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

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# 12 Abstract

- 13 A framework was developed to quantitatively assess the contribution of meteorology
- variations to the trend of fine particular matter (PM<sub>2.5</sub>) concentrations and to separate the
- 15 impacts of meteorology from the control measures in the trend, based upon an
- 16 Environmental Meteorology Index (EMI). The model-based index EMI realistically reflects
- 17 the role of meteorology in the trend of PM<sub>2.5</sub> and is explicitly attributed into three major
- 18 factors: deposition, vertical accumulation and horizontal transports. Based on the 2013-
- 19 2019 PM<sub>2.5</sub> observation data and re-analysis meteorological data in China, the
- 20 contributions of meteorology and control measures in nine regions of China were
- 21 assessed separately by the EMI-based framework. Monitoring network observations
- 22 show that the PM<sub>2.5</sub> 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
- 24 control measures were the dominant factor in the declining trend of China PM<sub>2.5</sub>
- 25 concentrations, contributing to about 47% of the PM<sub>2.5</sub> 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<sub>2.5</sub>
 concentrations, 5% on annual average and 10-20% for the fall-winter heavy pollution
 seasons.

### 5 1. Introduction

6 Recent observation data from the Ministry of Ecology and Environment of China (MEE) 7 has shown a steady improvement of air quality across the country, especially in particular 8 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 9 10 particulate matter with an aerodynamic diameter of less than 2.5  $\mu$ m (PM<sub>2.5</sub>) in 74 major 11 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 12 13 trend is critical to assess the reduction strategies implemented by the government and to 14 guide future air quality control policy. However, the assessment of the improvements of 15 air quality is a complicated process that involves the quantification of changes in the emission sources, meteorological factors, and other characteristics of the PM<sub>2.5</sub> pollution, 16 which are also interacting with each other. In order to separate the relative degree of 17 these factors, a comprehensive analysis, including observational data and model 18 19 simulation, is needed.

1	Researches have been done extensively on the impacts of weather systems on air
2	quality. Synoptic and local meteorological conditions have been recognized to influence
3	the PM concentrations at various scales (Beaver and Palazoglu, 2006;He et al., 2017a;He
4	et al., 2017b;Pearce et al., 2011a;Pearce et al., 2011b). For the atmospheric aerosol
5	pollution, the dynamic effect of the downdraft in the "leeward slope" and "weak wind
6	area" of the Qinghai Tibet Plateau in winter is not conducive to the diffusion of air
7	pollution emissions in the urban agglomerations of eastern China (Xu et al., 2015;Xu et al.,
8	2002). The evolution of circulation situation is an important factor driving the change of
9	haze pollution (He et al., 2018). The local circulations, such as mountain and valley wind
10	and urban island circulation, have significant impact on local pollutant concentration
11	(Chen et al., 2009;Yu et al., 2016). Previous studies also revealed that $PM_{2.5}$ concentration
12	is significantly correlated with local meteorological elements, such as temperature,
13	humidity, wind speed, and boundary layer height (He et al., 2017b;Bei et al., 2020;Ma et
14	al., 2019;He et al., 2016).
15	In the Beijing-Tianjin-Hebei (BTH) Region, a correlation analysis and principal
16	component regression method (Zhou et al., 2014) was used to identify the major
17	meteorological factors that influenced the API (Air Pollution Index) time series in China
18	from 2001-2010, indicating that air pressure, air temperature, precipitation and relative
19	humidity were closely related to air quality with a series of regression formulas. Yet, the
20	analysis was assumed a relatively unchanged emission whose impacts were not taken into
21	account. On a local scale, an attempt (Zhang et al., 2017) has been made to correlate the
22	air pollutant levels with a combination of meteorological factors with the development of
	3   Page

1	the Stable Weather Index (SWI) at CMA. The SWI is a composite index which includes the
2	advection, vertical diffusion and humidity and other meteorological factors that are
3	related to the formation of air pollutions in a specific region or city. A higher value of SWI
4	means a weaker diffusion of air pollutants. This index had some success in assessing the
5	meteorological impacts on air pollution, especially calibrated for a specific region, i.e.
6	Beijing. However, when applied to different areas where the emission patterns and
7	meteorological features are different, this index failed to give a universal or comparable
8	indication of meteorological assessment of pollution levels across the nation.
9	Using the Kolmogorov-Zurbenko (KZ) wave filter method, Bai et al (2015) separated
10	the API time series in three Chinese cities into short-term, seasonal and long-term
11	components, and then used the stepwise regression to set up API baseline and short-term
12	components separately and established linear regression models for meteorological
13	variables of corresponding scales. Consequently, with the long-term representing the
14	change of emissions removed from the time series, the meteorological contributions
15	alone were assumed and analyzed, pointing out that unfavorable conditions often lead to
16	an increase by 1-13 whereas the favorable conditions to a decrease by 2-6 in the long-
17	term API series, respectively. Though the contributions of emissions and meteorological
18	variations were separated by the research, it was only done by mathematical
19	transformations and far from the reality. The mechanisms behind the variation of the time
20	series were not investigated.

1 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. 2 Andersson et al. (2007) used a CTM to study the meteorologically induced inter-annual 3 variability and trends in deposition of sulphur and nitrogen as well as concentrations of 4 5 surface ozone  $(O_3)$ , nitrogen dioxide  $(NO_2)$  and PM and its constituents over Europe during 6 1958-2001. It is found that the average European interannual variation, due to meteorological variability, ranges from 3% for O<sub>3</sub>, 5% for NO<sub>2</sub>, 9% for PM, 6-9% for dry 7 deposition, to about 20% for wet deposition of sulphur and nitrogen. A multi-model 8 9 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 10 11 trend exceeds the natural variability for primary compounds, concluding that that 12 emission management strategies have had a significant impact over the past 10 years, hence supporting further emission reductions strategies. Model assessments of air quality 13 14 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 15 Comprehensive Air Quality Model with extensions (CAMx) plus the Particulate Source 16 17 Apportionment Technology (PSAT) to simulate the contributions of emission changes in various sectors and changes in meteorology conditions for the PM<sub>2.5</sub> trend from 2006 to 18 2013. It was found that the change of source contribution of PM<sub>2.5</sub> in Beijing and northern 19 Hebei was dominated by the change of local emissions. However, for Tianjin, and central 20 and southern Hebei province, the change of meteorology condition was as important as 21 22 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.

3 There is no surprise that previous studies could not systematically catch the 4 meteorological impacts across the whole nation as the controlling meteorological factors involving the characteristics of plenary boundary layers (PBL), wind speed and turbulence, 5 temperature and stability, radiation and clouds, underlying surface as well as pollutant 6 7 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 8 9 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 10 the individual impacts of meteorology and emission changes, based on a model-derived 11 12 index EMI, i.e., Environmental Meteorology Index, and observational data, providing a 13 comprehensive analysis of the air quality trends in various regions of China, with mechanistic and quantitative attributions of various factors. 14

#### 15 **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<sub>2.5</sub>) provides the basis for the trend analysis. Separating the

- 1 trend contribution from regional emission reduction and meteorological variation entails
- 2 a framework, which is discussed below.



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

- 3
- 4 Figure 1: Analysis region separation and definition.

# 5 **2.1.** Particular Matter (PM) Observation Data

- 6 The observational pollution data of PM<sub>2.5</sub> concentrations used in this study were from
- 7 the monitoring network of the Ministry of Ecology and Environment (MEE) of China
- 8 (http://english.mee.gov.cn/). From 2013 to 2019, the concentrations have shown a large
- 9 change in the country where most regions see a declined trend in the annual
- 10 concentrations. Data show that from 2013 to 2019, the national annual averaged PM<sub>2.5</sub>

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<sub>2.5</sub> reduction
rate from 2013 ranges from 35 to 53%. Detailed analysis will be given in the Results and
Discussions section.



#### 6

7 Figure 2: National and regional trend lines of PM<sub>2.5</sub> in China from 2013 to 2019.

8 It is noted that the PM<sub>2.5</sub> mass concentrations by MEE are now reported under 9 observation site's actual conditions of temperature and pressure from September 1, 2018 10 before which the values were reported under the standard state (STP), i.e. 273 K and 11 101.325 kPa. In order to maintain the consistence of the data series, the PM<sub>2.5</sub> 12 concentrations used in this study have all been converted according to the new standard 13 (MEE, 2012)(GB3095-2012) under actual conditions. Research has shown that after the

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change of reporting standard, the PM<sub>2.5</sub> concentration in most cities decreased, and the
 number of good days to meet the standard increased (Zhang and Rao, 2019).

3 2.2. Meteorological Data

Conventional meteorological data can provide qualitative assessment of the 4 contributions of meteorological factors to the changes of air quality. The data used in this 5 6 study are from 843 national base weather stations of the CMA from 2013 to 2019. The 7 wind speed (WS), day with small wind (DSW), relative humidity (RH) and haze days are 8 used to analyze the pollution meteorological conditions. When the daily average wind speed is less than 2 m s<sup>-1</sup>, a DSW day is defined. Since the haze formation is always related 9 10 to stable meteorological conditions and high aerosol mass loading, haze observation from 11 CMA is also used to analyze the haze trends and the impact of air quality on visibility. A 12 haze day is defined with daily averaged visibility less than 10 km and relative humidity less 13 than 85% (Wu et al., 2014), excluding days of low visibility due to precipitation, blowing 14 snow, blowing sand, floating dust, sandstorms and smoke.

The 2019 national annual averaged WS has increased by 4.5%, DSW dropped by 15.1%, and RH decreased by 3.9% compared with 2013, with regional differences (Table 17 1). Slightly changes occurred when compared with 2015 that WS has decreased by 0.7%, 18 DSW dropped by 11.3%, and RH decreased by 2.2%. Overall, it can be seen that the annual 19 haze days have a certain degree of correlations negatively with WS and positively with

- 1 DSW. Detailed analysis linking PM<sub>2.5</sub> and meteorology will be given in the Results and
- 2 Discussions section.

3 Table 1: National and regional environmental meteorology in 2019 and comparisons with

4 2015 and 2013

	Wind Speed			Days with Small Wind			Relative Humidity			Haze (days)		
Region	AVG (m s <sup>-1</sup> )	vs 2015 (%)	vs 2013 (%)	Days	vs 2015 (%)	vs 2013 (%)	%	vs 2015 (%)	vs 2013 (%)	Days	vs 2015	vs 2013
National	2.2	-0.7	+4.5	129.8	-11.3	-15.1	60.1	-2.2	-3.9	25.7	-19.0	-21.2
BTH	2.0	-8.6	-2.2	131.0	+14.7	+9.0	56.7	-2.6	-4.2	45.2	-20.4	-26.1
BTH+	2.0	-9.9	-1.0	114.4	+11.4	-5.6	58.3	-3.9	+0.6	54.5	-34.8	-30.3
YRD	2.1	+2.1	-4.7	114.1	-11.2	+5.2	76.3	-0.9	+5.5	34.0	-43.8	-54.9
FWP	1.9	+0.3	+10.9	122.8	-12.1	-25.2	59.9	-2.9	+3.3	51.6	-44.2	-43.8
PRD	2.0	+1.9	-10.4	118.5	+16.2	+14.4	79.7	-8.0	+10.3	3.1	-10.3	-34.3
NEC	2.7	+3.6	+12.9	55.8	-33.7	-38.4	61.6	-2.8	-5.8	13.6	-30.8	-12.4
CEN	1.8	+3.2	+0.4	172.1	-9.4	-2.8	77.9	-1.9	+6.9	30.3	-27.9	-23.2
SWC	1.7	+3.7	+12.2	180.7	-13.3	-16.3	74.7	-0.9	+5.7	11.1	-12.1	-12.4
NWC	1.9	-8.4	+4.3	146.8	-2.7	-9.5	58.5	1.5	+2.8	20.2	-14.7	-6.6

5

Note: "+" increased; "-" decreased

# 6 **2.3.** EMI – the Environmental Meteorological Index

7 Due to the complicated interactions of emissions, meteorology and atmospheric

8 processes, a single set of meteorological factors or a combination of them cannot

9 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:

5 
$$EMI(t) = EMI(t0) + \int_{t0}^{t} \Delta EMI * dt$$
  
6 (1)  
7 where the  $\Delta EMI$  is the tendency that causes the changes of pollution level in a time

8 interval dt defined as:

9 
$$\Delta EMI = iEmid + iTran + iAccu$$
 (2)

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 vice versa.

## 14 Mathematically, these factors are expressed as:

15 
$$iTran = \frac{1}{hC_0} \int_0^h \left( u \frac{\partial C}{\partial x} + v \frac{\partial C}{\partial y} + w \frac{\partial C}{\partial z} \right) dz$$

16 
$$iAccu = \frac{1}{hC_0} \int_0^h \left[ \frac{\partial C}{\partial x} \left( Kx \frac{\partial C}{\partial x} \right) + \frac{\partial C}{\partial y} \left( Ky \frac{\partial C}{\partial y} \right) + \frac{\partial C}{\partial z} \left( Kz \frac{\partial C}{\partial z} \right) \right] dz$$

17 
$$iEmid = \frac{1}{hC_0} \int_0^h [Emis - (V_d + L_d)] dz$$

18

(3)

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

5 It can be seen here that these key parameters account for the major 6 meteorological factors which control the air pollutant levels, including wind speed and 7 directions (u, v, w), turbulent diffusion coefficients  $(K_x, K_y, K_z)$  as well as dry and wet 8 depositions ( $V_d$  and  $L_d$ ). Therefore, under the conditions of an unchanged emissions 9 (Emis), the EMI variation reflects the impacts of meteorological factors on the levels of 10 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 11 12 each factor, which gives more detailed information to the meteorological influence on 13 the ambient pollutant concentration variations. It should be pointed out that the 14 current EMI index has only been accounted explicitly for three major physical processes 15 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 16 17 the three major physical processes are calculated from the concentrations of aerosols (C) as indicated in Equation (3). 18

For a period of time p (t0 to t1) when the averaged pollutant level (e.g. PM<sub>2.5</sub>) is compared with EMI(t)<sub>2.5</sub>, the time integral has to be done to obtain the averaged index for the period, such as:

1 
$$\overline{EMI(p)_{2.5}} = \frac{1}{t_1 - t_0} \int_{t_0}^{t_1} EMI(t)_{2.5} dt$$
 (4)

The relationship among the  $\Delta$ EMI, EMI(t)<sub>2.5</sub> and  $\overline{EMI(p)}_{2.5}$  is illustrated in Figure 3. It is clear that the EMI(t)<sub>2.5</sub> is a function of time and can be used to reflect the 4 pollution level at any time t, while the  $\overline{EMI(p)}_{2.5}$  is the area under the EMI(t)<sub>2.5</sub> from 5 time t0 to t1, which gives the averaged pollution levels for the period. The derivatives of 6 EMI(t)<sub>2.5</sub> are the  $\Delta$ EMI, which is a positive value when the pollution is being accumulated 7 and a negative value when the pollution is being dispersed.



8



10 Therefore, for the period p with n discrete steps from t0 to t1, the  $\overline{EMI(p)_{2.5}}$ 

11 represents the averaged meteorological influences on PM<sub>2.5</sub>, while the sum of the

12 positive  $\Delta$ EMI is the accumulation potentials and the sum of the negative  $\Delta$ EMI is the

#### 1 dispersing potentials as illustrated in Figure 3. The relationship between them is derived

2 as follows:

<sup>3</sup>
$$\overline{EMI(p)_{2.5}} = \frac{1}{n+1} [EMI(0) + EMI(1) + EMI(2) + \dots + EMI(n)]$$
<sup>4</sup>
$$= \frac{1}{n+1} [(n+1)EMI(t0) + n\Delta EMI(1)\Delta t + (n-1)\Delta EMI(2)\Delta t$$
<sup>5</sup>
$$+ (n-2)\Delta EMI(3)\Delta t + (n-3)\Delta EMI(4)\Delta t + \dots + \Delta EMI(n)\Delta t]$$

7 where *n* is the time steps in the period and the averaged EMI has been linked to the 8 starting point EMI(0) and the changing rates of EMI, i.e.  $\Delta$ EMI(*n*), at each time step. For 9 monthly simulations, the initial values EMI(t0) for each month was set up by the 10 averaged PM<sub>2.5</sub> concentrations for the first day from 2013 to 2019 divided by the 11 constant  $C_0$  (35 µg m<sup>-3</sup>).

## 12 **2.4.** Assessment Framework of Emission Controls

The EMI<sub>2.5</sub> index provides a way to assess the meteorological impacts on the changes of PM<sub>2.5</sub> concentrations at two time periods, i.e. January 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 changes in the observed concentration levels and meteorology factors  $\overline{EMI(p)_{2.5}}$  to quantify the changes caused by emission changes only at two time periods.

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

8 
$$\Delta EMIS = \frac{PM(m0,e1) - PM(m0,e0)}{PM(m0,e0)} \times 100\%$$
 (6)

9 where *PM* (*m*0, *e*1) is a hypothetically non-measurable quantity, indicating the PM
10 concentration at p1 with emission e1 and meteorology m0, that does not exist in
11 reality. An assumption is to be made to compute this quantity using the EMIs. It is
12 assumed that:

13 
$$\overline{EMI(p0)_{2.5}} / EMI(p1)_{2.5} = \frac{PM(m0,e1)}{PM(m1,e1)}$$
 (7)

which means that under the same emissions, the ratio of averaged EMIs under two
meteorology (m0, m1) equals to the ratio of PM concentrations under the same two
meteorology (m0, m1). Given the non-linear contributions of meteorology and
emissions to the ambient PM concentrations or simulated EMIs, this assumption is a
first-order approximation for the contributions of meteorology and emissions to the
observed concentrations. Substituting *PM (m0, e1)* calculated from Eq. (7) to Eq. (6) will

1 facilitate the estimate of percentage contribution of emission controls to the air quality

2 improvement at two periods of time, independent of meteorology variations.

3

2.5.

## Quantitative Estimate of EMI

Finally, a process-based method is developed to calculate the EMI and its 4 5 components, i.e. *iEmid*, *iTran* and *iAccu*. The main modeling frame-work used is the 6 chemical weather modeling system MM5/CUACE, which is a fully coupled atmospheric 7 model used at CMA for national haze and air quality forecasts (Gong and Zhang, 8 2008; Zhou et al., 2012). CUACE is a unified atmospheric chemistry environment with 9 four major functional sub-systems: emissions, gas phase chemistry, aerosol microphysics 10 and data assimilation (Niu et al., 2008). Seven aerosol components, i.e. sea salts, 11 sand/dust, EC, OC, sulfates, nitrates and ammonium salts are sectioned in 12 size bins 12 with detailed microphysics of hygroscopic growth, nucleation, coagulation, condensation, dry depositions and wet scavenging in the aerosol module (Gong et al., 13 14 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-15 16 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 17 sulfate and SOA production rates for the aerosol module and for the aerosol equilibrium 18 module ISORROPIA (Nenes et al., 1998) to calculate the nitrate and ammonium aerosols. 19 20 This is the default method to treat the secondary aerosol formations in CUACE. For the

1	EMI application of CUACE, another option was also adapted to compute the secondary
2	aerosol formations by a highly parameterized method (Zhao et al., 2017), that computes
3	the aerosol formation rates directly from the pre-cursor emission rates of SO <sub>2</sub> , NO <sub>2</sub> and
4	VOC. This option was added to facilitate timely operational forecast requirements for
5	CMA. Both primary and pre-cursor emissions of PM are based on the 2016 MEIC
6	Inventory ( <u>http://www.meicmodel.org/</u> ) developed by Tsinghua University for China.
7	In order to quantitatively obtain each term defined in Equation 3, the CUACE
8	model was modified to extract the change rates for the processes involved. Driven by
9	the re-analysis meteorological data, the new system CUACE/EMI can be used to
10	calculate each term in $\Delta$ EMI at each time step ( $\Delta$ t).
11	In summary, this section presents a systematic platform to separate and assess the
12	impacts of the meteorology and emissions on the ambient concentration changes. The
13	$\overline{EMI(p)_{2.5}}$ and $\Delta$ EMIS form the basis for the assessment. In the Results and Discussions
14	section, the application of the platform is presented to assess the fine particular matter
15	(PM <sub>2.5</sub> ) changes in China.

# 16 **3. Results and Discussions**

# 17 **3.1.** Validation of EMI by Observations

- 18 Under the conditions of no changes in annual emissions for PM<sub>2.5</sub> and its precursors,
- 19 the daily EMI<sub>2.5</sub> was computed by CUACE from 2013 to 2019 on a 15×15 km resolution **17** | P a g e

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., 2011) variation
was applied to the emission inventory for computing the EMI<sub>2.5</sub>, which is more
realistically reflecting the meteorology contributions to the PM<sub>2.5</sub> concentrations.

6 To evaluate the applicability of EMI<sub>2.5</sub>, the index was compared with the 7 observed PM<sub>2.5</sub> concentrations. Figure 4 shows the spatial distribution of correlation 8 coefficients between PM<sub>2.5</sub> and EMI<sub>2.5</sub> for 2017 for all China. The correlation coefficients 9 between EMI<sub>2.5</sub> and PM<sub>2.5</sub> 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 10 correlation was found in Western China, possibly due to complex terrain and less 11 12 accurate emission data over there. Furthermore, due to the uncertainty in emissions 13 and the difference in model performance for year-to-year meteorology simulations, the correlation coefficients may differ for different years. Overall, the good correlation 14 between them merits the application of EMI<sub>2.5</sub> to quantify the meteorology impact on 15 PM<sub>2.5</sub>. 16 17

19

18



2 Figure 4: Correlation coefficients (R) between the EMI<sub>2.5</sub> and the observed PM<sub>2.5</sub> daily

3 concentrations across China for 2017 and for typical regions averaged between 2013 and 2019.

4

5 To further illustrate the applicability of EMI<sub>2.5</sub>, the difference of various 6 conditions between December 2014 and December 2015 in BTH region was also 7 analyzed when a significant change of air quality and meteorological conditions 8 occurred. The winter of 2015 was accompanied by a strong El Nino (ENSO) event, 9 resulting in significant anomalies for meteorological conditions in China. Analysis shows that the meteorological conditions in December 2015 (compared to December 2014) 10 had several important anomalies, including that the surface southeasterly winds were 11 significantly enhanced in the North China Plain (NCP) and the wind speeds were 12 decreased in the middle-north of eastern China, while slightly increased in the south of 13 eastern China. Study suggests that the 2015 El Nino event had significant effects on air 14 15 pollution in eastern China, especially in the NCP region, including the capital city of

1 Beijing, in which aerosol pollution was significantly enhanced in the already heavily

2 polluted capital city of China (Chang et al., 2016). 3 Figure 5 shows the monthly average EMI<sub>2.5</sub>, PM<sub>2.5</sub> and the contribution of sub-4 index to total EMI<sub>2.5</sub> in December 2014 and 2015 over BTH region. The monthly average 5 EMI<sub>2.5</sub> increases about 54.9% from 2.1 in December 2014 to 3.2 in December 2015, 6 indicating worsening meteorological conditions for PM2.5 pollution. The increase of 7 EMI<sub>2.5</sub> is mainly contributed by adverse atmospheric transport conditions (Fig. 5c), which 8 results in the increase of EMI<sub>2.5</sub> reaching 3.2. With the increase of background 9 concentration, the deposition and vertical diffusion also increase, and offset the impact of adverse transport conditions to some extent. 10



11

12 Figure 5: (a) the monthly averaged EMI<sub>2.5</sub> and (b) monthly PM<sub>2.5</sub> for Decembers of 2014 and

13 2015 over BTH. (c) contributions of sub-index to the EMI<sub>2.5</sub> change and (d) contributions of

emission and meteorology changes to PM<sub>2.5</sub> change for Decembers from 2014 to 2015,
 respectively.

4	The worsening meteorological conditions represented by EMI <sub>2.5</sub> were also
5	supported by the observations for the two periods. The observed day with small wind
6	(DSW, wind speed less than 2 m s <sup>-1</sup> ) reveals that, except for part of southern Hebei
7	province, the DSW increases 5-15 days for 2015 in most meteorological stations in BTH
8	region (Fig. 6a), which indicates a large decrease of local diffusion capability. The
9	comparison of wind rose map shows that the decrease of northwest wind and the
10	increase of southwest and northeast wind occurred in December 2015 (Fig. 6b). The
11	change of wind fields indicates more pollutants were transported to BTH region from
12	Shandong, Jiangsu, Henan, and Northeast China. These variations indirectly validate the
13	conclusions of adverse atmospheric transport conditions with high iTran in December
14	2015.





2 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.

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

1 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 2 al. (2019) who found out that the monthly emission strengths for PM<sub>2.5</sub>, SO<sub>2</sub>, NOx, VOCs 3 and NH<sub>3</sub> in 2015 were reduced by 22.0%, 6.9%, 2.5%, 2.5% and 2.5%, respectively, as 4 5 compared with 2014. The sensitivity and the nonlinear response of  $PM_{2.5}$  concentrations 6 to the air pollutant emission reduction in the BTH region (Zhao et al., 2017) have been 7 estimated to be about 0.43 for both primary inorganic and organic PM<sub>2.5</sub>, 0.05 for SO<sub>2</sub>, -0.07 for NOx, 0.15 for VOCs, 0.1 for NH<sub>3</sub>. Combining the emission reduction percentages 8 9 between Decembers 2014 and 2015 and the nonlinear response of emissions to the PM<sub>2.5</sub> 10 concentrations results in an approximately 10.2% ambient PM<sub>2.5</sub> concentration reduction 11 due to the emission changes. This is very close to the estimate of emission reduction 12 contribution to the December PM<sub>2.5</sub> concentration difference of about 9.4% between 2014 and 2015 by the EMI framework. 13

The applicability of EMI to assess the meteorology and emission changes is also 14 evaluated by results from a full chemical transport model (MM5/CUACE) and 15 observational data for PM<sub>2.5</sub> in China for Novembers of 2017 and 2018. The averaged 16 EMI<sub>2.5</sub> and observational data for the two months were used to estimate the emission 17 change ratio (E-Ratio in Table 2) by Equations 6-7 from 2017 to 2018. In order to evaluate 18 the correctness of this emission change estimate, the E-Ratio was used to adjust the 19 20 emissions for November 2018 from the base emissions of the same month for 2017, which were then implemented in the MM5/CUACE to simulate the PM<sub>2.5</sub> concentrations for the 21 22 two months, respectively. If the simulated concentration differences (M-Ratio) for the 23 | Page

two months were comparable with the observed concentration differences (O-Ratio), it can be concluded that the emission change estimated by the EMI framework was reliable and could approximately represent the actual emission changes. Table 2 summarizes the analysis results of this evaluation for six typical cities. It is clear that the O-Ratios for the six cities are very comparable with M-Ratios, indicating that the EMI framework can be reasonably used to estimate the emission changes over time.

Table 2: Comparison of PM<sub>2.5</sub> Concentrations in Novembers of 2017 and 2018 from Ambient
 Observations and from CTM Simulations by EMI-derived Estimated Emission Changes

City	EMI <sub>2.5</sub>		Observations			Emission Changed	CI	「M Simu	lated
	2017	2018	2017	2018	O-Ratio	E-Ratio	2017	2018	M-Ratio
Beijing	1.8	3.6	45.7	72.8	1.59	0.80	42.3	67.5	1.59
Shanghai	2.7	2.6	42.0	40.1	0.95	1.00	52.7	51.2	0.97
Jinan	3.3	4.9	57.1	85.8	1.50	1.02	62.4	90.9	1.46
Xian	2.4	2.7	94.8	84.7	0.89	0.79	95.1	86.9	0.91
Zhengzhou	4.3	6.2	73.9	100.4	1.36	0.96	80.4	91.1	1.13
Shenyang	1.8	2.7	40.2	48.9	1.21	0.82	73.3	120.1	1.63

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## **3.2. PM**<sub>2.5</sub> Trends and Meteorological Contributions

12 The annual averaged PM<sub>2.5</sub> concentrations in China have been decreased

- 13 significantly from 2013 to 2019. Figure 7 shows the observed spatial distribution of
- 14 national PM<sub>2.5</sub> concentrations from 2013 to 2019, respectively. These spatial
- 15 distributions are consistent with those of primary and precursor emissions of PM<sub>2.5</sub>

1	(Wang et al.), pointing out the fundamental cause of the air pollution in China. From the
2	spatial distributions, it is clear that the regions of BTH, FWP, CEN and NWC had the
3	highest $PM_{2.5}$ concentrations among the 9 regions. Even though the national
4	concentrations have been reduced significantly from 2013 by reducing emissions, the
5	pollution center of particular matters has not been changed very much, locating at the
6	southern Hebei Provence and indicating the macroeconomic structure has not been
7	gone through a great change yet. Another phenomenon can be seen from the
8	distribution is that in the North-west China, especially in some cities of the Xinjiang and
9	Ningxia Provinces, the $PM_{2.5}$ concentrations were on an increasing trend, due to certain
10	migrating industries from developed regions in East China.
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1

2 Figure 7: Regional annual PM<sub>2.5</sub> concentration distributions from 2013 to 2019.

Averaged for the nation, 9 focused regions and Beijing, the PM<sub>2.5</sub> trend lines 1 2 were shown in Figure 2. It is seen that all regions have had a large reduction of more than 35% in surface PM<sub>2.5</sub> concentrations in 2019 as compared with those in 2013. The 3 averaged national annual concentration at 36 µg m<sup>-3</sup> has been very close to the national 4 5 standard of 35  $\mu$ g m<sup>-3</sup> 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. 6 7 Regionally, the largest drop percentage of PM<sub>2.5</sub> was seen in NEC and NWC regions (Fig. 8 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 9 10 around 35%.





Figure 8: Annual averaged PM<sub>2.5</sub> 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.

1

2 As one of the key factors in controlling the ambient PM<sub>2.5</sub> concentration 3 variations, the annual meteorological fluctuations, i.e. EMI<sub>2.5</sub>, from 2014 to 2019 with 4 2013 as the base year, are shown in Figure 9 for nine regions. Generally, the annual 5 EMI<sub>2.5</sub> shows a positive or negative variation, reflecting the meteorological features for 6 that specific region. Except for a couple of regions or years, most of the fluctuations are 7 within 5% as compared with 2013 and have a no definite trend. It can be inferred that 8 the meteorological conditions are possibly responsible for about 5% of the annual PM<sub>2.5</sub> averaged concentration fluctuations from 2013 to 2019 (Fig. 9 middle). This is consistent 9 with what has been assessed in Europe by Andersson et al. (2007). 10 11 The variations in meteorological contributions (EMI<sub>2.5</sub>) to PM<sub>2.5</sub> for the heavy 12 pollution seasons of fall and winter (October 1 to March 31) generally follow the same 13 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. 14 15 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 16 17 meteorology variations in fall-winter would exercise more controls to the heavy pollution episodes than the annual averaged concentrations, signifying the importance 18 of meteorology in regulating the winter pollution situations. 19 20 It is found that though most of the regions have a fluctuating EMI<sub>2.5</sub> in the

pollution season during the 2014-2019 period (Fig. 9 bottom), the YRD and FWP show a

**28** | Page

consistent favorite and unfavorite meteorological conditions, respectively. BTH has
witnessed the same unfavorite conditions as FWP except in 2017. In other words, in BTH
and FWP, the decrease in ambient concentrations of PM<sub>2.5</sub> from 2014 to 2019 has to
overcome the difficulty of worsening meteorological conditions with larger control
efforts.



6

- 7 Figure 9: Annual averaged EMI<sub>2.5</sub> in 2013 (top) and corresponding changing rates for annual
- 8 average (middle) and for fall-winter seasons (bottom) from 2014 to 2019 as compared with 2013
- 9 in 9 regions.

10



#### 1 3.3. Attribution of Control Measures to the PM<sub>2.5</sub> Trend

2 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 3 emissions and meteorology contributions to the pollution level reduction entails a combined 4 5 analysis of them. The analysis in Section 3.2 shows that from 2013 to 2019, the national 6 averaged PM<sub>2.5</sub> as well as those for 9 separate regions were all showing a gradual decline trend 7 (Fig. 8). By 2019, 45% - 50% of reductions in surface PM<sub>2.5</sub> concentrations were achieved while the meteorology contributions did not show a definite trend as from 2013, clearly pointing out 8 9 the contribution of emission reductions in the trend. Using the analysis framework for separating emissions from meteorology based on the monitoring data of PM<sub>2.5</sub> and EMI<sub>2.5</sub> 10 (Section 2.4), the emission change contributions are estimated. 11 Figure 10 shows the 2013 base emissions of PM<sub>2.5</sub> (Zhao et al., 2017) and the annual 12 13 changes in the emission contributions to the PM<sub>2.5</sub> concentrations from 2014 to 2019 as 14 estimated from the EMI<sub>2.5</sub> and observed PM<sub>2.5</sub>. For the emissions, it is found that the unit area emissions match better with ambient concentrations of PM<sub>2.5</sub> in regions than the total 15 emissions and the high emission regions are BTH, BTH+, YRD, PRD and FWP in 2013. Nationally 16 by 2019, the emission reduction contributions to the ambient PM<sub>2.5</sub> trend accounted for 17 18 ranging from 32% to 52% of the total PM<sub>2.5</sub> decrease percentage, while in BTH and BTH+ regions the reduction was more than 49% from 2013 base year emissions, leading the national 19 emission reduction campaign. The emission reduction rates clearly illustrate the effectiveness 20 of the national-wide emission control strategies implemented since 2013 and the emission 21

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



6

Figure 10: Annual PM<sub>2.5</sub> emissions (total and per unit Km<sup>2</sup>) for 2013 (top) and corresponding changing
 rates (bottom) from 2014 to 2019 as compared with 2013 in 9 regions.

9



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 unfavorite meteorological conditions during this period.

4 The year of 2015 is a special year in the history of China air pollution control. Though the 5 systematical and network observations of PM<sub>2.5</sub> started in China from 2013, it took about two 6 years (until 2015) to evolve to the current status in terms of spatial coverage and observational 7 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 8 9 effect in January, signalizing the stage of lawfully control of air pollution. For the regulation 10 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. 11

12 Table 3 summarizes the PM<sub>2.5</sub> difference between 2019 and 2015 and the relative 13 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<sub>2.5</sub> concentrations are all reduced from 14 15 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 unfavorite meteorological conditions with positive 16 17 EMI<sub>2.5</sub> 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 18 19 to achieve the observed reductions in ambient PM<sub>2.5</sub> concentrations. On the contrary, for the 20 regions of FWP and SWC, the emission control impacts were to deteriorate the concentrations, 21 implying an increase in emissions to restrain the PM<sub>2.5</sub> concentration decrease by favorite 22 meteorological conditions. For other regions, both meteorology and emission controls

- 1 contributed to PM<sub>2.5</sub> decrease from 2015 to 2019, with the control measures contributing from
- 2 -7.9% in NWC to -68.4% in NEC (Table 3).
- 3 Therefore, due to the diversity of meteorological conditions and emission distributions in
- 4 China, their impacts on ambient PM<sub>2.5</sub> concentrations display unique reginal characteristics.
- 5 Overall, the emission controls are the dominant factor in contributing the decline trend in China
- 6 from 2013 to 2019. However, in certain regions or certain period of years, emissions were
- 7 found to be increased even with favorite meteorological conditions, which means the design of
- 8 national control strategies has to take both meteorology and emission impacts simultaneously
- 9 in order to achieve maximum results.
- Table 3: Observed PM<sub>2.5</sub> 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 <sub>2.5</sub> Diffe	rence	Attributions						
			Meteorolog	y (EMI)	Emission Controls				
	(µg m <sup>-3</sup> ) (%)		(µg m⁻³)	Relative %	(µg m⁻³)	Relative %			
National	-10	-21.7	-4.1	-40.9	-5.9	-59.1			
ВТН	-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			

12

Note: "+" increased; "-" decreased

#### 1 4. Conclusions

2 Based on a 3-D chemical transport model and its process analysis, an Environmental Meteorological Index (EMI2.5) and an assessment framework have been developed and applied 3 4 to the analysis of the PM<sub>2.5</sub> trend in China from 2013 to 2019. Compared with observations, the 5 EMI<sub>2.5</sub> can realistically reflect the contribution of meteorological factors to the PM<sub>2.5</sub> variations 6 in the time series with impact mechanisms and can be used to as an index to judge whether the 7 meteorological conditions 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<sub>2.5</sub>-based framework has been used to 8 9 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<sub>2.5</sub> 10 11 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 12 13 range of 46% to 53% while in FWP, PRD and YRD, the reduction was from 45% to 35%. It is 14 found that the control measures of emission reduction are the dominant factors in the PM<sub>2.5</sub> 15 declining trends in various regions. By 2019, the emission reduction contributes about 47% of the PM<sub>2.5</sub> decrease from 2013 to 2019 on the national average, while in BTH region the 16 emission reduction contributes more than 50% and in YRD, PRD and SWC regions, the 17 18 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 19 20 emission amounts were increased for certain years. The contribution by the meteorology to the surface PM<sub>2.5</sub> concentrations from 2013 to 2019 was not found to show a consistent trend, 21

- 1 fluctuating positively or negatively about 5% on annual average and 10-20% for the fall-winter
- 2 heavy pollution seasons. It is noted that the estimate of emission control contributions was
- 3 made under a first-order approximation of emission and meteorology, which should be
- 4 improved in the future implementations.

## 5 Code/Data availability

6 No

## 7 Author contribution

Sunling Gong proposed the EMI concept and led the research project, and Hongli Liu
implemented the EMI concept in the modeling system. Bihui Zhang and Hengde Zhang led the
operational implementation in the national center and provided the meteorological
observational data. Yaqiang Wang, Jianjun He and Lei Zhang led the modeling activities and
result analysis. Shuxiao Wang provided the emission change data and Jie Wang provided the
observational data of pollutants.

14

## 15 Competing interests

16 No

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