Dear Editors and Referees:

Thank you very much for your constructive suggestions and helpful comments for improving our manuscript acp-2019-758. We have accordingly made the careful revisions. Revised portions are highlighted in the revised manuscript. In the following we quoted each review question in the square brackets and added our response after each paragraph.

Responses to Referee #2

[General comments.

In this manuscript, the authors present an observational analysis to characterize the unique features of meteorological conditions that account for the heavy air pollution events in Wu Han, a metropolis in the Yangtze River Middle Basin, China. and then use a Lagrange particle dispersion model to quantify the percentage contribution of regional transport to such heavy pollution events. They found that $PM_{2.5}$ concentrations show a positive correlation with wind speeds and no stable atmospheric boundary conditions are required to support the accumulation of air pollutants when 24-hr average $PM_{2.5}$ concentrations are higher than 150.0 $\mu g \cdot m^{-3}$. Regional transport driven by strong wind speed contributed more than 65% increase in surface $PM_{2.5}$ concentrations during the development of air pollution events in this region. The study represents a great interest to air quality community given the unique features which are very different from those presented in the textbooks. This version is improved to some extent as compared to the first submission. Part of my comments have been addressed but not all. Especially, the manuscript structure is not re-organized as suggested, a lot of grammar errors or typos need to be corrected throughout the manuscript. In addition, I have several major concerns with the authors' arguments during their analyses and discussion. Thus, a major revision is still required before it is accepted for publication.]

Response 1: Many thanks for the encouraging comments and constructive suggestions on our manuscript acp-2019-758. According to the suggestions of referee, we have re-organized

the manuscript structure with detailed descriptions of observational and modeling methods, concerns of technical methodology as well as corrected the grammar errors and typos over the entire manuscript (please find them in the following responses and the highlighted revisions in the revised manuscript).

[Major comments

1. It is strongly recommended to re-organize the structure of the manuscript. Both Methodology and Results/Discussion parts are mixed together in the current version. For instance, it is suggested to move "Model Description (Section 3.2.1)", "Model Configuration (Section 3.2.2)", and the way of calculating "contribution rates" (Lines 360-375 in Section 3.3) to a new section like "Data and methods" (say Section 2 in the new version), and then move part of current Section 2.1, Sections 2.2 and 2.3 to Section 3 like "Results and Discussion" in the revised or new version something like that.]

Response 2: Following the referee's suggestions, we have re-organized the structure of the paper. In the revised manuscript, the descriptions of observational data from various sites and FLEXTPART-WRF (Sect. 3.2.1,Sect. 3.2.2 and the way of calculating "contribution rates" in Section 3.3) are placed in Section 2. And Results and Discussions, including the analysis of the observational data and modeling study, are placed in Section 3*"Results and Discussion"* .

[2. The East Asian winter Monsoon was mentioned at least 10 times throughout the manuscript to highlight its importance in driving the regional transport during development of heavy pollution events observed in Wuhan. As we know, the East Asian Monsoon represents a seasonal mean behavior and its temporal scale is much longer than that of air pollution events which usually have a scale of one to several day(s) but not longer than one week according to the authors' argument. The authors need show some scientific evidences to support their arguments on how the East Asian winter Monsoon can drive the regional transport which may lead to the development of heavy pollution events. Otherwise the readers may get confused when they read

Fig.9b in where the regional transport was from East China other than North China. My suggestion is to limit the emphasis of the East Asian winter Monsoon in this study.]

Response 3: We totally agree with the referee's comments and suggestions. Following the them to correct this misunderstanding on the East Asian Monsoons, we have changed "the East Asian monsoon" to "the cold air activity of East Asian winter monsoon over central-eastern China" to limit the emphasis of the East Asian winter Monsoon in this study (please see the highlighted revisions in the revised manuscript.

[3. Estimate of percentage contribution of regional transport to the heavy pollution events in the YRMB region is one of the major works proposed by this study. As described in Eq.1 and 2, simulation of residence time of $PM_{2.5}$ is critical to do such calculations. Please define residence time. How does the FLEXPART simulate the residence time? A little bit more details are helpful for our readers to understand the percentage contribution of regional transports to the three different episodes.]

Response 4: Thanks for the comments. In the revised manuscript, we have clarified the quantification of regional transport contributions with utilizing the model FLEXPART-WRF in the revised manuscript as followings:

In the model FLEXPART-WRF, the trajectory of a large number of particles released from a source is simulated with consideration of the processes of tracer transport, turbulent diffusion, wet and dry depositions in the atmosphere. With Lagrangian method, it could result in a Jacobian matrix (footprint), in unit of mass per volume per unit flux. Stohl et al. (2005) mathematically derived the residence time for particles out of FLEXPART. Generally, in the backward trajectory of FLEXPART modeling, a large number of particles is released at a receptor and transported backward in time. Then the residence time (not the lifetime) of all particles, normalized by the total number of released particles, is determined

on a uniform grid. In this study for the receptor of Wuhan, the residence time for a thickness of 100 m above the surface was calculated and considered the ‘‘footprint’’ (in unit of s). By multiplying the residence time with the air pollutant emission flux in the respective grid cell (in unit of μg m^{-2} s^{-1}) calculated from the Multi-resolution Emission Inventory of year 2016 for China (MEIC, http://www.meicmodel.org/), the emission source contribution (in μg m^{-2}) from this grid cell to the receptor could be estimated (Stohl, 2003; Stohl et al., 2005;Ding er al.,2009), yielding a so-called potential source contribution map, which is the geographical distribution of the regional transport contribution rates (%) of the emission source grid cell to $PM_{2.5}$ pollution at the receptor of Wuhan (Fig. 9).

References

Stohl, A., Forster, C., Eckhardt, S., Spichtinger, N., Huntrieser, H., Heland, J., Schlager, H., Wilhelm, S., Arnold, F., and Cooper, O.: A backward modeling study of intercontinental pollution transport using aircraft measurements, Journal of Geophysical Research: Atmospheres, 108, https://doi.org/10.1029/2002jd002862, 2003..

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, Atmospheric Chemistry & Physics, 5, 2461-2474, https://doi.org/10.5194/acp-5-2461-2005, 2005.

Ding, A., Wang, T., Xue, L., Gao, J., Stohl, A., Lei, H., Jin, D., Ren, Y., Wang, X., and Wei, X.: Transport of north China air pollution by midlatitude cyclones: Case study of aircraft measurements in summer 2007, Journal of Geophysical Research: Atmospheres, 114, https://doi.org/doi:10.1029/2008JD011023, 2009.

[4. Lines 323-329: I assume that the FLEXPART simulations were driven by the WRF outputs rather than ECMWF or NCEP reanalysis data. If this is the case, please make clarification and delete lines 323-325.]

Response 5: Following the referee’s suggestion, we have made clarification with deleting lines 323-325 in the revised manuscript as follows:

In this study on the fine and multiscale modeling of air pollutant sources and regional transport, FLEXPART was coupled offline with the Weather Research and Forecasting Model (WRF) to effectively devise the combined model FLEXPART-WRF.

[5. Fig.5b: We can see that the heavy air pollution events had stronger winds within the 1-km layer but weaker winds above the 1-km layer. Does this mean that regional transport are mainly limited to the 1-km layer. Some discussions on this will be helpful.]

Response 6: Thanks for the referee's comment. We have accordingly added the following discussions in the revised manuscript:

Compared to the clean air period, the heavy air pollution events had stronger winds within the 1-km layer but weaker winds above the 1-km layer (Fig. 5b), indicating that regional transport of $PM_{2.5}$ was mainly limited to the 1-km layer, especially between 0.25 km and 0.8 km. These vertical structure of horizontal wind could conduce the downward mixing of the regionally transported air pollutants and produce the near-surface accumulations of air pollutants over the YRMB area with elevated ambient $PM_{2.5}$ concentrations, thus contributing to a heavy air pollution.

[6. Writing needs a heavy edit work. There are a lot of grammar errors or typos and many sentences need further improvement. Some of examples include "obviously differences (L107)", "relative high (L109)", "suffering under significant (L133)", "has significantly influence (L162)", "relatively to (L276)", "a horizontally resolution (L344)", etc. I am not going to list all of them since there are many.]

Response 7: Many thanks for the referee's careful review. We are so sorry for the grammar errors or typos, which have been corrected in the revised manuscript.

By the way, this revised manuscript was edited by Elsevier Language Editing Services to improve the English language.

Minor comments:

[1. L19: central China → Central China.]

Response 8: It has been changed.

[2. L20: I am not sure "excessive" is appropriate in this manuscript.]

Response 9: The "excessive $PM_{2.5}$ concentrations" has been changed to "hourly $PM_{2.5}$ concentrations" in the revised manuscript.

[3. L30: I did check "List of regions of China" at Wikipedia at https://en.wikipedia.org/wiki/List_of_regions_of_China, and didn't find "central-eastern China". So "Central China" should be better and sufficient.]

Response 10: I am sorry for the geographical misleading. In the revised manuscript, we have changed *"central-eastern China" to "Central and Eastern China"* covering the major anthropogenic pollutant sources over the vast flatlands from the eastern edges of the Tibetan Plateau and the Loess Plateau to China's Pacific coast for the regional transport of air pollutants toward to YRMB.

[4. L33: FLEXPART-WRF or WRF-FLEAPART? I would suggest the latter since it is WRF-driven FLEXPART. In addition, please define any abbreviated terms at its first appearance. Please check similar issue for other abbreviations throughout the manuscript.]

Response 10: We totally agree with the referee's comments. However, the Lagrangian particle dispersion model FLEXPART-WRF was developed by Brioude et al. (2013), therefore, we adopted the model name FLEXPART-WRF in the manuscript.

All the abbreviated terms were defined at its first appearance in the revised manuscript.

Refereces

Brioude, J., Arnold, D., Stohl, A., Cassiani, M., Morton, D., Seibert, P., Angevine, W., Evan, S., Dingwell, A., Fast, J. D., Easter, R. C., Pisso, I., Burkhart, J., and Wotawa, G.: The Lagrangian particle dispersion model FLEXPART-WRF version 3.1, Geoscientific Model Development, 6, 1889-1904, https://doi.org/10.5194/gmd-6-1889-2013, 2013.

[5. L155-157: Please define these abbreviations at their first appearances.]

Response 11: We have defined these abbreviations at their first appearances in the revised manuscript.

[6. L251: change "the atmospheric stability in the boundary layer" to "the stability of the atmospheric boundary layer"?]

Response 12: It has been changed.

[7. L272: are you sure "it is in Section 3.1"?]

Response 13: It is "3. Regional transport of PM2.5 in heavy air pollution periods".

[8. L342: Please change to (30.61°N, 114.42°E).]

Response 14: It has been changed.

[9. L232-235: I feel a "jump" when I read this sentence.]

Response 15: We have modified this sentence in the revised manuscript as follows:

The meteorological drivers of air quality change are complicated by a series of physical and chemical processes in the atmosphere especially the formation of secondary air pollutants with strong hygroscopic growth in the humid air environment overlying the dense water network (see Fig. 1b) in the YRMB region (Cheng et al., 2014, He et al., 2012, Huang et al., 2014),

References

Cheng, H., Gong, W., Wang, Z., Zhang, F., Wang, X., Lv, X., Liu, J., Fu, X., and Zhang, G.: Ionic composition of submicron particles ($PM_{1.0}$) during the long-lasting haze period in January 2013 in Wuhan, central China, Journal of Environmental Sciences, 26, 810-817, https://doi.org/10.1016/s1001-0742(13)60503-3, 2014.

He, K., Zhao, Q., Ma, Y., Duan, F., Yang, F., Shi, Z., and Chen, G.: Spatial and seasonal variability of $PM_{2.5}$ acidity at two Chinese megacities: insights into the formation of secondary inorganic aerosols, Atmospheric Chemistry and Physics, 12, 1377-1395, https://doi.org/10.5194/acp-12-1377-2012, 2012.

Huang, R. J., Zhang, Y., Bozzetti, C., Ho, K. F., Cao, J. J., Han, Y., Daellenbach, K. R., Slowik, J. G., Platt, S. M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S. M., Bruns, E. A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z.,

Szidat, S., Baltensperger, U., El Haddad, I., and Prevot, A. S.: High secondary aerosol contribution to particulate pollution during haze events in China, Nature, 514, 218-222, https://doi.org/10.1038/nature13774, 2014.

[10. L352: "The simulated meteorology" → "The simulated meteorological fields".]

Response 16: It has been changed.

[11. L373-374: Change "by calculation of the $PM_{2.5}$ contribution rates with Eq (1)" to "by using the $PM_{2.5}$ contribution rates calculated with Eq.1" something like that.]

Response 17: Thanks for careful editing. We have been accordingly changed in the revised manuscript.

[12. L309-313: I do not think this paragraph is necessary since it does not provide any useful information. Similar issue can be found in other places of the manuscript.]

Response 18: Following the referee's suggestion, we have deleted the unnecessary sentences and paragraphs.

[13. L338-339: What are the horizontal resolutions of the NCEP reanalysis data?]

Response 19: *the horizontal resolutions of the NCEP reanalysis data* is 1°×1°, which has been added in the revised manuscript.

[14. L419-423: Does this paragraph represent any significant findings or conclusions obtained from this study? I am not sure this paragraph is really needed here.]

Response 20: We have accepted the suggestion of referee and deleted this paragraph in the revised manuscript.

[15. L424-426: We know this already and I don't think you need iterate this sentence here. It does not provide any more useful information to me.]

Response 21: Following the suggestion of referee, we have deleted the sentence (L424-426) and modified the paragraph as follows:

This study of environmental and meteorological observations in the YRMB region revealed a unique "non-stagnant" meteorological condition of the boundary layer characterized by strong wind, no inversion layer and a more unstable structure in the atmospheric boundary layer associated with heavy air pollution periods with excessive $PM_{2.5}$ concentrations in the YRMB region, which facilitates understanding of the air pollutant source-receptor relationship of regional air pollutant transport. The study represents a great interest to air quality community given the unique features of air pollution meteorology which are very different from those "stagnant" meteorological conditions presented in the textbooks.

[16. L625-629: Please define WS, T, P, and RH in the description of Table 1 and Table 2.

Response 22: WS, T, P, and RH have been defined with wind speed, air temperature, air pressure and relative humidity respectively in the revised manuscript.

[17. Fig.1b: The font size of those cities shown in Fig.1b is too small. Is it possible to add the locations of 10 sites presented on Page 5 at Lines 101-103 in this plot?]

Response 23: We have added the locations of 10 sites in Wuhan in the supplemental file (Fig. s1).

[18. Fig.9: I believe that the values of the percentage contribution rates are not correct.]

Response 24: Thanks for the comments. We have confirmed that the values of the percentage contribution rates in the Fig.9 are correct. Fig.9 is composed by 151×161 grid points, and the total contribution rate of all grid points is 100%.