Comments of Anonymous Referee #1

The manuscript "Dramatic changes in atmospheric pollution source contributions for a coastal megacity in Northern China from 2011 to 2020" by Baoshuang Liu et al. reports the long-term variations of major air pollutants. By applying a Random Forest method, Theil-Sen regression, and dispersion normalization, the authors separated the contribution of meteorology and that of clean air actions to the air pollution mitigation. I am so sorry that I have uploaded a wrong comment file during the first round of the review. After noticed that, I went through both the original and the revised manuscripts. In general, I think they are well written and provide valuable information to the community. I recommend the publication after some minor revisions. Please noted that my following comments are referring to the revised manuscript.

General comments

1. The authors essentially applied two methods (i.e., RF and VC) to decouple the influence of meteorology. It would be interested to know to which extent the corrected temporal variations agree to each other. Also, it is better to give a short explanation of using VC instead of RF correction for the analysis in Section 3.3 and 3.4.

Response: Thanks for your advice. (1) The RF-based weather normalization method can well decouple the overall weather effects, while the VC-normalization can only decouple the local dispersion. VC-normalization is relatively simple but needs VC measurement data to be known a priori, while RF-based weather normalization needs a large size of data to well training the model before de-weathering. *The fact that there is a big difference in the size and time-resolution between the two datasets (routine air quality data versus PM chemical composition data)*, we therefore chose two methods rather than a "one-size-fits-all" approach to decouple the "weather effects" based on the strengths and limitations of methodologies. To this end, we chose RF-based weather normalization to de-weathering for air quality data that measured in 2015-2020, and used VC-normalization for offline filter-based measured chemical compositional data. A comparison of the two methods sounds desirable but does not make physical sense cause both methods fit their own purposes. (2) In this study, we added related explanation of using VC instead of RF normalization in the revised manuscript (on the lines 203-211). The more details as following:

Although the RF-based weather normalization method can well decouple the overall weather effects, it needs a large size of data to well training the model before de-weathering. The fact that there is a big difference in the size and time-resolution between the routine air quality data and the offline filter-based measured $PM_{2.5}$ chemical compositional data. However, the meteorological dispersion can be quantified by the ventilation coefficient (VC) (Kleinman et al., 1976; Iyer and Raj, 2013). Although the VC-normalization that needs VC data to be known a priori can only decouple the local dispersion, it is relatively simple and useful to decouple the impact of dispersion (Ding et al., 2021). Therefore, this normalized approach is very suitable for the offline data with small size and poor continuity.

2. In section 3.3.2, the authors used VC for correcting the meteorological influence on the source apportionment results. How would this be compared with the source apportionment derived from the VC corrected PM concentrations?

Response: Thanks for your advice. The reviewer's suggestion is very good. Source apportionment can be conducted by the PM composition data after dispersion normalization, we have carried out relevant research in the early stage (as shown in Dai et al. (2020)), and constructed the dispersion normalized PMF (DN-PMF); firstly, we normalized the PM composition data using the VC data during the study period, and then carried out the source apportionment by these normalized data so that the apportioned results can more accurately reflect the impact of emission sources. In contrast, this study mainly used the VC data to normalize the results of source analysis to correct the impact of meteorological conditions, to better reflect the impact of emission sources. In fact, we are carrying out relevant studies on the comparison of the two methods, and the relevant results will be published in the future. However, the related analysis was obviously beyond the scope of this study. The purpose of this study was to use the mature normalized methods to analyze the changes in emission sources in Qingdao in recent 10 years.

Dai, Q. L., Liu, B. S., Bi, X. H., Wu, J. H., Liang, D. N., Zhang, Y. F., Feng, Y. C., and Hopke, P. K.: Dispersion Normalized PMF Provides Insights into the Significant Changes in Source Contributions to $PM_{2.5}$ after the COVID-19 Outbreak, Environ. Sci. Technol., 54, 9917-9927, https://doi.org/10.1021/acs.est.0c02776, 2020.

Specific comments

Line 102 -103, Page 4: It is better to give a quantitative description (e.g., AQI or $PM_{2.5}$ changes) on “greatly improved".

Response: Thanks for your advice. We added the quantitative descriptions on “greatly improved” in the revised manuscript (on the lines 102-103). The more details as following:

Up to now, the air quality in Qingdao has been greatly improved, the annual mean concentrations of $PM_{2.5}$ and PM_{10} all decreased by 38% from 2015 to 2020 based on the air quality monitoring data.

Line 147, Page 6: Are the sampling instruments home-built or commercial? Please also specify the size of the sampling filter and the sampling flow rate.

Response: Thanks for your advice. The sampling instruments are commercial and the more details on the instrument corporations, sampling filter and the sampling flow rate are shown in Table S3.

Table S3. Details of sampling instruments and filters during different sampling years.

Year	Instrument	Model	Corporation	Country	Flow rate (L min^{-1})	Filter diameter (mm)	Filter category	Corporation	Country
2011-2012	Four channel air particulate matter sampler	TH-16A	Wuhan Tianhong Instrument Co., Ltd	China	16.7	47	Polypropylene/ Quartz	Beijing Synthetic Fiber Research Institute/Pall Life Sciences	China/ USA
2016	Multichannel ambient air particulate sampler	ZR-3930D	Qingdao Junray Intelligent Instrument Co., Ltd	China	16.7	47	Polypropylene/ Quartz	Munktell	Sweden

2019	Multichannel ambient air particulate sampler	ZR-3930D	Qingdao Junray Intelligent Instrument Co., Ltd	China	16.7	47	Polypropylene/ Quartz	Pall Life Sciences	USA

Line 386-389, Page 14: While the enhancement of atmospheric oxidation can certainly cause O_3 increase, the strong decrease of NO_2 (by almost the same percentage as that of O3 increase) indicating a weakened "NOx titration effect" which may also result in higher O_3 levels, especially during cold seasons when photooxidation is usually weak.

Response: Thanks for your advice. We very agree with the reviewer's suggestion. The related explanations have been added in the revised manuscript (on the lines 386-388). The more details as following:

Meanwhile, the markedly decrease of NO_2 during the full lockdown can also weaken "NOx titration effect", further resulting in higher O_3 level during this period.

Line 416-418, Page 15: The unit for VC should be "$m^2\ s^{-1}$"

Response: Thanks for your advice. The unit of VC has been modified in the revised manuscript (on the lines 414 and 416).