Referee Report for "Assessment of meteorology vs control measures in China fine particular matter trend from 2013-2019 by an environmental meteorology index" by Sunling Gong et al. (manuscript #: acp-2020-348)

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 $PM_{2.5}$ 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 $PM_{2.5}$ 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.

(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 $PM_{2.5}$.

Given the large contribution of secondary formation to $PM_{2.5}$ 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 ($PM_{2.5}$). 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 SO_2, NO_2, 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.

(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 $PM_{2.5}$ 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 ($\Delta EMI = iTran + iAccu + iEmid = \frac{1}{c_0}\int_0^h (Emis)dz = \frac{1}{c_0} Emis \cdot h$, where c_0, $Emis$, and h 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 h, 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 $PM_{2.5}$ 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.

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).
(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.
(3) What is h in Eq. (3)? Is it boundary layer height or not?
(4) What kind of data were used for the correlation in Fig. 4? Monthly? Or Daily?
(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.

Reference

Seinfeld, J. H., and Pandis, S. N.: Atmospheric chemistry and physics: from air pollution to climate change, John Wiley & Sons, Inc, Hoboken, New Jersey, 2016.

Huang, R., Zhang, Y., Bozzetti, C. et al. High secondary aerosol contribution to particulate pollution during haze events in China. Nature 514, 218–222, https://doi.org/10.1038/nature13774, 2014.