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 #1

[Yu et al investigated the impacts of regional transport to the heavy haze pollution in January 2016 over Wuhan, a city located over the Yangtze River Middle Basin in the central part of China. This study characterized unique "non-stagnant" conditions (e.g., high winds, no inversion layers) associated with extreme high levels of $PM_{2.5}$ concentrations (e.g., strong correlation between $PM_{2.5}$ concentrations greater 150 μg m^{-3} and wind speed), which differed significantly from traditional haze pollutions with low near-surface winds and inversion layers found in the literatures. The authors employed both observational and modeling analyses to prove the importance of the contribution of regional transport to the excessive $PM_{2.5}$ concentrations over Wuhan. This is an interesting study to demonstrate the complexity and challenge of the severe haze pollution over central-eastern China during wintertime, with research scope aligned with topics suitable for ACP. However, the current format of the manuscript is not accepted, due to ambiguous structure of the manuscript, lack of detailed descriptions of observational and modeling methods, concerns of technical methodology as well as numerous grammar errors and typos over the entire manuscript. A major revision is needed for this manuscript before further consideration of publication in ACP. My comments for the manuscript are shown as follows.]

Response 1: Many thanks for the encouraging comments and constructive suggestions on our manuscript acp-2019-758. Accordingly, we have restructured the manuscript 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. Research Methodology and Results/Discussions for the paper are not clear I have difficulty in following the paper's research methodology/results. The authors mix the research methodology and results in the same section. I highly recommend that the authors should re-organize the structure of the paper. The descriptions of observational data from various sites and FLEXTPART-WRF (Sect. 3.2.1 and Sect. 3.2.2) should be placed in Section 2. And Results and Discussions, including the analysis of the observational data and modeling study, should be placed in Section 3.]

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 and Sect. 3.2.2) are placed in Section 2. And Results and Discussions, including the analysis of the observational data and modeling study, are placed in Section 3.

[2. The descriptions of the data used in this study are not adequate and needed to be expanded to provide a more detailed and rigorous documentation.

We don't know the spatial locations of the observational sites for $PM_{2.5}$ measurements, especially ten sites over Wuhan, which need to be presented. A spatial map of WRF modeling domain, with $PM_{2.5}$ measurement sites inserted, will be very helpful. Moreover, what is the measurement technique used for $PM_{2.5}$? What is the measured frequency/quality data control method, and measurement uncertainty associated with $PM_{2.5}$ concentrations and other meteorological parameters for each site? How do you represent Wuhan's hourly $PM_{2.5}$ concentrations out of the ten measurement sites? And how do you calculate the correlation coefficients between $PM_{2.5}$ concentrations and wind speed/temperature over Wuhan in January 2016 out of ten measured sites?]

Response 3: According to the referee's suggestions, we have added the spatial locations of the ten observational sites for $PM_{2.5}$ measurements over Wuhan in the supplemental (Fig. s1). Besides, the $PM_{2.5}$ data used in this study were collected from the national air quality monitoring network operated by the Ministry of ecology and environmental protection of China, The mass volume concentrations of surface $PM_{2.5}$ are operationally hourly observed with the instrument of the Thermo Fisher Scientifi. The observation data are under quality control based on the China's national standard of air quality observation before released by the Ministry of ecology and environmental protection of China. The source of the data has been added in the revised manuscript. At last, the surface $PM_{2.5}$ concentrations averaged over 10 observation sites in Wuhan are used to calculate the correlation coefficients with the changing meteorological drivers (wind speed/temperature etc.) over Wuhan in January 2016 to investigate the local meteorological influences on hourly changes of surface $PM_{2.5}$ concentrations in Wuhan.

[3. In terms of quantification of regional transport contributions for PM2.5 over Wuhan, the authors have utilized FLEXPART-WRF model. However, I have concerns about the convolution of FLEXPART-WRF residence time with the PM2.5 bottom-up emission fluxes from MEIC. Firstly, what is the definition of residence time here? Is it the PM2.5 lifetime? With Lagrangian method, it will result in a Jacobian matrix (footprint), in unit of mass per volume per unit flux. It is helpful for the authors to mathematically derive the residence time for particles out of FLEXPART, the product of the residence time and the bottom-up emission flux, and ultimately the regional transport contribution rate in the "Research Methodology" Section. The authors should insert the unit for each variable out of FLEXPART modeling. Meanwhile, please help the readers about the purpose of the WRF model here. Further, FLEXPART does not consider chemistry and deposition in the model, the only part it accounts for is the transport, driven by reanalysis data. PM2.5 contains a significant portion of secondary organic and inorganic aerosols, which come from important and complex physiochemical processes in the atmosphere. How this methodology (FLEXPART-WRF) is proven robustness to quantify the regional transport contribution? What is the uncertainty range here?]

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).

A need for further multiscale modeling and analysis has encouraged new developments in FLEXPART-WRF, a FLEXPART version that works with the Weather

Research and Forecasting (WRF) mesoscale meteorological model (Brioude et. al., 2013). For the refined simulation of air pollutant sources and transport, FLEXPART modeling driven by mesoscale meteorology from WRF modeling has been widely used to investigate the potential sources of air pollutants in consideration of air pollution change.

In this study, the $PM_{2.5}$ contributions of regional transport to air pollution in the downwind receptor region could be approximately estimated based on the product of the residence time of air particles during regional transport simulated by FLEXPART-WRF, and the $PM_{2.5}$ emission flux over the source grid in Central and Eastern China. The potential source contribution is estimated based on transport alone, ignoring chemical and removal processes. We also understand that the physical and chemical processes such as complex deposition and chemical conversion for the formation of secondary particles are not introduced in the FLEXPART-WRF emulation, which could represent the basic features of contribution and patterns of regional $PM_{2.5}$ transport over central and eastern China, when limited to the primary $PM_{2.5}$ particles highlighted in this study. Considering less precipitation in the winter monsoon season, how this methodology (FLEXPART-WRF) is proven robustness to quantify the regional transport contribution with the uncertainty range here could mostly rely on a portion of secondary organic and inorganic aerosols, which are resulted from important and complex physiochemical processes in the atmosphere.

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.
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.

Minor Comments

[1. Line 48: The order of the references is messed up, which should follow the order of the first letter of the first author for each reference alphabetically, and should be "An et al., 2019; Fuzzi et al., 2015; Nel, 2005" for this case. Please check the entire manuscript.]

Response 5: We have corrected all the similar errors in the revised references.

[2. Line 50: The definition of PM2.5 "particulate matter with an aerodynamical diameter equal to or less than 2.5 micrometers".]

Response 6: It has been revised.

[3. Line 99: change "humid environment. (see Fig. 1b)" to "humid environment (see Fig.1b)". There are so many similar typos across the entire manuscript. Please CHECK!]

Response 7: We have corrected all the similar errors in the revised manuscript.

[4. Line 101: The associated temporal variations of $PM_{2.5}$ concentrations for the study period out of ten sites in Wuhan are strongly recommended to be plotted and placed in the Supplemental.]

Response 8: According to the referee's suggestions, we have added the temporal variations of $PM_{2.5}$ concentrations for the study period out of ten sites in Wuhan in the supplemental file (Fig. s2 and s3).

[5. Line 107: Change "obviously" to "obvious".]

Response 9: It has been revised, "obviously" has been changed to "obvious".

[6. Line 124: "heavy $PM_{2.5}$ pollution the over central-eastern China" should be revised as "heavy PM2.5 pollution over the central-eastern China".]

Response 10: It has been revised.

[7. Line 128: The number and unit should be separated (75 μg m^{-3}). Similar changes should be applied for the entire manuscript.]

Response 11: All the similar errors have been corrected in the revised manuscript.

[8. Line 146: "at same day." should be changed to 'at the same day,".]

Response 12: "at same day." has changed to 'at the same day".

[9. Lines 147-Line 149: The authors use "am" and "a.m." interchangeable. Please be consistent for the entire manuscript. Similar for "pm" and "p.m.".]

Response 13: All the similar errors have been corrected in the revised manuscript.

[10. Lines 161-165: Grammar error here. Please re-write this sentence. And what is the logical relationship between this sentence and the previous one? Do you try to demonstrate the reasons for this result? If so, probably it is better to begin the sentence with "There are several reasons associated with this result. Firstly, ……".]

Response 14: We are so sorry for the grammar error here. Following the referee's suggestion, we have re-written the sentence as follows:

There are several reasons associated with this result. Firstly, the lower near-surface wind speed could alter the concentrations of air pollutants with a weaker advection of cold air, in conjunction with strong subsidence and stable atmospheric stratification, easily producing a stagnation area in the lower troposphere with resulting in regional pollutant accumulations for the development of haze events.

[11. Line 165: what is "CEC" here?]

Response 15: The CEC stands for Central-eastern China; and it has been revised in the manuscript.

[12. Lines 165-170: There are many typos and grammar errors in this sentence. And I am confused by this sentence as well, which looks very odd to me. Is this your statement or conclusion? Several references to support your statement will be necessary.]

Response 16: We are so sorry for the typos and grammar error here. Following the referee's suggestion, we have modified the sentence with adding the relevant references to support our statement as follows:

Secondly, in the presence of high soil moisture, strong surface evaporation could increase the near-surface relative humidity, which is also conducive to hygroscopic growth of participles for haze formation (Dawson et al., 2014; Xu et al. 2016). High air temperature and strong solar radiation could enhance chemical conversions for the formation of secondary aerosols in the atmosphere (He et al., 2012; Huang et al., 2014). Furthermore, precipitation could alter the emissions, and depositions of air pollutants (Dawson et al., 2007; Cheng et al. 2016).

References

Cheng, X., Zhao, T., Gong, S., Xu, X., Han, Y., Yin, Y., Tang, L., He, H., and He, J.: Implications of East Asian summer and winter monsoons for interannual aerosol variations over central-eastern China, Atmospheric Environment, 129, 218-228, https://doi.org/10.1016/j.atmosenv.2016.01.037, 2016.

Dawson, J., Adams, P., and Pandis, S.: Sensitivity of $PM_{2.5}$ to climate in the Eastern US: a modeling case study, Atmospheric chemistry and physics, 7, 4295-4309, https://doi.org/10.5194/acp-7-4295-2007, 2007.

Dawson, J. P., Bloomer, B. J., Winner, D. A., and Weaver, C. P.: Understanding the Meteorological Drivers of U.S. Particulate Matter Concentrations in a Changing Climate, Bulletin of the American Meteorological Society, 95, 521-532, https://doi.org/10.1175/bams-d-12-00181.1, 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.
Xu, X., Zhao, T., Liu, F., Gong, S. L., Kristovich, D., Lu, C., Guo, Y., Cheng, X., Wang, Y., and Ding, G.: Climate modulation of the Tibetan Plateau on haze in China, Atmospheric Chemistry and Physics, 16, 1365-1375, https://doi.org/10.5194/acp-16-1365-2016, 2016.

[13. Lines 184-185: There should be spaces between references, which should be "(Miao et al., 2018; Xu et al., 2016b). There are many cases (e.g., Line 187, 254, 263 and etc) like this. Please check over the entire manuscript.]

Response 17: We have corrected all the similar errors in the revised manuscript.

[14. Line 210: "the stagnation meteorological conditions" should be revised as "meteorological conditions of the stagnation".]

Response 18: It has been revised as suggested by the referee.

[15. Lines 233-234: References relevant to secondary organic and inorganic aerosols study over Wuhan?]

Response 19: Following the referee's suggestion, we have modified the sentence with adding the references relevant to secondary organic and inorganic aerosols study over Wuhan 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.

[16. Line 276: Change "relatively" to "relative".]

Response 20: It has been changed.

[17. Lines 296-299: First of all, there are grammar errors in this sentence (e.g., ...by winter monsoonal winds the from Tongling and Hefei to Wuhan (...). Second of all, the site numbers of Tongling and Hefei are 6 and 5 respectively, as indicated by legend of Figure 6a?)]

Response 21: In the revised manuscript, the grammar errors have been corrected, and the site numbers have been modified as indicated by legend of Figure 6a.

[18. Lines 311-313: It seems that this sentence belongs to the beginning of Section 3.2.]

Response 22: Yes, this sentence (Lines 311-313) has been moved to the beginning of Section 3.2.

[19. Lines 331-333: I recommend that the authors make a plot associated with the modeling domains, which demonstrates the regions with the coarse and finer horizontal resolutions (refer to my major comment #2).]

Response 23: We have added the modeling domains with the coarse and finer horizontal resolutions in the supplemental file (Fig.s4).

[20. Lines 341-342: I have concerns about the release of the number of particles in FLEXPART-WRF. Firstly, for particles from FLEXPART, it is not PM2.5 particles, it is just particles to represent the air parcels. Secondly, can you double check that the model release 50,000 particles per hour? From my understanding, for each hourly mean PM2.5 observation at the receptor list, the release of particles in the 48-h backward trajectory simulation in FLEXTPART just happens in the first hour, with the rest of the time tracking the routes/transport of the particles over the simulation domain?]

Response 24: Yes. Many thanks for the kind review. We have carefully checked our model configuration, and accordingly corrected the errors in the revised manuscript as follows:

For particles from FLEXPART, it is not $PM_{2.5}$ particles, it is just particles to represent the air parcels, and the release of particles in the 48-h backward trajectory simulation in FLEXTPART just happens in the first hour, with the rest of the time tracking the routes/transport of the particles over the simulation domain.

[21. Line 374: Change "Eq (1)" to "Eq. (1)".]

Response 25: It has been corrected.

[22. Lines 634-637: For "K km^{-1}", it should be "K km^{-1}".]

Response 26: It has been revised.

[23. Lines 640-645: There are many typos for Figure 1. For Y-axis title in Figure 1a, it should be "Latitude". Moreover, both units of X-axis and Y-axis in Figure 1a are missing. In Line 643, "YPD" is a typo. And where is the description of PRD here?]

Response 27: Many thanks for the careful review of referee. We are sorry for the typos, which have been corrected in the revised manuscript.

[24. Lines 663-664: The solid line for heavy $PM_{2.5}$ pollution and the dash line for clean air period are missing in the caption for Figure 5.]

Responses 28： We have modified the caption for Figure 5.

[25. Lines 679-680: Why there are no "comma" among "P1 P2 and P3". I suggest changing the caption of the last part of the caption of Figure 7 as "…. pollution periods of P1 (upper panel), P2 (middle panel) and P3 (lower panel), respectively, in January 2016".]

Response 29: We are sorry for the typos. We have add a "comma" between P1 and P2 as well as modified the caption for Figure 7.