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).

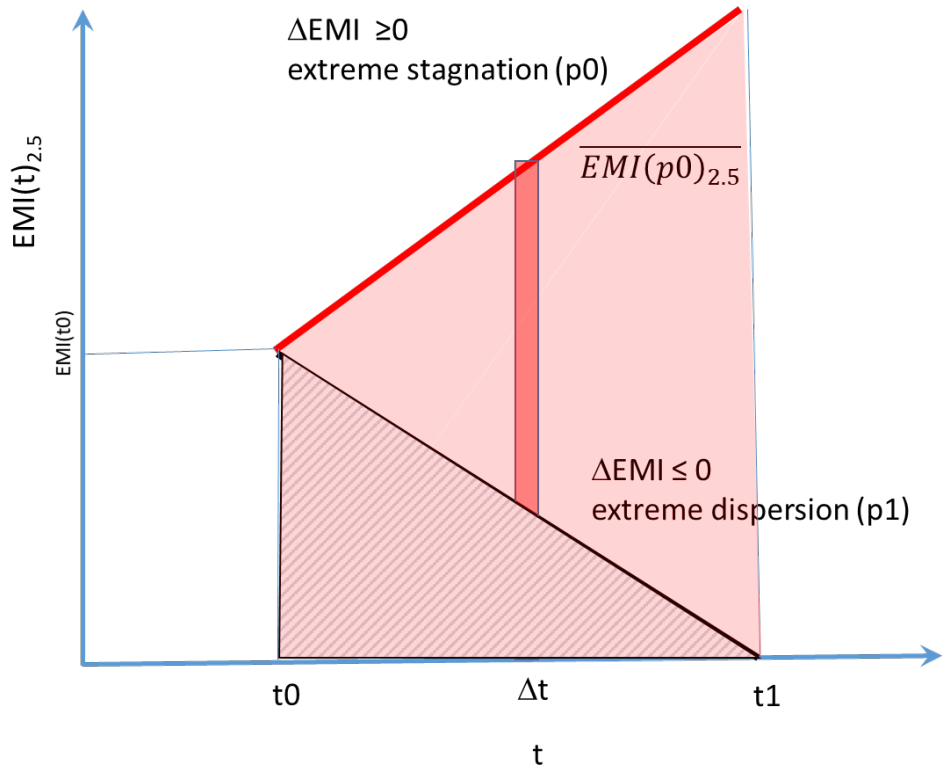

Figure 1: Illustration of relationship between 3 levels of EMI definition for the cases suggested by Referee #2.

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.