

Thanks for the referee's comments on this manuscript. After carefully reading the comments, we have made following point-to-point replies:

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 ( $E_{mis}$ ) in the  $\Delta EMI$  and EMI calculation.

**Reply:** First of all, the EMI was defined to reflect the impact of meteorology on the PM pollution in different regions and should be comparable with the in-situ observations. The addition of emissions term in the EMI was made to show the impact difference of meteorology on PM at different regions. We also would like to use an extreme case to explain the necessity to include emission term in EMI in order to compare with observations at different locations. Still under the hypothetical case with extreme air stagnation conditions as suggested by the referee, we choose two locations: one with an emission of  $E_{mis}$  as a constant and another without emission, i.e.  $E_{mis}=0$ . In reality, the location with a constant emission would be subject to a heavy pollution episode while the location without emissions would be pollution free. If we used the suggestion by the referee without emission, the EMIs at both locations would be equal, which could not reflect the difference of the real situations at these two locations and could not be compared with real observations. By introducing the emissions in the EMI, the difference would be clearly shown: the location with emissions would be experiencing an accumulation of pollution with time as indicated by  $\Delta EMI=1/C_0 E_{mis}$ ; the location without emission would be pollution free with time as indicated by  $\Delta EMI=0$ , which mimics the real situations as EMI was intended to be.

As we understood from pollution formation mechanisms, the pollutant emission was the fundamental cause of any pollution and the meteorology was the external force to modulate the pollution strength. The EMI was defined to show the impact of meteorology on PM pollution under a constant emission for any locations by including the emissions. If there were no emissions, there were no pollution at all and no meaning to define an EMI.

As a matter of fact, there exists no incorrectness or correctness in defining an EMI, all depending on the purposes of the EMI applications and the targets to compare.

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,  $\Delta EMI$  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),  $EMI(t)$  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

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

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).

**Reply:** Thanks to the referee. The referee was right here that we did use an incorrect time scaling factor as we missed one initial step in Equation 5 using the time step from 0 to n-1 to represent n time steps. This has been corrected in the revised manuscript.

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

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

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

**Reply:** This is exactly what EMI is intended to be used: using the same initial conditions to compare the EMI for two segments of time periods at the same location. One case study in the manuscript was the comparison of January of 2013 ( $p_0$ ) and January of 2016 ( $p_1$ ), where the same initial conditions and emissions were used to quantify the meteorological impact. Please note that in the model simulations, we have set the  $EMI_{t_0}$  and  $EMI_{t_2}$  equal to each other and differences in simulated averaged  $EMI(p_0)$  and  $EMI(p_1)$  would indicate the impacts of meteorology on pollutants (Fig. 1). If the same meteorology occurred in  $p_0$  and  $p_1$ , the  $EMI(p_0)$  and  $EMI(p_1)$  would be the same as expected.

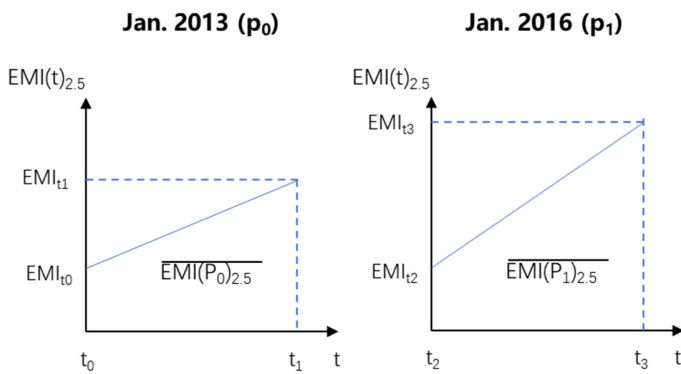


Figure 1: EMI simulation schemes for two periods of time.

The above contradiction derives from the incorrect inclusion of  $E_{mis}$  in  $iEmid/EMI$  as pointed out in the previous review. Let's see what will happen in the same stagnation case if excluding  $E_{mis}$  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,  $EMI(t) = EMI(t_0)$  becomes a horizontal line with a constant value  $EMI(t_0)$  for all time  $t$  (Fig.2). Furthermore,  $\frac{EMI(p_0)_{2.5}}{EMI(p_1)_{2.5}} = \frac{EMI(t_0)}{EMI(t_0)} = 1$  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

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

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

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.

**Reply:** Again, if the case was the extreme air stagnation conditions, our formulation would arrive at the same conclusions by including the same emission for the two periods, i.e.,  $EMI(p_0)_{2.5} = EMI(p_1)_{2.5}$ , as same initial conditions were used for  $n=m$ . However, we would not see any PM concentration change between  $p_0$  and  $p_1$  as the emissions, initial conditions and meteorology were all the same. This is what we should expect for the two periods. If no emissions were included, we would not see any increase of EMI with time and the acclamation of any pollutant in any period

would not be reflected in EMI, which contradicted to the real PM situations and made the EMI not comparable with PM observations.

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 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.

**Reply:** If the same degree of extreme dispersion occurred for both  $p_0$  and  $p_1$ , we should also arrive at  $EMI(p_0) = EMI(p_1)$  but with decreasing trends for both periods as the same initial conditions and emissions were used for both  $p_0$  and  $p_1$ . The Fig. 1 in the last version of response showed a monotonically decreasing line simply due to the assumption of a constant extreme dispersion for the sake of explaining the framework. In reality, the decreasing line is definitely variable as controlled by the time variant concentration gradients as indicated by the referee.

Finally, it is evident that the inclusion of emission term in the EMI was not a flaw but a necessity for the right application of EMI. However, we would thank the effort of the referee whose comments and suggestions have made this manuscript much more scientifically accountable.