Dear Editor,

We appreciate the prompt reviews and would like to thank the reviewer for insightful comments and suggestions on our manuscript entitled "Contributions of meteorology and anthropogenic emissions to the trends in winter $PM_{2.5}$ in eastern China 2013–2018" (MS No.: acp-2022-304). We have carefully considered all comments and suggestions. Listed below are our point-by-point responses to all comments and suggestions of this reviewer (Reviewer's points in black, our responses in blue).

Anonymous Referee #1

This paper presents a MLR statistical attribution of the 1985-2018 $PM_{2.5}$ trends in three megacity clusters in China, using visibility data as proxy for pre-2013 $PM_{2.5}$ data. It finds a large meteorological (non-emission) contribution to the trend, and argues that previous MLR analyses of the 2013-2018 trend using the actual $PM_{2.5}$ data starting in 2013 and attributing the trend to emissions are not robust. The paper makes some good points about the difficulty of sorting out meteorological effects when interpreting short (post-2013) trends. However, I believe that it may be (1) flawed in its reconstruction of the 1985-2018 $PM_{2.5}$ record which is the basis for most of the argumentation, (2) mistaken in claiming that attribution of recent $PM_{2.5}$ trends to emissions is not based on mechanistic knowledge, and (3) annoying in belaboring trivial statistical points that are well known to any trained scientist. I don't think that this paper is publishable in ACP in current form.

Response:

The reviewer made three general criticisms. Our responses are listed below one by one.

(1) Our reconstruction of the 1985–2018 $PM_{2.5}$ record is based on a method that converts observed visibility data to the concentrations of $PM_{2.5}$. This method has been shown in many previous studies to be credible (Shen et al., 2016; Liu et al., 2017; Gui et al., 2020; Li et al., 2020; Li et al., 2021). The first two references are already cited in

our paper. Given the serious concern of this reviewer, in Figure R1 below we compare the winter $PM_{2.5}$ record derived in our study (black line) to winter haze days derived from observed visibility data in Beijing by Li et al. (2021) (green line). The winter $PM_{2.5}$ concentrations are expected to be well correlated with the number of winter haze days. Indeed the correlation coefficient between the green line and black line is quite high at 0.7. Also shown in Figure 1 are $PM_{2.5}$ concentrations observed by the US Embassy in Beijing (blue line, 2009–2018) and those observed by CNEMC in BTH (red line, 2013–2018). The crucial bulge-2013 shows up consistently in all data sets. The correlation coefficients between our $PM_{2.5}$ values and those of the US Embassy and CNEMC are also very high at 0.6 and 0.9, respectively. These high correlation coefficients suggest that our reconstruction of the 1985–2018 $PM_{2.5}$ record from observed visibility data is credible.

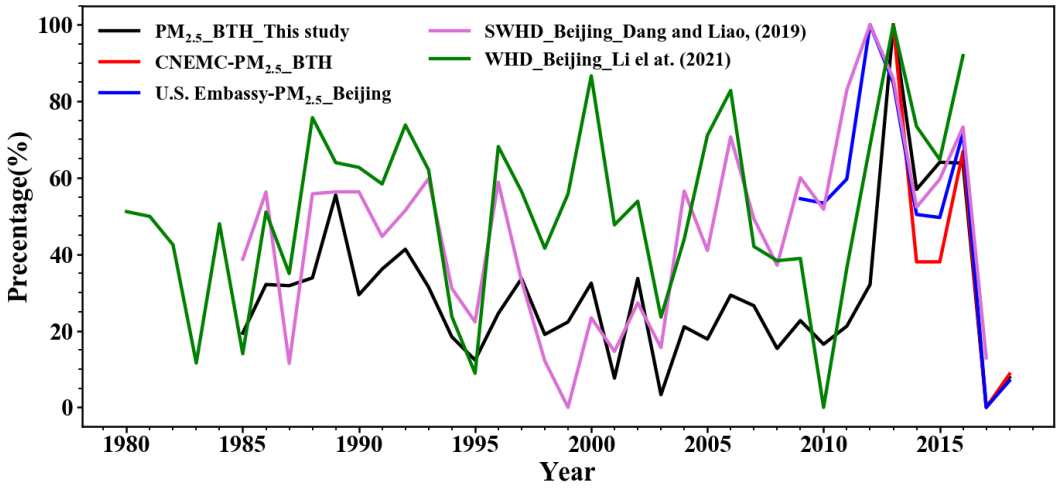

Figure R1. Temporal variations of winter inversed $PM_{2.5}$ concentrations in BTH of this study (black, 1985–2018), simulated $PM_{2.5}$ concentrations in Beijing by Dang and Liao (2019) (purple, 1985–2017), $PM_{2.5}$ concentrations observed by the US Embassy in Beijing (blue, 2009–2018) and those observed by CNEMC in BTH (red, 2013–2018).

(2) In our paper we didn't state "that attribution of recent $PM_{2.5}$ trends to emissions is not based on mechanistic knowledge". What we did state was that *quantitative attribution* of recent $PM_{2.5}$ trends to emissions is not based on *realistic/credible mechanistic models*. There is a significant difference between mechanistic knowledge

and realistic/credible mechanistic models. Only realistic/credible mechanistic models have the capability of making quantitative attribution of recent $PM_{2.5}$ trends. However, it is extremely formidable to make a multi-year realistic/credible simulation of the winter mean $PM_{2.5}$ in the megacity clusters in China. In our opinion, the most realistic multi-year mechanistic model simulation is the study by Dang and Liao (2019), who made a 33-year (1985–2017) model simulation study of severe winter haze days in BTH (purple line in Figure R1). There is an excellent agreement between the purple line and $PM_{2.5}$ concentrations observed by the US Embassy in Beijing (blue line, 2009–2018). The agreement with $PM_{2.5}$ concentrations observed by CNEMC in BTH (red line, 2013-2018) is also very good. For the entire period of 1985–2017, there are moderate mismatches near 1997–2002 and 2010 between the purple line (Dang and Liao, 2019) and green line (Li et al., 2021), but still has an acceptable overall correlation coefficient of 0.4. As cited in lines 201–202 of our paper, Dang and Liao (2019) "found that meteorology contributed significantly more than emissions to the linear trend", which is consistent with the result of our study.

(3) We accept the criticism of "belaboring trivial statistical points that are well known to any trained scientist." We will delete some of the repeated statements in the revised manuscript. We were surprised that previous MLR studies would have overlooked these trivial yet important statistical points, and tried to find an explanation. That leads us to realize the importance of the bulge-2013 (as noted by this reviewer in specific comment #1 below) and to suggest the "maximum possible contribution" as an alternative interpretation for the MLR results.

Specific comments

1. The 'bulge-2013' feature in Figure 1 (line 88) anchors much of the argumentation in the paper but it is very weird. It seems caused by the switch from the visibility proxy to the actual $PM_{2.5}$ data in 2013. The methods are buried in Supplementary Material. Is this 'bulge-2013' seen in the consistent long-term satellite AOD data record? I think that the authors would have to show that it is present in the AOD data in order to have

credibility.

Response:

We believe that Figure R1 and associated discussions above address this comment adequately. In response to the question about AOD, we compare below the winter satellite AOD data (MERRA2) in BTH to $PM_{2.5}$ and visibility (both from this study) in Figure R2. The correlation between AOD and $PM_{2.5}$ is fair (overall correlation coefficient 0.3) except some mismatches during two periods (2007–2009 and 2012–2013). As a result, only half of the bulge (2013–2018) can be seen in AOD. The reason for the mismatches is probably because surface $PM_{2.5}$ is sensitive to the height of mixed layer while AOD is not. In other words, changes of surface $PM_{2.5}$ due to changing mixing height are usually not detected in the AOD observations.

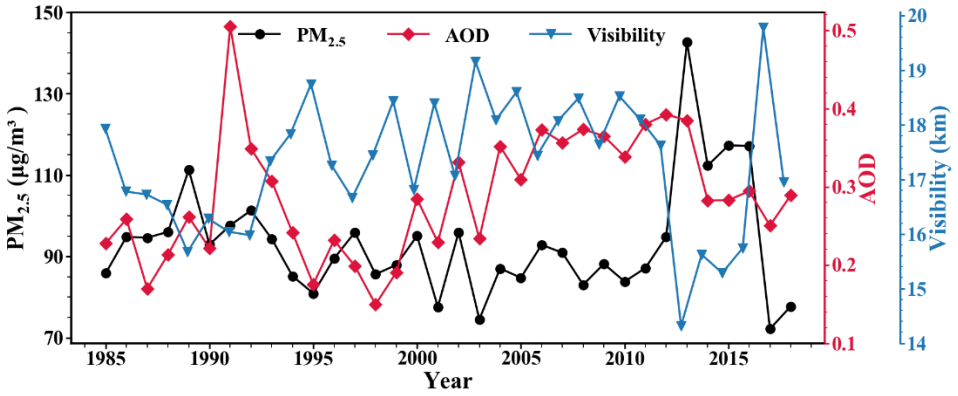

Figure R2. Time series of winter $PM_{2.5}$ concentration, visibility (both from our study) and AOD (from MERRA2) in BTH from 1985 to 2018.

2. What is the 'emission' in Figure 1? Of what species?

Response:

The 'emission' is composed of PM_{10}, $PM_{2.5}$, SO_2, NH_3, NOx, black carbon, and organic carbon in three sets of emission inventories (PKU inventory, MEIC inventory and PRD-EI inventory). Data and calculation methods for emissions are presented in Section 2.1.

As an example, Figure R3 shows the temporal variation of three emission inventories in PRD. They show generally consistent variations during overlapping periods.

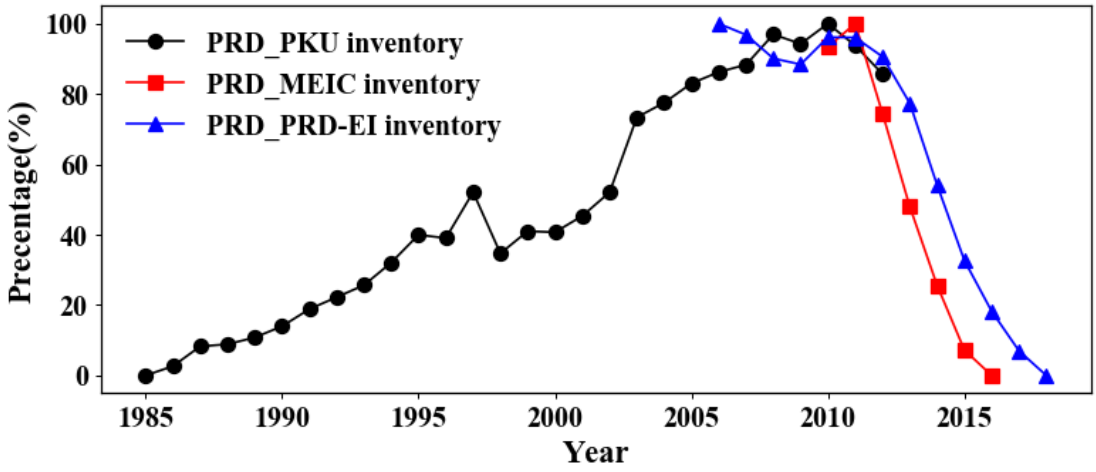

Figure R3. PKU emission inventory for winter 1985–2012, MEIC emission inventory for winter 2010–2016 and PRD-EI emission inventory for winter 2006–2018 for PRD. The raw data are normalized to the difference of the maximum value and minimum value.

3. Line 91: the Mao et al. 2019 reference which is intended to provide support for the authors' meteorological attribution of the trend is in fact grey literature involving some of the same authors.

Response:

Mao et al. (2019) is a peer reviewed article, not a "grey literature". The Mao et al. (2019) reference (Lines 352–353) is reproduced below:

Mao, L., Liu, R., Liao, W., Wang, X., Shao, M., Liu, S. C. and Zhang, Y.: An observation-based perspective of winter haze days in four major polluted regions of China, Natl. Sci. Rev., 6(3), 515–523, https://doi.org/10.1093/nsr/nwy118, 2019.

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4. Line 132, etc.: the mechanistic meteorological connection of ASI to $PM_{2.5}$ is not clear, and as the authors point out any meteorological variable with a suitable long-term trend

would do the trick. But there is in fact a strong mechanistic argument for emissions to be related to $PM_{2.5}$ (line 154), and there is strong independent evidence that Chinese emissions have decreased over the 2013-2018 period (emission inventories, satellite data). To claim that the connection of $PM_{2.5}$ to emissions has no mechanistic support strikes me as obviously wrong. In fact the authors cite Chen et al. 2019 in demonstrating the mechanistic connection in WRF-CMAQ but argue that the analysis is flawed because it did not consider the effect of the bulge-2013 (line 158). As pointed out above, I am very suspicious of this bulge-2013.

Response:

We have addressed extensively the issues raised here in general comment #2 (and #1 about the bulge-2013). Moreover, in lines 261–262 we already stated "there is little doubt that anthropogenic emissions make a significant contribution to the reduction trend of $PM_{2.5}$." We were only "skeptical of *those high* contributions by emissions obtained based solely on MLR models."

5. There is a lot of trivial stuff about the non-mechanistic basis of statistical models, correlation not implying causality, more years increasing the credibility of the model, etc., that is repeated again and again and does not rise above the level of a basic course in statistics.

Response:

As stated in our response to general comment #3, we accept the criticism of "belaboring trivial statistical points that are well known to any trained scientist," We will delete some of the repeated statements in the revised manuscript.

References

Dang, R. and Liao, H.: Severe winter haze days in the Beijing-Tianjin-Hebei region from 1985 to 2017 and the roles of anthropogenic emissions and meteorology, Atmos. Chem. Phys., 19(16), 10801–10816, https://doi.org/10.5194/acp-19-10801-2019, 2019.

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