-~~2.7~~+6.3~~	-9.5	58.5	~~-~~1.5~~-~~0.1	+2.8	20.2	~~-~~14.7~~-~~9.6	-6.6

Note: "~~+~~" ~~increased~~+" increased; "~~-~~" ~~decreased~~-" decreased

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2.3. EMI – the Environmental Meteorological Index

Due to the complicated interactions of emissions, meteorology and atmospheric processes, a single set of meteorological factors or a combination of them cannot quantitatively attribute the individual factor to the changes of concentration observed.

In order to quantitatively assess the impacts of meteorological conditions to the changes of air pollution levels, an index EMI (Environmental Meteorological Index) is defined as follows. For a defined atmospheric column (h) at a time *t*, an EMI is defined as an indication of atmospheric pollution level:

$$\mathrm{EMI}(t) = \mathrm{EMI}(t0) + \int_{t0}^{t} \Delta\,\mathrm{EMI} * \mathrm{dt} \quad (1)$$

where the ΔEMI is the tendency that causes the changes of pollution level in a time interval dt defined as:

ΔEMI = iEmid + i*Tran* + i*Accu* (2)

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where the iEmid is the difference between emission and deposition, and i*Tran* and i*Accu* are the net (in minus out) advection transports and the vertical accumulation by

turbulent diffusion in the column, respectively. A positive sign of each factor indicates a net flow of pollutants into the column, and vise ~~visa~~versa.

Mathematically, these factors are expressed as:

$$iTran = \frac{1}{hC_0}\int_0^h \left(u\frac{\partial C}{\partial x} + v\frac{\partial C}{\partial y} + w\frac{\partial C}{\partial z}\right) dz$$

$$iAccu = \frac{1}{hC_0}\int_0^h \left[\frac{\partial C}{\partial x}\left(Kx\frac{\partial C}{\partial x}\right) + \frac{\partial C}{\partial y}\left(Ky\frac{\partial C}{\partial y}\right) + \frac{\partial C}{\partial z}\left(Kz\frac{\partial C}{\partial z}\right)\right] \mathrm{dz}$$

$$iEmid = \frac{1}{hC_0}\int_0^h [Emis - (V_d + L_d)]\, dz \qquad (3)$$

where the tendency is normalized by a factor C_0. For an application of EMI to the $PM_{2.5}$, C_0 is set to equal 35 μg m^{-3}, the national standard for $PM_{2.5}$ in China (MEE, 2012), and the EMI(t) is written as $EMI(t)_{2.5}$. If the $EMI_{2.5}$ is less than 1, the concentration level will reach or be better than the national standard.

It can be seen here that these key parameters account for the major meteorological factors which control the air pollutant levels, including wind speed and directions (*u, v, w*), turbulent diffusion coefficients ~~mixing~~ (K_x, K_y, K_z) as well as dry and wet depositions (V_d and L_d). Therefore, under the conditions of an unchanged emissions (*Emis*), the *EMI* variation reflects the impacts of meteorological factors on the levels of atmospheric pollutants. Furthermore, because of the inclusion of individual factors such as *iTran, iAccu* and *iEmid*, the variation of $EMI(t)_{2.5}$ can be attributed to the variation of

each factor, which gives more detailed information ~~on~~ to the meteorological influence ~~to~~ on the ambient pollutant concentration variations. It should be pointed out that the current EMI index has only been accounted explicitly for three major physical processes of iTran, iAccu, and iEmid that are closely related to the meteorological influences. However, the secondary formation of aerosols is only implicitly considered in the EMI as the three major physical processes are calculated from the concentrations of aerosols (C) as indicated in Equation (3).

For a period of time p (t0 to t1) when the averaged pollutant level (e.g. $PM_{2.5}$) is compared with $EMI(t)_{2.5}$, the time integral has to be done to obtain the averaged index for the period, such as:

$$\overline{EMI(p)_{2.5}} = \frac{1}{t1-t0}\int_{t0}^{t1} EMI(t)_{2.5}dt \quad (4)$$

The relationship among the ΔEMI, $EMI(t)_{2.5}$ and $\overline{EMI(p)_{2.5}}$ is illustrated in Figure 3. It is clear that the $EMI(t)_{2.5}$ is a function of time and can be used to reflect the pollution level at any time t, while the $\overline{EMI(p)_{2.5}}$ is the area under the $EMI(t)_{2.5}$ from time t0 to t1, which gives the averaged pollution levels for the period. The derivatives of $EMI(t)_{2.5}$ are the ΔEMI, which is a positive value when the pollution is being accumulated and a negative value when the pollution is being dispersed.

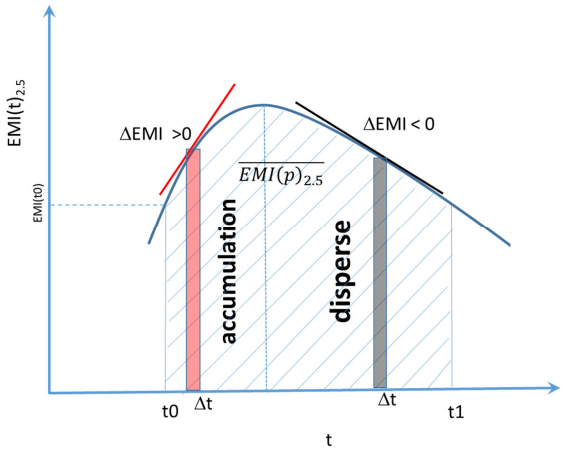

Figure 3: Relationship between the ΔEMI, EMI(t)$_{2.5}$ and $\overline{EMI(p)_{2.5}}$.

Therefore, for the period p with ~~n~~n discrete steps from t0 to t1, the $\overline{EMI(p)_{2.5}}$ represents the averaged meteorological influences on $PM_{2.5}$, while the sum of the positive ΔEMI is the accumulation potentials and the sum of the negative ΔEMI is the dispersing potentials as illustrated in Figure ~~2~~3. The relationship between them is derived as follows:

$$\overline{EMI(p)_{2.5}} = \frac{1}{n}[EMI(0) + EMI(1) + EMI(2) + \cdots + EMI(n-1)]$$

$$= \frac{1}{t1 - t0n}[nEMI(t0)\Delta t + (n-1)\Delta EMI(1)\Delta t + (n-2)\Delta EMI(2)\Delta t + (n-3)\Delta EMI(3)\Delta t + (n-4)\Delta EMI(4)\Delta t + \cdots + \Delta EMI(n-1)\Delta t] \quad (5)$$

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where n is the time steps in the period and the averaged EMI has been linked to the starting point EMI(~~t~~0) and the changing rates of EMI, i.e. ΔEMI(~~n~~n), at each time step. For monthly simulations, the initial values EMI(t0) for each month was set up by the averaged $PM_{2.5}$ concentrations for the first day from 2013 to 2019 divided by the constant C_0 (35 μg m^{-3}).

2.4. Assessment Framework of Emission Controls

The $EMI_{2.5}$ index provides a way to assess the meteorological impacts on the changes of $PM_{2.5}$ concentrations at two time periods, i.e. January ~~2103~~ 2013 (p0) and January 2016 (p1) under the assumption of unchanged emissions. However, due to the national efforts of improving air quality, the year-by-year emissions are changing rapidly and unevenly across the country. The changes in both emissions and meteorology are tangled together to yield the observed changes in ambient concentrations. For policy makers, the emission reduction quantification is critical to guide the further air quality improvements. The framework proposed here is to combine~~d~~ changes in the observed concentration levels and meteorology factors $\overline{EMI(p)_{2.5}}$ to quantify the changes ~~in~~caused by emission changes only at two time periods.

The observed concentrations at *p0* and *p1* are defined as PM~~PM~~ (*m0*, *e0*) and PM ~~PM~~ (*m1*, *e1*) where (*m0, e0*) and (*m1*, *e1)* indicate the meteorology and emission status

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at *p0* and *p1*, respectively. The contribution to the observed concentration changes between *p0* and *p1* by sole emission changes or control measures is defined as:

$$\Delta EMIS = \frac{PM(m0,e1)-PM(m0,e0)}{PM(m0,e0)} \times 100\% \quad (6)$$

where $PM\ (m0, e1)$ is an assumed concentration of pollutant under the conditions of unchanged meteorology at *p0* but with new emission at *p1*, which cannot be observed. Since ~~T~~the ratio of $\overline{EMI(p0)_{2.5}}/\overline{EMI(p1)_{2.5}}$ can be used to reflect~~s~~ the impact ratio of sole meteorology variations on the concentrations ~~from~~ between *p0* ~~to~~ and *p1* with the same emissions at ~~P~~p1. Therefore, $PM\ (m0, e1)$ is estimated from the averaged EMI ratio and the observed concentrations at *p1* as follows:~~:~~

$$\frac{PM(m0,e1)}{PM(m1,e1)}PM(m0,e1) = \frac{\overline{EMI(p0)_{2.5}}}{\overline{EMI(p1)_{2.5}}} \times PM(m1,e1) \quad (5~~6~~7)$$

~~which gives an assumed concentration of pollutant under the conditions of unchanged meteorology at *p0* but with new emission at *p1*. In a simple statement, the PM (*m0, e1*) is the concentration at *p1* only under the influence of emission change from *e0* to *e1* but with the same meteorology (*m0*). Consequently, the impact of only emission changes from *e0* to *e1* on the concentration changes can be expressed as:~~

$$\Delta EMIS = \frac{PM(m0,e1)-PM(m0,e0)}{PM(m0,e0)} \times 100\% \quad (6~~7~~)$$

~~which can be used to assess the impact of emission changes (control measures) on the air pollutants.~~

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2.5. Quantitative Estimate of EMI

Finally, a process-based method is developed to calculate the EMI and its components, i.e. *jEmid*, *jTran* and *jAccu*. The main modeling frame-work used is the chemical weather modeling system MM5/CUACE, which is a fully coupled atmospheric model used at CMA for national haze and air quality forecasts (Gong and Zhang, 2008;Zhou et al., 2012). CUACE is a unified atmospheric chemistry environment with four major functional sub-systems: emissions, gas phase chemistry, aerosol microphysics and data assimilation (Niu et al., 2008). Seven aerosol components, i.e. sea salts, sand/dust, EC, OC, sulfates, nitrates and ammonium salts are sectioned in 12 size bins with detailed microphysics of hygroscopic growth, nucleation, coagulation, condensation, dry depositions and wet scavenging in the aerosol module (Gong et al., 2003). The gas chemistry module is based on the second generation of Regional Acid Deposition Model (RADM Ⅱ) mechanism with 63 gaseous species through 21 photo-chemical reactions and 121 gas phase reactions applicable under a wide variety of environmental conditions especially for smog (Stockwell et al., 1990) and prepares the sulfate and SOA production rates for the aerosol module and for the aerosol equilibrium module ISORROPIA (Nenes et al., 1998) to calculate the nitrate and ammonium aerosols. This is the default method to treat the secondary aerosol formations in CUACE. For the EMI application of CUACE, another option was also adapted to compute the secondary aerosol formations by a highly parameterized method (Zhao et al., 2017), that computes the aerosol formation rates directly from the pre-cursor emission rates of SO_2, NO_2 and

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VOC. This option was added to facilitate timely operational forecast requirements for CMA. Both primary and pre-cursor emissions of PM are based on the 2016 MEIC Inventory (http://www.meicmodel.org/) developed by Tsinghua University for China.

In order to quantitatively obtain each term defined in Equation 3, the CUACE model was modified to extract the change rates for the processes involved. Driven by the re-analysis meteorological data, the new system CUACE/EMI can be used to calculate each term in ΔEMI at each time step (Δt).

In summary, this section presents a systematic platform to separate and assess the impacts of the meteorology and emissions on the ambient concentration changes. The $\overline{EMI(p)}_{2.5}$ and ΔEMIS form the basis for the assessment. In the Results and Discussions section, the application of the platform is presented to assess the fine particular matter ($PM_{2.5}$) changes in China. ~~It should be pointed out that the current EMI index has only been accounted explicitly for three major physical processes of iTran, iAccu, and iEmid that are closely related to the meteorological influences. However, the secondary formation of aerosols is only implicitly considered in the EMI as the three major physical processes are calculated from the concentrations of aerosols (C) as indicated in Equation (3).~~

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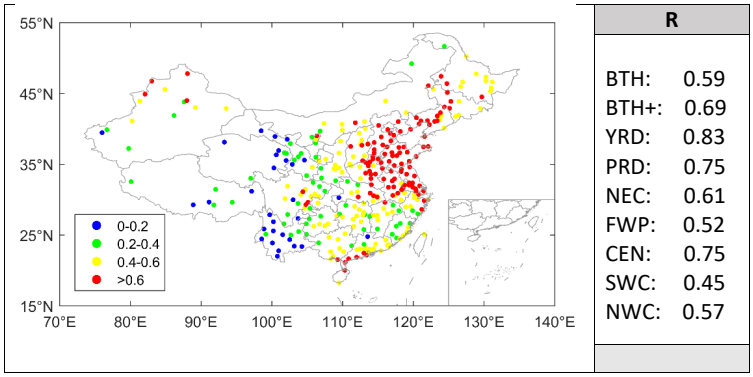
3. Results and Discussions

3.1. Validation of EMI by Observations

Under the conditions of no changes in annual emissions for $PM_{2.5}$ and its precursors, the daily $EMI_{2.5}$ was computed by CUACE from 2013 to 2019 on a 15×15 km resolution across China and accompanied by its contribution components: *iTran*, *iAccu* and *iEmid*. However, in order to reflect the significant changes of industrial and domestic energy consumptions within a year in China, a monthly emission (Wang et al.) ~~Wang et al.~~ (variation was applied to the emission inventory for computing the $EMI_{2.5}$, which is more realistically reflecting the meteorology contributions to the $PM_{2.5}$ concentrations.

To evaluate the applicability of $EMI_{2.5}$, the index was compared with the observed $PM_{2.5}$ concentrations. Figure 4 shows the spatial distribution of correlation coefficients between $PM_{2.5}$ and $EMI_{2.5}$ for 2017 for all China. The correlation coefficients between $EMI_{2.5}$ and $PM_{2.5}$ concentrations are greater than 0.4 for most of the Eastern China and greater than 0.6 for most of the assessment regions. Less satisfactory correlation was found in Western China, possibly due to complex terrain and less accurate emission data over there. Furthermore, due to the uncertainty in emissions and the difference in model performance for year-to-year meteorology simulations, the correlation coefficients may differ for different years. Overall, the good correlation between them merits the application of $EMI_{2.5}$ to quantify the meteorology impact on $PM_{2.5}$.



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BTH:	0.59
BTH+:	0.69
YRD:	0.83
PRD:	0.75
NEC:	0.61
FWP:	0.52
CEN:	0.75
SWC:	0.45
NWC:	0.57

Figure 4: Correlation coefficients (R) between the $EMI_{2.5}$ and the observed $PM_{2.5}$ daily concentrations across China for 2017 and for typical regions averaged between 2013 and 2019.

To further illustrate the applicability of $EMI_{2.5}$, the difference of various conditions between December 2014 and December 2015 in BTH ~~rejoin~~ region was also analyzed when a significant change of air quality and meteorological conditions occurred. The winter of 2015 was accompanied by a strong El Nino (ENSO) event, resulting in significant anomalies for meteorological conditions in China. Analysis shows that the meteorological conditions in December 2015 (compared to December 2014) had several important anomalies, including that the surface southeasterly winds were significantly enhanced in the North China Plain (NCP) and the wind speeds were decreased in the middle-north of eastern China, while slightly increased in the south of eastern China. Study suggests that the 2015 El Nino event had significant effects on air pollution in eastern China, especially in the NCP region, including the capital city of

Beijing, in which aerosol pollution was significantly enhanced in the already heavily polluted capital city of China (Chang et al., 2016).

Figure 5 shows the monthly average $EMI_{2.5}$, $PM_{2.5}$ and the contribution of sub-index to total $EMI_{2.5}$ in December 2014 and 2015 over BTH region. The monthly average $EMI_{2.5}$ increases about 54.9% from 2.1 in December 2014 to 3.2 in December 2015, indicating worsening meteorological conditions for $PM_{2.5}$ pollution. The increase of $EMI_{2.5}$ is mainly contributed by adverse atmospheric transport conditions (Fig. 5c), which results in the increase of $EMI_{2.5}$ reaching 3.2. With the increase of background concentration, the deposition and vertical diffusion also increase, and offset the impact of adverse transport conditions to some extent.

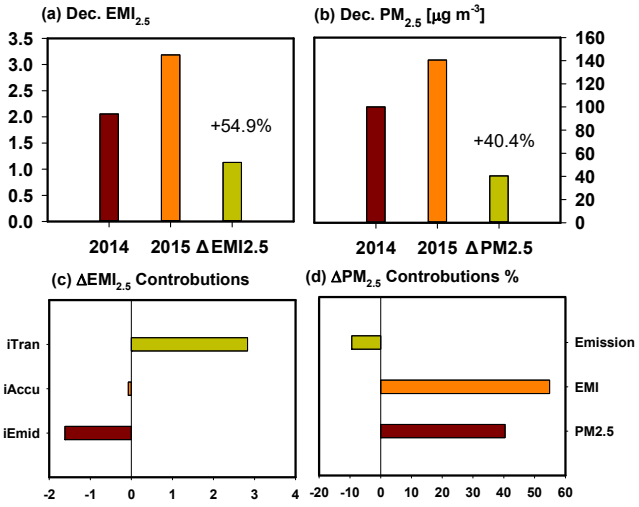

Figure 5: (a) ~~The~~ the monthly averaged $EMI_{2.5}$ ~~(a),~~ and ~~(b)~~ b) monthly $PM_{2.5}$ ~~(b)~~ for Decembers of 2014 and 2015 over BTH. (c) contributions of sub-index to the $EMI_{2.5}$ change and (d)

contributions of emission and meteorology changes to $PM_{2.5}$ change for Decembers from 2014 to 2015, respectively.

~~and the contributions of sub-index (c) and contributions to the $PM_{2.5}$ changes (d) in Decembers 2014 and 2015 over BTH.~~

The worsening meteorological conditions represented by $EMI_{2.5}$ were also supported by the observations for the two periods. The observed day with small wind (DSW, wind speed less than 2 m s^{-1}) reveals that, except for part of southern Hebei province, the DSW increases 5-15 days for 2015 in most meteorological stations in BTH region (Fig. 6a), which indicates a large decrease of local diffusion capability. The comparison of wind rose map shows that the decrease of northwest wind and the increase of southwest and northeast wind occurred in December 2015 (Fig. 6b). The change of wind fields indicates more pollutants were transported to BTH region from Shandong, Jiangsu, Henan, and Northeast China. These variations indirectly validate the conclusions of adverse atmospheric transport conditions with high iTran in December 2015.

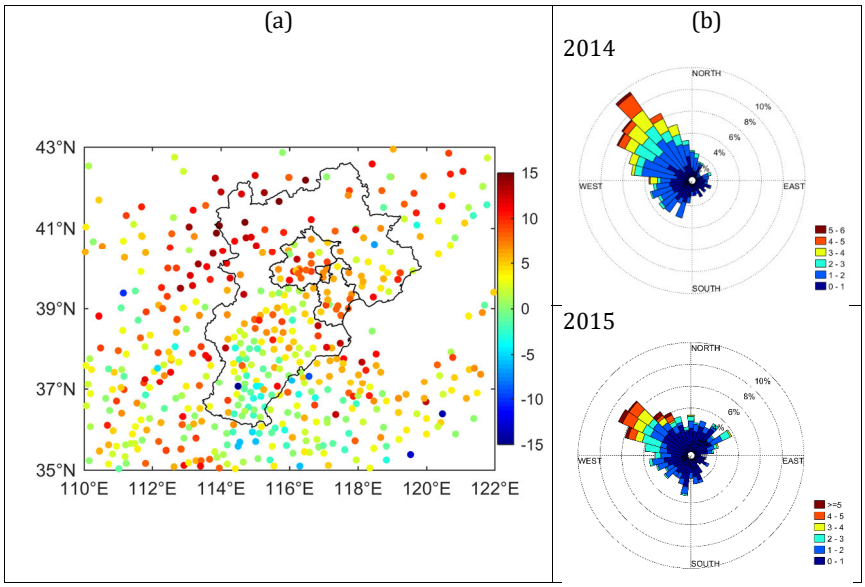

Figure 6: (a) The change of DSW (days) from December 2014 to December 2015 (December 2015 – December 2014) and (b) Wind rose maps in December 2014 and December 2015 over BTH region.

Based on the assessment method of emission contribution to the observed trend (Eqs. ~~5~~ 6 and ~~6~~7), the emissions reduction in December 2015 as compared to 2014 was estimated to contribute about 9.4% (Fig. 5d) to the $PM_{2.5}$ concentration decrease, compensating the large increase caused by meteorology, which is comparable with previous studies of about 8.6% reduction in emissions (Liu et al., 2017;He et al., 2017a) for the same two months. In other words, without the regional emission reduction efforts, the observed $PM_{2.5}$ concentration in December 2015 would have had a similar rate of 54.9% increase as the worsening meteorology conditions would bring about as compared with

December 2014. This assessment of emission reduction is supported by the estimate of emission inventories for the BTH region in the Decembers of 2014 and 2015 by Zheng et al. (2019) who found out that the monthly emission strengths for $PM_{2.5}$, SO_2, NOx, VOCs and NH_3 in 2015 were reduced by 22.0%, 6.9%, 2.5%, 2.5% and 2.5%, respectively, as compared with 2014. The sensitivity and the nonlinear response of $PM_{2.5}$ concentrations to the air pollutant emission reduction in the BTH region (Zhao et al., 2017) have been estimated to be about 0.43 for both primary inorganic and organic $PM_{2.5}$, 0.05 for SO_2, -0.07 for NOx, 0.15 for VOCs, 0.1 for NH_3. Combining the emission reduction percentages between Decembers 2014 and 2015 and the nonlinear response of emissions to the $PM_{2.5}$ concentrations results in an approximately 10.2% ambient $PM_{2.5}$ concentration reduction due to the emission changes. This is very close to the estimate of emission reduction contribution to the December $PM_{2.5}$ concentration difference of about 9.4% between 2014 and 2015 by the EMI framework.

3.2. $PM_{2.5}$ Trends and Meteorological Contributions

The annual averaged $PM_{2.5}$ concentrations in China have been decreased significantly from 2013 to 2019. Figure 7 shows the observed spatial distribution of national $PM_{2.5}$ concentrations from ~~2103~~ 2013 to 2019, respectively. These spatial distributions are consistent with those of primary and precursor emissions of $PM_{2.5}$ (Wang et al.), pointing out ~~to~~ the fundamental cause of the air pollution in China. From the spatial distributions, it is clear that the regions of BTH, FWP, CEN and NWC had the

highest $PM_{2.5}$ concentrations among the 9 regions. Even though the national concentrations have been reduced significantly from 2013 by reducing emissions, the pollution center of particular matters has not been changed very much, locating at the southern Hebei Provence and indicating the macroeconomic structure has not been gone through a great change yet. Another phenomenon can be seen from the distribution is that in the North-west China, especially in some cities of the Xinjiang and Ningxia Provinces, the $PM_{2.5}$ concentrations were on an increasing trend, due to certain migrating industries from developed regions in East China.

	A	B

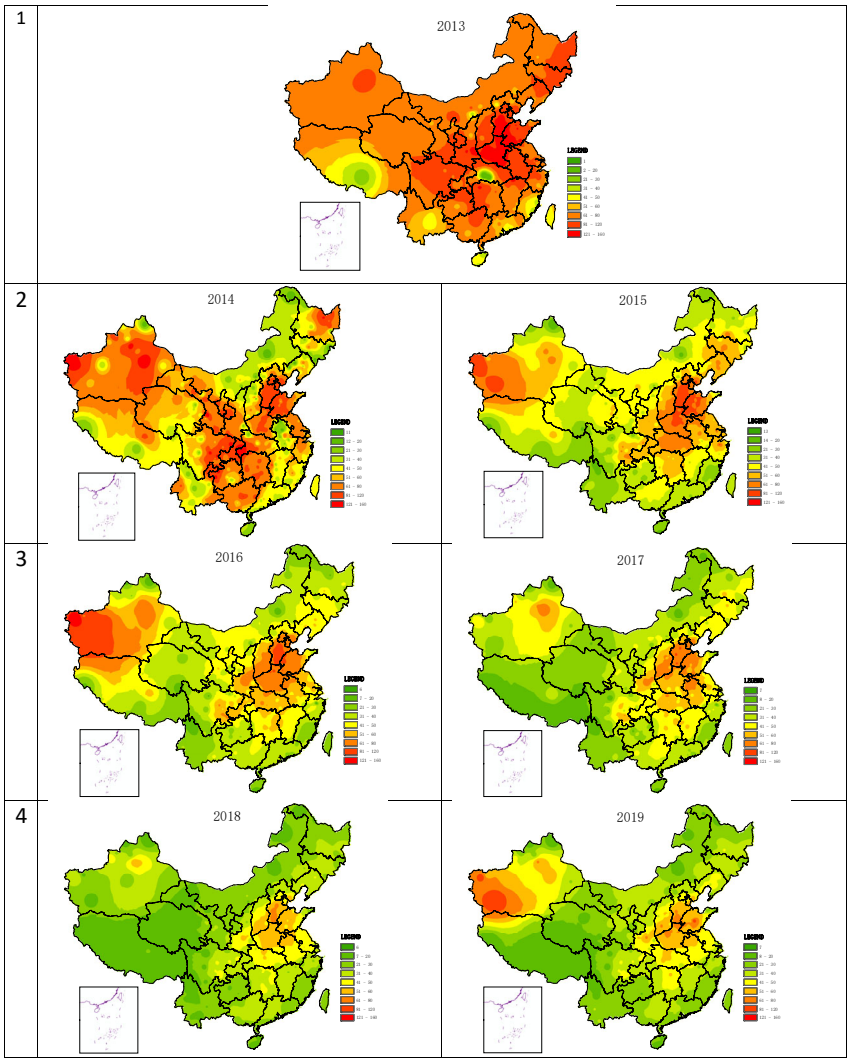

Figure 7: Regional annual $PM_{2.5}$ concentration distributions from 2013 to 2019.

Averaged for the nation, 9 focused regions and Beijing, the $PM_{2.5}$ trend lines were shown in Figure 2. It is seen that all regions have had a large reduction of more

than 35% in surface $PM_{2.5}$ concentrations in 2019 as compared with those in 2013. The averaged national annual concentration at 36 μg m^{-3} has been very close to the national standard of 35 μg m^{-3} while the concentrations in PRD, SWC and NEC regions have been below the standard. Regions above the standard are BTH+, BTH, YRD, CEN and FWP. Regionally, the largest drop percentage of $PM_{2.5}$ was seen in NEC and NWC regions (Fig. 8), reaching over 50% compared with 2013. In the BTH, BTH+, FWP and CEN regions, the reduction was in the range of 45% to 50% while in YRD and PRD the reduction was around 35%.

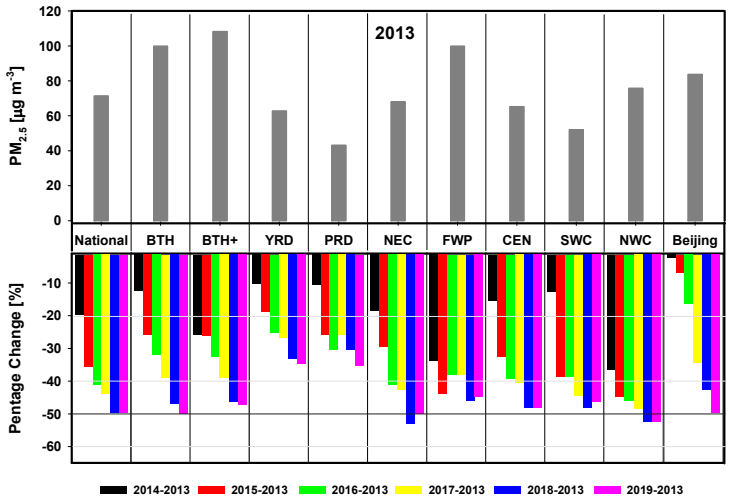

Figure 8: Annual averaged $PM_{2.5}$ concentrations in 2013 (top) and corresponding changing rates (bottom) from 2014 to 2019 as compared with 2013 for the nation, 9 regions and Beijing City.

As one of the key factors in controlling the ambient $PM_{2.5}$ concentration variations, the annual meteorological fluctuations, i.e. $EMI_{2.5}$, from 2014 to 2019 with 2013 as the base year, are shown in Figure 9 for nine regions. Generally, the annual $EMI_{2.5}$ shows a positive or negative variation, reflecting the meteorological features for that specific region. Except for a couple of regions or years, most of the fluctuations are within 5% as compared with 2013 and have a no definite trend. It can be inferred that the meteorological conditions are possibly responsible for about 5% of the annual $PM_{2.5}$ averaged concentration fluctuations from 2013 to 2019 (Fig. 9 middle). This is consistent with what has been assessed in Europe by Andersson et al. (2007).

The variations in meteorological contributions ($EMI_{2.5}$) to $PM_{2.5}$ for the heavy pollution seasons of fall and winter (October 1 to March 31) generally follow the same fluctuating pattern as the annual average but are much larger than the average (Fig. 9 bottom), over 5% for most of the regions and years. For specific regions and years, e.g. BTH, YRD, NEC, SWC and CEN, the variations are between 10-20% as compared with 2013. Since the $PM_{2.5}$ concentrations are much higher in the pollution season, the larger meteorology variations in fall-winter would exercise more controls to the heavy pollution episodes than the annual averaged concentrations, signifying the importance of meteorology in regulating the winter pollution situations.

It is found that though most of the regions have a fluctuating $EMI_{2.5}$ in the pollution season during the 2014-2019 period (Fig. 9 bottom), the YRD and FWP show a consistent favorite and ~~un-favorite~~unfavorite meteorological conditions, respectively.

BTH has witnessed the same ~~un-favorite~~unfavorite conditions as FWP except in 2017. In other words, in BTH and FWP, the decrease in ambient concentrations of $PM_{2.5}$ from 2014 to 2019 has to overcome the difficulty of worsening meteorological conditions with larger control efforts.

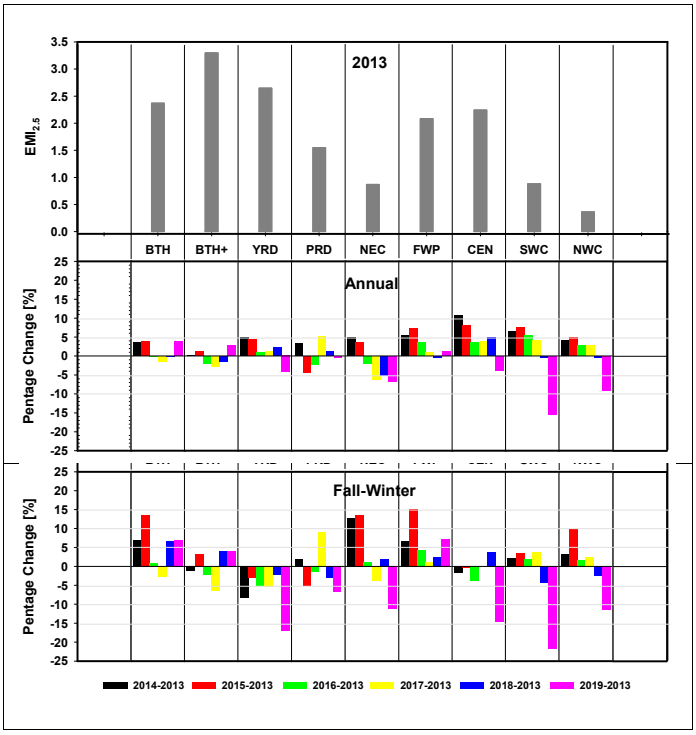

Figure 9: Annual averaged $EMI_{2.5}$ in 2013 (top) and corresponding changing rates for annual average (middle) and for fall-winter seasons (bottom) from 2014 to 2019 as compared with 2013 in 9 regions.

3.3. Attribution of Control Measures to the $PM_{2.5}$ Trend

As it is well known that the final ambient concentrations of any pollutants are resulted from the emission, meteorology and atmospheric physical and chemical processes. Separating emissions and meteorology contributions to the pollution level reduction entails a combined analysis of them. The analysis in Section 3.2 shows that from 2013 to 2019, the national averaged $PM_{2.5}$ as well as those for 9 separate regions were all showing a gradual decline trend (Fig. 8). By 2019, 45% - 50% of reductions in surface $PM_{2.5}$ concentrations were achieved while the meteorology contributions did not show a definite trend as from 2013, clearly pointing out the contribution of emission reductions in the trend. Using the analysis framework for separating emissions from meteorology based on the monitoring data of $PM_{2.5}$ and $EMI_{2.5}$ (Section 2.4), the emission change contributions are estimated.

Figure 10 shows the 2013 base emissions of $PM_{2.5}$ (Zhao et al., 2017) and the annual changes in the emission contributions to the $PM_{2.5}$ concentrations from 2014 to 2019 as estimated from the $EMI_{2.5}$ and observed $PM_{2.5}$. For the emissions, it is found that the unit area emissions match better with ambient concentrations of $PM_{2.5}$ in regions than the total emissions and the high emission regions are BTH, BTH+, YRD, PRD and FWP in 2013. Nationally by 2019, the emission reduction contributions to the ambient $PM_{2.5}$ trend accounted for ranging from 32% to 52% of the total $PM_{2.5}$ decrease percentage, while in BTH and BTH+ regions the reduction was more than 49% from 2013 base year emissions, leading the national emission reduction campaign. The emission reduction rates clearly illustrate the effectiveness of the national-wide emission control strategies implemented since 2013 and the emission

reduction is the dominate factor for ambient $PM_{2.5}$ declining trend in China. Taking the analysis data of $PM_{2.5}$ and $EMI_{2.5}$ from this study for BTH+ region from 2013 to 2017, it is found that control strategy contributed more than 90% to the $PM_{2.5}$ decline. Chen et al (2019) has estimated that the control of anthropogenic emissions contributed to 80% of the decrease in $PM_{2.5}$ concentrations in Beijing from 2013 to 2017.

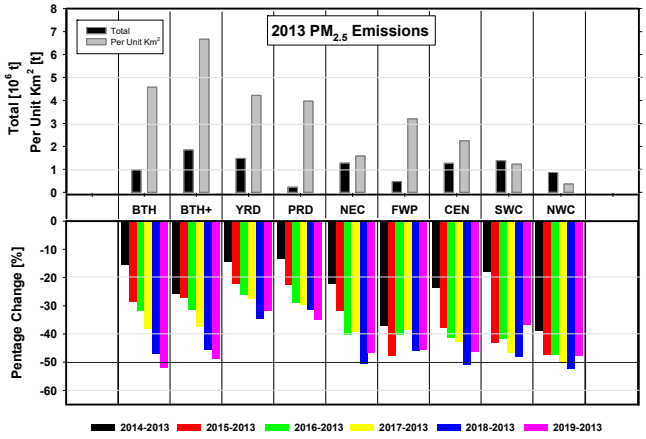

Figure 10: Annual $PM_{2.5}$ emissions (total and per unit Km^2) for 2013 (top) and corresponding changing rates (bottom) from 2014 to 2019 as compared with 2013 in 9 regions.

Regionally, the emission reduction trends from 2014 to 2019 display some unique characteristics. For the regions of BTH, BTH+ and PRD, the year-by-year reduction rate is consistent, indicating that regardless of fluctuations in meteorology, these regions have had an effective emission control strategy and maintained the emission reduced year by year since 2014. However, in some regions such as FWP, NEC, SWC and NWC, the emission reduction rates were fluctuating from 2014 to 2019, implying the emissions in these regions were increased in

certain years. Especially in FWP from 2016 to 2017, the emissions were estimated to be increased by about 10%, and then decreased in 2018 and 2019, despite of the factor that FWP has experienced ~~un-favorite~~unfavorite meteorological conditions during this period.

The year of 2015 is a special year in the history of China air pollution control. Though the systematical and network observations of $PM_{2.5}$ started in China from 2013, it took about two years (until 2015) to evolve to the current status in terms of spatial coverage and observational station numbers, establishing a consistent and statistically comparable national network. At the same year, the Environmental Protection Law of People's Republic of China was taken into effect in January, signalizing the stage of lawfully control of air pollution. For the regulation assessment point of view, the impact by emission changes from 2015 was relevant to the interests of management to show how effective the law was.

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Table 2 summarizes the $PM_{2.5}$ difference between 2019 and 2015 and the relative contributions of meteorology and emission changes to the difference for all China, Beijing and nine regions. Once again, as of the end of 2019, the $PM_{2.5}$ concentrations are all reduced from 2015, ranging from -1.8% in FWP to -46.2% in Beijing. During this period of time, regions of BTH, BTH+, PRD and Beijing had encountered ~~un-favorite~~unfavorite meteorological conditions with positive $EMI_{2.5}$ changes, which indicated that for these regions, emission reductions were not only to maintain the decline trend but also to offset the un- favorite meteorological conditions in order to achieve the observed reductions in ambient $PM_{2.5}$ concentrations. On the contrary, for the regions of FWP and SWC, the emission control impacts were to deteriorate the concentrations, implying an increase in emissions to restrain the $PM_{2.5}$ concentration decrease by favorite meteorological conditions. For other regions, both meteorology and emission

controls contributed to $PM_{2.5}$ decrease from ~~2105~~ 2015 to 2019, with the control measures contributing from -7.9% in NWC to -68.4% in NEC (Table 2).

Therefore, due to the diversity of meteorological conditions and emission distributions in China, their impacts on ambient $PM_{2.5}$ concentrations display unique reginal characteristics. Overall, the emission controls are the dominant factor in contributing the decline trend in China from 2013 to 2019. However, in certain regions or certain period of years, emissions were found to be increased even with favorites ~~and the~~ meteorological ~~dominance did occur~~conditions, which means the design of national control strategies has to take both meteorology and emission impacts simultaneously in order to achieve maximum results.

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Table 2: Observed $PM_{2.5}$ difference between 2019 and 2015 as well as its attributions to meteorology and control measures for all China, Beijing and nine regions.

Regions	Observed $PM_{2.5}$ Difference		Attributions			
			Meteorology (EMI)		Emission Controls	
	($\mu g\ m^{-3}$)	(%)	($\mu g\ m^{-3}$)	Relative %	($\mu g\ m^{-3}$)	Relative %
National	**-10**	**-21.7**	**-4.1**	**-40.9**	**-5.9**	**-59.1**
BTH	**-24**	**-32.4**	**+0.1**	**+0.4**	**-24.1**	**-100.4**
BTH+	**-23**	**-28.8**	**+1.2**	**+5.4**	**-24.2**	**-105.4**
YRD	**-10**	**-19.6**	**-4.0**	**-39.7**	**-6.0**	**-60.3**
PRD	**-4**	**-12.5**	**+1.4**	**+36.0**	**-5.4**	**-136.0**
NEC	**-14**	**-29.2**	**-4.4**	**-31.6**	**-9.6**	**-68.4**
FWP	**-1**	**-1.8**	**-3.6**	**-362.2**	**+2.6**	**+262.2**
CEN	**-12**	**-23.1**	**-5.5**	**-45.5**	**-6.5**	**-54.5**
SWC	**-4**	**-12.5**	**-8.5**	**-211.5**	**+4.5**	**+111.5**
NWC	**-6**	**-14.3**	**-5.5**	**-92.1**	**-0.5**	**-7.9**
Beijing	**-36**	**-46.2**	**+3.4**	**+9.4**	**-39.4**	**-109.4**

Note: "~~+" increased~~+" increased; "; "~~-" decreased~~-" decreased

4. Conclusions

Based on a 3-D chemical transport model and its process analysis, an Environmental Meteorological Index ($EMI_{2.5}$) and an assessment framework have been developed and applied to the analysis of the $PM_{2.5}$ trend in China from 2013 to 2019. Compared with observations, the $EMI_{2.5}$ can realistically reflect the contribution of meteorological factors to the $PM_{2.5}$ variations in the time series with impact mechanisms and can be used to as an index to judge whether the meteorological conditions ~~whether~~ are favorite or not to the $PM_{2.5}$ pollutions in a region or time

period. In conjunction to the observational trend data, the $EMI_{2.5}$-based framework has been used to quantitatively assess the separate contribution of meteorology and emission changes to the time series for 9 regions in China. Results show that for the period of 2013 to 2019, the $PM_{2.5}$ concentrations have been dropped continuously throughout China, by about 50% on national average. In the regions of NWC, NEC, BTH, BEIJING, CEN, BTH+, SWC, the reduction was in the range of 46% to 53% while in FWP, PRD and YRD, the reduction was from 45% to 35%. It is found that the control measures of emission reduction are the dominant factors in the $PM_{2.5}$ declining trends in various regions. By 2019, the emission reduction contributes about 47% of the $PM_{2.5}$ decrease from 2013 to 2019 on the national average, while in BTH region the emission reduction contributes more than 50% and in YRD, PRD and SWC regions, the contributions were between 32% and 37%. For most of the regions, the emission reduction trend was consistent throughout the period except for FWP, NEC, SWC and NWC where the emission amounts were increased for certain years. The contribution by the meteorology to the surface $PM_{2.5}$ concentrations from 2013 to 2019 was not found to show a consistent trend, fluctuating positively or negatively about 5% on annual average and 10-20% for the fall-winter heavy pollution seasons.

Acknowledgements

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