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

In the response to my previous review, the authors added more information to try to support the validity of their EMI assessment framework. I appreciate their efforts and clarifications, but these additional information only confirms my previous concern about the fundamental flaw in this EMI framework. To illustrate my point more clearly, here I use two figures below to show why it's incorrect to add the emission term (*Emis*) in the ΔEMI and EMI calculation.

Again, let's revisit the first hypothetical case with extreme air stagnation conditions. In this case, we assume no advection (*iTran*=0), no turbulent diffusion (*iAccu*=0), no precipitation and wet deposition ($L_d = 0\ in\ iEmid$), and negligible dry deposition compared with a constant emission ($Emis \gg V_d\ in\ iEmid$). Therefore, ΔEMI becomes

$$\Delta EMI = iTran + iAccu + iEmid = \frac{1}{hC_0}\int_0^h (Emis)dz = \frac{1}{c_0}Emis,$$

according to their updated Eq. (3) in the revised manuscript.

Based on this derivation and Eq. (1), $\mathrm{EMI(t)}$ would monotonically increase at a constant rate/slope of $\frac{1}{c_0}Emis$ from an initial value $EMI(t_0)$ along with time *t* (Fig. 1).

$$EMI(t) = EMI(t_0) + \int_{t_0}^{t} \Delta EMI \times dt = EMI(t_0) + \Delta EMI \times (t - t_0).$$

Thanks to the clarification in the response, now $\overline{EMI(p_0)_{2.5}}$ becomes

$$\overline{EMI(p_0)_{2.5}} = \frac{1}{t_1 - t_0}\int_{t_0}^{t_1} EMI(t)dt = \frac{1}{2}[EMI(t_0) + EMI(t_1)] = \frac{1}{2}[EMI(t_0) + EMI(t_0) + \Delta EMI \times (t_1 - t_0)] = EMI(t_0) + \frac{n}{2} \times \frac{1}{c_0}Emis \times \Delta t,$$

and $\overline{EMI(p_1)_{2.5}}$ becomes

$$\overline{EMI(p_1)_{2.5}} = \frac{1}{t_3 - t_2}\int_{t_2}^{t_3} EMI(t)dt = \frac{1}{2}[EMI(t_2) + EMI(t_3)] = EMI(t_2) + \frac{m}{2} \times \frac{1}{c_0}Emis \times \Delta t,$$

given $p_0 = t_1 - t_0 = n \times \Delta t$ and $p_1 = t_3 - t_2 = m \times \Delta t$.

Note that the equations for $\overline{EMI(p_0)_{2.5}}$ and $\overline{EMI(p_1)_{2.5}}$ in the response are wrong due to incorrect time scaling factors ("$\frac{n}{2} \times \Delta t$" and "$\frac{m}{2} \times \Delta t$" here vs "$(n-1) \times \Delta t$" and "$(m-1) \times \Delta t$" in the response).

If assuming the same time interval length for p_0 and p_1 ($n = m$) as the authors did in the response, then

$\frac{\overline{EMI(p_0)_{2.5}}}{\overline{EMI(p_1)_{2.5}}} = \frac{EMI(t_0)+\frac{n}{2}\times\frac{1}{c_0}Emis\times\Delta t}{EMI(t_2)+\frac{n}{2}\times\frac{1}{c_0}Emis\times\Delta t}$, which can't be equal to 1 as they claimed in the response unless $\mathrm{EMI}(t_0) = \mathrm{EMI}(t_2)$.

However, the only way that satisfies $\mathrm{EMI}(t_0) = \mathrm{EMI}(t_2)$ is $t_2 = t_0$ because EMI(t) is a monotonically increasing function in this case based on the equations from the manuscript.

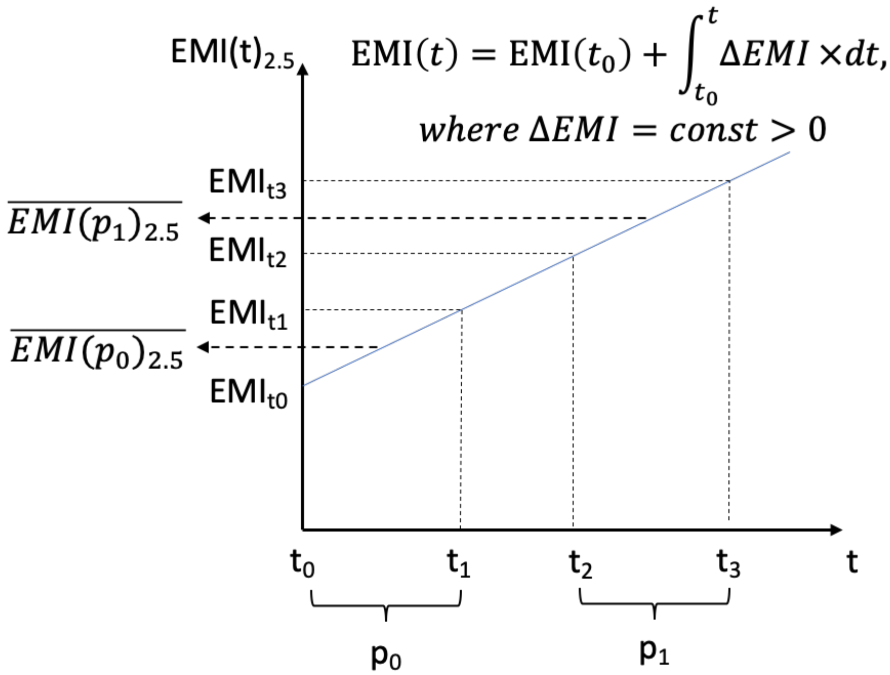

Figure 1 the illustration of the EMI(t) *function based on Eqs. (1)-(3) in the manuscript.*

The above contradiction derives from the incorrect inclusion of Emis in iEmid/EMI as pointed out in the previous review. Let's see what will happen in the same stagnation case if excluding Emis in iEmid/EMI.

In this way, $iEmid = \frac{1}{hC_0}\int_0^h -(V_d + L_d)dz = 0$, and $\Delta EMI = iTran + iAccu + iEmid = 0$. Therefore, $\mathrm{EMI(t)} = \mathrm{EMI}(t_0)$ becomes a horizontal line with a constant value $\mathrm{EMI}(t_0)$ for all time *t* (Fig.2). Furthermore, $\frac{\overline{EMI(p_0)_{2.5}}}{\overline{EMI(p_1)_{2.5}}} = \frac{\mathrm{EMI}(t_0)}{\mathrm{EMI}(t_0)} = 1$ is satisfied for any time period p_0 and p_1. By applying this equation to Eqs. (6)-(7) in the manuscript, we can obtain

$PM(m_0, e_1) = \frac{\overline{EMI(p_0)_{2.5}}}{\overline{EMI(p_1)_{2.5}}} \times PM(m_1, e_1) = PM(m_1, e_1)$,

and $\Delta EMIS = \frac{PM(m_0,e_1)-PM(m_0,e_0)}{PM(m_0,e_0)} \times 100\% = \frac{PM(m_1,e_1)-PM(m_0,e_0)}{PM(m_0,e_0)} \times 100\%$.

These two equations imply that the observed PM concentration change between p_0 and p_1 ($PM(m_1, e_1) - PM(m_0, e_0)$) is purely (100%) from the emission contribution in $\Delta EMIS$, while the contribution from meteorology ($1 - \Delta EMIS$) is zero. This result is as expected considering all the assumptions in this case.

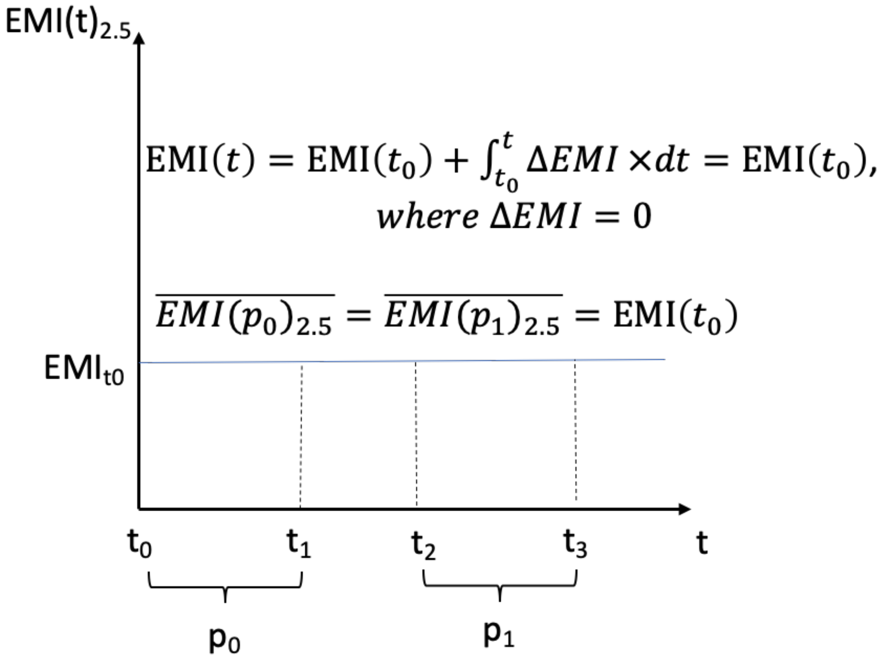

Figure 2 the illustration of the proposed EMI(t) *function without the inclusion of Emis in iEmid/EMI.*

The same problem also exists in the second extreme dispersion case, for which the illustration in Fig. 1 is still incorrect in the response. In this case, the EMI(t) function can't be a monotonically decreasing line with a constant slope as shown by Fig. 1 in the response because of the time variant concentration gradient in *iTran*. There are some other problems such as the arbitrary reset for the initial value of $\mathrm{EMI}(t_0)$ in each year/month (see the answer to the second technical comment in the response). The low sensitivity of EMI to the initial value may result from highly variable meteorological conditions in the real atmosphere.

I'll stop here without further review, but I think the above examples are self-evident that the current EMI framework is flawed in its basis (mostly in *iEmid*). I suggest the authors think about their framework carefully and reconsider the submission of the manuscript in the current form.